FINAL BOOK of ABSTRACTS

MARC XII

Twelfth International Conference on Methods and Applications of Radioanalytical Chemistry
April 3 -8, 2022

an International Topical Conference
Sponsored by the American Nuclear Society

February 25, 2022

METHODS AND APPLICATIONS OF RADIOANALYTICAL CHEMISTRY
KAILUA-KONA, HAWAII

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The organizers of the Twelfth International Conference on Methods and Applications of Radioanalytical Chemistry (MARC XII) conference are pleased to provide the following summary of abstracts for reference by attendees of the conference. These materials are a supplement to the actual Program, which provides complete details of the conference and sequence of presentations. MARC XII is expected to be a very successful MARC conference with over 400 abstracts submitted during challenging times.

The following document is over 200 pages containing the abstracts submitted for presentation at the MARC XII conference as of February 25, 2022. The list will only be provided electronically and only a small number of hard copy abstract summary books will be available for reference at the conference at the desk. Individual copies will not be provided.

If you have any questions, please contact the program chair, Sam Glover (sam.glover@uc.edu). Please visit the MARC website for additional details at www.marcconference.org.

Log 202. IN SITU SYNTHESIS OF SILVER NANOPARTICLES IN PECTIN MATRIX USING GAMMA IRRADIATION FOR THE PREPARATION OF ANTIBACTERIAL PECTIN/SILVER NANOPARTICLES COMPOSITE FILMS

Ardjoum, N., Shankara, S., Salmieria, S., Lacroix, M. (P) INRS Centre Armand Frappier Health & Biotechnology. (P) Presenting Author.

The present study reports in situ synthesis of silver nanoparticles (AgNPs) in pectin matrix using γ-irradiation at 2.5 and 5 kGy and preparation of antibacterial pectin-based nanocomposite films. The FE-SEM micrographs showed that the neat pectin film was smooth but became a little rough after forming AgNPs in the pectin films. The UV-visible spectroscopy analysis exhibited a surface plasmon resonance (SPR) band about 430 nm confirming the formation of AgNPs in the pectin films. The Hunter color values of nanocomposite film changed after the formation of AgNPs. FTIR and thermogravemetric analysis results revealed no structural and thermal stability changes in the pectin films after the synthesis of AgNPs by γ-irradiation. However, mechanical properties and water vapor permeability of the composite films improved after AgNPs synthesis. The pectin-AgNPs nanocomposite films exhibited potent antibacterial activity against Escherichia coli and Salmonella Typhimurium. The developed antibacterial pectin-based nanocomposite films incorporated with AgNPs using γ-irradiation have a potential to increase the shelf life of packaged food.

Log 203. IDENTIFICATION OF URANIUM HEXAVALENT COMPOUNDS USING X-RAY PHOTOELECTRON SPECTROSCOPY

S. A. Dunn (1) (P), P. Roussel (1), C. Poile (1), C.Puxley, M. Higginson (1), P. Kaye (1), T. Shaw (1), M. Gilbert (1), J. F. Watts (2). (1) AWE, Aldermaston, Reading RG7 4PR, UK. (2) Department of Mechanical Engineering Sciences, University of Surrey, Guildford, Surrey GU2 7XH, UK. (P) Presenting Author.

X-ray photoelectron spectroscopy (XPS) has applications in many fields ranging from development of thin films for semi-conductors to post failure analysis of organic coatings and structural adhesives. The current work expands on that versatility by applying XPS to the growing field of nuclear forensics. This was achieved by the synthesis and characterisation of several uranium compounds, predominantly in the hexavalent state associated with the nuclear fuel cycle, by X-ray diffraction and Raman spectroscopy prior to XPS. Spectral characteristics for each compound are discussed, and interpretations made through observations in the binding energy of the U4f region and counter ions as well as secondary energy loss features such as shake up satellites. The interpretation of such features is related to the stoichiometry, oxidation state and crystal structure of a range of uranium compounds. As XPS is typically insensitive to structural (crystallographic) variations, a rationale is provided for the relationship between structural variations, as measured by Raman and X-ray diffraction, and the XPS satellite to parent peak intensity of uranium compounds, providing a novel and useful approach for uranium compound characterisation. In addition to the novel approach described, Wagner chemical state plots have also been generated to provide another comparison tool. (UK Ministry of Defence © Crown Owned Copyright 2021/AWE)

Log 204. APPLICATIONS OF A DUAL COLUMN TECHNIQUE IN ACTINIDE SEPARATIONS
Miting Du. Oak Ridge National Laboratory

Ion exchange methods are widely used in the separation and purification of various actinides. Recovery of specific individual actinide typically requires multiple separation using multiple ion-exchange column runs. The proposed improved method uses stacked columns of different resins to minimize the number of separation steps required to achieve the desired actinide purity in a quick and efficient manner. This presentation will give examples of the application of a dual column separations method for recovery and purification of individual actinide such as Bk, Pa and U respectively. The advantages and required conditions of using this technique will be discussed.

Log 205. ELECTROCHEMICAL INVESTIGATION OF HIGHER OXIDATION STATES OF ACTINIDES
A. V. Gelis, N. H. Cicchetti (P). (P) Presenting Author.

Tens of millions of gallons of high-level radioactive waste remain from US plutonium production. In 2003, leaks in waste storage tanks at the Hanford site were discovered to be releasing hundreds of gallons per year into the environment. The treatment and disposal of this waste has therefore become a priority to protect public health. The waste contains significant amounts of actinides under alkaline conditions. Unfortunately, the behaviors of many of these elements in base are poorly understood. Any treatment approach will require significantly more information about the speciation, solubility, and reactivity of actinides in base than is currently available. This presentation will detail the use of electrochemical techniques to characterize solutions of neptunium under various conditions. Cyclic voltammetry and square wave voltammetry were used to observe the Np(VII/VI/V) and Pu(VII/VI/V) couples and measure the redox potentials in concentrated alkali, better defining values on which there has been substantial disagreement. Voltammograms of Pu(VII/VI) are reported for the first time. The approach shows great promise for the further characterization of neptunium, plutonium, and other actinides in base to provide better information for the treatment and disposal of problematic legacy waste.

Log 206. RADIONUCLIDE METROLOGY – CONFIDENCE IN RADIOACTIVITY MEASUREMENTS
S. Pommé, Joint Research Centre

Radionuclides, whether in natural quantities or artificially produced, are readily detected through their particle and photon emissions following nuclear decay. Radioanalytical techniques use the radiation as a looking glass into the composition of materials, thus providing valuable information to various scientific disciplines. Absolute quantification of the measurand often relies on accurate knowledge of nuclear decay data and detector calibrations traceable to the SI units. Behind the scenes of the radioanalytical world, there is a small community of radionuclide metrologists who provide the vital tools to convert detection rates into activity values. They perform highly accurate primary standardizations of activity to establish the SI unit becquerel for the most relevant radionuclides and demonstrate international equivalence of their standards through key comparisons. The trustworthiness of their metrological work crucially depends on a painstaking scrutiny of their methods and the elaboration of a comprehensive uncertainty budget. Through

meticulous methodology, rigorous data analysis, innovation, education and training, performance of reference measurements, and organization of proficiency tests, they help the user community to achieve confidence in measurements for policy support, science, and trade. The author dedicates the George Hevesy Medal Award 2020 to the current and previous generations of radionuclide metrologists who devoted their professional lives to this noble endeavor.

Log 207. **TRACE ANALYSIS BY TRLIF, TRLIC, RIMS, INAA AND ICP-MS**

Izosimov, I.N. (1,P); Saidullaev B.D. (2); Strashnov I. (3); Vasidov A. (2). (1) Joint Institute for Nuclear Research, 141980 Dubna, Russia. (2) Nuclear Physics Institute, Tashkent, Uzbekistan. (3) The University of Manchester, School of Natural Sciences, M13 9PL, UK. (P) Presenting Author.

Combination of the Instrumental Neutron Activation analysis (INAA), Inductively Coupled Plasma Mass Spectrometry (ICP-MS), and laser spectroscopy (TRLIF, TRLIC, RIMS) is efficient both for element and isotope composition analysis of the samples. The multi-step excitation of species and time-resolved registration of resulting luminescence (TRLIF) and chemiluminescence (TRLIC) for Pu, U, and Sm detection are considered. In the next step, we combine the resonance ionization laser spectroscopy with mass spectrometry detection (RIMS). The trace amount detection has been demonstrated for Kr isotopes (including 81Kr) of radiogenic and cosmogenic origin. Multi-step RIMS approaches have been extended to U and other radioisotopes from solid and liquid samples. The development of a suitable excitation/ionization schemes for both RLIF/TRLIC and RIMS is of a high priority allowing more complex sample characterization. We have applied both INAA and ICP-MS methods and analyzed the elemental composition (64 elements) of bones of dinosaurs, mammoths, prehistoric bear and archanthropus as well as the samples of surrounding soils; everything collected in different parts of Uzbekistan. A high concentration of uranium we detected in the bones of dinosaurs (122 mg/kg), South mammoth (220 mg/kg), prehistoric bear (24 mg/kg) and archanthropus (1.5 mg/kg) compared to surrounding soils (3.7-7.8 mg/kg) and standard bones (<0.01 mg/kg) is a bit of a puzzle.

Log 208. **WHAT EVER HAPPENED TO FOOD IRRADIATION: STILL AN ALTERNATIVE?**

Butterweck, J. Aerospace and Environmental Medicine Group, Ltd.

In the 1980's, when the commercial single use medical product industry adopted ionizing radiation to assure sterility in their products, the food industry decided try it to reduce pathogens in meat products. Our USA culture is always trying to improve our products under the Continuous Total Quality Improvement concept. The three legged stool of (1) regulators (2) consumers and (3) meat industry all had to buy into the technology. After awhile it became obvious that this was not going to happen. However, today what are the alternatives? The presenter will present alternatives based upon time allotted.

Log 209. **DEVELOPMENT OF A SENIOR/GRADUATE COURSE IN NUCLEAR FORENSICS**
Landsberger, S. (P); Haas, D. A. (P) Presenting Author.

A senior undergraduate course in Nuclear Forensic was developed with a grant through the Department of Homeland Security. Besides the usual lectures on the basics of nuclear phenomena, a successful effort was made to include a significant amount of historical background. In addition, several guest lecturers from national laboratories and universities were added to the curriculum resulting in a more meaningful and current course. As well due to COVID-19 restrictions, all the laboratories were given virtual. A detailed overview of the course will be given.

Log 210. **OPTIMIZATION IN THE DETERMINATION OF SEVERAL RARE-EARTH ELEMENTS USING EPITHERMAL NEUTRON ACTIVATION ANALYSIS**
McKay, K. (P); Landsberger, S. (P) Presenting Author.

Analysis of rare earth elements (REEs) using NAA can normally be difficult due to a variety of factors including spectral interferences, high Compton continuum backgrounds and the production of the identical isotopes due to uranium fission. To optimize the evaluation of REE concentrations within geological samples, epithermal NAA was used in conjunction with Compton suppression techniques to calculate the concentrations of Dy, Sm, Gd, and Ho.

Log 211. **NATURAL OCCURRING RADIOACTIVE MATERIAL (NORM) IN THE OIL AND GAS EXPLORATION: FIELD TRAINING IN HEALTH AND SAFETY PRACTICES**
Landsberger, S. G. (P); Landsberger, S., Graham, G. (P) Presenting Author.

The discovery of radioactivity in oil exploration dates back to the early 1900's. In the 1980's a lot of reseach began to emerge on the measurement of Ra-226 and other radioactive products in oil exploration field. This short course 4-day includes introduction to radiation, definitions and characterization of NORM, geological information of oil and gas production, basic chemistry, radioactive decay, radiation units of measurement, radiation calculations and dose limits, worker safety, survey methods, how to detect gamma rays, ALARA (as low as reasonably achievable) principle, health effects, waste and disposal, NORM regulations, worker protection guidelines for handling NORM and transportation.

Log 212. OPTIMIZATION OF NEUTRON ACTIVATION ANALYSIS OF RARE-EARTH ELEMENTS

Reis, C. (P); Landsberger, S. (P) Presenting Author.

During neutron activation analysis (NAA), if the material being analyzed is partially composed of a lanthanide content optimization of rare-earth analyses is crucial. To enhance these procedures a database was created to help assess the likelihood that a certain lanthanide isotope and gamma ray is indeed the best one to be determined by NAA. First, the most common isotopes through the (n,γ) reaction of each element was determined, along with the strongest and most abundant gamma rays to be used, and a chart was formulated with their relevant properties. A weighting factor was constructed based on seven characteristics of the properties of the lanthanides: thermal cross section, resonance integral, natural abundance of the element itself, half-life of the activation product, the branching ratio, the Igamma(relative) value, and relative detector efficiency of the photon spanning from 80 – 1596 keV. The seven individual characteristics were then added together and divided by the total possible value to get their total weighting factor. In addition, the application of gamma- gamma coincidence and Compton suppression was also evaluated to determine which radionuclides may be better evaluated.

Log 213. A METHOD FOR FISSION PRODUCT YIELD DETERMINATION USING GAMMA-GAMMA COINCDIENCE

De Luna, B.A. (1,2,P); Haas, D.A. (1). (1) The University of Texas at Austin. (2) Sandia National Laboratories. (P) Presenting Author.

Gamma-gamma coincidence spectroscopy was applied to the analysis of the fission product yields of short to medium-lived fission products from an epithermal irradiation of uranyl acetate. SCALE modeling was employed to extract the buildup and decay curves of the radionuclides of interest where a correction factor for the observed number of decays was determined. Uncertainties in the model were derived using the Sampler module on the Couple and Origen module processes. An MCNP detector model was designed to extract gamma-gamma coincidence efficiencies for select radionuclides. Preliminary results of this method were agreeable with the cumulative yields of the following fission products: ^{132}I, ^{134}I, ^{135}I and ^{92}Y. This method is expected to be expanded to applications in cyclic irradiation to improve uncertainties in the yields determined. A model is then discussed for the prediction of independent fission product yields from the data generated using this method.

Log 214. REDUCING UNCERTAINTIES IN GAMMA-RAY BRANCHING RATIOS OF Cd-115M

Gordon, E.M. (1,P); De Luna, B.A. (1); Johnson, W. (2); Haas, D.A. (1). (1) The University of Texas at Austin. (2) Air Force Technical Applications Center. (P) Presenting Author.

Current procedures for detecting and quantifying Cd-115m, a fission product relevant to the verification of the Comprehensive Nuclear Test Ban Treaty, rely on beta spectroscopy because of high uncertainty in gamma ray branching ratios for the isotope. Reducing the uncertainty in the intensity values for characteristic gamma rays of Cd-115m could facilitate a change to gamma counting, reducing the complexity of the procedure by removing the chemical processing necessary for beta spectroscopy. A recent task at UT Austin aimed to reduce the uncertainty in Cd-115m gamma branching ratios. In the experiment, a sample of enriched Cd-114 was irradiated at UT Austin's TRIGA reactor. The sample was surrounded by a cylinder of boron nitride and placed in a canister lined with cadmium which resulted in a neutron flux spectrum representative of a Watt fission spectrum at the sample location. The activated cadmium sample was analyzed using both a gamma-gamma coincidence system and standard gamma spectroscopy.

Log 215. MODELING THE USE OF MOBILE MODULAR GAS SAMPLERS IN NEAR-FIELD DETECTION USING HYSPLIT

Gordon, E.M. (1,P); Adhikari, P. (1); Haas, D.A. (1). (1) The University of Texas at Austin. (P) Presenting Author.

The Wireless Independent Noble Gas Sampler (WINGS) is a mobile, modular gas sampling system designed for use in low infrastructure environments. WINGS units operate on battery power and communicate by radio connection. In the case of a suspected underground nuclear explosion (UNE), WINGS units could be deployed to detect and identify noble gases emanating from the explosion site or identify gases entering the local area from offsite. This work uses the atmospheric transport modeling tool inline WRF-HYSPLIT to determine the ideal deployment configuration for WINGS units around a local area using the location of the 2014 Integrated Field Exercise in Jordan as a test case with a hypothetical upwind medical isotope production facility providing a nuisance background. HYSPLIT is a computer model for atmospheric transport and dispersion. Inline WRF-HYSPLIT integrates data from the WRF-ARW meteorological model, generating dispersion and deposition models with finer spatial resolution than achievable with an offline approach. These more accurate models enable determination of sampler network effectiveness as a function of sampler density, sample collection duration and interval, and distance from the emission point from a UNE.

Log 216. **PORTABLE MODULAR GAS SAMPLERS FOR NUCLEAR EXPLOSION MONITORING**

Shah, K.A. (1, P); Gordon, E.M. (1); Adhikari, P. (1); Allen, M.I. (1); Barth, B.S. (1); Bekker, J.P. (1); Byers, M.E. (1); Fowler, E.P. (1); Hudson, C.C. (1); Kaitschuck, N.M. (1); Wiser, R.S. (1); Wiser, W.S. (1); Haas, D.A. (1). (1) The University of Texas at Austin. (P) Presenting Author.

A gas sampler system has been designed, built, and deployed that may contribute to near-field detection of underground nuclear explosions. The system is inexpensive, portable, and autonomously collects samples for subsequent lab analysis. Each gas sampler unit contains seven sample containers. Samples are collected with a small air compressor allowing for delayed sample collection after emplacement. Collection duration and interval parameters can be adjusted based on the intended collection site. The system is designed to remain in standby mode for up to two months and can be operated remotely by radio, or with preset sampling times and durations. Two gas sampler units were deployed at Lawrence Livermore National Laboratory's Site 300 for an atmospheric chemical explosive field experiment. Collected samples were later analyzed on a gas-chromatograph mass-spectrometer. Following deployment, several design optimizations were introduced, including replacing the hand-made sampling containers with off-the-shelf compressed gas tanks. Additionally, software optimizations to upgrade the wireless radio communications network to a WiFi communications network have been made to use cellular hotspots for telemetry. Radioactive gas samples can be analyzed in- or near-field with NaI(Tl) or HPGe detectors, or with a xenon separation and analysis system such as the SAUNA. A 2"x4"x16" NaI(Tl) detector was efficiency calibrated with the sample containers to facilitate in-field analysis of the gas samples. The goal of the gas sampler systems is to deploy ten units in a field experiment releasing radioactive noble gas tracers to improve understanding of atmospheric dispersion of gas over distances of several kilometers.

Log 217. **DEVELOPMENTS IN ACTINIDE ANALYTICAL CHEMISTRY APPROACHES AT AWE**

Higginson, M (1, P); Dawkins, B (1); Kaye, P (1); Taylor, F (1); Shaw, T (1); Dunn, S (1); Ingman, L (1); Cross, S (1) and Taylor, T (1). (1) AWE, AWE Aldermaston, RG7 4PR, UK. (P) Presenting Author.

The actinide analysis team at AWE provide actinide analytical chemistry capabilities in support of customer requirements. This talk will give an overview of recent advances in our methodologies, demonstrating improvements in our process efficiency, safety and flexibility. Examples will be presented on the direct measurement of actinide samples, automation of processes, improved chemical purification techniques and novel, rapid prototyped sample containments for enabling analysis of samples outside of traditional containments; expanding our analytical toolkit

Log 218. ADVANCES IN URANIUM RADIOCHRONOMETRY TO ADDRESS CHALLENGES FOR NUCLEAR FORENSIC ANALYSIS AND INTERPRETATION

Higginson, M. (1); Dunne, J. (1); Gilligan, C. (1); Cross, S. (1); Kayzar-Boggs, T. (2); Gaffney, A. (3); Kaye, P.(1). (1) AWE Aldermaston, RG7 4PR, UK (2) Nuclear and Radiochemistry Group, Chemistry Division, Los Alamos National Laboratory, Los Alamos, NM 87545, USA. (3) Nuclear and Chemical Sciences Division, Lawrence Livermore National Laboratory, 7000 East Avenue, Livermore, CA 94551, USA. (P) Presenting Author.

The model age of a nuclear material is an important measurement to answer the when type questions for nuclear forensics. Measurements of decay progeny in samples by a variety of analytical techniques can be used to produce model ages for interpretation.

These measurements of trace elements require considerable effort. A variety of challenges remain to improve our ability to make defensible measurements and interpret discordant chronometer pairs relative to samples produced under controlled conditions. This work is required to identify if repeatable processing signatures are available and to prove that radiochronometry has utility for samples with complex processing histories.

This talk will cover improvements to measurement traceability and making our analytical methodologies more efficient and robust. We will then report an applied study with the aim to understand the impacts of uranium casting technologies on apparent model ages.

Log 219. CERTIFICATION OF NEW REFERENCE MATERIALS FOR URANIUM ISOTOPE ANALYSIS AT JRC-GEEL

Richter, S. (1); Venchiarutti, C. (1), Hennessy, C. (1), Truyens, J. (1), Bujak, R. (1), Aregbe, Y. (1), S. Pommé (1,P); Hexel, C. (2). (1) European Commission, Joint Research Centre, Geel, Belgium, (2) Oak Ridge National Laboratory (ORNL), Oak Ridge, TN, USA. (P) Presenting Author.

The European Commission's Joint Research Centre provides a wide range of certified nuclear reference materials to the safeguards authorities, the nuclear industry and to the geochemical and cosmochemical sciences. For several recently completed projects the certification strategies and applied measurement procedures is discussed: the IRMM-3000 series of HEU solutions, the extended IRMM-2019-2030 series of LEU solutions, and the IRMM-183-187 series of LEU solutions, which is currently being re-certified. Most emphasis is dedicated to the IRMM-3000-series, a set of five highly enriched uranium nitrate solution reference materials with the U-235 enrichment varying from 20 percent to 90 percent. The solutions were prepared by dissolving U3O8 materials, which were prepared by gravimetrically mixing oxides, highly enriched in U-235 and U-238, in a similar way as performed earlier for the IRMM-072, IRMM-074, IRMM-075 and IRMM-3636a,b series. The U-235/U-238 isotope ratios of the IRMM-3000-series were calculated based on the weighings of the two starting materials and the measured isotope compositions and impurities of both. Verification measurements for the U-235/U-238 isotope ratios were performed using standardized TIMS methods such as the double spike and the Modified Total Evaporation (MTE) methods. The minor isotope ratios U-234/U-238 and U-236/U-238 were directly characterized by TIMS-MTE. All certified ratios were successfully verified by external laboratories at the IAEA and ORNL. The developed strategies are suggested for the certification of further reference materials.

Log 220. **PREPARATION OF HIGH SPECIFIC ACTIVITY ACTINIDE TARGETS BY VAPOR DEPOSITION**
Loveland, W., Pica, A., Chemey,A. (P). Oregon State University. (P) Presenting Author.

We describe the preparation of targets of 233U, 237Np, 239Pu, 240Pu and 242Pu by vapor deposition. A dual glove box system is used for the preparation of the fluorides of these actinides and the subsequent vapor deposition.The preparation of the actinide fluorides is done by an aqueous precipitation procedure with yields of greater than 90%. The yields of the vapor deposition are less than 3%. We compare and contrast targets made by vapor deposition and molecular plating. We discuss the special problems of making targets of very high specific activity

Log 221. **THERMAL NEUTRON MEASUREMENTS WITH AN UNPOWERED, MINIATURE, SOLID-STATE DEVICE**
Hossain, T. (1,P); Fullwood, C. (1); Flanagan, W. (1,2); Hedlesky, P. (2); Rabaey, J. (2);Block, S.(2); Medcalf, A. (2); Tipping, T (3) . (1) Cerium Laboratories. (2) The University of Dallas. (3) The University of Texas at Austin Nuclear Engineering Teaching Laboratory. (P) Presenting Author.

A prototype neutron detector has been created through modification to a commercial non-volatile flash memory device. Studies are being performed to modify this prototype into a purpose-built device with greater performance and functionality. This presentation will describe a demonstration of this technology using a thermal neutron beam produced by a TRIGA research reactor. With a 4x4 array of 16 prototype devices, the full widths of the beam dimenstions at half maximum are measured to be 2.2cm by 2.1cm.

Log 222. **TWO CHLORINATION METHODS FOR CONVERTING UO_2 INTO UCl_3 USING $ZrCl_4$**
Chamberlain, J.L. (1,P); Simpson, M. F. (1). (1) The University of Utah. (P) Presenting Author.

A scalable method for conversion of UO_2 into UCl_3 is needed for simultaneously reducing spent nuclear fuel waste forms while providing a fuel source for molten salt reactors. It has been previously reported by Japanese researchers that this can be accomplished via reaction with the chlorinating agent $ZrCl_4$. The problem with this process is that $ZrCl_4$ is highly volatile for the appropriate temperature range. Two novel methods for introducing ZrCl4 into the molten salt were explored in this study and found to be simple and effective. The first in-situ method was achieved by combining NiCl2 with LiCl-KCl eutectic salt and submerging a zirconium rod into the chlorination cell. The second method volatilizes $ZrCl_4$ in a heated argon stream which was bubbled into the molten eutectic salt. A stainless-steel basket containing the UO_2 material is suspended in the molten salt bath for both approaches. Samples taken periodically from the molten salt bath and analyzed with ICP-MS revealed the increasing concentrations of uranium and zirconium with time. Cyclic voltammetry (CV) was used confirm $ZrCl_4$ and UCl_3 in the salt mixture and to estimate concentrations. Also, the open circuit potential (OCP) recorded during the chlorination confirmed the uranium to be present in the salt as UCl_3. Post chlorination urania particles were mounted, polished and imaged under a SEM and EDS. The cross sectioned particles revealed a skeletal product layer forming around an unreacted UO_2 core.

Log 223. NEUTRON ACTIVATION ANALYSIS USING GAMMA-GAMMA COINCIDENCE TO ELIMINATE SPECTRAL INTERFERENCES

Martinez, F. (P); Landsberger, S. (P) Presenting Author.

Over the past two decades coincidence and anti-coincidence gamma-ray spectrometry has been used in many areas of low-level counting, fission products and neutron activation analysis (NAA). It is well has known that NAA still has some limitations with the identification of specific gamma ray peaks due to spectral interferences, no matter how good the detector resolution. The implementation of gamma-gamma coincidence techniques using the XIA Pixie-16 system has demonstrated the ability remove spectral interferences. A series of experiments was performed on different NIST standards to assess radionuclides that are commonly difficult to identify due to spectral interferences. The experiments have showed that the Pixie-16 system can eliminate spectral interferences (or increase counting statistics) and therefore, successfully identify Hf-181, Lu-177m,124Sb-124, Tb-160, Yb-175 and Sm-153.

Log 224. CHARACTERIZATION OF A SODIUM BISMUTHATE PAN RESIN FOR THE SEPARATION OF AMERICIUM FROM CURIUM

Labb, S.A. (1,P); Bombard, A. (2); Bond, E.M. (3); Sudowe, R. (1). (1) Colorado State University. (2) TriSKem International. (3) Los Alamos National Laboratory. (P) Presenting Author.

Successful and efficient americium (Am) and curium (Cm) separations are necessary in support of experiments related to stockpile stewardship science. However, the efficiency of these separations suffers from the similar chemical properties of these radionuclides. Differences in redox chemistry can be exploited because americium can be oxidized to higher oxidation states while curium cannot. Recently, the ion exchange properties of sodium bismuthate were demonstrated, which led to the development of a chromatographic method for the separation of Am and Cm resulting in a greater than 97% recovery and purity. This method, however, suffers from poor adsorption capacity and flow rate properties, gradual material dissolution, and gas production in nitric acid. Based on this previous finding, this project focuses on evaluating and characterizing a novel resin from TriSKem that incorporates sodium bismuthate onto polyacrylonitrile. The bismuthate coating will ensure that Am is always present in higher oxidation states, while its incorporation onto the resin will allow for the coupling of the oxidation state and continuous separation process. This talk will highlight the batch extraction studies that explored the dependency of nitric acid concentration, resin preconditioning time, radionuclide contact time, and temperature as it related to retention on the resin. In addition, other radionuclides have been characterized on this resin to explore potential application in the nuclear fuel cycle.

Log 225. A PROTOCOL FOR VERY HIGH PRECISION CHEMCIAL BURNUP DETERMINATIONS FOR HIGH BURNUP URANIUM FUELS

Giaquinto, J.M. (1, P); Bevard, B. B. (1); Delashmitt, J.S. (1); Hexel C.R. (1); Ilas, G. (1); Roach, B.D (1); Rogers K. T. (1); Zirakparvar, N. (1) Oak Ridge National Laboratory. (P) Presenting Author.

Oak Ridge National Laboratory (ORNL) is performing high precision radiochemical assays (HP-RCA) for evaluations of high burnup uranium (HBU) fuel irradiated in commercial nuclear power reactors. The goal of the work is to provide data sets for key actinides and fission products with the lowest achievable total measurement uncertainties to fill gaps or improve on existing experimental RCA data in the international Spent Fuel Isotopic Composition (SFCOMPO) database. These data sets are essential to validate the computational methods and tools for predicting nuclide inventories in used nuclear fuel, with large impact in the safety analyses for used nuclear fuel transportation, storage, and disposal applications. A critical component of the HP-RCA work is the ability to provide high confidence chemical burnup determinations. The Nuclear Analytical Chemistry Section within the Chemical Sciences Division at ORNL are experts in performing chemical burnup determinations and have incrementally improved upon its analytical methodologies over the years to achieve relative 2-sigma total uncertainties for calculated burnups of less than 0.5 %. Presented will be the key steps developed to allow for this level of precision.

Log 226. THE FISSION FINGERPRINT OF RUTHENIUM ISOTOPES: A FORENSIC TOOL OF SAMPLE PROVENANCE AND AGE DETERMINATION

Sanborn, M.E. (1,P); Hanson, S.K. (1); Trellue, H.R. (1); Kinman, W.S. (1). (1) Los Alamos National Laboratory. (P) Presenting Author.

The use of the stable Ru isotopic composition of nuclear materials is a potentially valuable new asset in the nuclear forensics toolkit. In particular, in materials where fission produced Ru-106 is detected, the measurement of stable isotopes of Ru produced as end products of the fission beta decay scheme can provide insight into the fissiogenic processes (e.g., irradiation conditions and starting material). Additionally, measurement of the Pd-106 daughter of Ru-106 decay can provide high-resolution radiochronometry information on the generation date of the material. To this end, we utilized a commercially available nominally single isotope Ru-106 standard (multiple individual lots) to investigate the feasibility of utilizing Ru isotopes to determine fissiogenic process and chronometry information. In this work, we show that Ru isotopes are indeed a potentially robust tool for nuclear forensic applications with the stable Ru isotopic composition markedly different than natural. Utilizing Ru isotopes we were able to determine the generation processes across different lots of the Ru-106 standard (by comparison with nuclear reactor simulations) and determine difference in the age of the materials at a resolution of approximately 0.05 years. The results of this study indicate that where the Ru-106 fission product is detected, the stable isotopic composition Ru could provide an enduring record of nuclear activities. As such, use of Ru isotopes in investigating the origin and history of a novel sample has potential significant implications for nuclear treaty monitoring.

Log 227. THE FISSION FINGERPRINT OF RUTHENIUM ISOTOPES: MEASUREMENTS OF NUCLEAR DEBRIS SAMPLES

Hanson, S. K. (P), Sanborn, M., Meininger, D., Miller, J. L., Inglis, J., Kinman, W. Los Alamos National Laboratory. (P) Presenting Author.

The measurement of stable isotopes of ruthenium produced as end members of the fission product beta decay scheme could be a valuable tool for nuclear treaty monitoring. Ruthenium isotopes have cumulative fission yields that vary with the fissioning fuel type and neutron energy, and the element can be released in anthropogenic nuclear activities. Recently, we have shown the applicability of measuring stable Ru isotopic composition in a Ru-106 isotopic standards to determine the production process. In this talk, we apply this same forensic tool to investigate a nuclear debris sample by chemically separating Ru from an environmental matrix and measuring its isotopic composition by multi-collector inductively coupled plasma mass spectrometry (MC-ICP-MS). Here, we report the Ru isotopic composition in several nuclear debris samples from the historic Trinity test. The results show a clearly distinct stable isotopic composition compared to the natural Ru isotopic composition. The measurement results will be discussed in the context of potential applications in the area of nuclear treaty monitoring.

Log 228. CHARACTERIZATION OF THE NEUTRON SPECTRUM AND PNEUMATIC TRANSFER SYSTEM FOR A FAST NEUTRON CYCLIC NEUTRON ANALYSIS BEAM-PORT FACILITY

Pasman, M.P. (1,P); De Luna, B.A. (1);Gordon, E.M. (1); Haas, D.A. (1). (1) The University of Texas at Austin. (P) Presenting Author

A facility is being developed at The University of Texas at Austin which consists of a 10 cm neutron beam that resembles a Watt fission spectrum with a maximum flux of 3.75×106 n cm^{-2}s^{-1} at a reactor power of 950 kW. This facility has been characterized to accurately determine the neutron flux spectrum using MCNP6 and STAYSL. A pneumatic system, with transfer times of approximately one second, has been developed and verified to transfer a sample consistently to perform cyclic neutron activation analysis to ultimately determine fast neutron induced fission yields. Initial cyclic irradiations using gamma-gamma coincidence have been performed with aims to reduce the uncertainty of fission yields for short-lived fission products of interest for post-detonation nuclear forensics.

Log 229. A RADIOCHEMICAL METHOD FOR THE RAPID SEPARATION OF COPPER FROM FISSION PRODUCTS AND MATRIX ELEMENTS

Melinda S Wren (1,P); Iain May (1); Susan M K Hanson (1). (1) Chemistry Division, Los Alamos National Laboratory, Los Alamos, NM 87545. LA-UR-21-31946. (P) Presenting Author.

Activation products that arise from neutron capture reactions on different elements are important signatures in post-detonation nuclear forensics. The ability to identify the presence or absence of short-lived activation products of interest can facilitate the rapid characterization of a nuclear fission event. Here we describe the development of a method to separate copper from interfering fission product radionuclides and matrix elements. Two anthropogenic radioisotopes of copper are of particular interest; 64Cu and 67Cu. If present, they might occur in low concentrations relative to fission products, and in a matrix of fused glassy debris or soil which makes their separation especially difficult. This work outlines a rapid carrier based radiochemical separation scheme for copper which can provide vital information in the event of a post-detonation nuclear event.

Log 230. UNCERTAINTY BUDGET FOR URANIUM ISOTOPE RATIO ANALYSIS USING THE LS-APGD IONIZATION SOURCE COUPLED TO AN ORBITRAP MS

Goodwin, J.V. (1); Manard, B. T. (2); Ticknor, B.W. (2), K.T. Rogers (2), C.R. Hexel (2), Cable-Dunlap, P. (2); Marcus, R. K. (1,P). (1) Clemson University. (2) Oak Ridge National Laboratory. (P) Presenting Author.

The liquid sampling-atmospheric pressure glow discharge (LS-APGD) ionization source has provided excellent performance in challenging uranium isotope ratio measurements when coupled to Orbitrap mass spectrometers. A major impetus for the development of the microplasma/Orbitrap coupling is the potential for dramatic reductions in sample manipulations and analysis time required in the case of traditional methods of isotope ratio determination such as TIMS or MC-ICP-MS. The ultra-high resolution (> 70k) Orbitrap eliminates the need for the chemical separations that are often required to remove isobaric interferences in TIMS and ICP-MS isotopic analysis. The reduced sample volumes with more straightforward processing procedures is of interest to the nuclear forensics and nuclear safeguard communities. To gain a deeper understanding of measurement uncertainties inherent in U isotope ratio determinations with the LS-APGD/Orbitrap system, International Standards Organizations (ISO) guidelines for the expression of uncertainty in measurement (GUM analysis) have been applied to generate an uncertainty budget to identify significant sources of uncertainty. This will allow for a better understanding of measurement uncertainty and identify areas for potential improvements. By applying the laws of propagation of distributions, a standard uncertainty value can be calculated. Initial results point to the blank measurement being the largest source of error, to which alternatives to the conventional data acquisition system will now be investigated to improve the method further.

Log 231. FIRST MASS SPECTROMETRIC CHARACTERIAZATION OF Pu USING THE LS-APGD MICROPLASMA ION SOURCE AND ORBITRAP MASS ANALYZER

Goodwin, J.V. (1); Manard, B. T. (2); Ticknor, B.W. (2); Cable-Dunlap, P. (2); C. R. Hexel (2), B. D. Roach (2), S. C. Metzger (2), Marcus, R. Kenneth (1,P). (1) Clemson University. (2) Oak Ridge National Laboratory. (P) Presenting Author.

While mass spectrometric determinations of Pu using TIMS and ICP-MS are for the most part routine, the chemical and spectrometric complexity of these measurements remains onerous. In both approaches, samples must be extensively purified to remove potential isobaric (same mass) interferents, even for measurements made on multicollector instruments. We describe here the first measurements of Pu using the LS-APGD/Orbitrap instrumentation, providing far greater simplicity in terms of sample size and waste and factor-of-ten higher mass resolution. Extensive previous studies have demonstrated that high-precision isotope ratios can be obtained for U using this instrument, crossing all relevant levels of enrichment/depletion.   Initial developments towards the application for Pu analyses, were undertaken using the long-lived 242Pu isotope. Critical plasma operation and ion sampling parameters were optimized towards producing the highest ion intensities. The mass spectrometric tuning, including use of collisional dissociation, focused on solely obtaining the atomic Pu ionic species (i.e., rather than the dioxide form prevalent for U). The ability to make the measurements with a mass resolving power of >70 k and mass accuracy of <0.001Da virtually eliminates any possibility of interferents.   We report here the results of these first experiments, providing insights into the ionization processes and analytical figures of merit, and benchmarking those results versus the previous works with U.

Log 232. COMPARING THE MORPHOLOGY AND SOLID-STATE STRUCTURE OF UTc$_3$ TO URu$_3$
Libero, J. (1, P); Koury, D. (1); Poineau, F. (1) (1) UNLV. (P) Presenting Author.

Ruthenium and technetium are high-yield fission products of uranium that likely form metallic phases in spent nuclear fuel. Solid-state structures of the binary An-Tc alloys are not well characterized. However, the structure of some An-Ru compounds, like URu$_3$'s Pm-3m space group, are well characterized. Since ruthenium is often used as a surrogate for technetium, the URu$_3$ alloy serves as a comparable material for UTc$_3$. In this study alloys of URu3 and UTc3 were prepared by arc-melting solid uranium metal discs with pressed pellets of the ruthenium or technetium metal powder. After annealing at 800 oC for 10+ days, cross-sections of the alloys were studied using scanning electron microscopy (SEM) and energy-dispersive x-ray spectroscopy (EDS) to characterize the growth of microstructures and areas of non-homogeneity, as well as analyze the varying elemental compositions contained within the alloys Structural analysis was performed on homogenous samples using powder x-ray diffraction (PXRD). These PXRD patterns were used to experimentally solve the samples' solid-state structures. The resulting structural information was used to compare the solid-state structure of UTc3 to the known Pm-3m structure of the URu$_3$ alloy. Furthermore, the Pm-3m structure was investigated for UTc$_3$ by substituting technetium for ruthenium and performing density functional theory (DFT) calculations to optimize the lattice parameters. This theoretical structure was ultimately used to simulate a PXRD pattern which was subsequently compared to the experimental pattern. This study provides valuable information regarding the solid-state structures of Uranium-

Technetium alloys compared to uranium-ruthenium alloys and builds techniques to study other An-M alloys.

Log 233. **Boron-doped diamond resilience in chloride and fluoride molten salt fuel systems**

Patenaude, H.K. (1,P); Rakos, R.(1); Olney, R. (1); Damjanovic, N. (1); Branch, S.D. (2); Rusinek, C.A. (1). (1) The University of Nevada, Las Vegas. (2) Pacific Northwest National Laboratory. (P) Presenting Author.

Generation IV Molten Salt Reactors' (MSRs) liquid salt fuel system and fast reactor technology simultaneously address concerns over waste, safety, and nonproliferation. However, work is needed to understand the impacts of the fuel salt systems and subsequent corrosion of their containment. Electrochemical methods can probe several fundamental aspects of molten salt chemistry so long as the electrodes can withstand the molten salt environment for prolonged periods of time. Boron-doped diamond (BDD) is expected to be resilient against harsh environments, given its sp3-carbon structure. As such, BDD could potentially be used for in situ corrosion control and monitoring in molten salt containment vessels. However, the corrosion of BDD in molten salt is not yet fully understood. In this study, we investigated the corrosion resistance of BDD by soaking the material in chloride and fluoride molten salts at 600 C for over 250 hours. Scanning electron microscopy and Raman spectroscopy were used to identify topographical changes in diamond crystal structures. Electrochemical techniques like cyclic voltammetry and chronoamperometry were used before and after salt soaking to determine the following redox and thermodynamic properties: formal reduction potential, electron transfer stoichiometry, diffusion coefficients, and electroactive surface area of the BDD. In general, little corrosion or change to the BDD was observed. this work advances the understanding of MSR chemistry, while simultaneously proving the applicability of BDD as electrode material for harsh environments.

Log 234. DIRECT ANALYSIS OF URANIUM AND PLUTONIUM FROM COTTON SWIPES BY MICROEXTRACTION-ICP-MS

Ticknor, B.W. (1, P); Manard, B.T. (1); Metzger, S.C. (1); Rogers K. T. (1); Hexel C.R. (1); Cable-Dunlap, P. (1). (1) Oak Ridge National Laboratory. (P) Presenting Author.

Collection and analysis of environmental sample (ES) swipes is a critical component of international nuclear safeguards, particularly for detecting undeclared nuclear material and activities. Bulk analysis is a destructive analysis technique for ES that requires ashing and digesting the entire swipe and utilizes high precision mass spectrometry for measuring the uranium and plutonium present on the sample. The process is extremely sensitive and provides very precise and accurate results, but is time consuming, labor intensive, and expensive. The work presented here investigates an approach to directly extract the analytes from the environmental swipe surface and determine isotopic abundances via inline inductively coupled plasma-mass spectrometry (ICP-MS) detection. This methodology, microextraction-ICP-MS, lowers a microextraction probe head onto the environmental swipe, seals on the sample surface, and delivers an extraction solvent (i.e. 2% HNO_3) to subsequently desorb target analytes. The flowing solvent, with extracted analyte, is directed into the ICP-MS for actinide isotopic determination. Results for swipes doped with uranium, plutonium, and mixed U-Pu will be presented.

Log 235. OPTIMIZATION OF A REACTOR BASED FAST NEUTRON IMAGING SYSTEM

Bisbee, M.B. (1)(3)(P); Oksuz, I. (1); Cao, L.R. (1); Cherepy, N. (2); Champley, K. (2). (1) The Ohio State University. (2) Lawrence Livermore National Lab. (3) DOE Nuclear Energy University Program Fellow. (P) Presenting Author.

A fast neutron tomography station has been successfully designed, built, and tested at The Ohio State University 500 kW Research Reactor, using the newly constructed fast neutron beam line with peak neutron flux ~5.4 x 10^7 n·cm-2·s-1 at 1.6 MeV. The imaging apparatus is composed of a water-cooled EMCCD, a front-surface mirror and a MeV-neutron-sensitive PVT scintillator, with a customized mobile design and a special on-line focusing feature. A total of 5 motion stages are built into the system to give XYZ and rotational freedoms for sample positioning, and another linear stage to fine tune the distance between the camera and the mirror. An optimized shielding design has greatly reduced the non-Poisson noise such as saturated pixels showing as white spots/strips due to the direct hit of neutrons and gamma rays on the silicon chip inside the CCD camera. A customized Python code and user-friendly GUI allows a fully automated imaging acquisition not requiring user interaction but allows tracking of progress. A complete fast neutron tomography dataset with 360 projections can be acquired in less than 2 hours, with 20 second projections. The on-line focusing is accomplished with a dielectrically actuated liquid lens. The focusing algorithm is written into the code to search for the best spatial resolution by adjusting the applied voltage to the lens. Finally, tomographic reconstructions have been visualized using both Octopus and Livermore Tomography Tools (LTT) software packages. Spatial resolution of ~ 0.50 mm has been achieved.

Log 236. **SIMULATED NAA DATASETS FOR IMPROVED DISCRIMINATION BETWEEN CHEMICAL FINGERPRINTS**

Johannes H. Sterba (1,P). Center for Labelling and Isotope Production, TRIGA Center Atominstitut, TU Wien, Vienna, Austria. (P) Presenting Author.

Neutron Activation Analysis (NAA) is a powerful chemical fingerprinting tool for provenancing or authenticity applications. In general, a similarity measure is defined and applied to the comparison of the chemical fingerprints, resulting in groupings. While this approach has been demonstrated in many applications, one important, but often unspoken, premise is that the natural variation within one group or sample is much smaller than the difference between two groups. This is not necessarily true for all elements considered in a chemical fingerprint, which in turn can influence the similarity measure applied. Thus, in scenarios for which discrimination between two very similar groups relies on differences in only a few elements, authoritative grouping without some a-priori information may be challenging. Using simulated NAA-datasets, it is possible to provide chemical fingerprints with additional a-priori information on, e.g. natural variation of elemental concentrations. Applying established (statistical) methods and similarity measures to this simulated datasets, the a-priori information can be used to verify the outcome of the analysis. This enables exploration of borderline-cases for which statistical methods or similarity measures might need to be adjusted. With this experience, real datasets can be better understood and their interpretation better supported.

Log 237. **DISTRIBUTION OF TH-234 IN FRESHLY CAST URANIUM**

LaMont, S.P. (P); Boswell, M.; James, M.R.; Luitjohan, K.E.; Imhoff, S.D.; Kayzar-Boggs, T.M. Los Alamos National Laboratory.

A unique aspect of casting radioactive uranium metal is the presence of uranium progeny, which may or may not behave similarly to uranium. For example, there has long been evidence that thorium fractionates from uranium during casting, and is usually concentrated at the top of the casting. Understanding this behavior is key to interpreting radiochronometry data for uranium metal, where discordant model ages for Th-230/U-234 and Pa-231/U-235 are often observed. Given the long half-lives of Th-230 and U-234, it takes months for enough Th-230 to grow in to reliably measure the Th-230/U-234 ratio in depleted uranium by mass spectrometry. However, the U-238 daughter, Th-234, is also present, should behave identically to Th-230 during casting, and with a 24.1 day half-life is easily measurable by gamma-ray spectrometry. The feasibility of characterizing the distribution of Th using gamma-ray spectrometry was tested on a freshly cast cylinder of depleted uranium. A portable Ortec Detective HPGe and PHDS Co. NP Imager HPGe were used with collimators to identify heterogeneity in the Th-234 distribution. The system was built on an hydraulic table so that the detector could be moved vertically and allow collection of spectra at various points along the vertical axis of the cylindrical casting. Results show a higher Th-234 activity at the top of the casting, with a more consistent lower Th-234 activity through the main body of the casting.

Log 238. **COMPARING SYNTHETIC TRINITITE TO AUTHENTIC AND REMELTED TRINITITE SAMPLES**

Williams, L.P. (1,P); Koury, D. (1); Poineau, F. (1); Libero, J. (1). (1) University of Nevada, Las Vegas. (P) Presenting Author.

In the event of a nuclear attack, the analysis of fallout debris will be crucial to the timely attribution of the nuclear materials used. As such, the synthesis and analysis of realistic, urban, melt glass has been the focus of many projects in recent years. Traditionally, these synthetic materials have been prepared using furnaces to heat and cool the glass over several hours. A novel preparation method which uses a vacuum arc welder to simulate a nuclear fireball plasma allows for the instantaneous heating of materials to temperatures over 3000C, while in a reducing environment. In this study, the vacuum arc welder was used to prepare synthetic trinitite, as well as remelt a sample of authentic trinitite. Additionally, a sample of authentic trinitite was remelted using the more traditional MTI furnace. An authentic sample of trinitite along with the prepared samples of trinitite were analyzed and compared via SEM, EDS, and XRD analysis.

Log 239. **EXPERIMENTAL VERIFICATION OF THEORETICAL STATISTICAL UNCERTAINTIES WITH VRF HOLISTIC NON-LINEAR LEAST-SQUARES ANALYSIS OF GAMMA SPECTRA**

Lasche, G.P. (1,P); Metzger, R.L. (2). (1) Snakedance Scientific, LLC. (2) Radiation Safety Engineering, Inc. (P) Presenting Author.

Typical analysis of gamma spectra begins with a search for peaks. The area and uncertainty of area of each peak is typically calculated with a function that applies Poisson statistics to the fitted peak area and to estimates of the local underlying continuum. We have developed an entirely different approach for analysis of gamma spectra in an application, known as "VRF", in which we fit a single, self-consistent function to all of the spectral data with non-linear least-squares fitting. This "holistic" method derives the uncertainties of activities from the inverse of the final second-derivative matrix (also known as the "error matrix"). By treating all of the data as a single unit to be fitted with minimized chi-squared, and by including the correlated data from the off-diagonal "covariance" elements of the matrix, the statistical (or "counting") uncertainties of activities of radionuclides are often significantly smaller than uncertainties derived from the Poisson variances of individual peak and their local continua. Although the uncertainties with the VRF method are derived from sound mathematical theory, there remains a need to confirm theory with experiment. In this study we report the results of a numerical experiment in which we compare statistical uncertainties that were calculated from the distribution of apparent activities of a large number of spectra to uncertainties that were calculated with the VRF non-linear least-squares method. Comparison is also made with uncertainties calculated with typical techniques from an initial search for individual peaks.

Log 240. AUTOMATED RAPID ANALYSIS AND REPORTING OF LARGE NUMBERS OF COMPLEX GAMMA SPECTRA FOR EMERGENCY MANAGEMENT WITH VRF

Metzger, R.L. (1,P); Lasche, G.P. (2). (1) Radiation Safety Engineering, Inc. (2) Snakedance Scientific. LLC. (P) Presenting Author.

Emergency management in the aftermath of a radiation accident or incident requires analysis of a large number of samples within 24 hours of the event. While HPGe spectrometers with sample changers can count many samples with a 10 to 20 minute count time, the rapid analysis and accurate reporting of a large number of samples is difficult, particularly when the spectra are complex. Samples with fresh mixed fission products or SNM have overlapped peaks that must be separated to extract the required information. Software that relies on conventional peak-search algorithms cannot deconvolute overlapped peak complexes and iterative approaches require significant time requirements for the analyst. A method for automated rapid analysis of large numbers of complex spectra with non-linear least squares fitting has been developed using a new batch mode in the gamma spectrum analysis code VRF. The method is described and the results of an automated analysis of a large number of samples using this method are presented.

Log 241. FEASIBILITY OF THE USE OF COMPTON SUPPRESSION SPECTROMETRY TO DETERMINE Pu-239 IN SOIL

Egozi, C. (1,P); Landsberger, S. (1); Charlton, W.C. (1); Betts, S.E. (2); Winkler, R. (2). (1) The University of Texas at Austin. (2) Los Alamos National Laboratory. (P) Presenting Author.

Typically determination of Pu-239 using gamma ray spectrometry is done at relatively high activity levels for reprocessing, safeguards verification purposes, and for nuclear forensics applications. For environmental levels (tens of Bq/g or lower) other destructive assay methods are used. However, there is a region where gamma ray spectrometry can be used for environmental Pu-239 contamination. A digital Compton suppression system with a high efficiency (79%) HPGe detector was evaluated for the purpose of measuring Pu-239 in a soil reference material at approximately 185 Bq/g (75 ng/g). Statistical analyses for detection limits and uncertainties for various Pu-239 photopeaks were determined. In addition, because many of the photopeaks were of low energy, self attenuation properties were also taken into consideration.

Log 242. SOLAR PHOTOVOLTAIC DEVICES AND SOLAR FARM FOR POST-DETONATION MONITORING

Kandlakunta, P. (1,P); Van Zile, M. (2); Panaccione, W. (1); Cao, L. R. (1,2). (1) Department of Mechanical and Aerospace Engineering, The Ohio State University. (2) Nuclear Reactor Laboratory, The Ohio State University. (P) Presenting Author.

Solar photovoltaic (PV) devices can serve as potential sensors for identification of nuclear detonation and provide valuable information for post-detonation nuclear forensics. The inherent sensitivity of a PV material such as silicon (Si) to gamma-rays and neutrons makes the PV device responsive to nuclear radiation emitted in a nuclear explosion. In addition to neutron scattering and energy threshold reactions of fast neutrons with Si, thermal neutron interaction with boron in p-type Si may make a Si solar cell further sensitive to the neutron spectrum of nuclear detonation. A transient response may potentially be induced in solar PV devices by the prompt ionizing radiation pulse that is characteristic of a nuclear detonation. In this study, we first evaluated the feasibility of using solar PV panels as sensors of nuclear radiation using steady-state X- and gamma-ray measurements with solar cells. Pulsed X-ray and laser measurements were performed to understand the fundamental nature of the transient response of solar cells to ionizing radiation. Monte Carlo simulations in Geant4 were performed to evaluate solar cell sensitivity to gamma-rays and neutrons and support the experiments. The steady-state measurements indicated good response of solar cells to X- and gamma-rays. The pulsed measurements showed that the solar cells can produce a proportional response to a fast transient radiation. The steady-state and transient response of solar cells to neutrons is currently being investigated using the fast neutron beam facility of The Ohio State University research reactor.

Log 243. **ESTABLISHING CAPABILITIES FOR MOLTEN SALT RESEARCH USING THE SODIUM-POTASSIUM CHLORIDE SYSTEM**

Swinhart, M. (1,2,P); Lonergan, J. (1); Sudowe, R. (2). (1) Pacific Northwest National Laboratories. (2) Colorado State University. (P) Presenting Author.

As part of the MSR Campaign to establish a database of the thermophysical and chemical properties of binary and tertiary salt systems, Pacific Northwest National Lab chose to focus on establishing capabilities using a sodium and potassium chloride system. Chlorine salts are less toxic than their fluorine counterpart, more proliferation resistant, and have more compatibility with mixed and used fuels. This study focuses on the sodium and potassium-chloride (ClNaK) series and the methodologies developed within will be used for future actinide containing systems. Determination of heat capacity, enthalpy, volatilization, and viscosity have been well established as experimental methods. Many of these properties need to be understood to optimize models and prepare for successful reactor operations. Heat capacity experiments performed with differential scanning calorimetry and drop calorimetry have shown good alignment with literature values and compare well to systems used in applied solar. Leaching in containment vessels through experimental parameters suggests high corrosion and volatility. The volatility will need to be a primary focus in case of extra headspace requirements in the reactor core or subsequent systems, such as the heat exchangers. The fission products are often in ionic form that can cause a runaway reaction if there is sufficient mass loss. Thus, studying the fundamentals of these base fuel salts is crucial to reactor function and design.

Log 244. **CHARACTERIZING UNCERTAINTY IN THE THERMAL NEUTRON CROSS SECTION FOR THE Ca-40(n,α)Ar-37 REACTION**

Shah, K.A. (1, P); De Luna, B.A. (1); Kaitschuck, N.M. (1); Haas, D.A. (1). (1) The University of Texas at Austin. (P) Presenting Author.

Argon-37 is a signature of interest for nuclear explosion monitoring because its longer half-life compared to radioxenon isotopes provides a longer detection window and it is produced in underground nuclear explosions (UNEs) with high enough activity to be detected using current detection systems. 37Ar is formed via the (n,a) reaction with 40Ca, the dominant isotope in natural Ca, which is found in most common rock types. The thermal neutron cross section for the 40Ca(n,a)37Ar is poorly characterized, with the Evaluated Nuclear Data File (ENDF) and the Japanese Evaluated Nuclear Data Library (JENDL) disagreeing on what occurs at lower energies. We conducted a sensitivity study using Monte Carlo N-Particle code and the SCALE code system to predict the yield of 37Ar from the 40Ca(n,a) reaction from fission neutrons activating various rock types. This study shows resolving the differnces in the cross section models for the 40Ca(n,a)37Ar reaction and understanding variations in the elemental makeup of different rock types are important to estimating the amount of 37Ar produced in a UNE. A preliminary experiment is being designed to measure the thermal neutron cross section of the 40Ca(n,a)37Ar reaction. A thin sample of Ca in a cylindrical vaccum chamber will be irradiated in a thermal neutron beam and alpha spectroscopy will be performed to measure the thermal neutron cross section.

Log 245. **A COMPARATIVE ANALYSIS OF THREE Ti-44/Sc-44 GENERATOR SYSTEMS FOR BENCH-TOP SCALE APPLICATIONS**

Nicole Ahrens (1,P), Ralf Sudowe (1). (1) Colorado State University. (P) Presenting Author.

Scandium-44 is a positron-emitting radionuclide that has been identified as an efficacious Positron Emission Tomography (PET) imaging agent for radiopharmaceutical and theranostic applications. Theranostics is gaining momentum in the treatment of various cancers, with Sc-44 as a diagnostic imaging agent when paired with Sc-47 as the therapeutic agent. An easy way to obtain Sc-44 for laboratory experiments is through the use of a Titanium-44/Scandium-44 radionuclide generator. The long-lived Ti-44 is usually produced via proton-induced nuclear reactions on natural scandium. In this work, three different Ti-44/Sc-44 generator systems were being evaluated for Sc-44 recovery, Ti-44 breakthrough, and elution volume required. The first generator system utilizes extraction chromatography (TEVA resin) and dilute hydrochloric acid/oxalic acid, the second system employs anion-exchange chromatography (AG1-X8 resin) and dilute hydrochloric acid/oxalic acid, and the final system utilizes cation-exchange chromatography (AG50-X8 resin) and dilute hydrochloric acid/oxalic acid. Activities of Sc-44 and Ti-44 in the eluent were measured by gamma-spectrometry using a high-purity germanium (HPGe) detector.

Log 246. **COMPARISON OF VARIOUS EXTRACTION CHROMATOGRAPHIC RESINS FOR RADIUM ANALYSIS IN FLOWBACK WATERS**

Coupannec, M. (1); Happel, S. (2); Sudowe, R.(1) (1) Colorado State University. (2) TrisKem International. (P) Presenting Author.

A major concern arising from hydraulic fracturing is the generation of a large volume of flowback water potentially containing various amounts of dissolved naturally occurring radioactive material. Up to four million gallons of water-based fluid is injected per well, of which 10–70% is subsequently recovered as flowback. Improving the measurement and separation of "Technologically Enhanced Naturally-Occurring Radioactive Material" (TENORM) is therefore a priority. Radium has gained attention as a source of potential environmental contamination in process waters from hydraulic fracturing sites and bodies of water affected by the mining industry. Providing accurate radium measurements is an essential step for appropriate disposal of the brine water as regulated or non-regulated NORM-containing wastes. The oil and gas industry produces water with high dissolved solid material content leading to a significant scientific challenge in achieving separations of radium due to the high concentration of chemical analogs such as barium, strontium, and calcium. Multiple extraction chromatographic resins developed by TrisKem Chemical were investigated at different nitric acid concentrations to quantify and enhance the separation of radium from the chemical analogs. Identifying a resin capable of extracting radium from the fracking water would also allow the reduction of costs associated with handling, treatment, and disposal of the wastewater.

Log 247. Irradiation of NaCl-KCl-UCl₃ Salt with Depleted U for Total Mass Accounting in Advanced Liquid-Fueled Reactors

E. Gordon (1,2), P. Kandlakunta (2), M. Van Zile (3), A. Kauffman (3), S. Li (4), L. R. Cao (2,3). 1. Chemical Engineering Program 2. Nuclear Engineering Program 3. Nuclear Reactor Laboratory 4. University of Utah. (P) Presenting Author.

This research aims to validate a radioactive tracer dilution (RTD) method for irradiated fuel-bearing molten salt mass determination at a small scale to evaluate the possibilities of its deployment in nuclear material accountancy (NMA) scenarios, for example molten salt loops in liquid-fueled molten salt reactors (LFMSRs). The irradiation of a fuel-bearing salt sample with a radioactive tracer of known activity at a University Research Reactor provides a critical step for validating this NMA concept in the design of LFMSRs. This paper is focused on the initial efforts of evaluating the RTD method for mass determination in an irradiated salt. High purity (99.99%) NaCl and KCl in equal molar amounts with a sample of 10 mg of U-235 in 5 g of DU provides a risk-mitigation step for the first irradiation experiment. 22Na has been demonstrated as the first radioactive tracer to test the RTD concept. This tracer was chosen because it undergoes beta+ decay which won't interfere with the fission products, it has known chemical compatibility with actinides and fission products in molten salt chloride and fluoride salts, it emits a 1274.54 keV gamma-ray that is outside of the Compton plateau of many spectrums of fission products, and it has an identified overlapped peak at 1274.43 keV from 154Eu. The measured gamma-spectrum will be used to evaluate the potential spectrometer inferences from fission products.

Log 248. INVESTIGATING THE ROLE OF CARRIER IN THE ADSORPTION OF SCANDIUM ON DGA UTILIZING A Ti-44/Sc-44 GENERATOR

Brown, M.L. (1); Boron-Brenner, L.P. (2); Sudowe, R.(1,P). (1) Colorado State University. (2) University of Nevada Las Vegas. (P) Presenting Author.

Scandium is an isotope of major interest when it comes to theranostic applications. There are two main isotopes of scandium utilized in medical applications, Sc-44 and Sc-47. Sc-44 is a positron emitter and is used for imaging, while Sc-47 is utilized for tumor treatment. Together, the pair make a theranostic agent. This research, however, focuses mainly on Sc-44. In order to utilize this isotope, it must first be produced, separated from the irradiated target material, and purified. An efficient and frequently used method for separating radioisotopes is extraction chromatography. The goal of this research is to understand and possibly improve separation of scandium from titanium, while employing a variety of different chromatographic resins. In the past, several groups have examined the uptake of scandium and titanium on an extraction chromatographic resin based on a tetraoctyl diglycolamide, DGA, with both stable and radioactive scandium. While the uptake of titanium was consistent between the studies, all groups have reported different values for the uptake of scandium. The aim of this part of the work is to compare both the uptake of stable and carrier-free scandium to further elucidate the discrepancies between the studies reported in the literature. Radioactive Sc-44 for tracer studies was obtained by "milking" a Ti-44 generator in regular intervals.

Log 249. INVESTIGATION OF A HEXACYANOFERRATE(II) NANODIAMOND ADSORBENT FOR THE PRECONCENTRATION OF CESIUM FROM WATER SAMPLES

Deak, A.T. (1); Sudowe, R. (1,P). (1) Colorado State University. (P) Presenting Author

The determination of low levels of radioactivity in the environment is challenging, because the sample amount required to meet low minimum detection limits can be difficult to handle and can give rise to self-shielding which may lead to inaccurate measurements. Consequently, preconcentration techniques such as precipitation or evaporation are often used prior to radiochemical separations and/or sample measurement. In this work the use of a hexacyanoferrate(II) nanodiamond adsorbent is investigated to preconcentrate dissolved Cs in contaminated solutions in order to improve the counting efficiency and accuracy for low activity samples. The majority of the preconcentration studies yielded over 98% Cs-137 adsorption with adsorbent amounts of as little as 5 mg.

Log 250. TECHNETIUM COMPLEXATION WITH HALIDES

Nathalie A. Wall

Technetium-99, a fission product of concern for nuclear wastes, can be found in aqueous systems known to contain halide salts. While Tc(VII) is present under oxic conditions, Tc(IV) is also observed in the environment due to different reductive pathways. Quantification of thermodynamic parameters associated with the formation of the Tc(IV)-halide complexes in solution is important to understand the metal behavior in relevant environmental water systems. Our results show that the fluoride complexation is endothermic with inner-sphere characteristics, occurring as an overall entropy-driven reaction for the addition of three fluorides. The chloride and bromide systems are exothermic with outer-sphere characteristics; the reactions for the formation of the 1:0:1 complexes are enthalpy-driven.

Log 251. NEW NEUTRON IMAGING DETECTORS USING ASTRONOMY CAMERAS AND 3D PRINTED DETECTOR HOUSINGS

B. Schillinger (1,P), T. Juenger (1), T. Neuwirth (2), S. Sebold (1). (1) Heinz Maier-Leibnitz Zentrum (FRM II), Technische Universitaet Muenchen, Germany. (P) Presenting Author.

Classic neutron imaging detectors employ a high-end CCD or CMOS camera in detector housings with massive shielding at considerable cost. Lead and boron shielding is required to protect the camera from gamma rays and scattered neutrons originating from the sample or detector housing which may cause white spots or even damage on the chip. Due to size and mass, these detectors are not overly flexible. At the FRM II reactor of Technische Universitaet Muenchen, with the advent of new high-quality compact astronomy cameras, we developed a new modular concept that holds the camera in a housing as small as possible, with a cutout lead block only at the front of the camera that allows external stacking of shielding. The camera box has a flange that can be connected to various mirror-and-screen arrangements of different sizes. Members of our group have become very proficient in 3D printing, so we printed various detectors for different fields of view from standard imaging to a very high resolution microscope setup. The

new concept allows for low-cost but high-quality detectors to be built in very short time. First test measurements will be presented.

Log 252. NEW MEASUREMENTS ON BORATED NEUTRON IMAGING SCREENS

Burkhard Schillinger (1,P), Bill Chuirazzi (2), Steven Cool (3), Aaron Craft (2), Alessandro Tengattini (4).   (1) Heinz Maier-Leibnitz Zentrum (FRM II), Technische Universitaet Muenchen, Germany. (2) Idaho National Laboratory, USA. (3) DMI/Reading Imaging, Reading, USA. (4) Institut Laue-Langevin, Grenoble, France

The most commonly used screens for neutron imaging consist of 6LiF+ZnS. This type of screen yields the highest light output per detected neutron. For high resolution, gadolinium-oxisulfide (Gadox) screens are employed, which have a much higher detection efficiency, but a light output so much lower than for LiF+ZnS that measurements are often limited by photon statistics. Screens using boron as neutron sensitive material have not been very successful in the past until a new preparation method was introduced recently that combines light output higher than Gadox with detection efficiency larger than LiF+ZnS. The talk will report about measurements of new borated screens at he NEXT facility at ILL, Grenoble, in comparison to a high resolution Gadox screen.

Log 253. INTEGRAL FISSION PRODUCT CHAIN YIELD MEASUREMENTS AT NCERC

Bredeweg, T.A. (1,P); Berger, J. (1); Boggs, M. (1); Bond, E.M. (1); Boswell, M. (1); Bowen, S.M. (1); Cox III, D.L. (1); Dembowski, M. (1); Dry, D.E. (1); Flanagan, D.C. (1); Gaunt, A.J. (1); Gooden, M.E. (1); Hanson, S.K. (1); Hudston, L.A. (1); James, M.R. (1); Kinman, W.S. (1); Lance, C.A. (1); Lee, G. (1);Marenco, A.M. (1); Margiotta, C. (1); May, I. (1); Meininger, D. (1); Miller, J.L. (1); Oldham, W.J. (1); Reilly, S.D. (1); Rendon, R.J. (1); Roman, A.R. (1); Romero, J.R. (1); Rundberg, R.S. (1); Smythe, N. (1); White, J.M. (1); Williams, J.W. (1); Wren, M.S. (1); Uhnak, N.E. (2); Haney, M.M. (2); Pierson, B. (2); Greenwood, L. (2); Friese, J. (2); Metz, L. (2) (1) Los Alamos National Laboratory. (2) Pacific Northwest National Laboratory. (P) Presenting Author.

Fission product chain yields were historically determined by chemical separation and beta counting of fissile samples irradiated in carefully controlled fission chamber experiments. These measurements provided the means to extract absolute fission product yields (yield per fission) that are included in the international nuclear data libraries, and ultimately used to model and characterize multiplying systems. However, re-evaluations conducted in the late 2010s for neutron-induced fission of U235 and Pu239 highlighted disagreement between several key measurements. This prompted several new, targeted experimental programs, both in France and in United States, to understand and resolve these disagreements. It has also sparked a renewed interest in creating a new fission product yield evaluation that includes new energy differential and energy integral fission product data. In this presentation we will highlight recent efforts to address discrepancies and data gaps using energy-integral measurements for U235 and Pu239. This work relies on the historical combination of fission chambers and radiochemical analyses of samples irradiated on several of the critical assemblies located at the National Criticality Experiments Research Center (NCERC) at the Nevada National Security Site. Details will be provided on the design and performance of the fission chambers, and results from actinide

sample irradiations on the Flattop and Godiva critical assemblies. We will conclude with an outline of our plans to complete the work over the next several years.

Log 254. **Long Term Stability of Ag/AgCl Reference Electrode in Molten Chloride Salt**

S. Choi (1,P); J. Steppan (2); M. F. Simpson (1). (1) Unviersity of Utah. (2) HiFund, LLC. (P) Presenting Author.

Precise monitoring of reduction and/or oxidation potential is necessary for using molten salts as fuel compositions and coolants in molten-salt reactors as well as for pyrochemical processing of spent fuel. However, due to the high working temperature and the effects of the corrosion, there is no commercial reference electrode available for molten salts. A stable reference electrode is essential for this application. In this work, we focused on measuring the stability of high-temperature molten chloride reference electrodes (RE) using mullite and magnesia tubes as reference electrode membrane materials in molten CSP salt (MgCl2-KCl-NaCl). Two electrochemical techniques were used to characterize the long-term performance of REs at 500°C: Open Circuit potential (OCP) versus magnesium and cyclic voltammetry (CV) for up to 31 days. The OCP values and the onset potentials of magnesium reduction show similar behavior for the mullite and magnesia membranes.

Log 255. **IMAGE FUSION FOR NEUTRON IMAGING APPLICATIONS**

Chuirazzi, W.C. (1,P); Kane, J.J (1); Craft, A.E. (1); Schulthess, J.L (1). (1) Idaho National Laboratory. (P) Presenting Author.

Image fusion, the process of combining different images together, can be useful to create a more complete picture. This technique is commonly used in security and medical applications. Previous work has fused neutron and X-ray radiographs to utilize the advantages of both techniques. In this work, image fusion is applied to neutron tomography of nuclear fuel with the goal of enhacing the information obtained during the measurement of the sample. Thermal and epithermal neutron images were taken and fused together to show more details of the fuel sample than either tomograph would separately. Thermal neutrons offer more contrast as they interact with the sample, but epithermal neutrons can penetrate through the entire sample and offer more detail of thick or dense regions of the sample. Different reconstruction methods, such as FDK filtered backprojection and Simultaneous Reconstruction Technique (SIRT), emphasize different details of a reconstructed object. The FDK reconstruction can preserve edges in the image, while the SIRT algorithm significantly reduces noise. By combining separate reconstructions using each algorithms the edges of an image of the nuclear fuel sample were fairly well preserved, while the noise was reduced. then fused together to get a dataset exhibiting the desired features of each reconstruction. Discussion on technique enhancements and future applications for the neutron imaging community are also part of this work.

Log 256. IMAGING THE PLANT UPTAKE OF RADIONUCLIDES ON THE SINGLE-CELL SCALE USING SIMS AND rL-SNMS

Mandel, M. (1); Holtmann, L. (P)(1); Raiwa, M. (1); Wunnenberg, A. (1); Riebe, B. (1); Walther, C. (1). (1) Institute of Radioecology and Radiation Protection, Leibniz University Hannover. (P) Presenting Author.

In radioecological studies, the plant uptake and distribution of radionuclides are of major interest for risk assessment. In the present work, the elemental distribution within the tissue of plants is imaged at varying concentrations by resonant laser secondary neutral mass spectrometry (rL-SNMS). This technique combines a commercially available time-of-flight secondary ion mass spectrometry (TOF SIMS) with a home built laser system for ionization of sputtered neutrals. Due to the excellent suppression of molecular isobaric interferences by rL-SNMS, about 10^10 atoms of technetium suffice to image the Tc-99 distribution inside plant cells with ca. 300 nm spatial resolution. The distribution of technetium in the two plant species Daucus carota and Pisum sativum was determined. For reasons of radiation protection, the plants were labelled with Tc-99 at a concentration of 0.1 mM. Such a low concentration renders measurements by conventional SIMS impossible. SIMS measurements on the distribution of rhenium and iodine were performed for comparative purposes. Since suppression of isobaric interferences is not possible with SIMS, plants had to be labelled with much higher concentrations in the range of 10 mM.

Log 257. RADIOCHEMICAL EFFECTS OF THERMAL NEUTRON CAPTURE IN Cr(tmhd)$_3$

Pichler, V. (1,P); Welch, J.M. (1); Sterba, J.H. (1). (1) Center for Labelling and Isotope Production, TRIGA Center Atominstitut, TU Wien, Vienna, Austria. (P) Presenting Author.

The radiochemical effects of thermal neutron capture in tris(2,2,6,6-tetramethyl-3,5-heptandionato)chromium(III) (Cr(tmhd)3) have been investigated. A method has been devised to quantify the amount of the induced Cr-51 activity recoverable in the original chemical (i.e. parent) form and confirm that this "retention" is in fact in parent (Cr(tmhd)3) form. The method is based on a combination of chemical separation (column-chromatography and recrystallization) as well as analytical (radio-TLC, HPLC and gamma spectrometry) techniques. Cr(tmhd)3 samples were irradiated and purified in two steps after irradiation. We found that two of the (three) purification pathways yielded good agreement among final Cr-51 specific activities and retention values thus obtained. Furthermore, the corresponding samples appeared to be radiochemically very pure but chemically a little less so. The vast majority (> 90 %) of the Cr-51 activity induced by neutron irradiation is not in parent form and can be separated from the bulk material chromatographically, making Cr(tmhd)3 a potential candidate for use in a Szilard-Chalmers Cr-51 enrichment process.

Log 258. **CHARACTERIZATION OF NEUTRON IMAGING FACILITY AT PENN STATE BREAZEALE NUCLEAR REACTOR**

Kenges, A. (P); Unlu, K.; Beck, D.B.; Pennsylvania State University, The Radiation Science and Engineering Center. (P) Presenting Author.

The design features and results of characterization experiments that have been performed at Radiation Science and Engineering Center for the new Neutron Imaging Facility (RSEC - NIF) will be presented. The RSEC -NIF has been categorized as a Category I facility by ASTM designation of quality by means of collimator components and filters. The resulting spatial resolution of the RSEC – NIF is between 15 to 200 microns depending on factors such as the aperture-to-plate distance or the imaging plate that being used. Additionally, the effective collimation ratio (L/D ratio) of our current system is adjustable between 100-150 and the thermal flux at the exit surface of the neutron beam is equal to 5.4E+06 n/cm^2-s at 1MWth reactor power. These results allow us to use both direct and indirect methods of neutron radiography to image any given sample of interest in 2D format utilizing conventional films and modern phosphorous plates. In addition, the RSEC – NIF will also deploy a neutron tomography technique that allows to use the attenuation properties of neutrons for 3D imaging. The RSEC – NIF will be employed to solve the relevant issues such the study of emerging global concern of microplastic pollution of the environment. Preliminary experiments have shown that the content and spatial distribution of microplastics in the sand can lead to the research of beach sands and soil samples for deeper understanding of plastic contamination of waters and lands.

Log 260. **MEASUREMENTS OF 135/137 CESIUM RATIOS IN VARIOUS FORMS OF TRINITIE BY THERMAL IONIZATION MASS SPECTROMETRY**

Boggs, M.A. (1); Reinhard, D. (1); Inglis, J. (1) Lee, G. (1); Margiotta, C. (1). (1) Los Alamos National Laboratory

The 135 and 137 mass chains are produced in greater than 6% of the thermal fissions from 235U and 239Pu. Both of these decay chains result in the long half-life cesium isotopes 137Cs (30yr) and 135Cs (2.3x106 yr). However, because of differences in independent yield of 137Cs and 135Cs as well as the inherent volatility and varied half-lives of the cesium precursors (Sb, Te, I, Xe) the ratio of 135Cs to 137Cs is highly dependent on both the time after fission event and temperature at which the sample was formed. Thus, the ratio of 135/137-cesium offers a novel method to investigate the manner in which a piece of nuclear debris was formed and how it may be influenced by conditions directly after detonation. Debris from an above ground nuclear event comes in many different forms (glassy rock, aerodynamic beads, etc.) all of which are formed at different times, temperatures, and distances from the detonation site. Variations in the 135/137-cesium ratios between these sample types offer insights into not only nuclear debris formation but also into the broader question of how to use such values for treaty and safeguards applications. We will show high precision TIMS results on debris from the Trinity nuclear test and discuss how these results influence our understanding of how nuclear debris is formed, as well as relating them to broader nuclear forensics and treaty monitoring missions. LA-UR-21-32177

Log 261. **UTILITY OF ATONA AMPLIFIERS FOR ISOTOPIC ANALYSES BY THERMAL IONIZATION MASS SPECTROMETRY**

Reinhard, A.A. (1,P); Inglis, J.D. (1); Kara, A.H.S. (1); Goldstein,S.J.(1); Boggs, M.A. (1); Lopez, D.M. (1); LaMont, S.P. (1); Steiner, R.E. (1) (1) Los Alamos National Laboratory, Chemistry Division (P) Presenting Author.

For 50+ years thermal ionization mass spectrometers (TIMS) have relied on faraday cup detectors utilizing high-ohmage resistors as amplifiers. Recently Isotopx released the ATONA amplifier system which utilizes capacitance based amplification, resulting in faraday detectors with a larger dynamic range, greater gain stability, and lower noise. When installed on the IsotopX Phoenix TIMS this system provides a number of benefits for measuring actinides and fission products of interest for treaty monitoring and nuclear forensic applications. We demonstrate the capabilities of these amplifiers with isotopic analyses of three elements of interest Sr, Cs, and U. The dynamic range of the ATONAs enables the measurement of 88Sr on a faraday cup while simultaneously measuring 90Sr on the secondary electron multiplier (SEM), yielding precise determination of these isotopes in environmental samples containing less than 5fg of 90Sr. The combination of low noise and high abundance sensitivity allows for the measurement of 135Cs and 137Cs by either peak hopping on the SEM or by total evaporation with all Cs isotopes on faraday detectors. These methods allow for analyses of 135Cs/137Cs in larger Cs standards (30fg 137Cs) with very high precision 0.2% and small Cs standards (200ag 137Cs) with good reproducibility (1%). The ATONA amplifiers also allow for the precise and accurate analysis of the minor 234U/238U ratio (as well as 235U/238U) in isotopically natural standards using only faraday cups and in combination with the SEM, 236U/238U can be measured with high sensitivity. LA-UR-21-31990

Log 262. **ARCANA: THE NATIONAL NUCLEAR MATERIALS AND SIGNATURES DATABASE**

Robel, M. (1,P); Marks, N. E. (1). (1) Lawrence Livermore National Laboratory. (P) Presenting Author.

The National Nuclear Material Archive (NNMA) program identifies, samples, characterizes, and preserves nuclear materials of US origin to support attribution of materials discovered out of regulatory control. The program includes a physical archive of relevant materials; a robust materials characterization program; the compilation of information about the samples in a database and data repository; and an assessment and query response process including statistical tools and subject matter expertise. The NNMA sample characterization work will produce a large quantity of analytical data which must be captured and stored in a way that is secure, accessible to stakeholders, and available for use in assessments, query responses, and the identification of nuclear forensic signatures in archive materials. In support of this goal, we have developed Arcana, a bespoke relational database specifically designed for efficient upload, storage, retrieval, and queries of NNMA data. The Arcana database structure is implemented with Oracle SQL through the Oracle APEX development environment, enabling rapid development of web-browser interfaces for database uploads, queries, and data visualization. The flexible structure can accommodate detailed processing and signature information about each sample, as well as associations between samples representing similar processes. The Arcana database along with subject matter expertise will provide a timely response capability for queries

Log 263. **SMALL-BATCH PLUTONIUM METAL CREATION**

Ferrier, M. G., Henderson, R. A. (P), Lawrence Livermore National Laboratory. (P) Presenting Author.

Small-batch processes were used in the early days of Pu research into the properties of the element primarily because of the scarcity of the material. Baker (Baker, 1946) described a process that has been used as a basis for the current project. High purity 242Pu was purchased from the Oak Ridge National Laboratory for this purpose. The thermo-metallic bomb reduction process is used to create up to 1g of Pu metal. The excellent reference by Cleveland (Cleveland, 1979) serves as a basis for the chemical processing of the plutonium. The complete process includes the recycling of by-product material created during target fabrication.

Log 263. **APPLICATION OF ARTIFICIAL INTELLIGENCE METHODS TO MICROSCOPIC PARTICLE ANALYSIS**

Bickley, A.A. (1,P). (1) Air Force Institute of Technology. (P) Presenting Author.

The modern challenge of detecting nuclear proliferation activities by integrating trace materials analysis with machine learning to identify signatures of nuclear fuel cycle activities is addressed. Analysis of trace materials collected from nuclear fuel cycle processes has been conducted for generations; however, the laboratory and analytical procedures used are human resource intensive and time consuming. The introduction of modern automated materials analysis laboratory tools has resulted in an increase in the fidelity of the chemical and elemental data gathered, but evaluation of this data has been hampered by its sheer quantity. This is the type of problem at which machine learning and artificial intelligence models excel. In this work, the chemical synthesis conditions of a wide variety of uranium oxides are predicted via the application of traditional machine learning based classification methods trained on the observed morphological features of microscopic particles available from scanning electron microscopy (SEM) imagery. Separately, an artificial neural network is applied to elemental assay data measured using energy dispersive spectroscopy (SEM-EDS) of field collected samples. The accuracy of the neural network for determining whether two particles originate from the same sample based on their elemental composition is assessed. Preliminary results of efforts to enhance these capabilities through the use of data fusion will be discussed.

Log 264. RADIOCHEMICAL SEPARATIONS AND YIELD MEASUREMENTS FOR SHORT-LIVED NATURAL THORIUM FISSION PRODUCTS

Kuatbek, M. (1,P); Pierson B. D. (2); Lyons S. M. (2); Johnsen A. M. (1). (1) The Pennsylvania State University. (2) Pacific Northwest National Laboratory. (P) Presenting Author.

While significant research has been performed previously to experimentally determine yields for long- and intermediate-lived fission products, a number of short-lived fission products suffer from relatively high uncertainties. Additional measurements for short-lived fission product yields are needed for improved nuclear security applications, and there is particular interest in radiochemical separations at short times post-irradiation to facilitate the evaluation of isotopes with low-yield and low-energy gamma-ray emissions. In this work will be presented simulated and experimentally obtained results from the initial irradiation of thorium-232 targets in a fast fission neutron oriented irradiation fixture of the Penn State Breazeale Reactor (PSBR), as well as the subsequent radiochemical separations. The PSBR Fast Neutron Irradiator (FNI) is a permanent irradiation tube shielded with boral (borated-aluminum) and lead; our characterization results show a neutron flux spectrum where 59.4% of the neutrons are >0.5 MeV, 40.6% are between 0.56 eV and 0.5 MeV, and less than 0.05% are thermal neutrons. This spectrum presents a unique opportunity to analyze fission product yields from incident neutrons that are predominantly fast, but with a contribution from the resonance region group, which can significantly influence fission yield values. Compton suppression measurements and post-irradiation radiochemical separation techniques will be used to identify and quantify additional fission product isotopes that are obscured by the Compton continuum from higher energy gamma-ray emitters.

Log 265. NEUTRON TOMOGRAPHY OF A HIGHLY RADIOACTIVE SINQ SPALLATION TARGET ROD

Trtik, P. (1,P); Welte, J. (1); Yetik, O. (1); Grünberger, S. (1); Kalt, A. (1); Hovind, J. (1); Blau, B. (1); (1) Paul Scherrer Institut, Switzerland. (P) Presenting Author.

The neutron spallation source (SINQ) at the Paul Scherrer Institut produces free neutrons by means of a solid state target bombarded by an intense 590 MeV proton beam. The SINQ target comprises of more than 300 hermetically-sealed Zircaloy tubes filled with lead up to 90 per cents of its inner volume. The redistribution of the lead filling induced by many cycles of melting and solidification poses a potential risk of the tubes cracking and thus endangers the safety of the target. We performed a tomographic investigation of one of the most heavily perturbed and thus highly radioactive SINQ target rod (~350 mSv/h, approximately 18 months after the end of the 2-years long irradiation cycle) using Dy-based imaging plates at the NEURAP insertion device at the NEUTRA thermal neutron imaging beamline. The obtained neutron tomography dataset is pioneering in numerous ways and goes qualitatively beyond any tomography of highly radioactive objects made so far. It reveals in 3D the re-distribution of the lead filling. Likewise, the precise shape of the deformed target rod (manifested by the ~100 μm swelling of its centre part) is revealed. Thanks to the applied advanced image processing methods trustworthy values of the linear attenuation coefficients (LAC) of the materials are derived. The change in the LAC of the irradiated lead filling (likely due to the presence of the entrapped spallation products) in comparison with the pristine lead is quantified. The dataset is currently utilized as an input for possible design improvements of future SINQ targets.

Log 266. **A COMBINATION OF THERMAL AND EPITHERMAL INSTRUMENTAL NAA AS WELL AS CLOUD POINT EXTRACTION PRECONCENTRATION NAA FOR ALUMINUM IN CANADIAN DUPLICATE DIETS**

E. E. Sullivan, J. Dalziel, A. Chatt (P). Trace Analysis Research Centre, Department of Chemistry, Dalhousie University, 6274 Coburg Road, Room 212, PO BOX 15000, Halifax, NS, B3H 4R2, Canada. (P) Presenting Author.

Aluminum is a rather difficult element to determine by almost all analytical techniques. In this work a combination of instrumental neutron activation analysis (INAA) and epithermal INAA (EINAA) using the Dalhousie university SLOWPOKE-2 reactor facility was used for the determination of aluminum in diets. The 1779-keV gamma-ray of ^{28}Al produced by the reaction $^{27}Al(n,\gamma)^{28}Al$ was employed after correction by EINAA for the interfering reaction $^{31}P(n,\alpha)^{28}Al$. Our results for food reference materials analyzed by this INAA/EINAA method agreed well with the certified values. An independent preconcentration NAA (PNAA) method by cloud point extraction (CPE) using p-nonylphenol ether 7.5 as the surfactant and 8-hydroxyquinoline as the chelating agent was successfully developed for not only to concentrate aluminum but also to significantly reduce phosphorus. The results by two methods agreed well.

Log 267. **DETERMINATION OF LOW LEVELS OF ANTIMONY IN SEAWEEDS BY MICELLE-MEDIATED EXTRACTION FOLLOWED BY NEUTRON ACTIVATION**

Serfor-Armah, Y. (1,2); Carboo, D. (3); Akuamoah, R.K. (3); Chatt, A. (1,P). (1) Department of Chemistry, Dalhousie University, Halifax, NS, Canada. (2) School of Nuclear and Allied Sciences, University of Ghana, Atomic-Accra, Ghana. (3) Department of Chemistry, University of Ghana, Legon-Accra, Ghana. (P) Presenting Author.

Micelle-mediated extraction, commonly called cloud point extraction (CPE), is a good technique for preconcentration of trace elements. However, it has not been used that much in combination with NAA except in our and a few other laboratories. A simple one-step CPE method was developed for the preconcentration of antimony using a mixture of PAN and TAN chelating agents and PONPE-20 surfactant. The recovery of antimony under the optimum conditions of pH 6.4, [PAN/TAN]=1x10E-4 M, [PONPE-20]=0.1% (m/v), ionic strength=0.05 M KNO3, and a temperature of 41oC was >95%. Antimony was assayed using the 564.1-kev photopeak of 122-Sb (half-life=2.72 d). The detection limit of the CPE method was 2.8 µg/kg in seaweeds. The method was validated using several reference materials. It was applied to the analysis of seaweeds.

Log 268. A COMPARISON OF EXPANDED UNCERTAINTIES BY PSEUDO-CYCLIC EPITHERMAL INAA WITH ANTICOINCIDENCE COUNTING USING COMPARATOR AND K0 METHODS FOR MEASURING IODINE LEVELS IN GHANAIAN FOODS

Nyarko, B.J.B. (1,2); Akaho, E.H.K. (2); Fletcher, J.J. (3); A. Chatt (1,P). (1) Department of Chemistry, Dalhousie University, Halifax, NS, Canada. (2) Ghana Atomic Energy Commission, P.O. Box LG80, Legon-Accra, Ghana. (3) Physics Department, University of Cape Coast, Cape Coast, Ghana. (P) Presenting Author.

Several instrumental neutron activation analysis (INAA) methods, namely conventional INAA, epithermal INAA (EINAA), pseudo-cyclic INAA (PC-INAA) and pseudo-cyclic EINAA (PC-EINAA) using conventional as well as anticoincidence (AC) gamma-ray spectrometry and comparator and k0 standardization procedures, were developed for measuring iodine levels in Ghanaian foods. The PC-EINAA-AC method gave the lowest detection limit of 0.5 µg kg-1 and was used. The expanded uncertainties for determining iodine by this method using the comparator and k0 standardization procedures were evaluated following ISO GUM. The NIST RM 8415 Whole Egg Powder was selected for validating the method as well as for calculating expanded uncertainties. The value for the k0 procedure appears to be slightly influenced by the uncertainties in the nuclear constants and detector full photopeak efficiency determination.

Log 269. DETERMINATION OF NEUTRON ABSORPTION SELF-SHIELDING FACTORS FOR LANTHANIDE ELEMENTS DURING NEUTRON ACTIVATION ANALYSIS

Landsberger, S.(P);Kravitz, I.; University of Texas at Austin. (P) Presenting Author.

Neutron Activation Analysis (NAA) is a non-destructive method of analyzing the elemental composition of a sample with fine detection limits that can reach parts per billion for some isotopes. When conducting NAA, materials with high absorption cross-sections reduce the overall neutron flux within the sample by absorbing incident neutrons. This effect is known as neutron self-shielding. The implementation of correction factors that can account for these effects are important in increasing the accuracy of NAA.The objective of this research was to determine a wide variety of neutron self-shielding factors for each lanthanide element, with the exception of promethium, for a range of elemental concentrations. This was done by experimentally irradiating solutions of these elements and then using NAA to determine the activity of each sample.

Log 270. **CHARACTERISATION AND MONITORING OF UNDERGROUND LONG-LIVED CONTAMINANTS USING IN-GROUND ASSETS**

Elisio, S. (1); Joyce, J.M. (1); (1) Lancaster University (UK). (P) Elisio, S. (P) Presenting Author.

This research presents a resilient radiometric logging probe technology for underground monitoring of blind-tubes for long-lived radioactive contaminants, such as Cs-137 and Sr-90. The probe comprises a commercially-available 10x9.5mm cerium bromide scintillation crystal attached to a small, full-featured MCA Topaz-SiPM module. Key features include high gamma-ray detection efficiency, competitive energy resolution, high count-rate and radiation tolerance, small form factor and low-voltage operation, compatible with supply via laptop-based USB interface. The assessment of Cs-137 and Sr-90 in the surrounding soil formation (but close to the borehole) is achieved by detecting the gamma-rays from radioactive decay and bremsstrahlung photons (the latter from the deceleration of beta particles emitted by Sr-90 in the steel of the blind-tubes). Preliminary laboratory-based tests suggest effective gamma-ray spectroscopy performance and detector response linearity of the scintillator detector. The hypothesis of detecting beta-emitting Sr-90 indirectly, via bremsstrahlung, has been proven successfully with a source placed on outside wall of a steel-pipe based phantom. In the presence of Sr-90, an anomalous photopeak-to-scatter peak ratio is expected, relative to that without. Further investigations are underway to enable a wider diversity of scenarios to be considered including the study of detection limitations, and to calibrate the system in a soil-filled phantom arrangement replicating the in-ground blind-tubes used on legacy sites for site inspection.

Log 271. **RAPID RESPONSE IN VITRO BIOASSAY METHOD FOR THE DETERMINATION OF Pu ISOTOPES IN URINE SAMPLES**

Macsik, Z. (1,P); LaMont, S. P. (1); Riggins Cardon, A. M. (1); Hudston, L. A. (1); Wende, A. M. (1); Inglis, J. D. (1); Steiner, R. E. (1). (P) Presenting Author.

The Plutonium rapid response in vitro bioassay method was designed to determine Pu-238, Pu-239, and Pu-240 of urine samples from workers with potential internal contamination. Results provide confirmation of the contamination and information about its level and nature as quickly as possible to assist making further medical decisions. The radiochemical procedure can be performed within approximately 48 hours, including sample preparation, measurement(s) and data evaluation. Though it is a rapid method, it includes all steps of a traditional radiochemical procedure: overnight sample digestion and equilibration of the isotope dilution tracer, calcium phosphate pre-concentration, and Pu separation using AG MP-1M ion exchange resin. The samples are measured by alpha spectrometry and/or inductively coupled plasma mass spectrometry (ICP-MS). The procedure has some limitations: to be able to accommodate a complex radiochemical procedure in such a short time period, the sample volume is limited to a maximum of 200 mL, and the number of samples analyzed in one batch is 7 samples, 1 procedural blank and 1 quality control (QC) sample. The procedure has been validated using in-house QC samples prepared from urine with and without chelating agent (used in the treatment of patients with internal contamination by radioactive material).

Log 272. **PLUTONIUM MOBILIZATION FROM ESTRUARINE SEDIMENTS**

Balboni E. (1,P), Merino, N. (1), J. Begg (1,2), A.B. Kersting (1), M. Zavarin (1). (1) Lawrence Livermore National Laboratory; (2) AMPHOS; (P) Presenting Author.

Since 1952, authorized liquid radioactive effluents containing a range of actinides and fission products (e.g. Cs, U, Np, Pu and Am) have been discharged from the Sellafield plant, U.K. into the Irish Sea. In the Eastern Irish Sea, the majority of the transuranic activity has settled into an area of sediments (Mudpatch) off the Cumbrian coast. The radionuclides from the Mudpatch have been re-dispersed via particulate transport in fine-grained estuarine and intertidal sediments including the Esk Estuary saltmarsh. In this work we examine the factors affecting the mobility of plutonium (Pu) in redox stratified soils by conducting long term (9 months) remobilization experiments using contaminated Esk estuary sediments under both oxic and anoxic conditions. The experiments were periodically sampled to determine the amount of remobilized Pu and redox indicators and the microbial community composition was also characterized. We show that Pu remobilization is enhanced under anoxic conditions and in the top 10 cm of the soil column. We also show that Pu remobilization is independent of the Pu concentration in the soils. The results of this work provide information on the potential mobilization of Pu in biogeochemically dynamic/transient environments over long timescales. Understanding the behavior of actinides in such conditions remains a significant challenge and our lack of knowledge currently limits our progress in developing predictive conceptual and numerical models of actinide mobility in the environment. Work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344 LLNL-ABS-830061

Log 274. **TAIL HAIR AS A BIOMONITOR OF NUTRITIONAL STATUS OF BEEF CATTLE**

Moreira, G.R. (1); De Nadai Fernandes, E.A. (1); Mazola, Y.T. (1); Bacchi, M.A. (1); Sarriés, G.A. (2); Gonzaga, C.L. (1,P). (1) Nuclear Energy Center for Agriculture, University of São Paulo. (2) College of Agriculture Luiz de Queiroz, University of São Paulo. (P) Presenting Author.

Brazil consolidated its position in the international market as the second largest producer and the largest exporter of beef. Since beef quality is directly related to its mineral composition, the assessment of nutritional status of the animal in the rearing and finishing phases allows adjusting the mineral content in its body. The mechanism commonly used for this measurement is the blood test, which only reflects the conditions at the time of collection. Hairs have the advantage of accumulating chemical elements throughout the animal's life, providing a temporal record, in addition to non-invasive collection and easy storage. To verify whether the mineral composition of the tail hairs reflects the mineral composition of the beef, high-performance animals from Angus and Wagyu breeds were selected, originated from complete cycle system that ensures traceability of the productive chain. Tail hair and rump cap samples were analyzed by neutron activation analysis (NAA) and triple quadrupole inductively coupled plasma mass spectrometry (TQ-ICP-MS). From the 24 chemical elements determined, 13 are considered essential for bovine nutrition (Ca, Co, Cr, Cu, Fe, K, Mg, Mn, Mo, Na, P, Se and Zn). With the exception of Co and Na, all essential elements showed correlations between mass fractions in tail hair and in rump cap, indicating that tail hair is a potential non-invasive biomonitor of the nutritional status of beef cattle.

Log 275. **A DATA-DRIVEN APPROACH TO EVALUATION OF SUSTAINABLE USE OF AGRICULTURAL BYPRODUCTS FOR DIETARY SUPPLEMENTS**

Furlan, G.N. (1); De Nadai Fernandes, E.A. (1,P); Bacchi, M.A. (1); Sarriés, S.R.V (1); Sarriés, G.A. (2); Lima, R.C. (1). (1) Nuclear Energy Center for Agriculture, University of São Paulo. (2) College of Agriculture Luiz de Queiroz, University of São Paulo. (P) Presenting Author.

This study has the premise of working in alignment with the Zero Hunger Goal from the Sustainable Development Goals of United Nations, which aims to curb undernourishment around the world, by presenting the potential value of dietary supplements made of agriculture byproducts in Brazil. The focus is towards evaluating different types of dietary supplements commonly used in several regions, that could be a promising alternative to help fighting poor nutrition of children, women and elderly in vulnerable communities. A comprehensive sampling of alternative supplements and their ingredients, and commercial products was performed. Neutron activation analysis was used to determine the chemical profiles of the samples, which were evaluated by parametric, non-parametric and multivariate analysis, combined with supervised and unsupervised machine learning methods, to discriminate categories of products and ingredients. A bootstrapping technique was also applied to normalize distribution and perform confirmatory analysis. The results have evidenced that supplements made from agriculture byproducts are statistically different (p-value <0.0001) from similar conventional products regularly commercialized in the market. Five distinct machine learning algorithms (Multilayer Perceptron, Random Tree, Random Forest, Support Vector Machine, CART) provided over 80% accuracy on the classification models.The trace elements with the greatest impact on the agriculture byproducts classification was cesium, rubidium and zinc, while the industrialized type was better defined by its content of bromine, iron and sodium.

Log 276. **PET FOOD CATEGORIZATION BY NEUTRON ACTIVATION ANALYSIS AND DATA SCIENCE**

Lima, R.C. (1); De Nadai Fernandes, E.A.(1,P); Mazola, Y.T.(1); Bacchi, M.A.(1); Sarriés, G.A.(2); Furlan, G.N.(1). (1) Nuclear Energy Center for Agriculture, University of São Paulo. (2) College of Agriculture Luiz de Queiroz, University of São Paulo. (P) Presenting Author.

Recent studies have shown how pets bring benefits to human psychological and physical health, especially for children and the elderly. The United States, with a population of hundreds of millions of animals, is the country that spends the most on pets in the world. The pet care market is worth hundreds of billions of dollars a year, most of it for food. The importance of this market is also growing in Brazil, even in years of economic crisis. Pet food manufacturers often offer products classified in market categories such as Standard, Premium and Super Premium, however there are no clear and objective criteria to define their characteristics. A representative sampling of the commercial pet food available in the Brazilian market was carried out. The samples were analyzed by neutron activation analysis for determining chemical elements. Here, it was investigated whether the chemical composition associated with statistical tests and machine learning models can discriminate categories, brands and ingredients, thus understanding whether the categorization made by the manufacturers reflects the quality of the product. The results indicate that only the Standard category was significantly different from the other two categories. Cluster analysis separated the brands into similarity groups. The proportion of ingredients of plant and animal origin in the pet foods was not a preponderant factor for discrimination.

Log 277. **IN VITRO LUNG DISSOLUTION RATE AND PARTICLE SIZE DETERMINATION FOR PLUTONIUM-238 OXIDE INVOLVED IN AN INCIDENT AT THE LOS ALAMOS NATIONAL LABORATORY**

Macsik, Z. (1,P); LaMont, S.P. (1); Hudston, L.A. (1); Harris, M.N. (1); Tenner, T.J. (1); Naes, B.E. (1); Wurth, K.N. (1); Steiner, R.E. (1). (1) Los Alamos National Laboratory. (P) Presenting Author.

In mid-2020, an incident occurred at Los Alamos National Laboratory (LANL) resulting in the release of plutonium-238 and subsequent contamination of several employees. As part of the response to this incident, a study was conducted to determine the lung dissolution rate and particle size distribution of the airborne material. These material characteristics were used by LANL's internal dosimetrists to refine dose estimates for exposed workers. Results of the 100-day in vitro dissolution rate study show that the PuO_2 collected on air filters during the incident has behavior comparable to material previously studied, with approximately 1% of material dissolving quickly, and 99% of the material dissolving very slowly, with a half-time of approximately 120 years. Particle size distribution measurements indicate that particle sizes range from 0.10 – 6 microns, with most less than 2 microns. The experience and the lessons learned from this study will be used to establish to perform radionuclide lung dissolution rate measurements on a routine basis.

Log 278. **EVALUATION OF ALPHA INDUCED DAMGE BY ELECTRODEPOSITED Am-241 ON SIC SCHOTTKY DIODE AND SIMULATION WITH ALLPIX2**

Xue S. (1); Giglio. D. (2); Spitz, H. (3); Hoffman K. (2); Kandlakunta, P. (1); Vasil Hlinka (2) Cao, L.R. (1)

SiC devices show a great potential for high radiation field applications in industry and nuclear fuel cycle. The challenge is exemplified by the application of SiC in alpha voltaic batteries where the constant bombardment of alpha particles to the semiconductor energy transducer (e.g., Si p-n junction) could cause severe degradations and eventually device failure. A rad-hard wide bandgap semiconductor such as 4H-SiC could potentially alleviate this problem. The objective of this study is to evaluate the longevity of an alpha-voltaic battery based on 4H-SiC Schottky diode, which was fabricated by depositing Pt on 6 mm by 7 mm 4H-SiC (21 mm epitaxial layer) to form Schottky contact and Ni/Pt to form Ohmic contact.

The alpha emitting actinides such as americium-241 are electrodeposited on the surface of the Pt electrode of SiC Schottky diodes. With 17 nCi americium-241 directly coated on the top of a device, the 2-pi geometry could yield a high energy (5.486 MeV) and high fluence (up to 10^10 total particles for 6 months irradiation) of alpha particles' infusion. The forward, reverse I/V, dark current, are periodically measured to monitor key characteristics of device durability for any sign of degradation over time. Alpha spectrum of the electrodeposited actinides on the device is also monitored on a consistent timed count. The direct deposition of actinides on the surface of the device also enables the direct measurement of alpha particles energy spectrum in a 2-pi geometry. The device performance is simulated by Allpix 2. These measurements provide an insight into the high durability of 4H-SiC as an energy converter option for alpha-voltaic battery and for other nuclear fuel cycle monitoring applications.

Log 279. **CRITICAL EVALUATION OF THE NEW NEOMA MC-ICP-MS FOR ACTINIDE ISOTOPIC MEASUREMENTS.**

Hexel, C.R. (1,P); Dunlap, D.R. (1); Zirakparvar, N.A.(1); Manard, B. T.(1,). (1) Oak Ridge National Laboratory. (P) Presenting Author.

As laboratories adopt new product offerings to advance analytical capability, the new platforms n should be critically evaluated to determine if the data produced is equivalent, or better, than the previous system(s). In addition, it is important to re-validate community agreed-upon analytical norms such as mass-bias correction schemes and broader analytical approaches. Here we present the first actinide ratio measurement using the ThermoFisher Scientific Neoma MC-ICP-MS. The new MC-ICP-MS platform was experimentally tested to examine adherence to mass bias laws, hydride formation, ion counter stability, detector linearity, long term abundance sensitivity, ratio accuracy and precision using the JRC-Geel 2000 and 3000 series standards. Also, major, and minor ratio measurement uncertainty will be discussed in comparison to historical data collected using a ThermoFisher Scientific Neptune Plus MC-ICP-MS. Lastly, we will highlight how the new platform's features have affected existing workflows with regards to sample throughput and analytical flexibility.

Log 280. **SIMULATED GENERATION OF RADIOACTIVE MICROPARTICLES RELEASED BY THE FUKUSHIMA NUCLEAR ACCIDENT**

Inagaki, M.(1,P); Sekimoto, S.(1); Takamiya, K.(1); Oki, Y.(1); Ohtsuki, T.(1). (1) Kyoto University. (P) Presenting Author.

Water-insoluble microparticles called radiocesium-bearing microparticles (CsMPs) are known to be one of the forms of radioactive materials released by the accident at the Fukushima Dai-ichi Nuclear Power Plant. In this study, simulated CsMPs were generated in the laboratory to investigate the formation process. Referring to the composition of CsMPs reported in a previous study, we generated a glass that reproduced the composition of CsMPs. The glass was placed in a chamber and irradiated with an infrared laser, and the fume containing fine particles generated by the heating was collected by an impactor. As a result of analyzing the collected particles, it was found that spherical particles with a diameter of several micrometers, similar to actual CsMPs, were successfully generated. One hypothesis for the formation process of CsMPs is that the silica-rich materials were heated rapidly by contact with the high-temperature molten fuel, resulting in the formation of fumes containing CsMPs.

Log 281. **AUTOMATED FAST NEUTRON TOMOGRAPHY FOR COMPLEX OBJECTS AT A 500 KW RESEARCH REACTOR**

Oksuz, I.(1, P); Bisbee, B.(1, 3); Cherepy, N.(2); Hall, J.(2); Cao, L.(1). (1) The Ohio State University. (2) Lawrence Livermore National Laboratory. (3) DOE Nuclear Energy University Program Fellow. (P) Presenting Author.

Fast neutron computed tomography (nCT) allows examining of bulky materials rich in low Z-number elements, produces less activations, and provides more information compared to a single radiograph. Therefore, acquiring nCT data with sufficient quality in a reasonable time is desirable. In this study, fast nCT was performed using two imaging phantoms designed and fabricated by Lawrence Livermore National Laboratory at the Ohio State University Research Reactor's fast neutron beam facility (ϕ=~5.4E7 n·cm-2·s-1 @ 1.6 MeV (mean), beam diameter: ~3.2 cm, divergence: ~0.6°), employing an automated imaging station consisting of an Electron-Multiplying Charge-Coupled Device, a light-tight enclosure, a High-Light yield scintillator, a rotation stage, and an XYZ stage. The first phantom consists of a stainless-steel truss cylinder (H x D: 2.2 cm x 2.2 cm) and HDPE inserts, including a cylinder with horizontal through holes (D: 0.5, 1, 2, and 3 mm), a vertical rectangular through hole, and a solid rectangle. The second phantom, named MAS-1, includes a Lead-Antimony spherical shell (D: ~2.2 cm) with a grooved structure on the internal surface and a cylindrical HDPE insert with horizontal through holes (D: 0.5, 1, 2, and 3 mm). A total of 360 projections per each configuration were collected of the truss cylinder and MAS-1 with the nCT acquisition time of 3 and 6 hours, respectively. 3D reconstructions clearly revealed the internal structure of the truss cylinder, the grooved features of MAS-1, and the 3-, 2-, and 1-mm holes in both cylindrical HDPE inserts shielded by high-Z materials.

Log 282. **METHYLAMMONIUM LEAD IODIDE PEROVSKITE SOLAR CELLS FOR POST-DETONATION MONITORING AND HIGH-ENERGY PHOTON IRRADIATION RESPONSE**

Wyatt Panaccione 1, Praneeth Kandlakunta 1, Matthew Van Zile 2, Marlin Keller 1, Dai Xuezeng 3, Jinsong Huang 3, and Lei Raymond Cao 1,2

The metal halide perovskite solar cells, such as Methylammonium Lead Iodide (MAPbI3), have the potential to be used in post-detonation monitoring via prompt gamma photon detection, as an ubiquitous urban infrastructure. Utilizing the Cesium-137 (137Cs) and Cobalt-60 (60Co) sources at the Ohio State University's Nuclear Reactor Laboratory, we were able to measure the electrical transient and characteristic responses during gamma irradiation. The X-ray response was conducted via a benchtop X-ray tube with energy ranges from 10-50 keV. The current response and IV curve were measured during irradiation to measure key electrical characteristics of solar cells such as the Short-Circuit current (I¬SC¬ ¬) and Open-Circuit voltage (VOC), which can elucidate the state of performance of the solar cell. After irradiation distinct degradation of the shunt and series resistance of the cells cause a dramatic drop in the power output of the cell. For detonation detection and post detonation forensics analysis, the rate of degradation in different performance parameters is correlated with radiation dose. The reason for degradation is investigated through Monte-Carlo simulations to analyze the total energy absorption vs photoelectric interactions with the different photon energies investigated from the gamma and X-ray sources. While there are a multitude of theories surrounding the degradation mechanisms of the solar cells, like PbI3 generation, we discuss their applicability to high energy photon induced degradation in hopes for utilization of perovskite solar cells for prompt and post detonation detection and analysis.

Log 283. **EXERCISE CELESTIAL SKÓNIS: A READOUT FROM THE SIXTH AND LARGEST MATERIALS EXERCISE OF THE NUCLEAR FORENSICS INTERNATIONAL TECHNICAL WORKING GROUP (ITWG)**

Schwantes, J.M. (1,2,P); Corbey, J.F.(1); Marsden, O.(2,3); (1) Pacific Northwest National Laboratory. (2) Exercise Task Group Co-Chair, Nuclear Forensics International Technical Working Group. (3) AWE. (P) Presenting Author.

The ITWG conducted its sixth and largest Collaborative Materials Exercise, Celestial Skónis, in which laboratories from 20 countries and the European Commission, working with 15 law enforcement agencies, analyzed a set of samples containing special nuclear material as part of an elaborate mock nuclear forensics investigation. The nuclear materials used in the exercise included (1) depleted uranium and (2) stable cerium metal ingots contaminated on their surfaces with trace amounts of depleted uranium- and weapons grade plutonium-oxy-fluoride powders. This exercise also included traditional evidence (e.g., cut surfaces of pipes, toolmarks on plastic bags, and patent and latent fingerprints) comingled/contaminated with radioactivity. Participating laboratories processed traditional forensic evidence contaminated with radioactivity and analyzed nuclear materials in support of the mock investigation for the purpose of connecting people, places, things, and events over a two-month period. In all, a total of 36 nuclear forensic protocols, procedures, and analytical techniques were carried out by laboratories, the results of which were submitted in preliminary reports to the exercise facilitator after 24 hours and 1 week, with a final report due 2 months after receiving materials. These

results were used to evaluate the "State of Practice," defined as any analyses carried out by half or more of the participating laboratories within a particular reporting timeframe (i.e., 24 hours, 1 week, and 2 months) and identify promising emerging technologies in nuclear forensics.

Log 284. SEQUENTIAL LEACHING OF SINGLE "HOT" PARTICLES FROM THE CHERNOBYL EXCLUSION ZONE

Leifermann, L. (1)(P); Weiß, M. (1); Weissenborn, T. (1); Hanemann, P. (1); Raiwa, M. (1); Fraatz, P. (1); Schulz, W. (1); Walther, C. (1). (1) University of Hannover. (P) Presenting Author.

During the Chernobyl reactor accident on April 26, 1986, radioactivity was in part released in the form of nuclear fuel particles. These so-called "hot particles" have different structures and oxidation states, which behave differently in the environment. To study these particles individually, they are separated by flotation with a poly tungsten solution, located by radiometric scanning with a Geiger counter and extracted with a tungsten needle on a micromanipulator in a scanning electron microscope (SEM). Following various non-invasive analytical methods, including single particle gamma spectrometry, EDX-measurements, SIMS and SNMS-measurements, sequential leaching was performed on five particles. Each particle is sequentially leached in: ammonium acetate solution, hydrochloric acid, oxalic acid, nitric acid, and aqua regia. This method is based on Kashparov et al. (Kashparov V. 2019), performed on single particles rather than heterogeneous soil samples. The residual solutions were analyzed for fission products and breeding products by gamma spectrometry and ICP-MS. The oxidation state of the particles is confirmed to be consistent with the initially assumed state based on the SEM images. The initial steps of sequential leaching do not structurally attack the particles, however Cs-137 and other fission and breeder products can be separated in small amounts. The bioavailability of the particles is therefore estimated to be rather low.

Log 285. HIGH-PRECISION MEASUREMENT OF U-Pu-Np-Am-Li CONCENTRATIONS AND ISOTOPE RATIOS IN ENVIRONMENTAL REFERENCE MATERIALS BY MASS SPECTROMETRY

Goldstein, S.J. (1,P); Price, A.A. (1); Hinrichs, K.A. (1); LaMont, S.P. (1); Nunn, A.J. (1); Amato, R.S. (1); Cardon, A.M. (1); Gurganus, D.W. (1). (1) Nuclear and Radiochemistry Group, Los Alamos National Laboratory. (P) Presenting Author.

We report results of precise and sensitive mass spectrometric measurements of uranium, plutonium, neptunium, americium, and lithium concentrations and isotope ratios in a variety of environmental reference materials. Measurements were performed in a clean laboratory by isotope dilution, multi-collector thermal ionization and inductively coupled plasma mass spectrometry. In general, our results are in agreement with, but lower the uncertainty of, literature or certificate values for these materials. Our results for U in basalts also confirm previously reported data from our laboratory. In many cases our measurements of U-Pu-Np-Am-Li nuclides appear to be novel. NIST-4350b Columbia River sediment, and IAEA-385 Irish Sea sediment, have 241Pu-241Am model ages of 1954 and 1964, respectively, reflecting a mix of Pu operations at the nearby Hanford, WA and Sellafield, UK sites, and global atmospheric fallout from nuclear weapons testing. Large environmental fractionation between Pu and Np is observed for ocean, river, and lake sediment materials. Results for BHVO-2 and other Hawaiian basalts

indicate that those which erupted before or during the period of abundant atmospheric nuclear weapons testing (1950-1970) contain significant levels of Pu (~1E7 atoms 239Pu/g) with a global fallout Pu isotopic composition, compared to more recent eruptions which incorporated less Pu. Hence, Hawaiian basalts, and other ocean island basalts by inference, may provide an integrated temporal record of anthropogenic actinide fallout deposition from the atmosphere since eruption.

Log 286. **FEEDSTOCK-PROCESSING-STRUCTURE CORRELATIONS FOR TAGGED UO$_2$ TO SUPPORT INTENTIONAL FORENSIC TECHNOLOGIES**

Ulrich, T.L. (1,P); Kercher, A.K. (1); Wilson, B.A. (1); Sadergaski, L.R. (1); Spano, T.L. (1); Shields, A.E. (1); Nelson, A.T. (1); Scott, S.M. (2); Wellons, M.S. (2). (1) Oak Ridge National Laboratory (2) Savannah River National Laboratory. (P) Presenting Author.

Intentional nuclear forensics involves deliberately incorporating persistent elements ('taggants') into nuclear fuels to create unique specimens that are distinguishable from the parent material. Taggants of interest must be benign with respect to fuel performance, therefore, the impact on the microstructure and other material properties must be carefully evaluated. A successful taggant of a nuclear fuel must: (1) survive the ceramic processing conditions used for producing nuclear fuel forms, (2) yield a microstructure that preserves key nuclear fuel properties such as thermal conductivity and fracture behavior, and (3) maintain acceptable fuel performance throughout steady state and transient reactor conditions. This work aims to elucidate the interplay between taggant quantities and processing environments (sintering temperatures and times), on the properties of unirradiated UO2 nuclear fuel. Ni-, Ti-, Fe-, and Mo-tagged UO2 pellets were synthesized through reference laboratory scale ceramic processing methods using multiple variants of taggant feedstocks introduced at levels from 250-3000 weight parts per million. The impact of taggant chemistry and concentration on the resulting microstructure and baseline unirradiated properties will be discussed with an emphasis placed on quantification of taggant level throughout processing and high temperature sintering. These datasets will then be contextualized from the standpoint of nuclear fuel performance and an upcoming irradiation of these candidate tagged UO2 fuels.

Log 288. **HIGH SELECTIVE RADIOCHEMISTRY OF CAESIUM FOR THE DETERMINATION OF THE ^{135}Cs/^{137}Cs RATIO IN LARGE SOIL AND SEDIMENT SAMPLES**

Magré, A. (1,P); Boulet, B. (1); Pourcelot, L. (2); Roy-Barman, M. (3); Ardois, C. (1). (1) Laboratoire de métrologie de la radioactivité dans l'environnement (PSE-ENV/SAME/LMRE) (2) Laboratore d'étude et d'expertise sur la radioactivité de l'environnement (PSE-ENV/SEREN/LEREN). (3) Laboratoire des sciences du climat et de l'environnement (LSCE-IPSL). (P) Presenting Author.

The isotopic signature of radioelements is a tool for characterizing different sources of radioactive contamination in the environment. Thus, caesium isotopes were widely studied by measuring the 135Cs/137Cs isotopic ratio by mass spectrometry in highly contaminated environmental samples. In contrast, few data are available at the environmental level (< 1,000 Bq.kg-1 of 137Cs) due to the occurrence of interferents in the mass range of interests and very few radiocaesium atoms. This low radiocaesium content implies the need to analyse a very large sample. The current work aims at developing and validating a highly selective radiochemical protocol for Cs from a 100 g of ash sample to achieve the low levels of radioactivity found in soils and sediments. A first work of adaptation of the digestion to the very large sample size was carried out allowing a total digestion of radiocaesium. A Cs preconcentration step was then carried out with a method involving the ammonium molybdophosphate molecule. Once extracted from the sample matrix, caesium was then separated from its interferents by a sequence of ion exchange resins prior to mass spectrometry measurement. The chemical performances of the developed protocol are very satisfactory: the 137Cs recovery yield for 100 g of sediment, measured by gamma spectrometry, is about 80% and the decontamination factors of interferents allowing their contribution to the masses of interest to be negligible. Finally, the measurement of the purified fraction is performed by ICP-MS/MS under optimized reaction gas conditions to obtain the best signal/noise ratio.

Log 289. **SKAPHIA: PRESENTATION OF THE LATEST SHIELDED ELECTRON PROBE MICRO ANALYSER (EPMA)**

Robbes, A.S (1); Henderson, C (2); Choi, S (1); Peres, P (1); Defouilloy, C (1); Vuillaume, A (1,P); Jacobson, D (2). (1) CAMECA Gennevilliers, France. (2) CAMECA Inc., USA (P) Presenting Author.

Electron Probe Micro Analysis (EPMA) is a technique used for material analysis, allowing quantitative mapping of nearly all chemical elements at concentration levels down to few 10s ppm with a spatial resolution of about 1 um. At nuclear facilities, EPMA is mainly being used for nuclear fuel characterization and fission products, irradiated materials behavior investigation, Post Irradiation Examination, and radioactive waste management. CAMECA has been developing dedicated EPMA instrumentation for radioactive samples for more than 40 years. We will show the global conception of SKAPHIA with the loading of the hot samples, the safety capacities as well as the maintenance. Technical details to maintain the EPMA efficiency for detection limits of trace elements in the shielded environment will be reviewed. The instrument will be shown at our production facility and also integrated in various possible hotcell configurations. Finally, we'll present different type of applications served by instruments in operation worldwide.

Log 290. **TEACHING RADIOCHEMISTRY SIMULTANEOUSLY TO NUCLEAR ENGINEERS AND CHEMISTS**

Nathalie A. Wall, Donald E. Wall

The University of Florida Nuclear Engineering program has undergone a renaissance in the last 10 years, intending to educate the engineers of tomorrow. The program now includes 12 full-time core faculty, new facilities, and an updated curriculum. While the educational program and areas of research include the traditional components necessary for an ABET-certified BS and graduate degrees in nuclear engineering, it also incorporates courses and research in related fields, such as radiochemistry. However, the approaches to the teaching of radiochemistry to nuclear engineers and chemistry major students are vastly different. Even though nuclear engineering students possess an acute knowledge of decay schemes, radioanalytical instrumentation, and mathematics, they need to learn the fundamental chemistry background to understand radiochemistry processes. Meanwhile, the course is also open to chemistry major students, who need to acquire the basic knowledge of nuclear sciences. We will discuss how a radiochemistry course is adapted to deliver a cohesive education to seemingly disparate groups of students.

Log 291. **STUDIES IN DUAL DOPING OF URANIUM NITRIDE FOR USE AS ADVANCE TECHNOLOGY FUELS**

Gonzalez L.G. (1,P); Axhage E.(1); Hedberg M.(1); Retegan T.(1). (1) Chalmers Univeristy of Technology. (P) Presenting Author.

Uranium Nitride (UN) is a potential candidate for the advanced technology fuels (ATF) concept, as it has numerous benefits compared to conventional uranium oxide (UO_2) used today. Namely, higher thermal conductivity and uranium density. UN main drawback is its poor corrosion resistance in oxidizing environments. Doping of UN with oxide scale forming elements such as chromium could improve the resistance towards oxidation. In this work, the addition of chromium and aluminum in the UN matrix was accomplished using a sol-gel method to obtain a homogenous mixture of the metals. The effect caused by the introduction of these dopants in the properties of the UN material was studied. It was observed that addition of aluminum caused a change in the microstructure of the spheres. These changes are suspected to be responsible for the decrease in the reaction temperatures needed to oxidize the UN with air during the thermogravimetric analysis. Chromium content appeared to decrease during the synthesis of UN. Nonetheless, oxidation temperatures were increased only when chromium alone was added as dopant.

Log 292. **A CONTROLLED URANIUM METAL CASTING EXPERIMENT TO UNDERSTAND RADIOCHRONOMETRY SIGNATURES**

Kayzar-Boggs, T.M. (1, P); Luitjohan, K.E. (1); Imhoff, S.D. (1); LaMont, S.P. (1); Boswell, M. (1); James, M.R. (1); Hudston, L.A. (1); Denton, J.S. (1); Edwards, M.E. (1); Krajewski K.J. (1); Engel, J.R. (1); Wende, A.M. (1); Sanborn, M.E. (1). (1) Los Alamos National Laboratory. (P) Presenting Author.

Radiochronometry constrains the age of nuclear material by measuring the radiogenic ingrowth of progeny radionuclides (e.g., 230Th and 231Pa) from the decay of parent nuclides (234U and 235U). Age constraints are recognized as important forensic signatures and radiochronometry is routinely applied during nuclear forensic examinations. Ideally, uranium (U) production completely removes ingrown 230Th and 231Pa from the parent U and measured model ages accurately record the time elapsed since the U was chemically purified. However, during metal casting, U model ages indicate that 231Pa is separated from U less efficiently than 230Th, resulting in discordant 230Th/234U and 231Pa/235U model ages. Our ability to interpret discordant model ages is challenged by a fundamental lack of understanding of 230Th and 231Pa behavior during metal casting. To address this knowledge gap, we conducted a controlled vacuum induction melting cast of a 120 kg depleted U metal rod to investigate potential 230Th and 231Pa migration to the hot top, cast U surfaces, or mold surfaces during casting. We present our experimental design including casting parameters, autoradiography and gamma spectrometry used to characterize the cast metal, radiochemistry and mass spectrometry methods characterizing 230Th and 231Pa concentrations in the feedstock charge prior to casting, and sampling techniques for destructive analysis to constrain 230Th and 231Pa spatial migration within the cast U product. We also present a radiochemistry method developed to separate 231Pa from very young gram-scale depleted U samples.

Log 293. **Incorporating density functional theory modeling to predict the chemical behavior of high-yielding fission products (Cs-137, I-129) in a UO$_2$ framework in conjunction with experimental validity**

Montoya, E. (P)(1); Kim, E. (2); Czerwinski, K. (1); Lowe, R. (1); Swift, A. (1). (1) University of Nevada, Las Vegas (2) University of Texas at El Paso. (P) Presenting Author.

Predicting the chemical arrangement of molecular or crystal structures grants researchers a unique insight into the chemical behavior of many atomic frameworks. Those that have been studied extensively use empirical findings as a form of validation, improving the methodology for close accuracy and efficiency. These improvements are important for work on atomic systems that have yet to have been investigated experimentally due to legitimate safety concerns associated with radioisotopes. Density functional theory (DFT) is a tool used to simulate the ground state of an atomic system and is utilized in this research. By simulating bulk spent oxide fuel (UO2) and incorporating high-yielding fission products (FP) in the cell framework, we can observe the behavior that SNF may exhibit under natural or synthetic conditions. There is yet no experimental data that can be used to validate the experimental results, so this research aims at providing it as well. The validity of both theory and experiment is confirmed using various characterization methods, such as x-ray diffraction (XRD). DFT aims at solving for the Schrödinger

equation to reach a ground state in the [UO2] cell-matrix where the FP [Cs, I2, CsI] is introduced to the cell as a defect. As there has been extensive DFT research on UO2, we use multiple parameters for the DFT calculations due to the unique nature uranium exhibits as a Mott-insulator.

Log 294. INVESTIGATING O$_2$ CERAMIC SENSORS IN MOLTEN CaCl$_2$

Dale, O.R. (1, P); Felling, F. M. (1); Simpson, M. (1). (1) University of Utah - Materials Science Department. (P) Presenting Author.

Pyrochemical processes, including electrorefining and electroreduction, have an instrumental role to play in recycling advanced nuclear reactor fuels such as U metal fuel and U/TRU containing molten salts. These processes use molten salts such as LiCl-KCl or CaCl2 as the electrolyte and thus run at high temperatures in a corrosive environment. Real time measurement of O2-concentration in these salts can be beneficial for process monitoring. Solid electrolytes can operate as potentiometric sensors in these conditions and remain stable while collecting precise O2- concentration data in real-time. A sensor that could respond to a single element, such as oxygen, would be advantageous to a molten-salt electrorefiner or electrolytic reduction system and improve process performance/efficiency. Open circuit potential versus reference electrodes featuring different ceramics as membranes, such as mullite, magnesia, and Na-beta"- alumina, will be reported in experiments in molten CaCl2 salt, with varying CaO concentration. Results for electrochemical impedance spectroscopy (EIS) and surface analysis will also reported to investigate the ionic conductivity across these membranes.

Log 295. GRANDDAUGHTER RADIOCHRONOMETRY FOR NUCLEAR FORENSICS: MODEL AGES FOR CERTIFIED REFERENCE MATERIALS AND URANIUM SAMPLES

Denton, J.S. (1,P); Wende, A.M. (1); Edwards, M.A. (1), Sanborn, M.E. (1), Kayzar-Boggs, T.M. (1); Steiner, R.E. (1) (1) Los Alamos National Laboratory. (P) Presenting Author.

A key parameter to be determined in a nuclear forensics investigation is the age of the interdicted material. This can be accomplished through radiochronometry. To date, this has been commonly achieved using the parent-daughter radiochronometers (e.g. 230Th/234U and 231Pa/235U). Here we present research at Los Alamos National Laboratory to develop a parent-granddaughter (226Ra/234U and 227Ac/235U) age-dating capability by isotope dilution mass spectrometry. Calculating four model ages (compared to two) can increase confidence in the age if all ages are concordant. If the ages are non-concordant, then information about the behavior of radionuclides during the fuel cycle can be obtained. We discuss: (i) spike preparation, (ii) chemical purification procedure, and (iii) mass spectrometry methods. In addition, we report model ages for a suite of certified reference materials as well as selected uranium samples of various enrichments and chemical forms.

Log 296. MULTI-ANALYTICAL APPROACH TO PLUTONIUM ISOTOPIC STANDARD CERTIFICATION
Hilton, C.D. (1,P); Engelmann, M.D. (1); Anderson, K.K. (1); Darnell, L.P. (1); Greenwood, L.R. (1); Krogstad, E.J. (1); Soderquist, C.Z. (1). (1) The Pacific Northwest National Laboratory. (P) Presenting Author.

Properly certified isotopic standards are key to supporting routine nuclear safeguards measurements and related research and development. Total evaporation (TE) by thermal ionization mass spectrometry (TIMS) is a popular technique for isotopic certification due to the minimization of fractionation effects. However, this technique poses challenges for standards that contain a range of isotopic abundances. For example, the New Brunswick Laboratory's (NBL) C136, C137, and C138 suite of Pu standards have 238Pu isotopic abundances that range from 0.010 % to 0.268 %. Since these standards all have abundances of the neighboring 239Pu isotope of >77 %, the 238Pu is subject to a variety of peak-tailing effects, complicating TE measurements by TIMS. In this work, we explore the potential benefit of coupling multiple analytical techniques to measure the suite of Pu standards. These techniques include TE using a Thermo Scientific Triton Plus TIMS equipped with 1011 and 1012 Ω resistors, peak-hopping to correct for peak-tailing by a Thermo Scientific Neptune Plus multi-collector inductively coupled plasma mass spectrometer (MC-ICP-MS) equipped with 1011 and 1013 Ω resistors, and measurement of 238Pu/(239Pu+240Pu) by alpha spectroscopy.

Log 297. COMPARISON OF IMAGE ANALYSIS TOOLS DEVELOPED FOR NUCLEAR FORENSIC EXAMINATIONS
Middlemas, M. (1, P); Kips, R. (2); Kimura, Y. (3); Ross, A. (1), Yamaguchi, T. (3); Matsumoto, T. (3). (1) Los Alamos National Laboratory. (2) Lawrence Livermore National Laboratory. (3) Japan Atomic Energy Agency. (P) Presenting Author.

Image analysis is an important tool in nuclear forensic examinations because morphological parameters can give insights to the origin and process history of a material. When sharing of data between different agencies and institutions, it is important to ensure consistency between sample preparation methods, image analysis protocols, and output of the software packages used. Under an agreement between the Japan Atomic Energy Agency's (JAEA) Integrated Support Center for Nuclear non-proliferation and Nuclear Security (ISCN) and the U.S. Department of Energy's (DOE) Office of Nuclear Smuggling Detection and Deterrence (NSDD), a benchmarking study was conducted to evaluate the ability to acquire similar results between image analysis tools developed by each country. Using the same standard reference powder (SRM-1984), samples were imaged using scanning electron microscopy (SEM), and analyzed at the ISCN, Los Alamos National Laboratory (LANL), and Lawrence Livermore National Laboratory (LLNL). For given image sets, similar distributions of the morphological parameters (area, circumference, aspect ratio, and diameter) were achieved, demonstrating consistent output between the JAEA-developed image analysis tool and the Morphological Analysis for Material Attribution (MAMA) software used by the U.S. National Laboratories. The results also identify sources of variation between analyses, highlighting the importance of establishing common sample preparation and image analysis protocols.

Log 298. PARTICLE ANALYSIS IN NUCLEAR FORENSIC INVESTIGATIONS: THE SIGNIFICANCE OF SOURCE COMPOSITION AND SAMPLE SIZE.

Inglis, J.D. (1,P); Reinhard, A.A. (1); Tenner, T.J. (1); LaMont, S.P. (1); Steiner, R.E. (1); Wende, A.W. (1); Fisher, W.S (1). (1) Chemistry Division, Nuclear and Radiochemistry Group, Los Alamos National Laboratory, Los Alamos. (P) Presenting Author.

Many processes in the nuclear fuel cycle such as enrichment, reprocessing and reactor operation result in the release of radioactive particles. Measuring the isotopic composition of such particles can provide useful data on the history of its parent, as a particle's isotopic signature reflects the history of enrichment and irradiation it has witnessed. Although some parent materials have a complex composition, analysts are often tasked with making inferences about the parent from a limited number of particles. How can we be certain that small particle datasets adequately represents the compositional variation within the parent? To examine this, we created a synthetic dataset intended to emulate a large U particle population derived from a parent with a variable isotope composition. We used this parent population to assess the effect of sample size by repeatedly modeling a series of analyses between 2-200 particles randomly drawn from the larger population. Comparison of the compositional distribution between the modeled results and the parent allows us to estimate the number of particles required to have a reasonable probability of accurately detecting compositional variation in the parent. We demonstrate that the likelihood of missing a 1:10 fraction of the parent composition to be >10% for datasets of n=20 particles, and to grow considerably larger when n<20. While it is understood that analyzing large numbers of particles is often not possible, our data underlines the importance of understanding statistical adequacy when interpreting particle datasets. LA-UR-21-32102

Log 299. REFINING THE ISOLATION AND PURIFICATION OF PROTACTINIUM FROM URANIUM-NIOBIUM ALLOYS FOR 231PA-235U RADIOCHRONOMETRY FOR NUCLEAR FORENSICS

Chen, C.Y. (1, P); Kayzar-Boggs, T.M (2); Higginson, M. (3); Denton, J.S. (2); Dunne, J. (3); Edwards, M.A. (2); Eng, C. (1); Engel, J. (2); Gaffney, A.M. (1); Gilligan, C. (3); Morris, M.N. (1, 4); Rolison, J.M. (1); Sanborn, M.E. (2); Wende, A.M. (2). (1) Lawrence Livermore National Laboratory. (2) Los Alamos National Laboratory. (3) Atomic Weapons Establishment. (4) University of California, Merced. (P) Presenting Author.

The isolation and purification of actinide elements is central to radiochronometric age dating of nuclear materials for forensics investigations. In recent years, separation techniques for U decay products have been optimized for pure bulk U materials and successfully applied in nuclear forensics labs worldwide. However, U may be alloyed with other elements (e.g., Ti, Zr, Nb) for fuel cycle applications. The presence of percent-level Nb renders standard methods for ultratrace-level Pa purification from U ineffective due to the chemical similarity of Pa and Nb, posing a unique analytical challenge for 231Pa-235U radiochronometry. Here we present three viable strategies for the purification of Pa from U-Nb alloys. Method development supported a recent radiochronometry interlaboratory comparison by LLNL, LANL, and AWE, sponsored by the NNSA Office of Nuclear Forensics. Differences in instrumentation and workflow required that each lab devise a custom approach to Pa-Nb separations. Strategies included using pure HF as a sample loading solution with anion-exchange resin; modifying the concentration of trace HF in

HCl acid for Pa elutions; and utilizing silica gel, TK-400, and ZR resins. Ultimately, all approaches adequately purified Pa to enable 231Pa-235U measurements. Our results illustrate challenges in ad-hoc adaptations of protocols for untested materials and the importance of developing purification methods suitable for a diversity of U and U-alloy materials to ensure nuclear forensics lab preparedness. Prepared by LLNL under Contract DE-AC52-07NA27344, LLNL-ABS-830178.

Log 300. **REAL-TIME IN SITU MONITORING OF COMPONENTS IN A FLOWING MOLTEN SALT LOOP WITH INDUCTIVELY COUPLED PLASMA-MASS SPECTROMETRY**

Pamplin, K.L. (1,P); Babb, E.M. (1); Berry, A.M. (1); Bonamie, E.J. (1); for the NEXT Lab Collaboration. (1) Abilene Christian University (P) Presenting Author.

As humanity seeks to reduce its reliance on carbon-based energy, advanced nuclear power emerges as the safe, clean, sustainable, and reliable alternative to carbon, wind, and solar options. Among advanced nuclear power possibilities, liquid-fueled molten salt reactors (MSRs) offer some intriguing advantages, including the possibility of providing access to valuable medical isotopes in the salt. The long-term operation of MSRs will require operators to control and/or respond to a variety of parameters within the salt, including fuel, fission products, corrosion products, and trace components like oxygen, which plays a critical role in corrosion. Many researchers rely on inductively coupled plasma-mass spectrometry (ICP-MS) as a valuable tool capable of quantification of many components into the parts-per-trillion range. Typical procedures for salts with ICP-MS include dissolving the salt in aqueous solutions, which prevents the use of ICP-MS in a real-time monitoring mode with flowing molten salts and precludes the determination of oxygen in the salt. At the Nuclear Energy eXperimental Testing Laboratory (NEXT Lab) at Abilene Christian University, researchers demonstrated a patent-pending nebulizer-based interface between a flowing molten salt loop and an ICP-MS that allows for near real-time analysis of flowing molten salts without exposure to oxygen or water, potentially allowing for determination of trace oxygen and other species in the flowing molten salt. Research described here presents an update on the progress of the development of this loop-interface-ICP-MS system.

Log 301. **IMPROVEMENTS TO LIQUID SCINTILLATOR BACKGROUND CONTRIBUTIONS FOR ULTRA-LOW BACKGROUND LIQUID SCINTILLATOR COUNTING**

Lyons, S.M. (1,P), Arnquist, I.J.(1), H.O. Back (1), M. Bliss (1), M. Bronikowski (2), di Vacri, M.L.(1), Edwards, E.R.(1), Hackett, B.(1), Hoppe, E.W.(1), Rocco, N.D.(1), Rosero, R.(3), Seifert, A.(1), Swindle, A.(2), Yeh, M.(3). (1) Pacific Northwest National Laboratory. (2) Savannah River National Laboratory. (3) Brookhaven National Laboratory. (P) Presenting Author.

Liquid scintillation counting is a commonly used analysis technique for measuring trace levels of environmental radionuclides, the sensitivity of which is largely impacted by background count rates. An ultra-low background liquid scintillation counter (ULB-LSC) in Pacific Northwest National Laboratory's Shallow Underground Laboratory provides lower detection limits than are achieved by commercial instruments. Simulations estimate a background budget of roughly 20 cpd but initial measurements determined background rates of 250 cpd. Through the assay of a variety of commercial cocktails and their constituents via inductively coupled plasma mass spectroscopy

(ICP-MS), 40K in the surfactants were determined to be the highest background contributor. Efforts were made to both purify the commercial cocktails to remove 40K and to identify potassium-free surfactants. The purification technique achieved a factor of 40 reduction in the 40K, which resulted in a significant reduction of the ULB-LSC background. The improved background and sensitivity of the ULB-LSC system will be presented. (PNNL-SA-169243).

Log 302. **MASS TRANSPORT IN RENEWABLE ENERGY DEVICES BY NEUTRON IMAGING**

Borgschulte, A. (1,P); Nikolic, M. (1); Cesarini, A. (1); Billeter, E. (1); Terreni, J (1); Fumey, B. (1); Baldini, L. (1); Kaestner, A. (2); Trtik, P (2). (1) Empa, Swiss Federal Laboratories for Materials Science and Technology. (2) Paul Scherrer Institut, Laboratory for Neutron Scattering and Imaging. (P) Presenting Author.

The technical implementation of fundamental scientific results into a real working device is a challenging long-term journey. Operando neutron imaging allows the visualization of chemical processes inside running devices, such as reactors, and mass/heat exchangers. Sorption enhanced methanation is a novel process to reach high methanation concentration prerequisite for the feed of synthetic natural gas into the natural gas grid. Neutron imaging confirmed the specific macroscopic reaction kinetics resulting in the formation of a reaction front in the reactor, laying the foundation of a rational reactor design. For a heat storage application, we visualized the water uptake and distribution in a mass/heat exchanger by high-temporal resolution neutron imaging experiments, from which a new design with markedly enhanced performance was deduced. We will present concepts and discuss potential for future applications focussed in particular on the domain of renewable energy research.

Log 303. **HIGHTROUGHPUT HYDROGEN ANALYSIS OF ENERGY MATERIALS**

Nikolic, M. (1); Cesarini, A. (1); Billeter, E. (1); Terreni, J (1); Borgschulte, A. (1,P); Kaestner, A. (2); Trtik, P (2). (1) Empa, Swiss Federal Laboratories for Materials Science and Technology. (2) Paul Scherrer Institut, Laboratory for Neutron Scattering and Imaging. (P) Presenting Author.

Hydrogen is involved in numerous chemical reactions, in photosynthesis and the combustion of its products, as renewable energy carrier, and is crucial in catalysis and materials science. Despite its relevance in science and technology, the qualitative and quantitative determination of hydrogen in matter is challenging. The standard approaches are volumetric or gravimetric techniques to follow the hydrogenation reaction and to record pressure–concentration isotherms. In a pioneering work, we demonstrated the use of neutrons to quantify the amount of hydrogen in methanol catalysts. The results showcased the methodology on a single sample, however, far reaching conclusions must rely on larger materials parameter set. To do so, we developed a combinatorial setup for neutron imaging, capable of measuring up to 68 samples simultaneously under identical conditions (temperature/pressure). In this presentation, we highlight the concept along various application examples ranging from hydrogen quantification in metal alloy hydrides, hydrogen in oxides, and hydrogen in catalysts with particular focus on spillover phenomena. We give an outlook on the combinatorial search for shape-selective porous catalysts for the production of chemical energies.

Log 304. **RECENT DEVELOPMENTS ON TELLURIUM SOURCE TERM ANALYSIS IN SEVERE ACCIDENT CONDITIONS**

Pasi, A-E (1,P), (1) Chalmers University of Technology   (P) Presenting Author.

One of the understudied, yet significant, radionuclides in severe nuclear accident scenario is tellurium. Compared to the attention received by other elements such as iodine and cesium, the research around tellurium is relatively scarce. However, tellurium activities released during the two major accidents, Chernobyl and Fukushima, are comparable to that of iodine and cesium, and it is estimated that 132Te was a major contributor to the internal dose of public during the early stage of the Fukushima accident. In addition, it should not be overlooked that many of the released tellurium isotopes decay to iodine, and therefore contribute also to the iodine source term. Although tellurium has been part of several large severe accident experimental programs, and some individual studies, the overall information on the source term is still incomplete. The previous research has focused on the speciation and release of tellurium from the core, interactions with the cladding and transport to the containment. However, where the information becomes scarce is the behavior in the containment possibly leading to delayed releases. Due to the complex chemistry of tellurium, multiple oxidation states, metalloid properties and amphoteric nature, the possible reactions and important parameters affecting tellurium behavior are endless. Especially the potential volatile species, their management and mitigation should receive more attention. This work presents the techniques used to study the source term of fission products, the latest results on tellurium behavior, evidence of formation of volatile tellurium species and the uncertainties surrounding management of tellurium in severe accident conditions.

Log 305. **ACTINIS: SHIELDED SIMS FOR ANALYSIS OF HIGHLY RADIOACTIVE SAMPLES**

Peres, P. (1); Choi, S.(1); Defouilloy, C.(1); Renaud, L.(1); Touzalin, N.(1); Vuillaume, A.(1,P); Jacobson, D.(2). (1) CAMECA Gennevilliers, France. (2) CAMECA Inc., US. (P) Presenting Author.

Offering extreme sensitivity, high spatial resolution together with high throughput, dynamic SIMS (Secondary Ion Mass Spectrometry) proves extremely useful for a wide range of nuclear science applications. Derived from the field-proven CAMECA IMS 7f-Geo system, ACTINIS is designed to perform high precision elemental and isotopic analyses of highly radioactive samples in a safe environment. It is integrated in a set of biological protections and includes specific adaptations to minimize manual operations such as a full-security sample introduction system. ACTINIS benefits from the close collaboration between CAMECA and CEA Cadarache (France), and from improvements of technical design based on the experience from the first generation shielded instrument (IMS 6fR installed at CEA). ACTINIS offers depth profiling with excellent detection limits (ppb to ppm) and high depth resolution; elemental & isotopic information ranging from low mass (H) to high mass species (Pu and beyond); as well as unique sub-micrometer resolution 2D and 3D imaging capabilities. Studies performed with SIMS on irradiated nuclear fuel focus on three main axes: 1) the nuclear reactions occurring during in core irradiation which are characterized with isotopic ratio measurements, 2) the physical and chemical behavior of fission products which is evidenced by isotopic mapping, 3) the characterization of fission gases which

is carried out through depth profiling measurements. Different applications covered by ACTINIS for irradiated fuel analysis will be presented.

Log 306. ISOTOPIC URANIUM PARTICLE ANALYSIS BY LG-SIMS FOR NUCLEAR SAFEGUARDS PURPOSES

Peres, P. (1); Choi, S.(1); Defouilloy, C.(1); Fernandes F.(1); Vuillaume, A.(1,P); Jacobson, D.(2). (1) CAMECA Gennevilliers, France (2) CAMECA Inc., US (P) Presenting Author.

It has been demonstrated that Large Geometry Secondary Ion Mass Spectrometry (LG-SIMS) can be used efficiently to search for individual uranium particles among a large matrix of other particles by performing automated sample screening measurements. These screening measurements are typically followed by more precise and accurate micro beam measurements of both the major and minor uranium isotopes on individual particles. The quality of this work is essential when analyzing uranium aerosol particles from dust samples collected in nuclear facilities in the search for undeclared nuclear activities. This paper describes the latest developments that have been undertaken to enhance the detection limits and to improve the uranium isotope measurement accuracy and precision. It includes improvements in the analytical protocol as well as in the instrument acquisition software and data reduction method.

Log 307. DIRECT SYNTHESIS METHODS FOR SURROGATE ENVIRONMENTAL SWIPE SAMPLES TO SUPPORT NUCLEAR SAFEGUARDS TREATY MONITORING ACTIVITIES

Ashlee Swindle (1), Spencer M. Scott (1), Michael G. Bronikowski (1), Benjamin E. Naes (2), Travis J. Tenner (2), Kimberly N. Wurth (2), Katherine Koh (3), Riane E. Stene (3), Timothy R. Pope (3), Stephan Vogt (3), Christopher A. Barrett (3) , Matthew S. Wellons (1,P) (1) Savannah River National Laboratory, Aiken, SC. (2) Los Alamos National Laboratory, Los Alamos, NM. (3) Pacific Northwest National Laboratory, Richland, WA. (P) Presenting Author.

International treaty monitoring activities involving nuclear operations and materials typically utilize environmental collection methods to capture nuclear materials representative of facility operations. Future supporting analytical operations for treaty monitoring could include a combination of field methods, mobile laboratory operations, and dedicated high fidelity instrumentation within permeant laboratories. Scenario-dependent nuclear material characterization requirements are also expected to encompass many potential analytes. Combining diverse analytical operations and characterization goals necessitates the generation of diverse test materials capable of exercising monitoring operational activities. It is advantageous for the test material form to be representative of field operation collections consisting of particulate-laden cotton swipes with targeted isotopic and elemental compositions. Previously developed colloidal- and aerosol-based nuclear reference particulate production techniques were adapted to enable both direct and in-direct deposition of particulates on swipe substrates. Surrogate swipe production parameters were explored via a combination of production operation parameters and swipe characterization activities. Postproduction surrogate swipes were chemically processed and characterized by various radiochemical analytical measurement methods. In the case of direct deposition, experiments demonstrated total particulate mass can be tuned from nanograms to micrograms of uranium per swipe. In-

direct sample preparation, where various surfaces are first primed with a mixture of radiological and non-radiological particles and then smeared, is shown to generate swipes more representative of field samples. A demonstration of surrogate swipe subsampling and subsequent automated particle secondary ion mass spectrometry microanalysis was conducted. The results demonstrate two new capabilities for manufacturing surrogate environmental swipes samples to assist future treaty monitoring analytical operations.

Log 308. **UO$_2$ AS A POINT LIGHT DETECTION DEVICE**

Karl Rickert (1); Timothy A. Prusnick(1); Matthew A. Streby(1); David B. Turner(2,P); Charles J. Reyner (3); J. Matthew Mann(3). (1) KBR, Dayton, OH, 45431. (2) Azimuth Corporation, Beavercreek, OH, 45431. (3) Air Force Research Laboratory, Sensors Directorate, Wright Patterson AFB, OH 45433, USA. (P) Presenting Author.

A functioning, efficient and sensitive U-based neutron detector could revolutionize both proliferation monitoring and scientific instrumentation because of the inherently high fission energy released from U, but such a device is not yet realized. This presentation covers some of the first tentative steps toward this goal with a prototype device constructed from hydrothermally grown UO2. Focused laser light is used as a surrogate for point neutron-induced fission events. Wavelengths of 325, 488, 532, and 633 nm with different powers are incident on the device for a selection of timescales to test the detection responses, resolution, and saturation behaviors. Non-amplified changes in the current are observable and presented. The illumination's diameter, power, and location are altered and the impact on the observed signals is discussed. Illumination event detection is independent of illumination surface area size and largely independent of the illumination location. The ramifications of these results with regard to the development of UO2 as a neutron detector are considered.

Log 309. **IDENTIFYING CRYSTALLOGRAPHIC FACES OF URANIA AND THORIA WITH ROTATIONAL POLARIZED RAMAN SPECTROSCOPY**

Phyllis K. Morgan (1); Timothy A. Prusnick (2); Michael A. Velez (3); Karl Rickert (2); David B. Turner (4,P); J. Matthew Mann (1). (1) Air Force Research Laboratory, Sensors Directorate, Wright Patterson AFB, OH 45433, USA. (2) KBR, Dayton, OH, 45431. (3) UES, Inc., Dayton, PH 45431. (4) Azimuth Corporation, Beavercreek, OH, 45431. (P) Presenting Author.

The forensic characterization of urania and thoria is often restricted to extremely small particle sizes, which limits the instrumentation that can be employed for a given investigation. Micron-Raman spectroscopy is invaluable in these endeavors in that it can be used to completely map a variety of sample sizes down to 1 micron particles. This ability has resulted in micron-Raman spectroscopy being widely applied to analyzing urania and thoria, but its potential is still not yet fully realized. Here, we report a systematic study of polarized Raman scattering of high-quality, hydrothermally grown, single crystals of urania and thoria. The peak intensity variations for as-grown major crystal planes, post-growth polished crystal planes, and a post-growth polished non-crystallographic plane are directly linked to crystallographic orientation and crystal rotation. The empirical results agree with computed models. Parallel polarized peak intensities are directly correlated with the fluorite structure's metal–oxygen–metal chains which can in turn be used to

determine both orientation and rotational alignment of a given crystal face (if sufficiently small rotational steps are applied). These results also suggest that Raman spectroscopy can identify non-crystallographic orientations that are not discernable by traditional means.

Log 310. SYNTHESIS, STRUCTURAL PROPERTIES, AND ELECTRICAL PROPERTIES OF SINGLE CRYSTALS OF Zr-DOPED UO₂

Karl Rickert (1); David B. Turner (2,P); Timothy A. Prusnick (1); J. Matthew Mann (3). (1) KBR, Dayton, OH, 45431. (2) Azimuth Corporation, Beavercreek, OH, 45431. (3) Air Force Research Laboratory, Sensors Directorate, Wright Patterson AFB, OH 45433, USA. (P) Presenting Author.

UO2 nuclear fuel is encased in Zr cladding in reactors as a standard practice, making the synthesis of (Zr,U)O2 of interest. Furthermore, doped and undoped UO2 have long been speculated as valuable semiconductors for niche applications such as rad-hard electronics, neutron detection, and thermoelectrics. In this talk we present the incorporation of tetravalent zirconium into UO2, which has some surprising electronic properties despite having the same oxidation state as U. The production and characterization of Zr:UO2 has been pursued in both powder and microcrystalline form, but we report the first large scale (~1 cm2 surface area) hydrothermal synthesis and subsequent analysis of single crystal Zr:UO2. This synthesis is made possible by following the previously reported procedure for growing UO2, samples of which are used for property comparisons. The presence of Zr is confirmed with TOF-SIMS and the minor structural differences observed with X-ray diffraction and Raman spectroscopy are discussed. μ-Raman analysis with a 532 nm laser is used to demonstrate the oxidation inhibition that results from Zr incorporation. Preliminary electronic measurements are presented, including I/V curves, resistance measurements, burn-in behavior, and Hall measurements. Reliable electrical contacts are needed for these measurements, and the challenges and solutions of this requirement are also discussed. In having large single crystal samples of both UO2 and Zr:UO2, the difference in properties are assessed and the results can be of use to both the nuclear energy and semiconductor industries.

Log 311. SPATIAL VARIATION OF FISSION NOBLE GAS RATIOS IN SPENT NUCLEAR FUEL

Willett, C.D. (1,P); Cassata, W.S. (1); Isselhardt, B.H. (1); Matzel, J.E. (1). (1) Lawrence Livermore National Laboratory. (P) Presenting Author.

Fission-derived noble gases from nuclear fuel irradiation have the potential to support nuclear safeguards and declarations verifications, as fission produces 11 noble gas isotopes whose proportions depend on reactor operating parameters. However, existing analytical approaches do not clearly address the effects of spatial variability of irradiation temperature on in-fuel gas migration and release. Because gas diffuses through solid as a strong function of temperature, a plenum gas mixture is likely derived primarily from the hottest portion of a pin. Therefore, plenum ratios may not be representative of all pin material and gas retained along the length of the pin will vary as a function of local conditions. In this work, we directly investigate the spatial variability of fission gas signatures via high-precision noble gas mass spectrometry. Samples comprise both plenum gas and retained gas from four locations along the axis of a single UO2 fuel pin irradiated in Belgian research reactor BR-3. Samples are particularly well-suited to this

problem because irradiation conditions, including fuel temperature, are well-documented. In retained gas samples, example flux-dependent ratio 136Xe/(131Xe+132Xe+134Xe) scales with gamma activity along the pin length. The plenum gas ratio, however, most closely matches the retained gas from the two highest-flux pin positions and overestimates the pin average, as calculated using all retained gas data. These results indicate that fission gas ratios from plenum and retained gas samples are most valuable—and applicable to supporting safeguards—when their interpretation includes consideration of spatial heterogeneity in irradiation conditions.

Log 312. **HARVESTING U-234 FROM HEAT SOURCE PLUTONIUM FOR USE AS A SPIKE SOLUTION**
Carver, Nell; Jump, Robert; Pardington, Paige; Porterfield, Donivan (P). Los Alamos National Laboratory. (P) Presenting Author.

A method to separate U-234 from a Heat Source Plutonium (HS-Pu) matrix via Eichrom UTEVA® resin was developed for use in PF-4, however, a commercially available and convenient tracer and/or spiking solution is not available. Ideally, a laboratory control sample (LCS) or blank spike (BS) is processed with each batch of samples to monitor the effectiveness of the separation process. A spiking solution was generated in-house by harvesting ingrown U-234 from HS-Pu residues. U-234 in-growth from Pu-238 is approximately 1% per year. Pu-238 + alpha à U-234 (Pu-238 t1/2 = 87.7 years). Blank spike recoveries using the freshly made U-234 solution were, on average 99.9% and matrix spike recoveries were 98.5%, on average.

Log 313. **COMPARING URANIUM RADIOCHRONOMETRY AGES DETERMINED BY SINGLE-COLLECTOR AND MULTI-COLLECTOR ICP-MS AT LOS ALAMOS NATIONAL LABORATORY**
Wende, A.M. (1,P); Denton, J.S. (1); Kayzar-Boggs, T.M. (1); Inglis, J.D. (1); Edwards, M.A. (1); Sanborn, M.E. (1); Steiner, R.E. (1) Los Alamos National Laboratory. (P) Presenting Author.

Radiochronometry is an important tool used during nuclear forensic investigations as model ages may constrain when a material was produced or last purified. Modern radiochronometric data are ideally obtained using high precision multi-collector (MC) mass spectrometers. However, there is interest in better understanding the capabilities of less costly single-collector (SC) mass spectrometers, and the impact of less precise isotope ratio measurements when applied to chronometry. Modeling of ^{230}Th/^{234}U age data for uranium certified reference materials (CRMs) suggest that the greatest impact on age precision from use of a SC instrument will be derived from increased uncertainty in the measured ^{234}U/^{238}U and ^{230}Th/^{229}Th ratios used in ^{234}U and ^{230}Th assay calculations. Modeling also indicates that a more subtle impact on age accuracy can be introduced by small measurement biases from low precision SC instruments. To evaluate the capability of SC relative to MC instruments, Los Alamos National Laboratory (LANL), Lawrence Livermore National Laboratory, and the China Institute of Atomic Energy collaborated on an interlaboratory, multi-instrument radiochronometry exercise to obtain ^{230}Th/^{234}U and ^{231}Pa/^{235}U model ages for uranium CRMs U030A, U850, and U900. Here, we present results obtained at LANL in which chemically identical aliquots were analyzed in triplicate on a ThermoScientific Neptune Plus MC-ICP-MS and Element XR SC-ICP-MS. We compare model ages determined by each instrument and describe method development to optimize Element XR measurement accuracy and precision.

Log 314. **OPTIMIZATION OF A SIMPLIFIED RADIOCHEMICAL METHOD BASED ON SR®-RESIN FOR MEASUREMENT OF 90-Sr IN NUCLEAR WASTE**

Baudat, E. (1,P); Gautier, C (1); Bagán, H(2); Tarancón, A(2); Colin, C(1); Laporte, E(1); Fichet,P(3);(1) Université Paris-Saclay, CEA, Service d'Études Analytiques et de Réactivité des Surfaces, 91191, Gif-sur-Yvette, France,(2) Department of Chemical Engineering and Analytical Chemistry, University of Barcelona, Marti i Franqués, 1-11, ES-08028, Barcelona, Spain. (3)CEA, DES/DDSD/DFDE/SGOF, Building 611, 91191, Gif-sur-Yvette, France. (P) Presenting Author.

As a caracteristic fission product with a half-life of 28.8 years, Sr-90 must be quantified in nuclear waste. Prior to its nuclear measurement, it is necessary to separate it from its matrix and interferents with radiochemical method. The extraction chromatographic resin, Sr-resin®, developed by Eichrom in the 1990's is widely applied. Its efficiency has been demonstrated for a wide range of samples even if pre-treatments are necessary depending on the nature of the sample. For nuclear waste, difficulties can be encountered especially with samples with high Pu content. A precipitation with ammonia can be used but this can lead to a Sr loss depending on the experimental conditions (ex: digestion step with HF acid or not) and interferents present. In this presentation, the robustness of an alternative pre-treatment using TRU-resin® will be highlighted. It has been proven to be efficient on all the nuclear waste samples investigated in the study, such as effluent, sludge, resin and concrete, even if they contain high Pu amount. Moreover, in this method, a plastic scintillation resin for Sr-90, PS-resin, developed by Bàgan et al., has been tested for nuclear waste samples. The advantages of this new resin is to reduce time of separation by eliminating elution step but mainly to avoid the use of scintillation cocktails. Due to their high toxicity, they are concerned by the REACH regulation and their implementation will become complicated in a near future. The PS-resin could be a suitable alternative for Sr-90 measurement by scintillation counting.

Log 316. **ANALYSIS OF TURTLE AND TORTOISE SCUTES CAN PROVIDE A TIME-CONSTRAINED RECORD OF RADIONUCLIDE RELEASES INTO THE ENVIRONMENT**

Inglis, J.D. (1,P); Conrad, C.N. (2); Mukundan, N. (1); Sanborn, M.E. (1); Wende, A.M. (1); Tenner, T.J. (1); Wurth, K.N. (1); Naes, B.E. (1); Fair, J.M. (3); Middlebrook, E.A. (3); Gaukler, S.M. (2); Whicker, J.J. (2); Tapia Aguilera, W. (4); Gibbs, J.P. (5); Wolf, B.O. (6). (1) Chemistry Division, Nuclear and Radiochemistry Group, Los Alamos National Laboratory, Los Alamos National Laboratory; (2) Environmental Protection and Compliance Division, Environmental Stewardship Group, Los Alamos National Laboratory; (3) Bioscience Division, Biosecurity and Public Health Group, Los Alamos National Laboratory; (4) Giant Tortoise Restoration Initiative, Galapagos Conservancy; (5) State University of New York College of Environmental Science; (6) Department of Biology, University of New Mexico. (P) Presenting Author.

Testudines reptiles, such as tortoises and turtles, are characterized by a shell (scute) composed of keratin tissue. Many Testudines species grow their scutes in sequential annual layers, and as each layer grows it records chemical signatures from the surrounding environment. Scutes may therefore provide a time-resolved archive of any chemical inputs or changes in the reptile's habitat. We postulated that if a habitat intersects with an area of nuclear release, the scute could accumulate some record of that release into the environment. Additionally, isotope analysis of individual growth layers from such a specimen could also provide detailed and time-constrained insight into the nature of the release. To investigate this idea, we analyzed the uranium concentration and isotope composition of several Testudines scute samples collected from areas with a known history of radionuclide releases. Our initial data shows that individual specimens can accumulate several ng/g of uranium in their scute layers, which allowed us to identify anthropogenic uranium signatures in 3 out of 6 samples measured. In addition, using a specimen of a box turtle collected from Oak Ridge (c.1962), we were able to determine quantifiable changes in the U isotope composition along individual layers of growth (representing seven years of life). We believe our study provides a new, time-resolved, tool for understanding the fate of radionuclide effluents in the environment. LA-UR-21-32103

Log 317. RADIOACTIVE ASBESTOS - A CHALLENGING MATERIAL FOR RADIOANALYTICAL INVESTIGATION

Köhler, F. (1,P); Heule, M. (1); Jäggi, M. (1). (1) Paul Scherrer Institute (PSI). (P) Presenting Author.

In the second half of the 20th century, asbestos has been widely used as construction material, especially with regard to thermal insulation. During this period, many nuclear facilities were built and contamination of asbestos with a wide variety of radionuclides is to be expected today. For dismantling, decommissioning, and disposal of asbestos containing material (ACM), a complete dissolution of ACM is necessary, which can be achieved with a maximum of 1.5 g of ACM and a lithium borate / lithium metaborate (80:20) flux at a maximum temperature of 1065° C, giving a complete dissolution of ACM. The molten glass is poured into 3 M HNO3 solution, containing polyehtylene glycol, where the latter forces the flocculation of SiO2-gel, thus avoiding a treatment with HF as well as removing this chemically inert yet interfering part of the sample matrix. The dissolved glass can be further processed in a multi-column-multi-step separation scheme. The separation is based on a combination of UTEVA/TRU/Sr resins, utilizing generic chromatographic separation methods. Recent results proved the functionality of the method for effective extraction of Am, Cm, Pu, U isotopes, as well as Sr-90 with recoveries ranging from 80 –100 % from the highly complex matrix of asbestos material. All laboratory work can done within 2 – 3 days, which gives a fast method and leaves the required measurement time as limiting factor for the overall analytical process; further asbestos types with higher contamination grade are currently under investigation.

Log 318. ^{230}Th-^{234}U AND ^{231}Pa-^{235}U RADIOCHRONOMETRY INTERCOMPARISON OF MULTIPLE ICP-MS INSTRUMENT PLATFORMS AT LLNL AND LANL

Rolison, J.M. (1,P), Denton, J. (2), Wende, A. (2), Lindvall, R. (1), Wimpenny, J. (1), Edwards, M. (2), Sanborn, M. (2), Kayzar-Boggs, T. (2), Gaffney, A. (1), Treinen, K. (1). (1) Lawrence Livermore National Laboratory. (2) Los Alamos National Laboratory. (P) Presenting Author.

Radiometric dating of nuclear material found outside of regulatory control provides valuable information about the apparent age of the material which can be useful in a forensic investigation. The radiometric age of a nuclear material is assumed to reflect the elapsed time since the parent radionuclide (e.g., 234U, 235U) was separated from its progeny isotopes (e.g., 230Th, 231Pa). The time dependent ingrowth of the progeny isotopes into a bulk nuclear material is a conceptually simple forensic signature that has found widespread utility in the nuclear forensic community. While conceptually simple, accurate and precise measurements of 230Th/234U and 231Pa/235U present technical challenges. Many nuclear forensic laboratories employ multi-collector inductively coupled plasma mass spectrometry (MC-ICP-MS) instrumentation for performing these interelement isotope ratio measurements. However, MC-ICP-MS instruments are more expensive and have a larger footprint than the more widely utilized single collector ICP-MS instrument platforms. This study aims to compare the accuracy and precision of multiple ICP-MS platforms during the radiometric dating of a variety of uranium certified reference materials. LLNL-ABS-830080

Log 319. **STABLE ISOTOPE TAGGANTS OF NUCLEAR FUELS**

Rolison, J.M. (1), Shollenberger, Q.R.(1,P), Said, M.(1), Marks, N.E.(1). (1) Lawrence Livermore National Laboratory. (P) Presenting Author.

Nuclear materials found outside of regulatory control are subjected to forensic analyses to determine their origin and intended use. Forensic techniques are generally labor and time intensive and are therefore difficult to incorporate into rapidly evolving law enforcement investigations. Nuclear fuels labelled with isotopically enriched taggants could provide unique identifiers that may expedite nuclear forensic timelines. Confidently detecting isotopic taggants in seized nuclear materials requires a fundamental understanding of how the isotopic composition of the tagged material could be altered during nuclear fuel cycle processes. For example, the taggant element will likely be diluted with isotopically natural material that is accumulated during normal processing and handling of the bulk nuclear material. Thus, the taggant element is unlikely to preserve its initial unique isotope ratios during the life of the nuclear material but is likely to obtain an isotopic composition which lies along a mixing line between the isotopic composition of the taggant and that of natural background material. It is also possible that bulk nuclear material may have non-natural isotopic compositions of the taggant element due to fuel cycle processes (e.g., enrichment) and as such a two end-member mixing assumption would be invalid. Here we will present an initial framework for deciphering isotopic taggants in nuclear materials and a survey study of a suite of LEU nuclear fuel to begin constraining stable isotope variations in potential taggant elements in nuclear fuels. LLNL-ABS-829899

Log 320. **FEASIBILITY OF EARLY FUEL CYCLE TAGGANT INCORPORATION FOR INTENTIONAL FORENSICS**

Spano, T.L. (P)(1), Ulrich, T.L.(1); Kercher, A.(1); Sadergaski, L.(1); Shields, A.E.(1) (1) Oak Ridge National Laboratory (P) Presenting Author.

To develop strategies for incorporating taggants into oxide fuels and understand how taggant candidates persist through early fuel cycle processes, synthetic procedures have been developed to produce intentionally tagged early fuel cycle intermediates. First, metal nitrate solutions of varying concentrations are introduced to aqueous uranyl nitrate (UN) to produce tagged UN. Then, uranyl peroxide is precipitated from UN. Tagged amorphous and alpha-UO_3 are produced by calcination of tagged uranyl peroxide at 180C and 350C, respectively. Taggant incorporation is monitored by inductively coupled plasma-optical emission spectroscopy (ICP-OES) to investigate how initial taggant concentration in the UN precursor influences obtained concentrations in later intermediate products. Structural influences of taggant incorporation are investigated using powder X-ray diffraction and Raman spectroscopy to provide insight into crystallographic modifications resulting from addition of tags to these early fuel cycle materials and elucidate the chemical form of tags introduced at these stages. The possibility of segregation of taggant species into discrete phases within uranium matrices will be examined using scanning electron microscopy-energy dispersive X-ray spectroscopy and Raman spectroscopic mapping with principal component analysis. Finally, production and analysis of tagged U_3O_8 as a precursor to UO_2 powders and compacts will proceed after confirmation of taggant incorporation in earlier

intermediates. Preliminary observations using Raman spectroscopy and ICP-OES indicate that introducing metal oxide powders immediately before UO2 synthesis results in tagged materials. Results from this study will inform strategies for optimizing taggant incorporation in UO2.

Log 321. CHARACTERIZATION OF SPECTRAL SIGNATURES AND PARTICULATE FORMATION FOR A BETTER UNDERSTANDING OF NUCLEAR FIREBALL CHEMISTRY

E.N. Weerakkody (1,P); D.G. Weisz (1); B. Koroglu (1); E. Balboni (1); N.G. Glumac (2). (1) Lawrence Livermore National Laboratory; (2) University of Illinois at Urbana-Champaign; (P) Presenting Author.

Spectroscopic methods can be used to better quantify and interpret experiments to understand post-detonation nuclear fireball condensation chemistry by developing signatures from relevant species. Uranium (U) and mixtures containing fission product surrogates (stable Cs, Sr, etc.), were excited using laser ablation, dust cloud explosion, high-temperature shock, and explosive detonation to determine the spectral signatures produced. Additionally, signatures from excitation of fission product surrogate mixtures emulating fast independent yields of fissionable materials, U235 and Pu239, were compared. Laser ablation, dust cloud, and shock tube experiments on depleted U elucidated atomic, ionic, and monoxide signatures in high-temperature conditions. U signatures did not manifest at temperatures lower than U's boiling point (4404K) or oxide volatilization (~3800K), providing insight into their evolution over 2000-10000K. Fission product surrogate mixtures were excited using flash powders and detonations with conventional explosives to study spectral signatures produced in high-temperature environments. Some s-block elements were found to be strong emitters and differentiable between mixtures tested. Finally, a plasma flow reactor, which facilitates study of kinetics in a plasma environment, was used to determine how species (U, Cs, Fe, Si, etc.) may fractionate when condensing from the plasma at variable rates. The resulting dataset shows that spectral signatures are highly influenced by plasma (or vapor) composition but can still be distinguished and interpreted under certain circumstances. This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344 and DTRA Grants HDTRA1-17-1-0026 and HDTRA1-20-2-0001. LLNL-ABS-830159

Log 322. INTEGRAL FUEL PERFORMANCE EVALUATION OF MATERIAL PROPERTIES AFFECTED BY FORENSIC TAGGANTS

Cheniour, A. (1,P); Wilson, B.(1); Sweet, R.T.(1); Nelson, A.T.(1); Shields, A.E.(1). (1) Oak Ridge National Laboratory. (P) Presenting Author.

The use of taggants in nuclear fuel for improved security and control of commercial fuel may potentially change the properties of UO2 fuel. Studies have shown that impurities (i.e. taggants) in UO2 can impact defect structure, grain size and density. These may impact fuel properties and, in turn, integral fuel behavior during irradiation. Changes to grain size modify fission gas release and can more generally affect the overall thermomechanical behavior of the fuel. To quantify the taggant effects on fuel behavior, simulations were developed to predict fuel behavior and to inform a future taggant-fuel irradiation in HFIR. The fuel performance code BISON was used to perform a sensitivity analysis of the thermomechanical behavior of UO2

as a function of grain size and fuel density based on expected changes to fuel microstructure from taggant additions. Simulations use irradiation conditions from a three-cycle PWR core applied to 2D axisymmetric fuel pellets and cladding. The results show that some of the considered models (specifically fission gas release and fission product swelling) are highly sensitive to either input parameters or both. To validate the BISON predictions, an irradiation of tagged fuel samples is planned for the upcoming fiscal year. Tagged fuel samples, both irradiated and unirradiated, will undergo a detailed material characterization regarding grain size, density, and thermomechanical properties to help inform the models and to quantify changes in fuel behavior during reactor operation.

Log 323. THE DEVELOPMENT OF METHODOLGIES FOR THE ANALYSES OF Ra-226 AND Ac-227 TOWARD THE APPLICATION OF NUCLEAR MEDICINE STARTING MATERIALS AT ORNL
Keever, T.J. (P); Wightman, H.H.; Chattin, M.R.; Giaquinto, J.M.; Ivey, W.P.

Oak Ridge National Laboratory (ORNL) is a producer of Ac-227 for Bayer Pharmaceutics for the production of the cancer treatment drug, Xofigo(Registered). This drug utilizes Ra-223 dichloride and is used to treat prostate cancer that no longer responds to hormonal or surgical treatment. The Radioactive Materials Analytical Laboratory within the NAS at ORNL in collaboration with the Bethel Valley Radiochemical Processing Group has developed the methodologies for the analyses of the Ra-226 target material and Ac-227 product material. Presented are the development of methods for the analyses of Ra-226 and Ac-227 isotopes by ICP-MS, along with potential impurities, both radiological and non-radiological. Also presented are the radiochemical analyses for the determination of Ac-227 via its daugher emitting radionuclides. In addition, radiochemical methods developed for the analysis of potentially interfering radionuclides, such as Sr-90, P-210 and Po-210 will be discussed. Safe handling practices for Ac-227 at ORNL will also be discussed.

Log 324. IDENTIFYING TRACER - ANALYTE DISEQUILIBRIUM IN PLUTONIUM BIOASSAY SAMPLES, AND A METHOD TO PROPERLY ESTABLISH EQUILIBRIUM
Harris, M.N. (1P); Hudston, L.A. (1); Zazueta, J.A.(1); Zuniga, M.M. (1); Lopez, D.M. (1); Eaton, S.J. (1); Debacker, K.B. (1); Inglis, J.D. (1); Lamont, S.P. (1); Steiner R.E. (1). (1) Los Alamos National Laboratory.

During 2019, the quality control (QC) samples included with each set of plutonium bioassay samples at Los Alamos National Laboratory consistently showed a low bias relative to known values. A method was established to compare isotopic ratios between ^{242}Pu tracer and plutonium analyte remaining in the supernatant portion of urine providing the ability to explore isotopic equilibrium imbalances between the tracer and analyte. The data illustrated a significant isotopic equilibrium differential, where the supernatant displayed over a 10-fold increase in ^{238}Pu/^{242}Pu, ^{239}Pu/^{242}Pu, and ^{240}Pu/^{242}Pu ratios when compared against the corresponding isotopic values measured in the precipitated fractions. This observation indicated that stable plutonium complexes can form within the urine matrix and are resistant to co-precipitation. Additional tests to better establish equilibrium between plutonium isotopes were performed by adding iron (II) and hydrogen peroxide prior to co-precipitation. Iron catalyzes the decomposition of hydrogen peroxide via metal-hydroxo species (e.g., heterogeneous iron-hydroxide precipitates) through

increased alkalinity from ammonium hydroxide during co-precipitation. Under alkaline conditions the rate of hydrogen peroxide decomposition is highly accelerated producing reactive oxygen species capable of degrading any chelators within the organic matrix in a matter of hours. Upon amending this technique, isotopic equilibrium between plutonium tracer and analyte was achieved and a low bias in plutonium isotopes in QC samples was no longer observed. This event demonstrates the need to incorporate methods to degrade the organic matrix within urine during co-precipitation, as chelators common in food and drink can interfere with isotope dilution methodologies commonly used in bioassay.

Log 325. **LANL METHODS AND RESULTS IN SUPPORT OF PLUTONIUM RADIOCHRONOMETRY CRM DEVELOPMENT**

Edwards, M.A. (1) (P); Kayzar-Boggs, T.M. (1); Wende, A.M. (1); Maassen, J.R. (1); Sanborn, M.E. (1); Denton, J.S. (1); Krajewski, K.J. (1); Steiner, R.E. (1). (1) Los Alamos National Laboratory. (P) Presenting Author.

Radiochronometry is a powerful tool for elucidating the processing history of nuclear materials including plutonium (Pu). Multiple daughter/parent and granddaughter/parent Pu chronometers are measured by forensics laboratories (e.g., 241Am/241Pu, 236U/240Pu, and 237Np/241Pu). Model ages obtained for preexisting Pu certified reference materials (CRMs) can be compared to production dates ("paper ages") or consensus ages obtained by multiple forensics laboratories. However, there is a critical lack of traceable Pu radiochronometry CRMs certified specifically for model age that can be used for quality control of radiochronometry measurements performed on Pu samples of unknown age. To address this need, SRM-946 (renamed CRM 136 by New Brunswick Laboratory) was purified at a known time in 2015 at the European Commission Joint Research Centre Karlsruhe, and subsamples of the purified Pu material were distributed for an interlaboratory analytical campaign to certify the material for multiple Pu radiochronometric model ages. This approach is analogous to that implemented in the development of uranium CRMs IRMM 1000a and 1000b. Here, we will present Los Alamos National Laboratory (LANL) methods employed in support of this interlaboratory reference material development effort and discuss some of the challenges associated with Pu radiochronometry measurements. We will also present the model ages obtained at LANL and compare our results to the known processing history of this reference material in development.

Log 326. ENGINEERED MICROPARTICULATES FOR THE INTRODUCTION OF INTENTIONAL FORENSIC SIGNATURES TO NUCLEAR FUELS

Scott, S.M. (1,P); Lawson, S.(1); Samperton, K.(1); Bronikowski, M.(1); Hoar, E.(1); Smith, R.(1); Wellons, M.(1). (1) Savannah River National Laboratory, Aiken, SC. (P) Presenting Author.

Intentional nuclear forensics is an emerging concept in which minute well-characterized additions of unique anthropogenic compositions of stable isotopes to nuclear materials, such as fuel, serve to encode signal within the host matrix. This unique embedded isotopic barcode could be used to readily identify the provenance of nuclear material with exceptionally high confidence. Recent developments in the generation of engineered microparticulates have demonstrated a route for the production of microparticulates with highly uniform, size, material phase, and elemental and isotopic composition to support international nuclear safeguards. As the result of their uniform nature, engineered microparticulates offer a promising route for the delivery of barcode materials to nuclear matrices of potential risk. Because each microparticle is identical in composition, even trace particulate quantities are capable of identifying the host material's origin. Initial efforts demonstrate the generation of microparticles with compositions of nickel oxide, molybdenum oxide, tungsten oxide, and Ni/Mo/W oxide combinations. All are potential stable isotope barcode materials if formulated with anthropogenic isotopic feedstocks. The generated particulates display a high degree of uniformity, indicated by monodisperse size distributions (geometric standard deviation < 1.20) at sizes near 1-micron in diameter. Further efforts have demonstrated both synthesis scale-up operations and explored the Ni/Mo/W oxide mixing compositional boundaries for which interparticle homogeneity may be maintained.

Log 327. STABLE NEODYMIUM ISOTOPIC COMPOSITION OF NUCLEAR DEBRIS SAMPLES

Patton, G.M. (1,P); Hanson, S.(1); Sanborn, M. (1); Inglis, J. (1). (1) Los Alamos National Laboratory. (P) Presenting Author.

Naturally occurring neodymium (Nd) isotopic compositions measured in continental material, with the exception of 143Nd/144Nd, are constant. Due to radioactive decay of 147Sm to 143Nd, the 143Nd/144Nd ratio is a function of the Sm concentration and age of the sample, which allows this ratio to be used as a tracer of provenance as this isotopic composition varies geographically in natural samples. However, fission reactions result in Nd isotope decay products which will alter the Nd isotopic compositions measured in nuclear debris. Trinitite samples measured by MC-ICP-MS show perturbations of Nd isotopes relative to Nd-142, the only isotope of Nd which is not influenced by fission product decay. By measuring the 144Nd/142Nd ratio and the total amount of Nd in the sample, it may be possible to quantify the total number of fissions in the sample.

Log 328. **APPLICATION OF MICRO X-RAY FLUORESCENCE BY SYNCHROTRON RADIATION TO INVESTIGATE THE MECHANISMS OF BIOFILM METAL BIOREMEDIATION**

Hauradou, T.(1,P); Pennafirme, S.(1); Lima, I.(1). (1) Federal University of Rio de Janeiro. (P) Presenting Author.

Sedimentary environments in highly urbanized areas are contaminated by metals at toxic levels. Bioremediation uses microorganisms to mitigate the effects of metallic contaminants on biota. The use of bacterial biofilms is an effective tool since they adsorb heavy metals from the site. Synchrotron X-ray microfluorescence (SRµ-XRF) is a non-destructive technique that allows us to generate 2D maps of the sample's elemental composition. This work aims to show the importance of using SRµ-XRF in the investigation of the mechanism of heavy metals bioremediation. Bacterial biofilms were tested for adsorption of Copper (Cu), Zinc (Zn), and Cadmium (Cd) at toxic levels (50 ppm -Cu and Zn; 25 ppm - Cd). From the analysis of the 2D maps of the metals concentrations, the bacterial biofilm was able to adsorb them in the following order of efficiency: Zn>Cu>Cd. The heterogeneous adsorption of Zn and Cu observed in the biofilm is because the organization of bacteria and exopolymeric substances (EPS) does not occur homogeneously, with a higher concentration of metals in the presence of a higher concentration of microorganisms and EPS. The formation of a dense biofilm was not observed in tests with Cd. The high levels of toxicity of Cd hindered a vast cell multiplication and subsequent formation of EPS at levels capable of greater efficiency, although some adsorption was still observed. The SRµ-XRF technique is a high-quality tool to analyze biofilms used in bioremediation.

Log 329. **NETWORK OPTIMIZATION FOR TREATY MONITORING**

Eslinger, P.W.(1,P), Miley, H.S.(1), Schrom, B.T.(1). (1) Pacific Northwest National Labotoratory. (P) Presenting Author.

Design of an effective monitoring network requires information on the type and size of releases to be detected, the accuracy and reliability of the measuring equipment, and the desired network performance. This work provides a scientific basis for selecting minimal-size networks of 133Xe samplers that achieve a desired detection probability for difference levels of release. The approach starts with a large number of fixed sampling locations and searches for optimal subsets of those locations for networks of different sizes. Although applied to a continental-scale network, the method can be applied to a local or global network. The approach is demonstrated using 120 hypothetical sampling locations and synthetic releases of 133Xe over a wide range of magnitudes at 1550 release locations in the continental United States. Separate modeled releases occur every 3 hours at each location to incorporate both short-term and long-term atmospheric variability. Three network performance metrics are calculated: the probability of detecting the releases, the expected number of stations to detect the releases, and the expected number of samples that detect the releases. The quantitative measures support picking optimal or near-optimal sampling locations for a network of a specified size and they identify the number of samplers required to meet design goals for a given release magnitude such as a target detection probability or the number of detecting stations.

Log 330.   **PROJECTED NETWORK PERFORMANCE FOR NEXT GENERATION AEROSOL MONITORING SYSTEMS**

Miley, H.S. (1,P), Eslinger, P.W. (1), Schrom, B.T. (1). (1) Pacific Northwest National Labotoratory. (P) Presenting Author.

Recently, Miley et al. (2019) showed that various combinations of increased air volume, an optimized 72-hour sample-decay-measure timeline, and larger detectors on both sides of a compact sample can plausibly maintain the same 140Ba minimum detectable concentration (MDC) with collection periods of less than 24 hours. This work considers the network performance of possible next-generation aerosol samplers if deployed at the radionuclide stations of the International Monitoring System. A large number of releases all over the globe were simulated eight times per day for a year. The global probability of detection was calculated for releases ranging from 109 Bq to 1016 Bq of 140Ba and 131I for both current and next-generation monitoring systems. The number of detecting stations and number of detecting samples were also calculated. Regardless of the sample integration time, the same MDC was used in all analyses. The shorter collection intervals substantially improve coverage in the tropical regions where travel times may exceed the design goal of 10 days, allowing for more decay and more time for rainout. The detection sensitivities for samplers with 12-, 8-, and 6-hour collection periods are quite similar for 140Ba and 131I, with both 8- and 6-hour results better than 12-hour configurations, and all three sampling intervals yield substantially better detection probability than 24-hour measurements. The improvement in the detection probability for 8- and 6-hour collection periods as compared to the 24-hour collection period is maximized at about 0.18 for releases of 1012 Bq, or 0.01% of the 140Ba produced in a nuclear explosion of 1 kt TNT equivalent.

Log 331. PHYSICOCHEMICAL PROPERTIES FOR VAPOR TRANSPORT THROUGH GEOLOGIC MATERIALS

Elizabeth H. Denis (1,P), Guohui Wang (1), Anjelica Bautista (1), Nicholas L. Huggett (1), Mark L. Rockhold (1), Carlos G. Fraga (1), Michael P. Foxe (1), April J. Carman (1). (1) Pacific Northwest National Laboratory, Richland, WA, USA. (P) Presenting Author.

Understanding the chemistry of gas-solid interactions under different temperature and humidity conditions can inform how volatile radiochemical compounds are transported through geological materials and to the surface. The inherent physical and chemical heterogeneity of many geologic materials can make prediction of sorption properties difficult. We are using inverse gas chromatography (IGC) to characterize and quantify the physicochemical sorption properties of geologic materials (e.g., soil, sand, salt, and zeolites) of various mineralogical makeup and particle sizes. The geologic material is probed using gases, such as volatile organic compounds and noble gases, which can serve as surrogates for additional compounds of interest (e.g., volatile radionuclides and environmental contaminants). Replicate measurements demonstrated higher sample variability of heterogenous materials, but also showed the ability to discern differences in physicochemical properties. Geologic materials can be differentiated based on heat of adsorption and partition coefficient. For diffusion coefficient, particle size may be more influential than material type. A goal is to integrate IGC-determined physicochemical parameters with subsurface transport modeling to help build a predictive framework for signature migration to the surface. Toward this end, IGC experiments were modeled with STOMP-GT (Subsurface Transport Over Multiple Phases-GeoThermal). This work demonstrates that IGC is effective in analyzing key physicochemical parameters used for modeling subsurface material transport.

Log 332. COMBINING AEROSOL AND NOBLE GAS SAMPLES IN SOURCE-LOCATION ANALYSES

Eslinger, P.W. (1,P), Miley, H.S. (1), Schrom, B.T. (1). (1) Pacific Northwest National Labotoratory. (P) Presenting Author.

Recently, Eslinger et al. (2019) introduced a new Bayesian algorithm that makes discriminating between possible types of releases (e.g., nuclear explosion, nuclear power plant, or medical isotope production facility) an integral part of a source-location analysis for samples that contain multiple xenon isotopes. The method can be applied to data sets containing both aerosol and noble gas samples that are measured with different frequencies. Using synthetic data, the method is applied to five estimation cases with station separation distances on the order of 1000 km where each release plume crosses two sampling stations. Releases for three xenon isotopes, 133mXe, 133Xe, and 135Xe, and three iodine isotopes, 131I, 133I, and 135I, are simulated for 235U fission. The average location discrepancy (estimated release location versus the simulated release location) decreases from 100 km (with any two isotopes) to 45 km with six detected isotopes. The posterior probability of selecting the correct release type increases with the number of detected isotopes and approaches 1 when four or more isotopes are detected.

Log 333. **LONG-TERM AND SEASONAL CYCLING OF RADIONUCLIDES IN A MONOMICTIC POND AT SAVANNAH RIVER SITE, S.C.**

Wasserman, N.L. (1, P); Merino, N. (1); Coutelot, F. (2); Kersting, A.B. (1); Powell, B.A. (2); Zavarin,M. (1). (1) Glenn T. Seaborg Institute, Lawrence Livermore National Laboratory. (2) Department of Environmental Engineering and Earth Sciences, Clemson University. (P) Presenting Author.

In the early 1960's, historical releases of 10's to 100's of MBq of Pu and 137Cs from Savannah River Site's R-Reactor were dispersed in local waterways and ponds that served as the secondary cooling loop for the reactor. One of the ponds, Pond B, provides a unique testbed to examine biogeochemical factors influencing long-term and seasonal Pu and 137Cs mobility in a wetland. In June 2019 and March 2020, water samples were collected at five locations along a transect and measured for physicochemical parameters, organic carbon, trace metals, anions, Pu, and 137Cs concentrations. The pond stratifies annually with respect to temperature and redox from March through October. In late fall, once it overturns, the water column is oxic. Plutonium and 137Cs concentration measurements indicate that these fluctuating redox conditions influence radionuclide cycling through iron oxyhydroxide dissolution in deeper waters and sediments, and sorption to microbially produced organo-Fe(III) complexes within the thermocline. High Pu concentrations correlate with localized accumulation of organic matter in the inlet and outlet of the pond pointing to the influence of organic matter complexation on Pu mobility. 240Pu/239Pu is consistent among location, depth, and season and identical to upstream sediments implying insignificant dilution of the Pu released by atmospheric fallout signatures. While seasonal stratification remobilizes radionuclides on a seasonal basis, much of the Pu and Cs persist in the Pond B system sixty years after release. Prepared by LLNL under Contract DE-AC52-07NA27344.

Log 334. **THE VLADIS INITITIVE: INCLUSIVE VIRTUAL ENGAGEMENTS IN RADIOCHRONOMETRY**

Brennecka G.A. (1, P), Treinen K. (1), Adena K. (2), Kayzar-Boggs T. (3), Bavio M. (4), Keegan E. (2), Kips R. (1), Davydov J. (3), Denton J. (3), Eppich G. (1), Inglis J. (3), M. Samperton K. (5) - (1) Lawrence Livermore National Laboratory (2) ANSTO, Australia (3) Los Alamos National Laboratory (4) International Atomic Energy Agency (5) Savannah River National Laboratory (P) Presenting Author.

The Virtual Laboratory on Age Dating for Investigation Support (VLADIS) is a consortium of researchers from the U.S. Lawrence Livermore National Laboratory (LLNL) and Los Alamos National Laboratory (LANL), the Australian Nuclear Science and Technology Organisation (ANSTO), and Argentina's National Atomic Energy Commission (CNEA). The VLADIS initiative was launched in October 2020 with the goal of creating and maintaining an online platform for practitioners of radiochronometry in the field of nuclear forensics and investigation support. This initiative was born out of LLNL and LANL's successful and long-running collaboration with ANSTO through the U.S. Department of Energy's Office of Nuclear Smuggling Detection and Deterrence (NSDD). VLADIS aims to facilitate discussion between subject matter experts, new staff, and partner agencies around the ever-evolving best practices of radiochronometry within the primarily online environment required by the pandemic. Whereas the group is organized by researchers at LLNL, it is facilitated by nuclear forensics technical analysts from all participating laboratories, including experts in chemistry, mass spectrometry, data analysis, and nuclear forensic signature evaluation. This initiative is not meant to replace in-person visits and technical exchanges between laboratories, rather, VLADIS will complement overall engagement between in-person meetings. VLADIS is being used as a pilot project for virtual information sharing and could be redesigned for use with other partner countries covering topics beyond radiochronometry.

Log 335. **GEOCHEMICAL CHARACTERIZATION OF WATER MASSES AND THEIR ROLE IN Cs-137 DISTRIBTUION AROUND THE EMPEROR SEAMOUNTS IN THE NW PACIFIC OCEAN**

Dulai, H. (1,P), (1) University of Hawai'i at Manoa. (P) Presenting Author.

A combination of radionuclides has been used to characterize water masses and their role in the distribution of anthropogenic radionuclides around 5 seamounts in the Emperor Seamount Chain. A total of 5 depth profiles were produced for Cs-137, Ra-226, and the Ra-228&Th-228 parent daughter pair along the flanks of 5 seamounts (Suiko, Nintoku, Yomei, Koko, and Jingu Seamounts). Cruise delays resulted in no measurable Cs-134 by the time of sampling. Cs-137 depth profiles were similar for Suiko, Nintoku and Jingu Seamounts showing presence to ~ 1000 m depth due to atmospheric inputs and ocean mixing. At Koko there was a well-mixed profile in the surface 500 m and Cs excess reached to 1500 m depth. Cs profiles were compared to Ra-226, which showed great similarity to published North Pacific profiles but with excess radium in the upper 500 m at Suiko and Yomei, and a significant deficit below 2000 m at Jingu. The profile at Koko showed a well-mixed surface water similar to Cs-137. Ra-228 patterns were greatly affected by the seamounts, which are sources of Ra-228. Notable were subsurface Ra-228 peaks in most profiles. Th-228, a product of Ra-228 is particle reactive and occurs in disequilibrium with Ra-228. Th-228 followed Ra-228 patterns in the surface water where Ra-228 has been added near shelf

regions, while "newly" added Ra-228 at the depth of the seamount peaks was not accompanied by Th-228 nor Cs-137. We will present the vertical profiles of these radioisotopes and illustrate how they were used to determine vertical and horizontal mixing patterns of water masses and Cs-137 along our transect.

Log 336. **UNCERTAINTY ANALYSIS OF NEAR-FIELD ANTINEUTRINO-BASED SAFEGUARDS**
Dunbrack, M.T. (1,P); Erickson, A.S. (1). (1) Georgia Institute of Technology. (P) Presenting Author.

The International Atomic Energy Agency (IAEA) currently relies heavily on surveying nuclear facilities and inventories to ensure that special nuclear material pathways are correct and complete. This process, conducted through on-site inspections, draws a significant amount of the limited resources from the IAEA. Through implementing near-field antineutrino detection systems, variations in reactor core fissile inventories can be detected without the need of expensive and invasive inspection. One possible system, the Reactor Evaluation Through Near-Field Antineutrinos (RETINA) system, compares the real-time antineutrino flux to previously simulated fluxes to continuously monitor reactors. This proposed system, however, still needs improvement to detect slight antineutrino spectra variations within three months of diversion. Uncertainty mitigation is one possible method to improve this system sensitivity. In this work, we analyze the uncertainty within the RETINA system for a diverse set of next generation reactor designs. Our results indicate that the leading causes of uncertainty include background uncertainty, antineutrino yield uncertainty, reactor power uncertainty, and systematic detector uncertainty. The magnitude of these relative uncertainties were highly depended on the antineutrino energy and the reactor design.

Log 337. **ACTINIDE AND FISSION PRODUCT ANALYSIS OF SPENT NUCLEAR FUEL PARTICLES**

M. R. Savina (1,P), B. H. Isselhardt (1), D. Z. Shulaker (1), M. Robel (1). (1)Lawrence Livermore National Laboratory. (P) Presenting Author.

Reconstructing the irradiation history of spent fuel can be daunting due to the high activity of the samples, coupled with widely varying isotopic compositions with position in the reactor. This places a premium on extracting maximum information from small samples. We measured the isotopic compositions of U, Pu, Am, Sr, Mo, and Ru in ten-micron cubes of spent nuclear fuel using a resonance ionization mass spectrometry method that removes or resolves isobaric interferences at, e.g., 238U/238Pu, 241Pu/241Am, and 90Zr/90Sr, without elemental separation. The Sr results linked the samples by showing that all came from the same discharge event, while correlations between the isotopic compositions of the three actinides showed trends in burnup and neutron capture correlated with sampling positions in the reactor core. All but Sr show a strong skin effect, in which the isotopic compositions of the core and near-edge regions of the fuel pellets differ due to differences in the incident neutron energy spectrum. While no direct elemental concentration measurements were made, the ability to measure the 238Pu in the presence of a 7400-fold excess of 238U in these small solid samples enabled an estimate of the enhancement in Pu production at the rim of the pellet due to the skin effect. This work performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344. Supported by the National Nuclear Security Agency Office of Defense Nuclear Nonproliferation Research and Development. LLNL-ABS-83016

Log 339. **LARGE DYNAMIC RANGE MEASUREMENTS VIA SIMS AND TIMS**

Cunningham, H.S. (1, P); Zimmer, M.M. (1); Peres, P. (2); Gajos, N. (1); Springer, K. (1); Denny, A. (1); Bowen, J (1); Sievers, N.E. (1); Carman, A (1). (1) Pacific Northwest National Laboratory. (2) Cameca Instruments. (P) Presenting Author.

[36]Cl produced during nuclear explosions has been measured globally in groundwater and glaciers. Traditionally quantification of 36Cl/Cl has been limited to accelerator mass spectrometry (AMS) due to relatively low abundances of 36Cl in the environment. We demonstrate that modern mass spectrometers including LG-SIMS (large geometry secondary ionization mass spectrometry) and thermal ionization mass spectrometry (TIMS) can quantify 36Cl/35Cl down to 1x10-10, well beyond the dynamic ratio that most analysts attempt. The limiting factor in these measurements is the resistance of the faraday cup and not instrumental capabilities such as mass resolving power, transmission, or sample ionization. In a pure matrix such as NaCl, we can measure on the LG-SIMS small volumes (400 μm3) with per mil precision on 37Cl/35Cl due to reasonable ionization efficiency (3-4%). We demonstrate the measurement of 36Cl/35Cl = 7x10-7 for irradiated NaCl and 1x10-10 (i.e. detection limits) for unirradiated NaCl. We model the effects of ionization efficiency, volume, and dynamic ratio necessary for analysis of other relevant nuclear systems.

Log 340. **BETWEEN INTERACTIVE SCREEN EXPERIMENTS AND FLIPPED CLASSROOM: IMPROVING THE TEACHING AND LEARNING OF RADIOCHEMISTRY AND RADIOPHYSICS THROUGH COOPERATIVE PROJECTS**

Dirk Brockmann-Behnsen, Lars Weiß, Jan Vahlbruch, Clemens Walther, Gunnar Friege

Two innovative approaches, a learning tool and a teaching concept, were implemented in cooperation projects by radiochemists, radiophysicists and science educators: The development of Interactive Screen Experiments and the use of a Flipped Classroom setting in a basic radioscience course. Interactive Screen Experiments are digital learning environments that in principle contain photorealistic recordings of all states of a real experiment (input: all possible parameter settings, output: all results). Learners can freely select the parameters of an experiment and study the result according to the chosen parameters. Interactive Screen Experiments are neither real experiments nor computer simulations. Unlike remote-controlled experiments, they can be used by many learners at the same time and require no maintenance costs. The criticism of a traditional lecture-exercise approach at universities and the hopes placed in the increasingly widespread Flipped Classroom Approach are great. The latter approach includes a self-learning phase for the students, often supported by digital tools, and a presence phase to deepen the content in which lecturers act as learning advisors. The Flipped Classroom Approach was implemented in a university radioscience course and evaluated. Conclusions for teaching, especially with regard to student teachers and science students, are drawn.

Log 341. **PRODUCTION AND CHARACTERIZATION OF STANDARD PARTICLES FOR rL-SNMS CALIBRATION**

Bister, S. (1); Hanemann, P.(1,P); Raiwa, M.(1); Reinhard, S.(1); van Eerten, D.(1); Walther, C.(1). (1) Institute of Radioecology and Radiation Protection, Leibniz University Hannover. (P) Presenting Author.

Resonant laser secondary neutral mass spectrometry (rL-SNMS) is a non-destructive method that combines high sensitivity and resolution of ToF-SIMS with high element selectivity of resonant laser ionisation. One main application is the determination of isotope ratios on individual micro particles of spent nuclear fuel from the Chernobyl exclusion zone (CEZ). Standard materials are needed for dealing with isobaric interferences such as ^{238}U and ^{238}Pu as well as differences in the laser ionisation and sputter efficiencies for different elements and isotopes. This work produced particles via Fe-coprecipitation to achieve homogeneous U and Pu bearing particles. Isotopes with different mass numbers were used to determine relative ionisation efficiencies without isobaric interferences. The homogeneity of U and Pu in the particles was confirmed by ToF-SIMS and EDX measurements. On several particles Pu-resonant measurement were performed to investigate the suppression of non-resonant U. It was shown that the suppression is high enough to be able to detect ^{238}Pu in a particle with a 10^5 higher ^{238}U content, as found in "hot particles" from the CEZ. RL-SNMS measurements of produced particles containing only different U isotopes allowed the investigation of isotope effects in the resonant laser ionisation.

Log 342. DEVELOPMENT OF ENGINEERED, NANO-POROUS ION EMITTERS (NANO-PIES) FOR ACTINIDE ANALYSIS BY TIMS

McHugh, K.C. (1, P); Makovsky, K.A. (1); Barpaga, D. (1); Sinwell, M.A. (2); Krogstad, E.J. (1); Brown, C.F. (1). (1) PAcific Northwest National Laboratory. (2) University of Iowa. (P) Presenting Author.

Results are presented for the development of engineered, nano-porous ion emitters (nano-PIES) for trace actinide analysis by thermal ionization mass spectrometry (TIMS). In this work, metal organic frameworks (MOFs) are used as precursors to generate high surface area structures with controlled synthetic chemistries. By capitalizing on the inherent tunability of MOFs (i.e., adjustable surface area, metal incorporation, and targeted sorption behaviors), they have potential to act as an all-in-one platform to construct nano-PIEs and deconvolute the tedious complexity of traditional techniques (e.g., resin beads, carburization, etc.). Preliminary results show promise for these materials to be customized for this application. Using neodymium as a proxy for actinides, initial experiments demonstrated analyte adsorption and a comparable performance to traditional loading techniques with the added advantage of time-saved in sample preparation. In most MOFs tested thus far, complete uranium (U) uptake occurred within 10 minutes. Sample utilization efficiency experiments have been designed to vary surface area, porosity, and metal composition of the MOF without controlling for sample retention on the filament. A comparison of MOFs with the same base structure (MOF-74 series) showed that, as expected, at the same pore volume/surface area, the MOF with the higher work function metal generated more ions. Additionally, for MOFs containing similar work function metals, increasing pore volume/surface area resulted in improved sample utilization efficiencies.

Log 343. CHARACTERIZATION OF PIPS DETECTORS FOR MEASUREMENT OF RADIOXENON

Sobel, P.W. (1,P); Biegalski, S.R.(1). (1) Georgia Institute of Technology. (P) Presenting Author.

The International Monitoring System arm of the Comprehensive Nuclear-Test-Ban Treaty Organization utilizes sampling of atmospheric radioxenon to scan the world for nuclear testing events. Radioxenon has a natural abundance in the atmosphere but is also a commonly released byproduct of fission reactions involving heavier atoms. Due to its inert nature and short half-life, the gas makes an excellent indicator of recent nuclear activity. Given that, differentiating the signatures released from nuclear power plants, nuclear testing, and medical isotope production facilities requires a fast, high resolution detection system. Current designs utilized in the field involve plastic scintillator cells run in coincidence with Sodium Iodide (NaI) or High Purity Germanium (HPGe) detectors. The use of plastic scintillators as electron detectors exhibit some issues in comparison to other materials. The energy resolution tends to be lower and the diffusion of gas into the plastic creates long-term signal degradation. Silicon detectors are analyzed as an alternative to plastic scintillators for beta detection based on their higher energy resolution and lowered memory effect. A radioxenon detection setup was created with the intent of measuring radioxenon samples using coincidence counting between silicon and NaI detectors as well as silicon and HPGe. The absolute efficiency of both setups is calculated and compared.

Log 344. **COINCIDENCE MEASUREMENTS OF RADIOXENON USING PASSIVE IMPLEMENTED PLANAR SILICON (PIPS) DETECTOR**

Wilson, C.R. (P). (P) Presenting Author.

The Comprehensive Test Ban Treaty (CTBT) which prohibits nuclear weapons testing, implemented the International Monitoring System (IMS), which uses isotopic ratios of radioactive noble gases, 135Xe, 133Xe, 133mXe, and 131mXe to verify global adherence to the treaty. Four different systems, ARIX, ARSA, SAUNA, and SPLAX have been designed to fulfill CTBT requirements for monitoring noble gases. All of these systems operate using beta gamma coincidence detection. The decay process of Xenon isotopes occurs in two steps: emitting a beta particle and gamma ray either simultaneously or in an extremely short time period and then emitting a conversion electron and an X-ray. The emission of the beta particle and X-ray also occurs simultaneously or in a very short time window. Coincidence counting between two detectors must be used to ensure detectors are only picking up these simultaneous counts that are characteristic of Xenon decay. The purpose of this experiment was to measure the gamma rays, beta particles, X-rays, and conversion electrons from all four isotopes of interest using a Passivated Implemented Planar Silicon Detector and NaI(Tl) detectors in coincidence mode. The experimental set up was configured to emulate systems used for treaty verification. From these measurements, the relative activities and concentrations of each isotope were determined.

Log 345. **ULTRASHORT-PULSED LASER FILAMENT EXCITATION OF CHLOROPHYLL FLUORESCENCE IN ALGAE AS A BIOSENSOR FOR NUCLEAR ACTIVITY**

Finney, L.A. (1,2,P), Peskosky, N.(1,2), Skrodzki, P. J.(1,2), Burger, M.(1,2), Nees, J.(2), Krushelnick, K.(1,2), Jovanovic, I.(1,2). (1) Nuclear Engineering and Radiological Sciences University of Michigan Ann Arbor. (2) Gérard Mourou Center for Ultrafast Optical Sciences University of Michigan Ann Arbor. (P) Presenting Author.

Living plants have been suggested as potential biosensors for nuclear activity monitoring because of their sensitivity to environmental conditions. We demonstrate that ultrashort-pulsed laser filament-induced fluorescence of chlorophyll in the green alga Chlamydomonas reinhardtii is a promising method for remote, in-field detection of stress from exposure to nuclear materials. This method holds an advantage over broad-area surveillance, such as solar-induced fluorescence monitoring, when targeted excitation of a specific plant would improve the detectability, for example when local biota density is low. We find that the chlorophyll fluorescence lifetime and carotenoid content increase upon exposure to uranium over 24 hours. The combination of increased carotenoid content with increased fluorescence lifetime signifies a decrease in non-photochemical quenching and production of radical oxygen species, as evidenced in the steady-state absorption spectrum. We identify the temporal profile of the chlorophyll fluorescence to be a distinguishing feature between uranium-exposed and unexposed algae. Discrimination of uranium-exposed algae is possible at a distance of 35 m with a single laser shot and a modest collection system, as determined through a combination of experiment and propagation of the distance-dependent uncertainty in the discrimination factor. Illustrating the potential for remote detection, we find that detection over 125 m would require 100 laser shots, commensurate with the detection time on the order of 1 s.

Log 346. **NOVEL GROSS ALPHA/BETA GROUND DEPOSTION METHOD**

Griggs, J. (1,P); Fournier, S. (2); Allen, M. (2). (1) National Analytical Radiation Environmental Laboratory. (2) Sandia National Laboratories. (P) Presenting Author.

The US EPA in partnership with US DOE and in partnership with and funding from DHS's Federal Emergency Managment Agency(FEMA) Nuclear Incident Response Team program has developed a novel method for measuring gross alpha/beta radioactivity in ground deposition samples taken after a radiological or nuclear incident Solids, as received from the field, are quickly dried and homogenized in a single container A hydraulic press is used to form an infinitely-thick pellet that is then analyzed by gas-flow proportional counting The method minimizes sample handling while maximizing the amount of sample measured. Direct measurement of solids in a large, reproducible test-source geometry facilitates representative, intercomparable results.

Log 347. MEASUREMENT OF CORROSION PRODUCT FILTRATION IN A FLUORIDE MOLTEN SALT SYSTEM USING INDUCTIVELY COUPLED PLASMA-MASS SPECTROMETRY

Head, T.L. (1,P); Babb, E.M. (1); Berry, A.M. (1); Bonamie, E.J. (1); Howe, R. (1); Kennedy, T. (1); Pamplin, K.L. (1); Sanchez, B. (1); Watson, T.S. (1); for the NEXT Collaboration (1); Robb, K.R. (2); (1) Abilene Christian University, (2) Oak Ridge National Laboratory, (P) Presenting Author.

The Nuclear Energy eXperimental Testing (NEXT) Lab at Abilene Christian University is advancing the technologies needed to build a Molten Salt Research Reactor (MSRR) to pave the way for a fleet of molten salt reactors that will meet the world's critical needs for energy, industrial heat for water purification, and medical isotopes. Advanced reactor technologies employing molten salts will use fuel more efficiently, and have other potential benefits such as the ability to harvest medical isotopes and provide industrial heat. Control of impurities in the salt both before and after introduction of salt into the reactor will require the ability to filter the salt mechanically. Effective filter designs require information about the filter hydraulic properties, as well as an understanding of what materials can be mechanically filtered effectively. This work describes an apparatus built at NEXT Lab to measure hydraulic properties of filter media with a eutectic mixture of LiF, NaF, and KF molten salt, and presents preliminary analysis of corrosion product mechanical filtering using Inductively Coupled Plasma Mass Spectrometry (ICP-MS).

Log 348. FORENSIC SIGNATURES FROM LASER ISOTOPE SEPARATION

Burns, H.S. (1,P); Biegalski, S.R. (1). (1) Georgia Institute of Technology. (P) Presenting Author.

It has long been recognized that laser isotope separation (LIS) poses a proliferation risk, since the lower energy requirements and smaller physical parameters associated with its efficiency makes an LIS facility harder to detect. This work introduces a theoretical analysis of the Atomic Vapor Laser Isotope Separation (AVLIS) process intended to assist regulatory agencies in detecting and inspecting AVLIS facilities that enrich uranium. Using the process model developed at LLNL, the stripping efficiency and nonselective pickup of a theoretical AVLIS device are calculated based on the product and tails assays. Assuming a feed of natural uranium, the product assays of U-234 and U-238 are calculated for enriched uranium with varying U-235 product assays. The analysis demonstrates that AVLIS-enriched uranium can be identified by this isotopic signature. Similarly, this work demonstrates that the feed assays of U-234 and U-238 can be calculated from the product assays, potentially identifying the feed source due to natural variations in U-234 assay. The decay of U-234 into Th-230 is used to estimate the time of last enrichment. These factors combine to help inspectors determine the origin of an intercepted sample of enriched uranium and ensure their ability to verify operational parameters at a hypothetical AVLIS enrichment plant.

Log 349. **SPATIAL AND TEMPORAL DISTRIBUTION OF ATMOSPHERIC TRITIATED WATER VAPOR IN MAINLAND CHINA**

Feng, B. (1, P); Steinhauser, G. (1); Zhuo W.H. (2); Chen, B.(2). (1) Institute of Radioecology and Radiation Protection, Leibniz University Hannover (2) Institute of Radiation Medicine, Fudan University. (P) Presenting Author.

The diminishing effects of the legacy of "bomb tritium" introduced by atmospheric weapon tests and the potential significant release from the future anthropogenic sources (e.g., Fukushima contaminated wastewater) together makes the current a golden window for studying environmental tritium baseline. China has been considered one of the most ideal regions for environmental tritium research in the world. Yet, the spatial and temporal profiles of current atmospheric tritiated water (HTO) vapor in this region remain unclear.

Here, we conducted an atmospheric HTO investigation within 34 cities in 30 provinces of mainland China during 2017-2018 by using passive sampling and liquid scintillation counting.

Overall, the atmospheric HTO levels in China ranged from lower than 0.18 Bq/L to 5.5 Bq/L. Spatially, the atmospheric HTO levels presented a strong positive correlation with latitude and the distance to the proximal coastline. Temporally, the atmospheric HTO levels peaked in the spring in most cities while reached to their trough in the summer season. Combining the quantitative datasets, including the stratosphere-troposphere exchange fluxes, the dynamic distance between the surface to the troposphere, and the fraction of oceanic or continental air mass, our correlation analysis suggested the atmospheric HTO dynamics are attributed to the combined effects of the above factors.

Given the importance and urgency of acquiring environmental tritium baseline, we appeal cohesive cooperation in the scientific community to reveal the global tritium baseline at current.

Log 350. REMOTE DETECTION OF ALPHA-EMITTING RADIONUCLIDES IN THE ENVIRONMENT

Klose, A. (1,P); Walther, C. (1); Krasniqi, F.S. (2); Luchkov, M. (2); Dangendorf, V. (2). (1) Leibniz University of Hannover   (2) PTB Braunschweig (P) Presenting Author.

Environmental contaminations by purely alpha emitting radionuclides are hard to detect. Independent of whether the release originated from an accident or happened on purpose, protective measures need to be based on knowledge of the contamination level. Measurements are performed by hand due to the short range of alpha particles in air. This long lasting procedure exposes the emergency team to considerable risk. In the project RemoteAlpha a new optical detection system is developed that should overcome these problems. Alpha particles in the atmosphere interact with its gases, mainly with nitrogen due its share of 78%. The ionization of the gases causes many secondary electrons, which can excite the electrons of the outer shell of nitrogen. By relaxation of the shell electron, a UV-photon is emitted. One alpha particle generates 19 UV-photons. These UV-photons can be measured with the new system.1 Mounted on a drone a large area can be scanned in a short time and a mapping of contamination hotspots is possible. We report on first tests of the optical system using well-calibrated samples ranging from commercially available certificated samples to environmental samples such as pitchblende. Sensitivity and selectivity of the detection system are iteratively tested and improved. 1.Environmental contaminations by purely alpha emitting radionuclides are hard to detect. Independent of whether the release originated from an accident or happened on purpose, protective measures need to be based on knowledge of the contamination level. Measurements are performed by hand due to the short range of alpha particles in air. This long lasting procedure exposes the emergency team to considerable risk. In the project RemoteAlpha a new optical detection system is developed that should overcome these problems. Alpha particles in the atmosphere interact with its gases, mainly with nitrogen due its share of 78%. The ionization of the gases causes many secondary electrons, which can excite the electrons of the outer shell of nitrogen. By relaxation of the shell electron, a UV-photon is emitted. One alpha particle generates 19 UV-photons. These UV-photons can be measured with the new system.1 Mounted on a drone a large area can be scanned in a short time and a mapping of contamination hotspots is possible. We report on first tests of the optical system using well-calibrated samples ranging from commercially available certificated samples to environmental samples such as pitchblende. Sensitivity and selectivity of the detection system are iteratively tested and improved.   1. Sand, J. et al. Stand-Off Radioluminescence Mapping of Alpha Emitters under Bright Lighting. IEEE Trans. Nucl. Sci. 63, 1777–1783 (2016).

Log 352. ELUCIDATING MOLECULAR SPECTRAL SIGNATURES IN NANOSECOND LASER-INDUCED PLUTONIUM SURROGATE PLASMAS

Kwapis, E.H. (1,P); Hartig, K.C. (1); (1) University of Florida. (P) Presenting Author.

A better understanding of the physical and chemical properties of actinides and nuclear-relevant materials released into the environment is essential to detecting and responding to nuclear explosion events. Optical spectroscopy techniques coupled with laser ablation provide a laboratory-scale method to study laser-produced plasmas (LPPs) that are representative of high explosives and nuclear fireballs. Specifically, laser-induced breakdown spectroscopy provides standoff, high-precision capabilities towards the elemental, molecular, and isotopically resolved characterization of actinides and nuclear explosions. However, interpreting results from optical spectra, which are sensitive to complex spatio-temporal conditions of the LPP, can be a challenging process. Chemical reactions within the plasma lead to the formation of molecular species, which are greatly influenced by the environment and can considerably impact particle formation and debris distribution in fireballs. Extensive studies into the spectroscopic characterization and plasma chemistry of uranium have been performed, yet there has been little investigation into the properties of plutonium LPPs despite marked differences in chemistry between the elements. To further development on remote sensing systems towards response readiness in the event of a nuclear detonation, the fundamental physics and chemistry of laser-produced plutonium plasmas needs to be studied further. Through the use of a common Pu surrogate (cerium), we will demonstrate the evolution of atomic and molecular species within cerium LPP plumes.

Log 353. **PREPARATION OF AN Ac-225 GENERATOR FROM AN OLD U-232/233 SAMPLE AND SUCCESSFUL Ac-225 RADIOLABELING OF A NOVEL DOTA 1,2,4,5-TETRAZINE DERIVATIVE**

Pressler, M. (1, 2, P); Denk, C. (2, 3); Scheibelreiter, V. (2); Kuba, W. (2); Mikula, H. (2).
(1) Institute of Atomic and Subatomic Physics, TU Wien, 1020 Vienna, Austria. (2) Institute of Applied Synthetic Chemistry, TU Wien, 1040 Vienna, Austria. (3) Center for Labelling and Isotope Production, TRIGA Center Atominstitut, TU Wien, 1020 Vienna, Austria. (P) Presenting Author.

Ac-225 has been shown to be a very promising candidate for targeted alpha therapy due to its outstanding decay characteristics but its limited availability remains a challenge for its application and further in-depth investigation. An U-233 sample also containing U-232 was used for the preparation of an Ac-225 generator. From this uranium sample the ingrown Th-229 (and Th-228 from U-232) were extracted using an anionic exchange process. The decay products of Th-229 (especially Ra-225 and Ac-225) and of Th-228 were allowed to build up on the generator for several weeks. To separate Ac-225 from the decay products of both decay series multiple ion exchange and absorption chromatography steps were undertaken. Additionally, to an Ac-225 fraction, a radium fraction was collected to give another Ac-225 fraction after several days. All fractions were investigated towards their radioisotopic purity using radioanalytical methods (alpha spectroscopy and gamma spectroscopy). The 1,2,4,5-tetrazine moiety is a powerful building block in bioorthogonal click chemistry, which allows fast, selective and biocompatible modification of targets in and ex vivo. This click reaction is of great interest in nuclear medicine, especially for pre-targeting approaches and was already successful utilized for both diagnostic and therapeutic isotopes. The synthesis and radiolabeling of a 1,2,4,5-tetrazine substituted DOTA derivative with Ac-225 will be presented and the results of the successful click reaction with minor-TCO-PEG4 (trans-cyclooctene) which was monitored using radio-TLC will be shown.

Log 354. COMPARISON OF ^{239}Pu- and ^{242}Pu-COLLOIDS IN THEIR STRUCTURE AND INFLUENCE OF RADIOLYSIS ON STABILITY

Reinhard, S. (1,P); Steppert, M. (1); Hedberg, M. (2); Persson, I. (3); Ekberg, C. (2); Walther, C.(1). (1) Leibniz University Hannover, Institute of Radioecology and Radiation Protection. (2) Chalmers University of Technology, Chemistry and Chemical Engineering, Nuclear Chemistry. (3) Swedish University of Agricultural Sciences, Department of Molecular Sciences. (P) Presenting Author.

Plutonium chemistry in aqueous solutions is amongst the most fascinating of all elements. For instance, up to four oxidation states can be present simultaneously. Solubility, mobility, sorption etc. strongly depend on the oxidation state. One "phase" of special interest are Pu colloids relevant for instance in the context of safety assessments of repositories for nuclear waste. Many studies have dealt with the characterization of colloids using a wide range of advanced physical and chemical methods, but with very different conclusions regarding the formation process, structures and stability. These studies initiated colloid formation by different routes, different acidic media and different plutonium isotopes. The latter influences the specific activities and hence radiolysis. This study aims at understanding these results with respect to the different conditions of Pu colloid formation. The effect of the specific activity on ageing processes and structural changes of the plutonium colloids is investigated by studying solutions of 242Pu and a mixture of 239Pu and 238Pu. Furthermore, the effects of the choice of acid and different formation methods like slow dilution of a Pu(IV) solution with weak acids on one hand or a harsh increase of the pH by adding NaOH on the other will be elucidated in this study. In order to get insight of the formation process and structures of plutonium colloids, EXAFS and M5 edge HERFD-XANES measurements are implemented. The ageing process is traced by UV/Vis spectroscopy.

Log 355. DIRECT SPECIATION OF RADIONUCLIDES IN PLANT PARTS: THE JOURNEY OF EUROPIUM IN SECALE CEREALE L. ANALYZED WITH MASS SPECTROMETRIC, SPECTROSCOPIC AND MICROSCOPIC TECHNIQUES

Stadler, J. (1,P); Vogel, M. (2); Steudtner, R.(3); Damböck, T.(1); Drobot, B.(3); Walther, C.(1). (1) Leibniz University Hannover, Institute of Radioecology and Radiation Protection. (2) VKTA, Strahlenschutz, Analytik & Entsorgung Rossendorf e.V.. (3) HZDR Innovation GmbH. (P) Presenting Author.

^{241}Am is an important radionuclide and currently the dominating α-emitter in the Chernobyl exclusion zone due to continuous buildup from its precursor 241Pu by beta decay. Am transfers into plants via contaminated soil and eventually enters the human food chain. For a better understanding of the uptake mechanism on a microscopic scale, its chemical speciation has to be investigated. In this work, Secale cereale L. was treated with Eu as homologue for the trivalent Am. A liquid nutrition medium was used to investigate the chemical form before the contamination process with theoretical calculations and Electrospray Ionization Mass Spectrometry (ESI MS) measurements. After harvesting the treated plant, direct speciation was done by using Raman spectrometry as imaging technique and time-resolved laser-fluorescence spectroscopy for the chemical environment analysis of Eu. These techniques are supported by plant extraction and speciation with ESI MS. For the first time possible Eu complexes with malate and citrate were identified after extraction from the plant root and there is indication for molecular bonding to protein and DNA/RNA. In addition, treatment with ubiquitous complexing agents in soil, such as EDTA and malonate, was indicating towards changes in the uptake mechanism of the Eu. With our methods, we provide first molecular-chemical insight into the direct uptake of Eu, illuminating previously unknown complexes. Further investigation will be carried out with Am, in order to provide detailed prove of extractions methods and measurement techniques for the actinide.

Log 356. **NEUTRON IMAGING OF LIQUID-GAS MENISCUS FOR PARA-XYLENE EXPOSED TO 100 BAR METHANE**

Vopicka, O. (1,P); Durdakova, T-M. (1); Cihal, P. (1); Boillat, P. (2); Trtik P. (2,P). (1) University of Chemistry and Technology, Prague, Czechia (2) Paul Scherrer Institute, Villigen, Switzerland. (P) Presenting Author.

Time-resolved neutron radiography enables the observation of dynamic processes in opaque high-pressure vessels. This contactless method allowed us to determine the shapes of normal and subcooled liquid bodies in pressurized cells. We report on the experiments in which a liquid body of perdeuterated para-xylene (C_8D_{10}) was exposed to a step of methane (CH_4) pressure from 1 bar to 100.4 bar. The system was studied at two temperatures: 7 °C (at which para-xylene is a subcooled liquid) and 20 °C (at which para-xylene is liquid). While the meniscus shape changed abruptly at short times, it remained practically constant over the time of the experiment (from 2 to 90 minutes) despite methane diffusion occurring through the interface. Thanks to the central symmetry of the experimental set-up, the tomographic reconstruction of the studied liquid, methane concentration and liquid density at the interface could be derived from the individual radiographies. Overall, we show that methane adsorption on the phase interface of the test liquid is a fast process that is independent of strong concentration gradients (and thus fluxes) in both normal liquid and subcooled liquid states.

Log 357. **NEUTRON IRRADIATION OF POLYDIMETHYLSILOXANE (PDMS): SWELLING IN XYLENE, RAMAN SPECTRA, MECHANICAL PROPERTIES**

Vopicka, O. (1,P); Durdakova, T-M. (1); Kral, M. (1); Hrdlicka, Z. (1); Vögele, A. (2); Eichler, R. (2); Trtik P. (2,P). (1) University of Chemistry and Technology, Prague, Czechia (2) Paul Scherrer Institut, Villigen, Switzerland. (P) Presenting Author.

PDMS (polydimethylsiloxane, silicone rubber) is a very radiation resistant polymer used, for instance, as a sealant. In the field of membrane separations, PDMS is used as a membrane-forming material. PDMS becomes brittle at high doses of gamma rays, which leads to a decrease of the (extractable) sol content. As a sensitive technique for detecting changes in the abundance of interchain cross-links, swelling in good solvents, here ortho-, meta-, para-xylene, was studied for samples irradiated (1.5 to 45 min) in the NAA irradiation facility in the SINQ spallation source at the Paul Scherrer Institute. Mechanical characteristics (dynamic mechanical analysis), sol content and Raman spectra did not show detectable changes of the polymer upon combined neutron irradiation (energies ranging from 1 meV to 100 MeV, total flux more than $1E13$ n cm-2 s-1) and gamma irradiation (energies ranging from 0.01 MeV to 100 MeV). However, the swelling in the xylenes rose (cross-links decayed) for short exposure times (from 1.5 to 30 minutes) while it decreased to approximately its initial value after 45 minutes of exposure (cross-links recovered). Interestingly, the solvent uptake differed for different individual xylene isomers thus enabling the enhancement of the para-xylene recovery for the industrially relevant processing of petrochemical reformate. Financial support of MEYS Czechia, projects A2_FCHI_2021_021 and 21 SVV/2021, is acknowledged.

Log 359. COMPARISON OF METHODS FOR MONITORING RECOVERY RATES IN Sr-SEPARATION

Blenke, T. (1,P), Schäfer,T. (1), Steinhauser, G. (1). (1) Leibniz University Hannover. (P) Presenting Author.

Strontium-90 ($T_{1/2}$= 29 a) is one of the most important fission products due to its high fission yield, long half-life, and health risks if ingested or incorporated in the human body. Conventional analytical techniques involve a chemical separation by using an extractive resin from other radionuclides, such as cesium-137, and β counting by liquid scintillation counting (LSC). Several methods are available to monitor the recovery rate (yield) of the chemical separation. In this work, we compare and discuss the various methods. These include the monitoring of stable strontium-88 via mass spectrometry (MS) or stronium-85 ($T_{1/2}$= 65 d) with simultaneous evaluation together with stronium-90 in the LSC. This requires an additional LSC measurement because strontium-85 needs to be evaluated right after the separation due to its short half-life and strontium-90 needs two weeks for the ingrowth of its daughter yttrium-90. Further possibilities to monitor the recovery rate include adding stable strontium and monitoring it by optical emission spectroscopy (OES) or strontium-85 and a gamma measurement (before and) after the separation. The choice of method should take into account aspects such as the half-life of the nuclides, availability of the instruments and quantity of strontium measurements.

Log 360. RADIONUCLIDE REMEDIATION USING NATURAL ASSOCIATION PROCESSES

Blenke, T. (1,P), Zok, D. (1,P), Steinhauser, G. (1). (1) Leibniz University Hannover. (P) Presenting Author.

The end of the nuclear energy production of Germany in 2022, as well as the decommissioning of old facilities led to a huge amount of nuclear waste materials. To minimize at least some part of a high volume but low activity component, the applicability of phytoremediation of soils is investigated. The RENA project - Biological radionuclide removal by using natural association processes – has been launched to this topic. The aim of the project is to develop a process for the ex situ treatment of radionuclide-contaminated soils in a dismantling area in Germany. In contrast to previous remediation projects, only natural association processes are used. To this end, the potential of solely biological organisms such as plants and fungi for mobilizing and removing radionuclides from soils is studied. The aim is a significant volume reduction of medium- and low-level radioactive waste for a final repository. Our investigations are based on already excavated and appropriately pre-treated soils. Additionally, there will be in situ test on an acre in the Chernobyl exclusion zone. The holistic approach, which combines soil microbiological, mineralogical, geo-chemical and radio-chemical aspects, will be prepared for implementation in a reactive transport model. With such a numerical approach, predictions about the efficiency, quantitative influencing factors and especially transferability to other soil materials from deconstruction projects should be possible.

Log 361. SILANIZATION FOR MINIMIZING CATION ADSORPTION IN SOLVENT EXTRACTION SYSTEMS

Maulden, E. (1,P); Wall, D.E.(1); Wall, N.A.(1). (1) University of Florida. (P) Presenting Author.

Tracer cationic metal species loss through retention onto glass vial walls can be problematic in solvent extraction systems. Methods for decreasing metal sorption, including low pH, may not offer viable options. Silanization is a technique used to increase hydrophobicity and decrease cation adsorption to glass materials and has been used sparingly in solvent extraction experimental work. The current work presents the behavior of Eu(III) tracer in components of solvent extraction systems contained in silanized glass vials. Our work demonstrates that silanization is most effective at near-neutral to slightly basic pH in decreasing Eu(III) sorption to glass walls. Polar or aromatic organic solvents outperform non-polar or aliphatic solvents in maintaining the hydrophobic coating. Extractants decrease Eu(III) sorption to glass walls, assuming the extractant is stable under the tested aqueous conditions. Finally, we observed little influence of ionic strength and electrolyte size within the alkali chlorides on the silane coating.

Log 362. SYNTHESIS AND TESTING OF FUNCTIONALIZED CLAY MATERIAL FOR THE SEQUESTRATION OF TECHNETIUM

Maulden, E. (1,P); Gager, E. (1); Nino, J.C. (1); Pearce, C. (2); Szecsody, J.(2); Wall, N.A.(1). (1) University of Florida (2) Pacific Northwest National Laboratory. (P) Presenting Author.

Nuclear waste repository designs require backfill buffer material to contain radionuclides. Technetium, as TcO4-, is an environmentally mobile fission product of concern given the long-lived nature of Tc-99. Commercially available functionalized clay can sequester Tc(VII). The current work focuses on the synthesis and performance of clays intercalated with quaternary amines and Fe for Tc retention. Results demonstrate increased Tc retention for clays functionalized with both Fe and quaternary amines, regardless of alkyl chain size.

Log 363. EVALUATION OF TWO EXTRACTION CHROMATOGRAPHY RESINS FOR SEPARATION OF SCANDIUM AND TITANIUM FOR MEDICAL ISOTOPE PRODUCTION

McLain, D.R. (1,P); Brossard, T.W.(1); De Kruijff, R.M.(1,2); Kankanamalage, P.H.A.(1); Rotsch, D.A.(1). (1) Argonne National Laboratory. (2) Delft University of Technology. (P) Presenting Author.

Scandium-47 can be used in nuclear medicine as a therapeutic-diagnostic, or "theranostic," radioactive medical isotope for malignant cancer tumor detection and treatment. The 47Sc isotope can be produced through the photonuclear reaction 48Ti(γ,p)47Sc by irradiating enriched 48Ti target material. The enriched target material necessary for production is costly; 48Ti – 48TiO2 costs \$1,550/gram, and targets can be >50g (\$77,500). In order to keep costs low, highly efficient separation of scandium from bulk titanium is desired, along with efficient methods for recycling the target material. This research evaluates two different commercially available extraction chromatography resins for their ability to separate scandium from bulk quantities of titanium.

Log 364. AGE DETERMINATION OF Se-75 GAMMA RADIOGRAPHY SOURCE MATERIAL

McLain, D.R. (1,P); Condon, N.J. (1); Brossard, T.W. (1); Tsai, Y. (1); Canaday, J.L. (1). (1) Argonne National Laboratory. (P) Presenting Author.

Two methods were used to evaluate the age of Se-75 sealed source material. Both methods utilized gamma spectroscopy to determine the quantity of parent Se-75 present, but they differed in the technique used to determine the quantity of daughter As-75. In one method, As-75 was quantified using inductively coupled plasma-mass spectrometry (ICP-MS) after using chromatographic techniques to separate the parent and daughter isotopes. In the other method, total As was measured using optical emission spectroscopy (OES), which did not require separation prior to measurement. The OES method was faster and gave slightly lower uncertainties, but required more daughter material to be present because of higher concentration requirements, making it unsuitable for young material. Both methods are hampered by the monoisotopic nature of As, but still gave reasonable model ages that were in agreement with each other.

Log 365. HIGH-ENERGY X-RAY IMAGING FOR NUCLEAR FORENSICS

McLain, D.R. (1,P); Almer, J.D. (1); Sharma, H. (1); DeAngeles, K.J. (1); Condon, N.J. (1). (1) Argonne National Laboratory. (P) Presenting Author.

The nuclear forensics community has long used traditional optical and scanning electron microscopy (SEM) techniques to observe morphological surface features. Recently, interest has increased in determining the sub-surface features of materials in three dimensions. High-energy X-ray 3D imaging techniques using a direct beam (micro computed tomography) or diffracted beam (high energy diffraction microscopy) are well suited to providing qualitative and quantitative sub-surface morphological information and have the added benefit of being non-destructive. This project is ongoing, and aims to produce morphologically different uranium oxide samples for characterization using high-energy x-ray techniques.

Log 366. **SIMULTANEOUS DETERMINATION OF LIGHT ACTINIDES BY ICP-QQQ-MS**

Schramm, L.-M. (1,P); Zok, D. (1); Steinhauser, G. (1). (1) Leinbiz University Hannover. (P) Presenting Author.

Growing applications of nuclear technology require nuclide specific analytical methods to ensure compliance with regulations and safety guidelines. Uranium and plutonium are frequently targeted in nuclear forensics for the determination of their nuclear background. Depending on their utilization purpose, their isotopic compositions may differ. Therefore, nuclear forensics methods allows for the distinction between sources. Owing to their half-lives, mass spectrometry is the method of choice for the determination of isotope ratios. We used ICP-triple quadrupole ICP-MS (ICP-QQQ-MS) for our purpose, in which the first and third quadrupole worked as a mass-filter and the second quadrupole worked as a reaction gas cell. This cell was necessary to reduce interferences of identical mass-to-charge ratios by adding carbon dioxide. Hereby, a mass shift of uranium to higher masses is achieved, while plutonium does not react and remains on the same mass-to-charge ratio. Implementation of chromatographic separations in addition to the instrumental separation inside the ICP-QQQ-MS, interferences from uranium and plutonium can be resolved and both elements can be measured simultaneously. We will present results on optimization parameters for the separation of both analytes and a discussion of the detection limits.

Log 367. **CLIMATE CHANGE STUDIES WITH ACCELERATOR MASS SPECTROMETRY.**

Povinec, P.P. (1,P); Kaizer, J. (1). (1) Comenius University in Bratislava. (P) Presenting Author.

Recent developments in accelerator mass spectrometry (AMS) have been crucial for analysis of cosmogenic radioactive isotopes Be-10 and C-14 in natural archives such as tree rings, ice cores and marine sediments, which have proved to be exceptionally important for better understanding of past climate changes caused by Sun-Earth orbital parameters, as well as by solar activity cycles, and recently also by increases of green-house gases in the atmosphere. The most visible past climate cycles were represented by Milankovitch cycles, as documented by observed Be-10 variations in ice cores. During the past 800 kyr the dominant cycle was due to the variations in the eccentricity of the Earth's orbit (125 kyr period). The exceptional case was a warm period during the Holocene (last 11 kyr), as documented from Be-10 and C-14 records in ice cores and in tree rings. Although the Holocene was generally a warm epoch, there were cool periods as well, such as a Little Ice Age which followed after the Medieval Warm Period. The cool period coincided partly with the Maunder solar activity minimum, confirming that solar cycles affected past climate. High precision C-14 data obtained by AMS analysis of annual tree rings (or even semi-annual rings represented by earlywood and latewood) will be useful source of detail information on past C-14 variations in the biosphere which could be associated with past solar activity cycles and climate changes at least during the Holocene epoch.

Log 368. **OPTIMIZATION OF A hBN-BASED NEUTRON RADIATION DETECTOR ARCHITECTURE FOR SPACE APPLICATIONS**

Almatouq, F.A.(1,P); Vira, A.(1); Connolly, P.(1); Jiang, Z.(1); First, P.N.(1); Orlando, T.M.(1). (1) Georgia Institute of Technology. (P) Presenting Author.

Radiation safety is one of the main concerns for manned space missions. To address this concern, boron-based materials have been proposed as critical components for radiation dosimetry applications in space as well as for radiation shielding, specifically due to their high cross-section to absorb thermal neutrons. Here, we report on developing and optimizing a neutron radiation detector architecture based on utilizing hexagonal Boron Nitride (hBN) and graphene field-effect transistors. To this end, single crystal growth of natural boron nitride (hNaBN) and isotopically enriched boron nitride (h10BN, h11BN) was done using Ni-Cr metal flux method. The device geometry and experimental parameters are optimized by Monte Carlo calculations using MCNP and GEANT4 codes. In the designed experimental setup, a 2.5 MeV D-D neutron generator (Starfire nGen-800) is used as the neutron source, with a neutron yield of 1010 neutrons/sec and neutron flux of 106 neutrons/(cm2.sec). Real-time resistance measurements are carried out upon irradiating device prototypes with fast neutrons as well as thermal neutrons produced by placing a 3-inch-thick block of high density polyethylene between the generator and the device.

Log 369. **THE MICROGE™: A NEW PORTABLE AND RUGGED HIGH-PERFORMANCE HPGE DETECTOR SUITABLE FOR CHALLENGING APPLICATIONS**

Ilie, G. (1, P); Masseron, J. (1, P); Marian, V. (1); Ralet, D. (1); Legras, J.B. (1); Ginsz, M. (1); Brocard, N. (1); Quirin, P. (1). (1) Mirion Technologies (P) Expected Presenting Author.

Long considered as too fragile to implement on the field, germanium gamma-ray spectroscopy technology was mostly restricted to controlled and stable environments such as laboratories. In recent years, Mirion Technologies has developed rugged germanium detectors that are suitable to use in challenging conditions. An example of such a detector is our new product called MicroGe™, which allow users to implement it for different applications and in various configurations. The MicroGe detector is an ultra-compact, lightweight (only 1.7 kg for the detection probe), and rugged state-of-the art high-purity germanium (HPGe) detector. Thanks to its physical features, the MicroGe brings the unmatched performance of HPGe to the most challenging environments such as enclosed spaces as well as for field applications. Its excellent detection properties make it an ideal alternative to room temperature semiconductor detectors, with better spectroscopic and counting capabilities: energy resolution of 0.82% @ 122.1 keV, 0.24% @ 661.7 keV, 0.17% @ 1332.5 keV; dose rate up to 0,1 Gy/h and even more with a collimator and shielding elements (10 to 100 times higher is expected). Its unique form factor permits an easy integration with other systems and even with robotic platforms, making the MicroGe particularly interesting for industrial applications: medical radionuclides production, nuclear industry (fuel production, power plant monitoring…). In this presentation, we will give feedbacks of applications and successful combination, as with the famous "SPOT" dog robot from Boston Dynamics.

Log 370. **SEPARATION OF SELECT TRANSITION METALS FROM A MIXED ACTIVATION AND FISSION PRODUCT SAMPLE**

Beck, Chelsie (1,P); Herman, Staci (1) ; Warzecha, Evan (1) ; Emerson, Hilary (1) ; Stene, Riane (1); Haney, Morgan (1) ; Metz, Lori (1) 1. Pacific Northwest National Laboratory. (P) Presenting Author.

The purpose of this work was to develop a chemical separation to isolate Be, Mn, Ni, Cs and Ir from a mixed fission and activation product sample. Stable tracers of the relevant elements were used for initial development. Actinides and trivalent elements are removed using UTEVA and TRU resin cartridges (Eichrom Technologies Inc.) with the analytes of interest remaining in the effluent. Resins of AG50W – x8 cation resin and AN™ resin (Eichrom Technologies Inc.) were evaluated individually and in combination. Various elution gradients were tested for the cation column but in all experiments the analytes of interest were insufficiently separated. However, by adding the ANTM resin before the cation resin there was the ability to separate Be and Mn, which are retained on the AN cartridge. The Ni and Cs were retained on the cation column and the Ir is not retained on either resin. Be and Mn were successfully co-eluted from the ANTM resin allowing for quantification of 7Be and 54Mn by gamma spectroscopy. The Cs and Ni are separated from each other on the cation column using HNO3 to elute Cs followed by HCl to elute Ni. Overall, the isolation of four fractions; Be/Mn, Cs, Ni, and Ir was achieved.

Log 371. **NBL Program Office Nuclear Reference Materials Plans**

Mason, P (1,P); Tourville, A (1); Watters, R (1); Holland, M (1); Englemann, M (2). (1) NBL Program Office, NNSA; (2) Pacific Northwest National Laboratory. (P) Presenting Author.

The NBL Program Office (NBL PO) is the US certifying authority for special nuclear reference materials. The NBL PO will provide an update on inventory management and shipping, review the status of new plutonium isotopic and metal standards, several uranium reference materials, and share plans for future materials. The NBL PO will discuss our important collaborations with a number of DOE and international collaborator laboratorys. Additionally, the NBL PO will provide an update on plans for the proficiency testing mission element.

Log 372. DESIGNING NOVEL CHEMICAL SEPARATIONS WITH THE USE OF DISTRIBUTION COEFFICIENTS OF NUMEROUS EICHROM RESINS

Ward, J.L (1,P); Snow, M.S.(1) (1) Idaho National Laboratory, (P) Presenting Author.

The ability to design new chemical separation flowsheets and approaches is becoming increasingly important to emerging radiochemical research and development efforts in a variety of fields including nuclear energy, nuclear medicine, isotope production, and nuclear forensics. To support the advancement of radiochemical separation science, we have developed a database of distribution coefficients for 41 elements on multiple Eichrom (actinide, DGA-branched, Cl-, etc.) and Bio Rad (AG1x4 and Ag50W) chromatographic resins at differing concentrations of nitric and hydrochloric acid. Using this database, we demonstrate a number of new, improved separations for a variety of different radiochemical, with separations ranging from simple binary to ternary elemental systems (including Sc-47 isolation from irradiated targets) up to extremely complex separations including isolation of radionuclides from fission product mixtures and environmental samples. The results from these studies, along with the potential benefits to diverse radiochemical applications, will be presented.

Log 373. SYNTHESIS OF AMERICIUM TRICHLORIDE IN LITHIUM CHLORIDE-POTASSIUM CHLORIDE MOLTEN SALT AND STUDY OF THE ELECTRODEPOSITION REACTION OF AMERICIUM

Bethany Kersten (1, P), Krista Hawthorne (2), Mark Williamson (2), Rohan Akolkar (1), Christine E. Duval (1). (1) Department of Chemical and Biomolecular Engineering, Case Western Reserve University, 10900 Euclid Ave, 44106 Cleveland, OH, USA.   (2) Chemical and Fuel Cycle Technologies Division, Argonne National Laboratory, Lemont, IL, USA (P) Presenting Author.

Reprocessing of used nuclear fuel (UNF) allows for more efficient use of fuel resources while lowering the radiotoxicity of nuclear waste for disposal. Electrorefining is a method of reprocessing UNF through electrodeposition of actinides in a molten salt electrolyte. In this work, we aim to understand the fundamental mechanisms that govern americium electrodeposition from $AmCl_3$-LiCl-KCl. The salt was synthesized by chlorinating AmO_2 with $ZrCl_4$ in LiCl-KCl at 500°C. The presence of $AmCl_3$ was confirmed through observance of both Am^{3+}/Am^{2+} and Am^{2+}/Am^0 redox couples in cyclic voltammetry (CV) measurements. Gamma spectroscopy of the dissolved chloride salt sample and insoluble oxide yielded a 20% reaction conversion. CV was performed to study the redox reactions of Am^{3+} to Am^{2+} and the deposition of Am^{2+} to Am^0. Analysis of the Am^{3+}/Am^{2+} peaks show linear relationship of peak current density with scan rate and 2.2 RT/nF difference between peak potentials, indicating the reactions are not kinetically limited. However, this analysis indicated the deposition reaction (Am^{2+}/Am^0) may be kinetically limited. To further investigate this behavior, a transient diffusion-reaction model was derived and simulations were performed using COMSOL Multiphysics. The model suggests the deposition reaction kinetics are slow, which allows time for Am^{2+} to diffuse away from the electrode surface before reducing to Am^0. Accounting for this behavior is important for developing accurate electrodeposition models and characterizing the overall efficiency of the actinide recovery process.

Log 374. **LOW LEVEL MULTI ION COUNTING FOR URANIUM ON NUTIMS FROM NU INSTRUMENTS**

Cohen, R.(1); Colucci, M.(1); Freedman, P.A.(1); Roberts, D.J(1,P). (1) Nu Instruments. (P) Presenting Author.

Low level signals and low abundance isotopes may benefit from the detector configurations possible on the NuTIMS instrument. Thanks to its different detector configurations the NuTIMS from Nu Instruments presents clear advantages when measuring low level signals and low abundance isotopes The mass spectrometer utilizes a zoom lens system, allowing the detector to be equipped with multiple full size ion counters ideal for extremely low signal measurements. In this work we present some low level (1cps or less) data run in both static and dynamic routines, showing the possibility for low sample sizes or extremely low abundance 236 measurements.

Log 375. **MULTISTATIC NEGATIVE ION MEASUREMENTS INSTRUMENTS**

Cohen, R.(1); Colucci, M.(1); Freedman, P.A.(1); Roberts, D.J(1,P). (1) Nu Instruments. (P) Presenting Author.

The Nu TIMS instrument allows ultimate peak alignment of all isotopes thanks to its unique Zoom Lens system. With 16 faradays present multistatic routines can be used to collect all isotopes on each magnet position. The collection of all isotopes on each measurement line allows for a reduced error when measuring small signals for online oxide corrections. Here we present some high precsion data for (Mo, W) using negative ions in MultiStatic routines.

Log 376. **OVERVIEW OF THE CONSEQUENCE MANAGEMENT LABORATORY ANALYSIS PROGRAM**

Fournier, S. (1,P); Shanks, S. (1); Allen, M. (1); Chapman, J. (2). (1) Sandia National Laboratories. (2) NNSA NA-84 Office of Nuclear Incident Response Nuclear Emergency Support Team. (P) Presenting Author.

The US DOE Consequence Management (CM) program is tasked with maintaining a readiness to respond to any significant release of radiological material. This is accomplished by developing processes, procedures, and guidance for other federal, tribal, state, and local response organizations through several working groups. This guidance drives the development of training materials, various tactics, and tools that are used during the response. The CM program maintains a staff of responders that, when called into action, partner with other response organizations to form the Federal Radiological Monitoring and Assessment Center (FRMAC). The FRMAC has several elements that work together to address requests for information from incident stakeholders including a large field presence that assists with environmental and agricultural sample collection, control, and laboratory analysis. A high level overview of the CM program mission and structure will be presented with an emphasis on the radiological sample control and analysis activities of the FRMAC laboratory analysis unit. The purpose of this presentation is to raise awareness of the CM program and the challenges we face. The goal of the engagement is to inspire creative solutions and form strategic partnerships with the larger radiochemical analysis community. SAND2021-15994 A

Log 377. **FREE WEB-BASED GAMMA SPECTROSCOPY TRAINING PROVIDED BY US DOE AND US EPA TO IMPROVE LABORATORY CAPABILITIES**

Fournier, S. (1,P); Enghauser, M. (1); Griggs, J (2); Litman, R. (3) Chapman, J. (4). Gill, J. (5). (1) Sandia National Laboratories. (2) US Environmental Protection Agency. (3) Environmental Management Support , Inc. (4) NNSA NA-84 Office of Nuclear Incident Response Nuclear Emergency Support Team. (5) FEMA Nuclear Incident Response Team. (P) Presenting Author.

Through a partnership with the Department of Homeland Security Federal Emergency Management Agency Nuclear Incident Response Team (DHS FEMA-NIRT) the US DOE Consequence Management program and US EPA have developed automated web-based training modules on gamma spectroscopy in an effort to assist laboratories across the country build future spectroscopists. Topics range from basic introductory material to more advanced concepts like performing parent-progeny decay corrections and identifying and correcting for true coincidence summing biases in measurements. This poster will highlight what training is available, how to access it, and open up a discussion with the radiochemistry community on what the needs are for training like this to help bridge the knowledge gaps left behind in this field. SAND2021-15994 A

Log 378. **DETERMINATION OF LITHIUM ISOTOPIC COMPOSITION AND CONCENTRATION IN ENVIRONMENTAL COLLECTIONS BY MC-ICP-MS**

Kimberly A. Hinrichs (1), Ronald S. Amato (1), Daniel W. Gurganus (1), Andrew J. Nunn (1), Steven J. Goldstein (1), Allison A. Price (1,P), Stephen P. LaMont (1). (1) Los Alamos National Laboratory. (P) Presenting author.

Lithium isotopes behave as geo/chemical tracers for a wide range of environmental and anthropogenic processes; therefore, the accurate and high precision measurement of lithium in environmental samples is of interest to many. We have established a high-yield lithium separation technique for low level environmental collections, which enables the Li element concentration and isotopic composition to be measured by multi-collector ICP-MS. Two aliquots of each sample, both containing 5-100 ng total Li, are measured, one of which is spiked with a high purity ^6Li tracer. The aliquots are purified with a single cation exchange column in dilute hydrochloric acid. We investigated how the presence of matrix elements in a sample affect Li elution, Na-Li separation, and chemical yield of lithium purified by cation exchange. Analyses are made on a modified Nu Plasma II equipped with a desolvating nebulizer. Masses 6 and 7 are collected simultaneously on two Faraday cups in static mode, and we utilize standard-sample-standard bracketing with a fractionation correction based on L-SVEC ratios. To test the accuracy of this method, we measured lithium concentrations and isotopic compositions for lithium carbonate material L-SVEC (NIST SRM 8545) and certified reference material USGS BCR-2, and compared results to those in the literature. Lastly, we explored potential lithium process blank sources, focusing on sample decomposition and subsequent chemical separation.

Log 379. MOLECULAR PLATING OF Am-241 ON A SEMICONDUCTOR DIODE TARGET

Hoffman, M.K. (1,2); Spitz, H.B.(2); Bissmeyer, P.(2); Mangu, S.(2); Xue, S.(1); Downing, R.G.(1); Hlinka, V.(1); Cao, L.R.(1,3). (1) AwareAbility Technologies, LLC. (2) University of Cincinnati. (3) The Ohio State University. (P) Presenting Author.

Utilizing wide bandgap semiconductor materials to convert the energy from radioactive decay into electric current is a continuously evolving area of research in both radiation detection and nuclear batteries. The distance between radioactive source and semiconductor material is one of many factors that affect the efficiency of this energy conversion. Minimizing this distance by attaching radioactive atoms as closely as possible to the semiconductor material could help optimize the efficiency. This paper describes a molecular plating method in which americium-241 (Am-241) was directly deposited onto the 4.8 mm x 4.8 mm contact surface of the semiconductor diode. Adapted from methods for preparing targets and alpha-counting sources, the non-aqueous plating solution consists of isopropanol and a small amount of dilute nitric acid. Plating was conducted for one hour using a high voltage ranging between 300V and 600 V to mobilize the radionuclide. A polyurethane mask with an aluminized mylar contact surface was used as a gasket in a traditional disposable electrodeposition cell to accommodate the small size of the diode. Deposition recoveries of 50-65% have been consistently achieved using 20-30 nCi of Am-241 with minimal damage to the diode surface or function. Natural uranium and strontium-90 (Sr-90) have also successfully been deposited on the semiconductor using this method.

Log 380. THE DISSOLUTION AND PURIFICATION OF IRIDIUM FROM IRRADIATED IRIDIUM FOILS USING POTASSIUM HYDROXIDE AND POTASSIUM NITRATE FUSION AND ION EXTRACTION CHROMATOGRAPHY

Marenco, A.M. (1,P); Bond, E.M.(1); Rusev, G.(1); Bredeweg, T.A.(1). (1) Chemistry Division, Los Alamos National Laboratory. (P) Presenting Author.

The purpose of this research is to develop a method for the dissolution of irradiated iridium foils from nuclear chemistry experiments. Methods were also developed and optimized to separate and purify the iridium from the foils. Iridium is important because it was used as a radiochemical detector in nuclear device tests to assist in the understanding of the neutron energy spectrum, and is still used today as a spectral index for energy integral nuclear physics measurements at reactor facilities. Iridium foils used in this research were irradiated at the University of Massachusetts Lowell. A series of fusions using potassium hydroxide and potassium nitrate were applied to the foils. Variables analyzed include mass of foil, fusion vessel, length of fusion, agitation during fusion and the amount of chemicals. Yields were determined using gamma spectroscopy. The iridium was separated and purified from the fused leachate using ion exchange chromatography and prepared for analysis by silicon drift detector. This coupled method provides Los Alamos National Laboratory with a new capability which allows the dissolution and chemical processing of irradiated iridium metal foils. LA-UR-21-32330

Log 381. **IMPROVEMENTS IN IRIDIUM TARGET CHEMISTRY**

Marenco, A.M. (1,P); Bond, E.M.(1); Rusev, G.(1); Bredeweg, T.A.(1). (1) Chemistry Division, Los Alamos National Laboratory. (P) Presenting Author.

The purpose of this research is to develop and optimize dissolution and separation chemistry of iridium from irradiated salt pellets containing potassium hexachloroiridate (K_2IrCl_6) mixed with potassium chloride (KCl) and prepare the iridium for analysis by a silicon drift detector. Iridium is important because it was used as a radiochemical detector in nuclear device tests to assist in the understanding of the neutron energy spectrum, and is still used today as a spectral index for energy integral nuclear physics measurements at reactor facilities. Current iridium chemistry techniques used on the irradiated salt pellets require extra safety precautions and planning due to the use of perchloric acid in the dissolution step. The current dissolution and chemistry is also tedious, lengthy and only produces one sample to be analyzed. The dissolution and separations method described in this work is safer, eliminating the use of perchloric acid, more efficient, uses only a portion of the total sample, and can successfully replace the current procedure. LA-UR-21-32346

Log 382. **LOS ALAMOS NATIONAL LABORATORY'S OVER 25 YEAR EFFORTS SUPPORTING STANDARD PRODUCTION AND CHARACTERIZATION FOR VARIOUS NATIONAL SECURITY PROGRAMS**

Tandon, Lav (1,P); Colletti, Lisa (1); Eiroa-Lledo, Cecilia (1); Kuhn, Kevin (1); Olson, Angela (1); Walker, Laurie (1); Essex, Richard(2); and Mason, richard (3). (1) Los Alamos National Laboratory, P.O. Box 1663, Los Alamos, NM 87545, USA; (2) National Institute of Standards and Technology, 100 Bureau Drive, Mail Stop 8462, Gaithersburg, MD 20899, USA; (3) New Brunswick Laboratory Program Office, 1 Science.gov Way, Oak Ridge, TN 37830, USA. (P) Presenting Author.

Since the Manhattan Project, Los Alamos National Laboratory (LANL) analytical chemistry operations have recognized the value of partnering with United States (US) metrology laboratories and various governmental entities to produce bulk plutonium (Pu) standards that are crucial to validating measurements made for a diverse portfolio of nuclear security missions. The Pu standards production process generally begins with certifying agencies reaching out to LANL for Pu material production, fabrication and chemical analysis expertise. Once Pu characteristics and fabrication parameters are defined, a material specification is issued with input from LANL. Then comes the challenge to produce standard materials that are fit-for-purpose; in other words homogenous, well-characterized, stable, and most importantly, that meet customer requirements. Often the final product material has to be encapsulated or sealed in specialized containers for long term storage and use. Encapsulation or containerization also presents a unique set of challenges for each project. This presentation aims to provide a glimpse of select historical and current projects for the production and characterization of Pu standards. Standards described will include reference materials of various types including standard, certified, working and nuclear forensics reference materials. One common factor that this presentation aims to highlight is the compositional attributes that are measured for almost all Pu standards including analyses for Pu assay, Pu isotopics, actinide decay products, and trace metallic elements. Unique characterization tools and techniques deployed to support these projects will be discussed, to include uncertainty requirements or treatments. LA-UR-21-32340

Log 383. **SCALE MODELING OF FOIL IRRADIATIONS AT WSU'S TRIGA WITH SENSITIVITY/UNCERTAINTY ANALYSIS**

Hinrichs, K.A. (1,P); Perfetti, C.M. (2). (1) Los Alamos National Laboratory. (2) University of New Mexico. (P) Presenting Author.

A conversion-type TRIGA reactor housed at the Washington State University (WSU) was modeled in the SCALE code system's KENO neutron transport code using dimensions obtained from an existing MCNP geometry. The model includes 119 fuel rods containing uranium-zirconium hydride (UZrH) fuel, a central boral control rod, 4 control blades, 5 irradiation ports, and 20 graphite reflector blocks within a rectangular aluminum grid box. The model was successfully used to simulate the irradiation of a depleted uranium foil and a highly enriched uranium foil, which had previously been analyzed for the relative production rates of 237U, 239U, and 99Mo at Los Alamos. The KENO model was able to provide the neutron flux distribution in the foils, as well as accurately determine the reaction rate ratio for 238U(n,2n)237U / 238U(n,gamma)239U and 99Mo production from fissions. The KENO-determined values are consistent with the laboratory experimental results. Finally, using the continuous energy TSUNAMI-3D module of SCALE, a sensitivity/uncertainty analysis was performed, which was used to determine the effects of various irradiation conditions on the reaction rate ratio and to explore its sensitivity to uncertainties in nuclear data throughout the system.

Log 384. **EVALUATING HYDROLOGIC PROCESSES IN THE EASTERN SNAKE RIVER PLAIN AQUIFER WITH URANIUM AND STRONTIUM ISOTOPES**

Rattray, G.W. (1); Treinen, K.C. (1,P); Paces, J.B.(2). (1) U.S. Geological Survey, Idaho National Laboratory Project Office; (2) U.S. Geological Survey, Geosciences and Environmental Change Science Center. (P) Presenting Author.

Waste constituents discharged to the eastern Snake River Plain (ESRP) aquifer at the U.S. Department of Energy (DOE) Idaho National Laboratory (INL) pose risks to aquifer water quality. To understand these risks, the U.S. Geological Survey, in cooperation with the DOE, is conducting geochemical studies to better understand the regional hydrologic processes of the ESRP aquifer underlying the INL. This study uses natural uranium activity (234U/238U) and strontium (87Sr/86Sr) isotope ratios of surface water and groundwater as tracers to identify the sources of water, the mixing of different source waters, and the flow directions in the shallow portions (upper 250 feet) of the aquifer. Additional samples were collected from surface water, groundwater, and springs to complement existing isotopic data. Uranium activity ratios and 87Sr/86Sr from groundwater were plotted on graphs within ternary mixing webs in which the three end members of the mixing web represent specific sources of recharge. The mixing webs were then used to estimate the percent recharge from specific recharge sources to groundwater at wells. Understanding and estimating source-water mixing behavior and groundwater flow paths can improve our understanding of waste constituent migration in the ESRP aquifer at the INL.

Log 385. **TOTAL MERCURY ANALYSIS IN RADIOACTIVE WASTE CONTAINING METHYL MERCURY**
Looney, B.B. (1); Brown, L.W. (1); Jones, M.A. (1); White, T.L. (1, P); (1) Savannah River National Laboratory. (P) Presenting Author.

A direct mercury analyzer (DMA) was used to analyze radioactive waste samples high in methylmercury and other forms of mercury with oxidation states of Hgo (elemental), Hg1+ (mercurous), and/or Hg2+ (mercuric) to determine total mercury. These samples were also analyzed using a Cold Vapor Atomic Absorption Spectroscopy (CVAAS) for comparison. The DMA instrument has been determined to be the preferred method in our laboratory based on both simulated and radioactive sample testing due to analytical results and, primarily, ease of use in a radiological containment unit. Researchers made several modifications to the DMA instrument and analysis method to limit the amount of time personnel are required to work in a radiological environment. Comparative statistical evaluation of the results from: spike solutions/simulants, radioactive samples, and performance test samples (Mixed Analyte Performance Evaluation Program (MAPEP)) for both instruments resulted in equivalent total mercury data within the uncertainty of both methods (2s ± 20%). Triplicate radioactive waste analyses on the CVAAS analyses resulted in an average value of 54.1 (0.7 %RSD) and 56.0 mg/L (2.1 %RSD) on the DMA. The primary advantage of the DMA is avoiding a multi-step, labor-intensive and time-consuming sample preparation protocol of the CVAAS in a radioactive environment.

Log 386. **PROTON NUCLEAR MAGNETIC RESONANCE (H NMR) OF GLYCOLATE IN REAL WASTE**
White, T.L. (1, P); Fondeur, F.F. (1); Coleman, C.J. (1); DiPrete, D.P. (1); Looney, B.B. (1); (1) Savannah River National Laboratory. (P) Presenting Author.

In preparation for implementing the Nitric-Glycolic (NG) acid flowsheet for the Savannah River Site (SRS) Liquid Waste System (LWS), analytical methods for determining glycolate at low concentration, below 10 mg/L in radioactive samples, are needed to support system management and safety. Use of H NMR for quantitative analysis, in this case to quantify glycolate at low concentrations in liquid waste, was developed and tested. The H NMR method uses a sample preparation protocol to lower sample dose and activity by stripping Cs and Sr from the samples. This step also removes other radioactive and paramagnetic elements leading to safer sample handling and improved sensitivity. Additionally, the sample viscosity is lowered by pH adjustment increasing signal sensitivity. Several variants of the H NMR method were developed and tested (providing a range of target sensitivities). In the most sensitive variants, samples are pH adjusted with nitric acid in heavy water (D_2O) to below 0.1 M total base and undergo multiple crystalline silicotitanate (CST)/monosodium titanate (MST) strikes with filtration through a polyethersulfone (PES) filter. Use of D_2O enables the instrument to overcome magnetic drift supporting a high number of scans per sample. Determination of a Limit of Detection (LOD) of 1 mg/L and a Limit of Quantitation (LOQ) of 5 mg/L was achieved using real waste samples.

Log 387. ENHANCING SENSITIVITY AND SELECTIVITY TO METASTABLE NUCLEAR ISOMERS IN MIXED FISSION PRODUCTS USING TEMPORAL LSC-GATED HPGE GAMMA SPECTROMETRY

Archambault, B.C. (1,P); Pierson, B.D.(1); Seiner, B.N.(1); Estrada, J.H.(1); Metz, L.A.(1);Friese, J.I.(1). (1) Pacific Northwest National Laboratory. (P) Presenting Author.

This work presents the investigation and development of novel data analytics techniques designed to enhance sensitivity to select radionuclides present in mixed fission and activation products samples by exploiting their metastable nuclear isomeric transitions. Measurements taken with a custom LSC-HPGe coincident detection system termed "Gaby" developed at PNNL are presented. Application to several radionuclides of interest in the post-detonation nuclear forensics community are detailed, including U-237, Zr-97, Tc-99m, and Ag-111. These radionuclides span the three distinct detection modalities developed, which include temporal pre-gating in which a photon feeding a metastable state is detected followed by the emission of a delayed conversion electron (e.g. U-237), anti-coincidence gating in which a photon is emitted from a long lived (i.e. seconds to hours) isomeric transition (e.g. Zr-97 and Tc-99m), and temporal post-gating in which a beta is emitted which feeds a metastable isomeric state followed by the delayed emission of a photon (e.g. Ag-111). Optimization of the timing of the temporal gates was performed for Ag-111, which has a delayed isomeric transition at 245.5 keV with a half life of 84.5 ns. Optimization of the temporal gate window found an optimal "delayed" beta-gate of 1100ns-2500ns after LSC detection yielding 50x background suppression and a greater than two reduction in MDA when compared to conventional coincident beta-gating alone.

Log 388. FORMATION OF CERIUM OXIDES THROUGH BENCHTOP LASER EXPERIMENTS IN CONTROLLED OXYGEN ATMOSPHERES

Auner, A.W. (1,P); Burton, M.A.(1); Weisz, D.G.(1); Crowhurst, J.C.(1); Knight, K.B.(1). (1) Lawrence Livermore National Laboratory. (P) Presenting Author.

High-temperature cerium chemistry, particularly vapor-phase oxidation chemistry, is understood to be complex. It is hypothesized that parameters such as cooling rate and oxygen availability affect cerium chemistry and that varying these parameters may induce different oxide stoichiometries and phases. Benchtop laser experiments using cerium targets provide controlled environmental conditions to study the products of high temperature reactions in situ. Toward this goal, we investigated the oxidation properties of cerium in high-temperature vapor phase chemistry experiments. In our study, we generated particulates via local ablation-vaporization with continuous wave (CW) and nanosecond pulsed (Nd:YAG 1064 nm) lasers in deficient (<1% O2) and abundant (>20%) oxygen atmospheres. Distinct cerium oxide structures are identified by Raman spectroscopy and SEM as CeO_x (where $1.5 \leq x \leq 2$) in both amorphous and crystalline structures. These results show that cerium oxide phase is dictated not only by cooling rate and peak temperature, but also by the specific oxygen deficient atmosphere facilitated by our benchtop experimental apparatus. This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344, is supported by LLNL Strategic Initiative LDRD project 20-SI-006.

Log 389. ADDITIVE MANUFACTURING FOR NUCLEAR NONPROLIFERATION

Cannon, N.L. (1,P); Biegalski, S.R. (1); Erickson, A. (1). (1) Georgia Institute of Technology. (P) Presenting Author.

Additive manufacturing (AM) is a rapidly developing technology that allows industries to automate and simplify the production of highly complicated items. Recently, AM techniques have been used in the fields of nuclear weapons and nuclear enrichment technology. Presently, there are very few international or domestic export controls that apply to AM's role in the nuclear industry, constituting an unmanaged proliferation pathway. Pre-existing export controls focus on general concepts and processes and do not take into account specific nuances of various techniques that are categorized as additive manufacturing. To introduce legislation and controls that will be effective in monitoring proliferation pathways, one must investigate and characterize AM techniques and their nuclear applications. This work involves categorizing 32 AM techniques and ranking them based on their potential impact on the nuclear fuel cycle. Through this method of characterization and categorization, export controls would address specific AM nuclear proliferation risks without disrupting the entire industry and fuel cycle. Additionally, legislation employing this method would identify loopholes in export controls because of its holistic approach to managing and monitoring proliferation pathways.

Log 390. RESULTS FROM THE FIRST DEPLOYED ULTRA-HIGH-RESOLUTION MICROCALORIMETER GAMMA SPECTROMETER

Croce, M.P (1,P); Carpenter, M.H. (1); Schreiber, K.A. (1); McNeel, D.G. (1); Ortiz, N.J (3) Wessels, A.L. (3); Becker, D.T (3); Imrek, J. (2); Gard, J.D. (3); Mates, J.A.B. (2); Bennett, D.A. (2); Schmidt, D. (2); Vale, L. (2); Swetz, D.S. (2); Ullom, J. (2). (1) Los Alamos National Laboratory, (2) National Institute of Standards and Technology, (3) University of Colorado. (P) Presenting Author.

Extensive technology development over the past several years has now led to the first deployed ultra-high-resolution gamma spectrometer based on low-temperature microcalorimeter detector technology. SOFIA (Spectrometer Optimized for Facility Integrated Applications) was developed to provide improved precision and accuracy for nondestructive isotopic analysis of radioactive and nuclear materials resulting from advanced fuel cycles and separations. The instrument provides typical energy resolution of 60-80 eV FWHM, nearly an order of magnitude better than high-purity germanium detectors, and count rates of several thousand events per second in a compact form factor designed for easy deployment in nuclear facilities and analytical laboratories. The performance of SOFIA was first evaluated in the laboratory for a series of samples from electrochemical and aqueous spent fuel separations and has been shown to provide significantly improved precision compared to germanium detectors. In 2021, SOFIA was deployed to the Los Alamos National Laboratory Plutonium Facility for a demonstration of nondestructive isotopic analysis capabilities on a range of items in a working nuclear facility. We will present results of these measurements and discuss experience from operating the instrument in the nuclear facility environment.

Log 391. **INVESTIGATION OF A FORMER LOS ALAMOS NUCLEAR REACTOR SITE VIA HYPERSPECTRAL X-RAY IMAGING OF PARTICULATE SAMPLES**

Croce, M.P (1,P); Carpenter, M.H. (1); Stein, B. (1); Schreiber, K.A. (1); McNeel, D.G. (1); Morgan, K.M. (3); Wessels A.L. (3); Becker, D.T (3); Imrek, J. (2); Gard, J.D. (3); Mates, J.A.B. (2); Bennett, D.A. (2); Swetz, D.S. (2); Ullom, J. (2). (1) Los Alamos National Laboratory, (2) National Institute of Standards and Technology, (3) University of Colorado. (P) Presenting Author.

Between 1956 and 1964, three different experimental nuclear reactors were operated at the present location of Technical Area 35 at Los Alamos National Laboratory. These included the Los Alamos Power Reactor Experiments (LAPRE) I and II, which used uranium-phosphoric acid fuel solution, and the Los Alamos Molten Plutonium Reactor Experiment (LAMPRE) which was fueled by a molten plutonium-iron eutectic alloy. The reactors have since been decommissioned and nearly all of the associated locations have been decontaminated such that they are released for normal operations. The building that housed LAPRE-1 and LAMPRE now serves as laboratory space for nuclear safeguards technology and analytical instrumentation development, including the development of Hyperspectral X-ray Imaging (HXI). HXI is based on an ultra-high-resolution microcalorimeter X-ray detector array coupled to a scanning electron microscope (SEM) to provide energy resolution better than typical wavelength-dispersive spectroscopy (WDS) with the wide dynamic range and high efficiency of energy-dispersive spectroscopy (EDS). The technology provides not only ultra-sensitive elemental composition measurements with the spatial resolution of the scanning electron microscope, but the potential for chemical speciation based on subtle features in the X-ray emission spectra related to chemical bonding. We will present results of an investigation based on sampling of particulates around the Technical Area 35 site, analysis using HXI and SEM, and our interpretation of measurement results based on documented history of the experimental reactors.

Log 392. **LESSONS LEARNED FROM THE ADAPTATION OF THE NUCLEAR AND RADIOCHEMISTRY SUMMER SCHOOL TO A VIRTUAL FORMAT**

Baisden, T. (1); Bechtel, H.(2); Bryan, J.(3); Cutler, C.S.(4); Deri, M.A.(5,6,P); Esfandiari, M.(7); Esker, N.E(7); Francesconi, L.C.(6,8); Maraschin, V.(7); Sanders, V.A(4); Van Wyngarden, A.(7). (1) National Ignition Facility and Photon Sciences, Lawrence Livermore National Laboratory, Retired. (2) Regional Network Consulting Director, IBM Global Technology Services, retired. (3) Department of Chemistry and Biochemistry, University of Wisconsin-La Crosse. (4) Collider Accelerator Department, Brookhaven National Laboratory. (5) Department of Chemistry, Lehman College of the City University of New York. (6) The Graduate Center of the City University of New York. (7) Department of Chemistry, San José State University. (8) Department of Chemistry, Hunter College of the City University of New York. (P) Presenting Author.

The Nuclear and Radiochemistry Summer School (NCSS) is a six-week intensive program for advanced undergraduate students that introduces them to fundamental nuclear science and its applications to fortify the nuclear workforce. As of 2021, the program has been in existence for 38 years and continuously funded by the US DOE since 1984. Over this time, 838 students have successfully completed the summer school. The program is currently hosted at San Jose State University and Brookhaven National Laboratory, each of which accommodates 12 students. The summer school normally consists of course lectures, hands-on counting and wet lab experiments, guest seminars, and field trips, but due to the COVID-19 pandemic the program was forced to go virtual for the summers of 2020 and 2021. This adaptation to an online program presented many organizational and practical challenges, but these were met with sufficient creativity and dynamism to enable two successful summers of online learning in extraordinary circumstances. In the adapted program, NCSS staff from both sites combined their efforts to run a single online program with 24 students. Course lectures and guest seminars were given over Zoom which enabled greater participation from experts in the field due to ease of access. Lab experiments incorporated prerecorded videos and were redesigned to focus on groupwork in understanding and processing provided data sets. Field trips were replaced with virtual tours and videos from several institutions. The lessons learned in the virtual format will continue to improve the in-person program we hope to re-establish in 2022.

Log 393. **BACKGROUND MEASUREMENTS OF Ar-37 SAMPLES USING THE ARGON-37 FIELD SYSTEM**

James C. Hayes (1,P), Bradley G. Fritz (1), Lance S. Lidey (1), Thomas R. Alexander (1), Emily K. Mace (1), Vincent T. Woods (1). (1) Pacific Northwest National Laboratory. (P) Presenting Author.

This presentation will highlight background measurements of Ar-37 samples that were conducted at several locations to better understand the atmospheric concentrations of Ar-37 using the Argon-37 Field System. The PNNL designed and built Argon-37 Field System has now processed and measured several hundred Ar-37 samples from both soil gas and from the atmosphere. The system was designed to process whole air samples from soil gas, the atmosphere, and from the output of radioxenon systems to detect Ar-37 in an above ground portable system. During these campaigns, samples were collected at locations, near radioxenon systems, and sent back to PNNL for processing and measurements as opposed to sending the system to a location and operating it remotely. Correlation with radioxenon measurements from the same air masses were also conducted and will be discussed. In addition to presenting the results of our atmospheric sampling campaign, we will foster a discussion on the path forward for better understanding Ar-37 and possible Ar-39 backgrounds in the environment.

Log 394. **HUNTING FOR NEW ELEMENTAL SIGNATURES FROM UNDERGROUND NUCLEAR EXPLOSIONS ON THE BENCHTOP**

Liezers M. (1, P) ; Carman A.J. (1). (1) Pacific Northwest National Laboratory. (P) Presenting Author.

Developments in analytical instrumentation in recent decades may allow for the use of new elemental signatures in the detection of underground nuclear explosions. While field-scale chemical explosive tests used as surrogates are expensive and require extensive planning, lab-scale explosive tests provide an appealing alternative. One such alternative is the use of an exploding wire that can generate mm-scale point source explosions in concrete or rock cores on the benchtop. The electrically driven explosion can generate fractures, even localized melting of the enclosing medium from an input energy of <1000 Joules (equivalent to <0.25g TNT). Different aspects of the work will be described along with studies to detect the propagation of elemental signatures produced by these miniature high energy events and how these might apply to the detection of underground nuclear testing now.

Log 395. ELECTROCHEMICAL ASSAYS OF URANIUM METAL IN MOLTEN FLINAK FUEL SALTS

Newton, M.L. (1,P); Simpson, M.F. (1). (1) The University of Utah. (P) Presenting Author.

Molten fluoride salt fueled nuclear reactors, which afford greater efficiency, improved safety, and less high-level waste, are some of the most promising reactors being researched. Existing methods of salt composition monitoring are slow and require samples to be removed from the reactor. Electrochemical sensors were identified as a possible method for monitoring real-time salt compositions that could improve reactor safety and efficiency. In this study, FLiNaK salt containing between 1-5 wt% U was tested using cyclic voltammetry with the objective of developing a method of measuring real-time uranium concentrations in fuel salts. These measurements could be used for improving process controls and nuclear material safeguards. Experiments were run in a Ni crucible using tungsten working and counter electrodes and a platinum quasi-reference electrode. The electrode surface areas were precisely controlled using a vertical translator. Solution resistance was measured using electrochemical impedance spectroscopy and compensated for using the potentiostat. Salt samples were analyzed using inductively coupled plasma mass spectrometry (ICP-MS) to verify uranium concentrations. Cyclic voltammetry revealed two reduction peaks at 1.05 V and 1.3 V versus the Na+/Na reduction potential. These peaks, which presumably represent the reduction of U4+ to U3+ and U3+ to U, respectively, exhibited a linear relationship between current density and the square root of the scan rate. A range of 10 to 500 mV/sec was used in these experiments. ICP-MS was used to develop a correlation between the slope of these plots and the uranium concentration in the salt. This correlation will be presented and discussed.

Log 396. NEUTRON ACTIVATION ANALYSIS AND ICP-MS FOR PROVENANCE OF HONEY COLLECTED FROM AMERICAN MIDWEST REGION

Weilert, T.M. (1, P); Gawenis, J. (1); Ray, C. (1); Brockman, J.D. (1, 2). (1) Department of Chemistry, University of Missouri – Columbia. (2) University of Missouri Research Reactor Center. (P) Presenting Author.

The elemental analysis of honey has been shown to be a useful tool for both regional and botanical authentication of honey. This analysis technique could be a valuable complement to other regional discrimination techniques such as palynology. A comprehensive database of elemental information on honeys with known regional and botanical sources is needed in order to actualize its usefulness for honey source provenance. Proven analytical techniques and quality control materials are needed to develop the database. In this study, honey samples collected from discrete sources in North Dakota, South Dakota, and Montana were analyzed by ICP-MS and INAA. Sucrose was measured by NMR to confirm that samples were not adulterated with sugar. Palynology was used to assess the pollen source. The elements Na, Mg, Al, K, Ca, Mn, Fe, Cu, Zn, Rb, Sr, B, P, Co, Cs and Ba were measured by ICP-MS, and the elements Br, Cl, Fe, Zn, Sc, Na, Fe, Co, Ag, Se, and Hg were measured by INAA. The duplicate elements from the two methods were compared. A principal component analysis was conducted to determine if the regional sources could be distinguished by their trace element content. The results have demonstrated that elemental analysis may be useful in discriminating honey sourced from Montana against honey sourced from North and South Dakota.

Log 397. **FAST NEUTRON DIRECTIONALITY FROM POSITION-SENSITIVE RECOIL AND CAPTURE RECONSTRUCTION USING BAYESIAN UPDATING**

Wu, T. C (1,P); Li, V. A. (2); Sutanto, F. (2); Dazeley, S. (2); Jovanovic, I. (1). (1): University of Michigan Department of Nuclear Engineering and Radiological Sciences, Ann Arbor, MI, USA; (2) Lawrence Livermore National Laboratory, Livermore, CA, USA. (P) Presenting Author.

Capture-agent-doped organic scintillation detectors capable of identifying both neutron recoils and captures are a promising candidate for spectroscopy of low-energy neutrons via capture-gated spectroscopy. The addition of position sensitivity in these detectors has the potential to provide neutron directionality information. This work assesses the feasibility of and methods for reconstructing directionality based on the location of proton recoil and the corresponding neutron capture. A Monte-Carlo simulation of 1.e6 DD (2.45 MeV) neutrons is used to create reference distributions for recoil and capture locations within a spherical PVT detector doped with 0.1% 6Li. Bayesian updating is then used to reconstruct incoming neutron angles with reduced statistics (<1000 of incident DD neutrons). The angular discrepancy between the estimated and simulated incoming neutron angle is used as a figure of merit. Where there is no energy threshold for recoil detection, the location of capture did not provide any significant improvement beyond what is estimated with only the detection of the location of recoil. In contrast, where there is an energy threshold applied to recoil detection, an improvement in the incident neutron angle estimate is observed: for a neutron detection threshold of 30% of the maximum proton recoil energy from DD neutrons, the angular discrepancy for the incoming neutron angle for 1000 DD incident neutrons incident improved from 6.5 to 4.1 deg., with the 1-sigma dispersion of the estimate improved from 2.7 to 1.2 deg. This functionality may be exploited in neutron scatter camera configurations fitted with scintillators that incorporate the neutron capture mode.

Log 398. DESIGN FOR LONG-TERM DETECTOR BACKGROUND MEASUREMENTS FOR GAS DETECTOR SYSTEMS

Perea, R.S. (P), Cooper, M.W., Mayer, M.F., Mendez, J.M., Slack, J. L. Pacific Northwest National Laboratory. (P) Presenting Author.

Several beta-gamma detectors are currently being utilized for long-term background measurements to monitor changes in natural background radiation. The resultant data will show if any variations (e.g.., day/nighttime differences, seasonal differences, changes over time, etc.) impact the background measurement used in radioxenon calculations. A typical detector background measures the natural radioactive background and is taken over a three-day period. Current background measurements may not be performed often enough to account for background variations. It is unclear how often these detector background measurements should be updated. Several beta-gamma detectors, each consisting of a NaI(Tl) and a plastic beta-cell, will be set-up in different locations at Pacific Northwest National Laboratory. One detector will be co-located with a SAUNA system in a smaller building, and for comparison, several others will be located inside a large building. The systems will continuously acquire background data and the resulting measurements will be compared over 2 to 3 years. These measurements will help determine how often new background measurements are needed. Having a good representative background will improve accuracy of radioxenon concentration measurements.

Log 399. PRELIMINARY RADIOCHEMICAL TRANSPORT MODEL OF THE MOLTEN SALT REACTOR EXPERIMENT

Shahbazi, S.(1,P); Fei, T.(1). (1) Argonne National Laboratory. (P) Presenting Author.

In contrast to nuclear reactors with solid fuel, reactors with flowing liquid fuel, such as molten salt reactors (MSR), must account for dynamic radionuclide transport and the associated chemical considerations as a function of both time and location. To this end, a radiochemical transport model of the Molten Salt Reactor Experiment (MSRE) was formulated based on the material derivative featuring system level 1-D flow providing temporal and spatial dependence. Radioactive decay, chemical transport, and removal due to neutron absorption are included in the material derivative that represents each custom volume of the reactor. The steady-state version of the model, which is based on a small system of ODE's, can be solved efficiently and is useful for sensitivity studies such as those employing Monte Carlo sampling. Properly accounting for all phenomena in a fully coupled model is necessary to understand the complexities of the transport and distribution of radionuclides across the primary loop of a reactor system, as well as the secondary systems. These are important considerations when modeling the mechanistic source term of an MSR, which is defined as the potential release of radioactivity from the reactor system to the environment. Knowledge of the radionuclide inventory in a specific location of an MSR at the time of a release, which is dependent on fuel burn-up, is key to understanding the radiological consequences of the accident.

Log 400. DETERMINATION OF THORIUM IN HEAT SOURCE PLUTONIUM

Jump, R K, Carver, N R, Matonic, J H, Rim, J.H.(1,P); and Dickson, F P. Los Alamos National Laboratory

A method to determine thorium (Th) in heat source plutonium oxide ($238PuO2$) has been developed. After dissolution in strong mineral acids, Th in a $238PuO2$ sample is isolated from the Pu matrix using TEVA resin. The separation is based on the fact that in a solution of 3M HNO3/ferrous sulfamate/ascorbic acid, Th is retained by TEVA resin whereas Pu is reduced to Pu(III) which is not retained and passes through. After treatment with TEVA resin, Th is determined by UV-vis spectrophotometric analysis of the colored complex formed by Asenazo III and Th; absorbance versus concentration of this complex is linear up to 1 ug Th / mL. Studies showed that this procedure was free of interference from added Np and Pu. A Pu / Th decontamination factor of 10,000 in heat source Pu is routinely achieved with this procedure. Blank spike data collected in 52 analytical batches over a 5 year period have averaged 102% recovery.

Log 401. ULTRA-SENSITIVE GAMMA SPECTROMETRY MEASUREMENTS USING THE ADVANCED RADIONUCLIDE GAMMA SPECTROMETER (ARGO) SYSTEMS

Burnett, J. L. (1, P); Myers, A. W. (1); O'Mara, R. P. (1) Sharma, M. (1). (1) Pacific Northwest National Laboratory. (P) Presenting Author.

Two ultra-sensitive Advanced Radionuclide Gamma spectrometer (ARGO) systems are being used to measure natural and anthropogenic radionuclides in a wide range of samples types, including air filters, meteorites, and irradiated materials. Both ARGO systems consist of two Broad Energy Germanium (BEGe) gamma-spectrometers located within low-background shielding within a Shallow Underground Laboratory at Pacific Northwest National Laboratory (PNNL). The detectors are capable of multiple simultaneous modes of operation, including single, sum, Compton suppressed and coincidence measurements. This provides multiple confirmatory measurements for a given acquisition, including absolute determination for many radionuclides using its unique coincidence gamma-emissions. The later approach has provided some of the most sensitive gamma-spectrometry measurements performed, with sensitivity improving by up to a factor of 300 for longer-lived radionuclides such as ^{106}Rh, ^{134}Cs and ^{144}Ce. To support the ARGO systems, including for advanced data processing and the calculation of the coincidence emission probability, a small-scale supercomputer is being developed. This will be integrated the ARGO systems, to provide unprecedented detection possibilities.

Log 402. **THE USE OF INTERNATIONAL MONITORING SYSTEM STATIONS TO DETECT EVIDENCE OF SEISMIC EVENTS THROUGH THE DISPLACEMENT OF NATURAL RADON ISOTOPES**

Burnett, J. L. (1); O'Mara, R.P. (1, P). (1) Pacific Northwest National Laboratory. (P) Presenting Author.

Research has been undertaken to examine whether measurements of natural terrestrial radionuclides (^{210}Pb and ^{212}Pb) made at stations of the Comprehensive Nuclear-Test-Ban Treaty's International Monitoring System (IMS) are useful for identifying seismic events such as earthquakes. It is hypothesized that such natural events could displace radon gas isotopes (^{222}Rn and ^{220}Rn), resulting in elevated ^{210}Pb (a decay product of ^{222}Rn) and ^{212}Pb (a decay product of ^{220}Rn) at IMS stations; similar processes may also occur for underground nuclear explosions. To understand natural fluctuations, data from all 71 active IMS radionuclide stations during 2010-2020 has been statistically analyzed and used to produce lognormal probability density models that are useful for identifying anomalous events. Ratios to other natural cosmogenic radionuclides such as ^{7}Be and ^{24}Na were also examined and found to be a useful discriminator of source terms. Five anomalous events were identified during 2019-2020 and were further investigated using atmospheric transport modelling to understand where the air mass sampled at the station may have originated. Two of these events were correlated to seismic activity and may be indicative of radon displacement from earthquakes.

Log 403. **IMPROVING THE DETECTION SENSITIVITY OF THE NEXT GENERATION OF RADIONUCLIDE AEROSOL SAMPLER/ANALYZER AIR SAMPLER**

Burnett, J. L. (1, P); Miley, H. S. (1); Myers, A. W. (1); Sharma, M. (1). (1) Pacific Northwest National Laboratory. (P) Presenting Author.

The Radionuclide Aerosol Sampler/Analyzer (RASA) is a high-volume air sampler combined with a single High-Purity Germanium (HPGe) gamma-spectrometer used for automated detection of aerosol radionuclides. It is used as part of the International Monitoring System (IMS) that supports the Comprehensive Nuclear-Test-Ban Treaty (CTBT). After over 20 years of reliable operations, the design of the successor RASA system is being considered. Significant improvements in sensitivity are achievable through the utilization of two HPGe gamma-spectrometers used for sample measurement. This effectively doubles the detection efficiency with near 4π coverage (when the measurement from both detectors is combined), and provides the capability for the ultra-trace measurement of certain radionuclides through their unique coincidence gamma-energy emissions (e.g. 140La, 60Co). Experiments to demonstrate these performance gains indicate an average improvement in minimum detectable activity of 52% for a standard 24h IMS measurement. The incorporation of an additional HPGe detector also improves operational resiliency acting as a backup should a detector fail.

Log 404. DEVELOPMENT OF A TECHNICAL DATA CENTER FOR THE PROCESSING AND ANALYSIS OF ENVIRONMENTAL RADIOACTIVITY MEASUREMENTS

O'Mara, R.P. (1, P); Burnett, J. L. (1) Pacific Northwest National Laboratory. (P) Presenting Author.

A new capability is being developed at Pacific Northwest National Lab (PNNL) to facilitate the processing and analysis of radionuclide data. PNNL's Technical Data Center (TDC) includes a growing number of data streams that allow near-real time monitoring of global radiological conditions. Examples of these data streams include particulate and xenon gas measurements collected by International Monitoring System (IMS) stations in support of the Comprehensive Nuclear-Test-Ban Treaty (CTBT), status-of-health indicators from IMS stations, environmental gamma radiation monitors and nuclear power plant stack monitoring data. An automatic processing, analysis and archiving pipeline is currently being implemented and already used to support scientific and radiological emergency response efforts. Work is currently being conducted to identify ways to implement additional remote sensing data into the TDC pipeline. Proposed projects that will be supported by the TDC range from applying machine learning techniques to IMS station measurements and status of health indicators for improved automatic anomaly detection to determining whether changes in climate are reflected in natural environmental radioactivity distributions.

Log 405. RAPID DISSOLUTION OF SURROGATE NUCLEAR DEBRIS AND ANALYSIS OF RARE EARTH ELEMENTS USING HPIC WITH GAMMA RAY SPECTROMETRY

Bradley, V. C., Brockman, J. D.

Techniques for rapid analysis of fission products in post detonation debris are needed for attribution analysis. A novel HPIC method for lanthanide separation with gamma ray spectroscopy detection was evaluated. Geological samples were digested using microwave and ammonium bifluoride fusion methods. Fresh fission products were produced through irradiation of a natural U standard at the University of Missouri Research Reactor to produce 7.3X 108 fissions. Rare earth elements (REE) and actinides were separated using a Dionex CS-5a mixed-bed cation and anion exchange column HPIC system with 1 M HNO_3, H_2O, diglycolic acid (DGA), and oxalic acid mobile phases. Actinides were eluted in 1 M HNO_3 while REEs were eluted in a mixture of oxalic acid and DGA. The REEs were eluted in order of increasing size. Fractions were collected at 1-minute intervals and counted on a HPGe detector. The total time of separation for all REE and actinides was 40 minutes. A geometry matched standard was prepared from the U standard. The recovery for the fission products were compared to the U standard and to a MCNP5/ORIGEN model prediction. 140La, 141, 143Ce, 147Nd, and 149, 151Pm were successfully separated. Recoveries were found to be between 60 % and 70 % when compared to ORIGEN predictions, and greater than 70 % when compared to the standard.

Log 406. **DESIGN AND VALIDATION OF A GAMMA-RAY SCANNING SYSTEM FOR MEASURING IRRADIATED NUCLEAR FUEL**

Ocampo Giraldo, L.A. (1,P); Holschuh, T.V. (1); Thompson, S.J. (1); Hix, J.D. (1); Johnson, J.T. (1); Chichester, D.L. (1). (1) Idaho National Laboratory. (P) Presenting Author.

A prototype gamma-ray scanning system was designed to perform post-irradiation measurements of nuclear fuel at Idaho National Laboratory (INL). The system is composed of a coaxial high-purity germanium (HPGe) detector, a collimator, and mechanical positioning stages that translate the fuel sample across the front of the collimator. This paper documents the design process, modeling and the laboratory characterization performed to validate the system. The modeling of the system was done using the Monte Carlo N-Particle code with vendor-supplied design specifications, dimensional measurements, and x-ray radiographs of the HPGe detector. The characterization included measurements of calibrated radiation sources in different geometries, and calculations for absolute efficiency and dead time. Benchmark measurements were performed by scanning an irradiated fuel rodlet containing ten pellets of 4.9% enriched UO_2 in zirconium alloy. These results were compared to simulated spectra to further help characterize the detector model. One goal of the system is to determine the number of fissions per gram of UO_2 in the fuel; comparison of the results from this prototype system's assay of the irradiated fuel rodlet are in agreement with previously estimated results.

Log 407. **RETROSPECTIVE ANALYSIS OF Pu ISOTOPES IN A BANDED CORAL CORE COLLECTED NEAR THE CACTUS CRATER CONTAINMENT STRUCTURE ON RUNIT ISLAND, ENEWETAK ATOLL**

Hamilton, T.F. (1); Ahren N.L (1,2); Shaw A.C. (1); Tumey S.J. (1) Brown T. (1). (1) Lawrence Livermore National Laboratory. (2) Colorado State University. (P) Presenting Author.

The Cactus Crater waste containment structure on Runit Island at Enewetak Atoll was built in 1979-80 and remains a controversial legacy of the U.S. atmospheric nuclear weapon testing program in the Marshall Islands. Scientists from the Lawrence Livermore National Laboratory (LLNL) in conjunction with the Office of Health and Safety (AU-10) at the U.S. Department of Energy (DOE) are embarking on an ambitious program to establish a long-term groundwater monitoring program at the site. The monitoring program is expected to provide a more accurate basis to assess long-term health or ecological impacts of contaminated groundwater flows leaving the site boundary and entering the nearby marine environment. Any outflow of Cactus Crater derived radioactivity through the groundwater aquifer into the marine environment will likely be compounded by the effects of sea-level rise and possible increases in the frequency and severity of major storm events. In the interim, we are exploring the use of banded corals as a retrospective tool to assess the time history of Pu contamination in the lagoon before and after construction of the waste containment site. Here we show results of analysis of Pu isotopes in a single banded coral core collected off Runit island in the immediate vicinity (app. 100 meters) of Cactus Crater.

Log 408. **OXIDATION OF CERIUM NITRATE AS A SURROGATE FOR PLUTONIUM IN A MICROFLUIDIC DETECTION AND CHARACTERIZATION SYSTEM**

Williams, B.L. (P), Gelis, A., Corne, F. (P) Presenting Author.

Working with actinides in fuel reprocessing or monitoring settings presents a unique health hazard due to the radiotoxicity of some radionuclides and their progeny. In recent years, microfluidics has presented a solution to this challenge by allowing scientists to develop systems capable of detecting and characterizing minute volumes of actinides and other key metals with high sensitivity and minimal exposure to the users. The characterization of plutonium during nuclear waste treatment is an ideal use of a microsystem. This project aims to extract Ce(IV) nitrate in microfluidic volumes of 30 percent tributyl phosphate (TBP) in dodecane by oxidizing Ce(III) to Ce(IV) in 1 M HNO3. Cerium is a readily available rare earth element and can be used as a surrogate for plutonium due to their similar ionic radii and shared ability to form complexes with TBP. Macroscopic oxidation and liquid-liquid extraction of Ce(IV) have been well documented. A microelectrochemical chip made from two titanium plates coated with a thin-film platinum electrode is used to perform electrolysis mediated separation of Ce(IV). The organic extractant is analyzed using inductively coupled plasma-optical emission spectroscopy (ICP-OES) and ultraviolet-visible (UV-Vis) spectroscopy to verify the extraction of Ce(IV). The concentration of Ce(IV) in the organic extractant is dependent on the flow rate of the organic and aqueous phases in the microelectrochemical chip. This proof of concept shows the viability of using a microfluidic system to extract radioelements.

Log 410. **ENVIRONMENTAL OBSERVATION OF NONSTANDARD RADIOXENONS**

Mayer, M.F. (1,P); Cooper, M.W (1); Ely, J.H. (1); Eslinger, P.W. (1); Foxe, M.P. (1), Hayes, J.C. (1); McIntyre, J.I. (1); Panisko, M.E. (1). (1) Pacific Northwest National Laboratory. (P) Presenting Author.

Testing of the Xenon International System at Knoxville, TN resulted in the detection of several previously unseen radioxenon isotopes in the atmosphere. The majority of these detected radioxenons can be created via neutron irradiation (125Xe, 127Xe, and 129mXe), but the isotope 122Xe and its daughter 122I was also observed which is produced via proton or alpha bombardment. These isotopes were periodically detected in the atmosphere from December 2019 until May 2021 when Xenon International concluded its testing. The radioxenons were observed using the beta-gamma coincidence detectors in the system. Atmospheric transport modeling suggested that the source of the radioxenons emanated from Oak Ridge National Laboratory's High Flux Isotope Reactor or Spallation Neutron Source. The production of 122Xe suggests the source of radioxenon to be off gassing of the mercury target used by the Spallation Neutron Source. The signatures produced by these atypical radioxenons can veil the signatures of the four radioxenons (135Xe, 133Xe, 133mXe, and 131mXe) used for nuclear explosion monitoring. Currently, there are only a few spallation sources worldwide, but it may be possible some of these isotopes may show up in nuclear monitoring systems due to the longevity of some of these isotopes.

Log 411. RADON QUANTIFICATION AND REJECTION LEVEL DETERMINATION

Mayer, M.F. (1,P); Cooper, M.W (1); Ely, J.H. (1); Hayes, J.C. (1). (1) Pacific Northwest National Laboratory. (P) Presenting Author.

Radon is the leading contaminant for radioxenon measurements and can suppress the sensitivity of the nuclear detectors used for atmospheric radioxenon sampling systems. The Pacific Northwest National Laboratory has developed a method to quantify radon levels in a beta-gamma detector system and determine the level of radon rejection needed to maintain optimal sensitivities. Atmospheric radioxenon measurement systems collect air and purify xenon from the sample to monitor for nuclear explosions. During the purification process, radon is chemically separated and removed from xenon to prevent spectral interference of radon with radioxenon isotopes. Some radon atoms can occasionally accompany the xenon and make it to the nuclear detector. The radon progeny decay then interferes with the radioxenon nuclear measurements due to the overlapping beta-gamma coincidence energy spectra. Accounting for the radon interferences results in increased measurement uncertainty of the radioxenon isotopes and reduced minimum detectable concentration values. Quantifying the amount of radon that enters the nuclear detectors and its impact on the detector sensitives will help place limits on a system's need for radon rejections.

Log 412. MEASUREMENT OF Pu-239 IN AUTOPSY BRAIN TISSUE FROM AN OCCUPATIONALLY EXPOSED WORKER USING ICP-MS

Wegge, D.L. (1,P); Tolmachev, S.L. (2); Brockman, J.D. (1,3). (1) The University of Missouri. (2) United States Transuranium and Uranium Registeries. (3) MU Research Reactor. (P) Presenting Author.

239Pu is a radiotoxic alpha emitter. The International Commission on Radiological Protection evaluated lifetime risk assessment of 239Pu exposure and lung, liver, and bone cancer mortality. There is interest in evaluating neurological outcomes associated with 239Pu exposure and in revision of the biokinetic models to include a brain compartment. This study presents the first work to measure Pu in brain tissue using ICP-MS. Two ICP-MS methods were developed to measure the radiotoxic isotope 239Pu in autopsy brain tissue of an occupationally exposed worker. The first method was a direct analysis of digested samples by quadrupole ICP-MS with quantification by using standard additions. Polyatomic interferences at m/z 239 were investigated. Hg, Pb, an U were identified to form polyatomic interferences in a sample prepared from bovine brain, with a 239/202 formation ratio of 8 x 10-6 from 202Hg, a 239/208 formation ratio of 3 x 10-6 from 208Pb, and a 239/238 formation ratio of 1 x 10-5 from 238U. A second analysis method was developed using extraction chromatography to separate Pu, Am, and U from the matrix followed by isotope dilution analysis of 239Pu. The instrumental LOD of 239Pu was 0.01 ng/kg. The results from the extraction chromatography method showed a 239Pu brain concentration of 0.69 ± 0.05 ng/kg. This result supports the use of isotope dilution ICP-MS to measure 239Pu in autopsy brain samples.

Log 413. **METHOD FOR MEASURING Pu, Am, AND U IN HAIR AND NAILS OF OCCUPATIONALLY EXPOSED WORKERS THROUGH EXTRACTION CHROMATOGRAPHY AND MC-ICP-MS**

Wegge, D.L. (1,P); Tolmachev, S.L. (2); Brockman, J.D. (1,3). (1) The University of Missouri. (2) United States Transuranium and Uranium Registeries. (3) MU Research Reactor. (P) Presenting Author.

Human biomonitoring of actinides is a potential tool that could aid in treaty compliance monitoring, forensic investigations of nuclear smuggling, and for determination of internal dose following a nuclear accident or use of a radioactive dispersal device or nuclear weapon. Currently, the ICRP biokinetic models for actinides do not include hair and nails as a compartment. This work describes an analytical method to measure of $^{235}U/^{238}U$, $^{236}U/^{238}U$, and $^{239}Pu/^{240}Pu$, as well as ^{235}U, ^{238}U, ^{239}Pu, ^{241}Am concentrations at ultra-trace levels by ICP-MS. Extraction chromatography resins TEVA, DGA, and UTEVA were used to separate Pu, Am, and U from the digested sample. The concentration of Pu and Am was measured using isotope dilution analysis with a multi-collector ICP-MS. The instrumental LOD was 1×10^{-5} ng/kg for ^{239}Pu and 3×10^{-5} ng/kg for ^{241}Am. The measured concentrations of ^{239}Pu in 18 hair and nail samples from 9 occupationally exposed individuals ranged from $1.8 \times 10^{-1} – 1.6 \times 10^{1}$ ng/kg. The $^{240}Pu/^{239}Pu$ isotopic ratios ranged from 0.029 to 0.075, indicating an exposure to weapons-grade Pu. The concentration of U was measured using a quadrupole ICP-MS and had an instrumental LOD of 1×10^{-3} ug/kg. The U levels ranged from $3.4 \times 10^{-5} – 6.0 \times 10^{-1}$ ug/kg. This work demonstrates that ^{239}Pu is excreted through hair and nail for years after the reported exposure and could be useful as a long-term biomonitor.

Log 414. **STUDY OF HISTORICAL BOMARC NON-CRITICAL WEAPON ACCIDENT DEBRIS**

Heffelfinger, A. J. (1), Varshney, G. (1,P), Bickley, A. A. (1) and Petrosky, J. C. (1)
(1) Department of Engineering Physics, Air Force Institute of Technology, 2950 Hobson Way, Wright-Patterson AFB, OH 45433, USA, (P) Presenting Author

The United States maintains an advanced nuclear forensics capability to identify the origins, source, and processing history of nuclear devices in case of a nuclear terrorism attack and other kinds of domestic nuclear detonations. To support establishing forensic signatures, the use of historical nuclear test debris is imperative for establishing physical and chemical processes that influence the method of formation and/or subsequent modification of the debris. This research focuses on historical environmental nuclear debris produced in the Boeing Michigan Aeronautical Research Center (BOMARC) accident (McGuire Air Force Base, 1960), which remained in the local environment for more than 50 years. Non-destructive analytical (NDA) techniques, including gamma spectroscopy and Scanning Electron Microscopy/Energy Dispersive X-Ray Spectroscopy (SEM/EDS) are employed to characterize the actinide-bearing micro-particles retrieved from the environs surrounding the accident, some 40 years after the accident. Here, we identify 70 microscopic particles (ranging in size from 1-34 microns) containing actinide remnants from the accidents and cataloged them using a lexicon for the development of databases. Elemental analysis indicates the presence of uranium in all 70 particles and identify trends of other major and trace elements within these particles. Our SEM analysis shows morphological trends that

indicate particles across the four evaluated size distributions have similar circularity, but we find three major particle classes present based on shape, angularity, and surface features.

Log 415. **FRACTIONATION OF NUCLEAR DEBRIS Cs-137 AND Sr-90 IN FAYETTEVILLE, AK RAIN 1973-1976**

Inn, K.G.W. (1,2,3,P); Kuroda, P.K.(3); Raines, W.(3); Flanders, M.(3); Carl Harris, C.(3). (1) K&E Inn Ovations, Inc. (2) National Institute of Standards and Technology, Retired. (3) University of Arkansas. (P) Presenting Author.

During the period of June 1973 and July 1976, the Chinese detonated four nuclear weapons in the atmosphere. The resulting fallout was spread worldwide and also fell at Fayetteville, AK where the University of Arkansas Department of Chemistry had rain collectors as well as air filter samplers. This study looked at the fractionation patterns from the Cs-137 and Sr-90 fallout. There was evidence of mixed production of Cs-137 and Sr-90 mixes from two energies of neutrons, likely fission from Pu devices, unexpectedly large fractionation of volatile fission products from refractory fission products and re-entrainment of previously deposited Cs-137 in storm systems. These methodologies could have application to current nuclear incident, environmental and radioecology studies.

Log 416. **RADIOCHEMICAL MEASUREMENT UNCERTAINTIES & CORRELATIONS – PHASE II [DEEPER INTO THE RABBIT HOLE]**

Inn, K.G.W. (1,2,P); McCroan, K.(3); Filliben, J(4). (1) K&E Inn Ovations, Inc. (2) National Institute of Standards and Technology, Retired. (3) US Environmental Protection Agency, National Analytical Radiation Environmental Laboratory. (4) National Institute of Standards and Technology. (P) Presenting Author.

The art of expressing measurement uncertainties can be a lifelong pursuit. The first two steps are the most difficult to do: 1) describing the measurement equation, and 2) list uncertainty components and estimating uncertainty values for each, This work provides starter lists of uncertainty components, and suggestions for addressing correlation of uncertainty components. The net effect of this effort is to assist the reader recognize a number of heretofore "dark uncertainty" components in their measurement process so that they may be successfully reduced, understood, and controlled.

Log 417. STATISTICAL & GRAPHICAL ANALYSIS OF VARIANCE IN MEASUREMENT ACCURACY ACROSS RADIOANALYTICAL LABORATORIES- 10 YEAR NIST RADIOCHEMISTRY INTERCOMPARISON PROGRAM (NRIP) ASSESSMENT

Inn, K.G.W. (1,2,P); Johnson, N.(3); Outola, I.(4); Filliben, J.(5). (1) K&E Inn Ovations, Inc. (2) National Institute of Standards and Technology, Retired. (3) University of Maryland, Baltimore County. (4) Formerly National Institute of Standards and Technology. (5) National Institute of Standards and Technology. (P) Presenting Author.

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The NIST NRIP has been assessing radioanalytical laboratory capabilities for a number of years. This study evaluates performance with respect to evaluation of measurement "outliers," 15 radionuclides, 20+ laboratories, 5 matrices, and over a span of 10 years. Results indicated: (1) the data is best characterized using medians and treating "outliers" as part of non-normal distributions; (2) radionuclides and then laboratory showed the largest variation in performance; (3) of all the radionuclides tested, Sr-90 showed the most variance, while alpha-emitter analysis were in good control; (4) overall laboratory performance improved after 2-3 years of experience in the program, and laboratories that tended to be in control continued to demonstrate consistent performance while some laboratories that needed to improve did so with continued participation in the program;   (5) spiked fecal and soil sample results were more problematic than acidified water, spiked urine and then glass fiber filters has the best results; and (6) there was little variation of overall laboratory performance over the years of testing. These results provides NRIP, the participating laboratories with much needed feedback to address problem areas that were uncovered, share advanced radioanalytical methods to improve lagging laboratory performance, and provide a platform for discussion of new radionuclides, matrices, interferences, and approaches for the pass/fail criteria and uncertainty reporting requirements to improve and expand the performance evaluation program.

Log 418. COMPARISON OF RADIOXENON ANALYSIS ALGORITHM RESULTS

Cooper, M.W.(P); Abromeit, B.L.; Ely, J.H.; Harper, W.W.; Hayes, J.C.; Mayer, M.F.; Panisko, M.E.; Slack, J.L. Pacific Northwest National Laboratory. (P) Presenting Author.

Pacific Northwest National Laboratory has recently developed a radioxenon matrix analysis method, called MITER, to account for interferences between 133Xe, 135Xe, and especially the two metastable xenons, 131mXe and 133mXe. The method was developed to provide more accurate results and to reduce the false positive/negative results when the metastable xenons are present. A comparison of the new MITER analysis to a 7-ROI net count calculation analysis will be presented using Xenon International data collected during field testing. The comparison will highlight the improvements gained by using the MITER analysis method.

Log 419. FORMATION OF URANYL PEROXIDE CLUSTERS UNDER IONIZING RADIATION

Felton, D.E. (1,P); LaVerne, J.A. (1); Burns, P.C. (1). (1)The University of Notre Dame. (P) Presenting Author.

Actinide peroxide nanoclusters self-assemble in aqueous solution across a wide pH range yet little is known about the mechanism of formation. Irradiation of water generates reactive species, including peroxide, that may produce conditions favourable for nanocluster formation. The impact of radiation on uranyl peroxide cluster formation is unexplored, although earlier studies have demonstrated the formation of the peroxide mineral studtite during irradiation. The three most readily synthesized uranyl peroxide clusters are U24, U28, and U60. This study aims to investigate how radiation impacts the formation of these uranyl peroxide clusters as well as their stability in a gamma radiation field after formation. Going further past this uranyl peroxide clusters were synthesized with radiolysis generated peroxide to study how high doses of ionizing radiation would affect uranyl peroxide nanocluster formation from a uranyl hydroxide solution. This study aims to determine which radiolysis products promote or hinder nanocluster formation as well as determining the impact of the uranyl ion on water radiolysis.

Log 420. CHARACTERIZATION OF ISOTOPE ABUNDANCE RATIOS OF Pu AND MOX BEARING-PARTICLES BY SECONDARY ION MASS SPECTROMETRY

Diacre, A. (1,2); Fauré, A.L. (1); Cornaton, M. (1); Pointurier, F. (1,P); Evrard, E. (2). (1) CEA, DAM, DIF, F-91297 Arpajon, France. (2) Laboratoire des Sciences du Climat et de l'Environnement (LSCE/IPSL), Unité Mixte de Recherche 8212 (CEA/ CNRS/UVSQ), Université Paris-Saclay, Gif-sur-Yvette, France. (P) Presenting Author.

Every industrial facility, industrial and nuclear accident emits micrometric fragments of material in the environment whose elemental and isotopic compositions are characteristics of the process or event. Thus particle analysis is a powerful tool to monitor nuclear activities in the framework of the Non-Proliferation Treaty and to assess the exposure of human beings to ionizing radiation. Initially, particle-scale isotopic analyses aimed at the determination of the uranium isotopic composition. Now focus is given on plutonium detection and characterization both in pure Pu particles but also in mixed uranium-plutonium particles. Such measurements are more challenging because of the presence of isobaric interferences induced by hydride ions, like $238UH+$ on $239Pu+$. Such ions are generated during ionization process when using Secondary Ion Mass Spectrometry. We described here the analytical methodology that has been developed to measure simultaneously the U and Pu isotopic composition and the Pu content of two MOx samples whose Pu content differs. Mass bias and hydride rate determination were determined on uranium standard particles. Applied corrections allow obtaining accurate results. The choice of the U and Pu ionic species to be analyzed and the influence of the Pu content on the accuracy are also discussed.

Log 421. MACHINE LEARNING APPLICATIONS IN THE CHARACTERIZATION OF RADIOXENON SIGNATURES

Hunter, C.J. (1,P); Powers-Luhn, J.R. (2); Hartig, K.C. (1). (1) The Unviersity of Florida. (2) Pacific Northwest National Laboratory. (P) Presenting Author.

The International Monitoring System (IMS) is a worldwide network of more than 300 continuously operating monitoring stations deployed by the Comprehensive Test Ban Treaty Organization (CTBTO) with the purpose of detecting nuclear detonations anywhere across the globe. Sensor technologies employed at these stations include seismic, infrasonic, hydroacoustic, and radionuclide detectors, and of these four, only the later provides a definitive means of confirming a nuclear detonation. Gaseous radionuclides, and particularly xenon-131m, 133, 133m, and 135 with their relatively long half-lives, are of special interest due to their ability to escape into the atmosphere independent of the detonation location. However, because the production and emission of these radionuclides are not exclusive to nuclear detonations and can be regularly found in air samples where stations are present, inherent interference signals exist and must be accounted for. Currently, radionuclide measurement analysis is performed in a generally automatic fashion, accounting for background and any other contributory signals to characterize a measurement with little effort to integrate modern computer enabled technologies. This work utilizes machine learning techniques to autonomously discriminate between IMS radionuclide gas detector measurements and enhance the confidence of event determination. This is done by exploiting convolutional neural networks (CNNs) to transform measurement data followed by unsupervised clustering methods for measurement differentiation.

Log 422. NOVEL APPROACHES OF ACTINIDE TARGET MAKING FOR NUCLEAR SCIENCE MEASUREMENTS

Khachatur Manukyan

The safe and economical production of robust, stable, actinide targets of desired thicknesses and uniformity has remained a challenge in nuclear science studies. Over the last few years, we established a new target laboratory infrastructure at the University of Notre Dame. Our research program investigates solution combustion synthesis (SCS) reactions between actinide metal nitrates with different organic compounds to ensure the production of robust and uniform targets. The SCS reactions coupled with spin-coating and sparing techniques produce isotopically pure targets with tunable thicknesses. This talk will present recent achievements in preparing actinide targets. It will discuss the relationships between process conditions, structure, and morphology of the resulting targets. Selected examples of thin-film target tests with charged particles will also be highlighted. The results on target characterization by X-ray fluorescence, X-ray photoelectron spectroscopy, Raman spectroscopy, alpha-particle emission spectroscopy, and high-resolution transmission electron microscopy before and after irradiation tests will also be presented. The significant relationship between the structure of targets and their stability will be discussed. The work was performed with financial support from the U.S. Department of Energy's (DOE) National Nuclear Security Administration (NNSA, Grant # DE-NA0003888).

Log 423. **MICRO-ANALYTICAL TECHNIQUES FOR FAST CHARACTERIZATIONS OF NUCLEAR MATERIALS IN THE FRAME OF NUCLEAR FORENSIC**
Pointurier, F (1, P); Fauré, A.L. (1); Cornaton, M. (1); Marie, O. (1), Hubert, A. (1); Humbert A.C. (1). CEA, DAM, DIF, F-91297 Arpajon, France. (P) Presenting Author.

In case of nuclear forensics events like the seizure of nuclear materials, information related to the isotopic, elemental, molecular and structural compositions of the materials must be reported as soon as possible to the authorities. Now handling of macroscopic amounts of nuclear and radioactive materials requires specific facilities and equipment and lengthy procedures. An alternative approach is to collect and analyze microscopic fragments of the materials, i.e., a tiny fraction of powdery materials or microparticles sampled on the surfaces of continuous solid materials or recovered after fragmentation of the objects. Therefore, the sampled amounts are low enough, typically in the ng to the µg range, to be safely and rapidly analyzed in available facilities and instruments usually dedicated to trace analysis in environmental samples. Moreover, an additional advantage of such an approach is to provide information on homogeneity of the materials, allowing possible identification of components of a mixture of several compounds. In this presentation, implementation of analytical techniques applicable to direct analysis of individual microparticles in the frame of nuclear forensic exercises will be described. Examples of applications of SIMS and laser ablation – ICPMS coupling for measurement of the isotope composition, and of Raman spectrometry coupled to SEM-EDX for identification of chemical phases, morphology and elemental compositions of the microparticles will be presented. Advantages and limitations of these techniques will also be discussed.

Log 424. **GEANT4 MODELING OF $CsPbBr_3$ AND $Cs_3Bi_2I_9$ GAMMA DETECTORS**
Han, C.(1); Barzilov, A.(1,P). (1) University of Nevada Las Vegas. (P) Presenting Author.

The wide-bandgap semiconductors CsPbBr3 and Cs3Bi2I9 with a high mobility-lifetime product are promising materials for the design of ambient temperature, high efficiency, high energy resolution gamma-ray detectors of large volume. The development of large-size inorganic perovskite detectors requires high-fidelity computational models. The performance of perovskite gamma detectors was studied using the stochastic code GEANT4 and the ROOT toolkit. Photon and charged particle interactions with matter, including event and track management, for various detector geometries was simulated by GEANT4, and ROOT was used to process and analyze the gamma energy distributions. It was determined that the energy resolution of the perovskite detector is comparable to that of a CZT detector of a similar geometry. For example, computed energy resolution and photo peak efficiency under the incident 662-keV gamma rays for 8 cm3 CsPbBr3 crystals are 1.1% and 29.2%, respectively. The cost-effective larger size perovskite detectors can be fabricated with a low defect density compared to CZT.

Log 425. **ISOTACHOPHORETIC SEPARATION AND CHARACTERIZATION OF f-BLOCK ELEMENTS**

Branch, S.D.(1,P); Lackey, H.E.(1); Lines, A.M.(1); Veldman, T.G.(1); Humble, P.H.(1); Arnold, E.S.(1); Schroeder, J.M.(1); Beck, C.L.(1); Katalenich, J.A.(1); Bottenus, D.R.(1); Seiner, B.N.(1). (1) Pacific Northwest National Laboratory. (P) Presenting Author.

Microfluidic devices provide ideal environments for small-scale, versatile processing and sample separations. Microfluidics also serve as a rapid, low-cost alternative to traditional lab-scale efforts. Previous efforts have demonstrated the applicability of a microfluidic chip used for the electrophoretic separation of 14 rare earth elements from a homogenous sample. This technique – isotachophoresis – relies on a two-buffer system that allows target species to stack at maximum concentrations between the two buffers. Coupling of the micro-scale system to downstream detection instrumentation, such as inductively coupled plasma mass spectrometry (ICP-MS) has been used to optimize the separation by using commercial off-the-shelf products to provide an interface that allows for immediate injection of the stacked sample on the fabricated chip into the ICP-MS, where the elemental composition of the sample is analyzed. The successful demonstration of isotachophoretic separation of the lanthanides on the chip has led to upscaling to a bench scale platform that allows for higher sample throughput and higher concentrations of analytes. Results presented will highlight the effectiveness of the up-scaled system.

Log 426. **EXTRACTIONS OF Th(IV) AND U(IV,VI) FROM NITRIC ACID SOLUTION USING ORGANOPHOSPHORUS REAGENT (HEH[EHP])**

Castillo, J. (1,P); Gelis, A.V.(1); (1) University of Nevada, Las Vegas. (P) Presenting Author.

Understanding actinide (An) speciation in lipophilic organic systems is a critical factor for developing novel liquid-liquid extraction processes for advanced nuclear fuel reprocessing and medical isotope separation. One such process (ALSEP) utilizes a combination of an organophosphorus acidic extractant, 2-ethylhexylphosphonic acid mono-2-ethylhexyl ester (HEH[EHP]) and a neutral extractant, N,N,N',N'-tetra(2-ethylhexyl)diglycolamide (T2EHDGA). The goal of this project is the speciation of An(IV, VI) in HEH[EHP]/hydrocarbon organic systems using techniques like 31P & 1H NMR, and FT-IR. Previous results collected at room temperature (293 K) showed coalescence of the expected two 31P peaks (corresponding to bound to unbound ligand dimers). More recent results collected at 203 K show two peak regions for Th(IV) and three for U(IV,VI). This further convolutes the interpreted system and a clear explanation about the speciation in the cases of U(IV,VI) extraction is still needed. So far, there were no differences in IR spectra between the blank and loaded organic phase. This is likely due to chemical matrix signals overpowering the nitrate signal. Higher-loading conditions (i.e. greater concentrations of metal and/or organic extractant) could extract more metal and (possibly) nitrates, resulting in a clearer FT-IR signal. Similar studies with U(IV), Pu(IV), and Np(IV,VI) can be used to further define the behavior of HEH[EHP] extraction across the An series.

Log 427. **IN-SITU POSITRON ANNIHILATION SPECTROSCOPY OF IRRADIATION INDUCED DEFECTS**

Chuting Tan (1,P), Paul J. Stonaha (2), Jagoda M. Urban-Klaehn (1), Andrew T. Smolinski (1), Kyrone D. Riley (1), Kyle Francis (3), Connor J. Harper (4), Tony Forest (4), Chase N. Taylor (1). (1) Idaho National Laboratory. (2) Physics Education and Consulting, LLC. (3) Walsh Engineering Services. (4) Idaho State University. (P) Presenting Author.

Providing direct measurements of material dynamics at the early stages of radiation damage will facilitate the advancement of nuclear systems, especially for early detection of material failure. Herein, we have commenced a proof of principle study for a new positron annihilation spectroscopy capability that uses neutrons to induce damage while generating positrons to analyze the defect evolution in nuclear fuels and materials. The science question we hope to answer using this technique is in nuclear materials and fuels, how point defects, specifically vacancies, are generated and evolve at the onset of irradiation damage (such as low as $10-5$ to $10-4$ dpa in studies concerning pressure vessel alloys). At present, only few methods can experimentally characterize vacancy or void type of defects at low dose stage, which features defects of small sizes and low concentrations across the bulk of irradiated samples. The resulting experimental data gap leads to uncertainty in modeling and simulation at larger length and time scales. PAS is sensitive to low vacancy concentration (~1014 cm-3) and subnanometer defect size. Therefore, it is capable of non-destructively probing the formation of vacancy defects under neutron doses well below 1 dpa. The capability will enable routine atomic defect examination of fuels and materials, potentially shorten fuel and material qualification process, and improve our under-standing of material damage in complex, fast changing and dynamic conditions. In this presentation, theory, design, and verification will be discussed.

Log 428. MATRIX-ASSISTED IONIZATION MASS SPECTROMETRY FOR URANIUM MEASUREMENT IN FIELD ENVIRONMENTS

Danielle Mannion (1,P), Kyle Samplerton (1), Elizabeth LaBone (1), Haley Lawton (1), Joseph Mannion (1), Wendy Kuhne (1), Binod Rai (1), Matthew Wellons (1). (1) Savannah River National Laboratory. (P) Presenting author.

Mass spectrometry is considered the "gold standard" in nuclear safeguards for the analysis of long-lived actinides such as uranium. However, conventional mass spectrometric analyses often require time-consuming sample preparation and significant laboratory infrastructure. Currently the nuclear safeguards community lacks the ability to rapidly determine uranium enrichment and assay of low-level samples (<100 ng U) while in the field. Matrix-assisted ionization (MAI) is a recently discovered atmospheric pressure ionization technique for mass spectrometry (i.e., MAI-MS) which holds promise as an enabling technology for overcoming many of the challenges associated with in-field mass spectrometry. MAI generates gas phase ions without the application of heat, electrons, photons, or high voltage. Without using a traditional ion source, MAI-MS enables sub-nanogram detection of molecular uranium species on the timescale of seconds. MAI-MS method development for uranium isotope ratio determination has demonstrated accurate and repeatable U-235/U-238 measurement using as little as 100 pg U-235 in low-enriched standards. Uranium calibration has been explored for the simultaneous measurement of enrichment and assay. Uranium calibration curves generated with respect to both a standard spike solution and the MAI matrix exhibit linearity with U-238 content; measured assays have uncertainties of ca. 20% and agree well with known values between 5-500 ng U. Research to understand the inorganic ionization mechanisms of MAI is ongoing and appears to differ from published theories of organic ionization mechanisms. Continued research into this novel ionization technique could be highly transferable to safeguards uranium measurement in the field.

Log 429. TRIGA MARK II PRODUCTION AND ISOLATION OF n.c.a. GOLD-199 - A RADIONUCLIDE WITH PROMISING PROPERTIES FOR MEDICAL APPLICATION

Denk, C . (1, P), Pressler, M. (1)
(1) Center for Labeling and Isotope Production, TU Wien, Atominstitut, 1020 Vienna, Austria. (P) Presenting Author.

Gold-199 has a set of promising properties for medical application: Its intermediate half-life of 3.14 days allows the use in combination with long circulating targeting vectors (e.g. antibodies), that are not compatible with commonly used short-lived radionuclides. The ß-max energy of 452 keV is well suited for radionuclide therapy, and accompanying medium energy gammas allow for imaging using SPECT or scintigraphy. Medium flux reactors are suitable to access 199Au in high activities and in non carrier added (n.c.a.) form. Based on literature procedures we produce [199]Au from natural platinum targets (using the [198]Pt(n,γ)[199]Pt reaction followed by ß[-] decay to [199]Au) The Pt/Au separation using liquid/liquid extraction has been improved, giving high yields of [199]Au with high radionuclidic purity within a short time span. Automation of the process to allow handling of high activities is currently under development. Within this conference contribution our results and experiences regarding [199]Au production will be presented.

Log 430. **ELECTROCHEMICAL SEPARATION OF Co-60 FROM ELECTROPOSITIVE RADIONUCLIDES**
Dutheil, P.(1,2,P); Heule, M.(1); Steinegger, P.(1,2). (1) Paul Scherrer Institut (PSI). (2) Swiss Federal Institute of Technology Zürich (ETHZ). (P) Presenting Author.

The quantification of radionuclides in nuclear waste is essential to provide good risk assessment during decommissioning of nuclear facilities. High activities of Co-60 in irradiated matrices (e.g., activated steel) can be an important source of interference during analysis of minor radionuclides by gamma-spectrometry or liquid scintillation counting. In that respect, the removal of Co-60 from complex samples containing other gamma- or beta-emitters (e.g., Sr-90 or Ni-63) must be carried out before the analysis. Thus, a fast and easy procedure is required to provide a reliable quantification of minor radionuclides in a mixture. Selective electrochemical reduction provides an easy way to separate elements based on their respective reduction potentials. Cobalt has a higher reduction potential than numerous other elements usually present in nuclear waste (e.g., Sr-90 or Cs-137). Investigations regarding the development of a separation method based on selective electrodeposition of cobalt are presented. Reduction potentials of cobalt at glassy carbon electrodes were characterized by cyclic voltammetry in various conditions in order to design a controlled-potential reduction procedure. Gamma-spectrometry analysis showed that quantitative electrodeposition of Co-60 and separation in the presence of various electropositive radionuclides such as Cs-137, Sr-85, and Ba-133 could be obtained within a few hours by applying -1.35 V (vs Ag/AgCl/3M KCl) to a glassy carbon electrode in a KCl electrolyte.

Log 432. **USE OF NEUTRON RADIOGRAPHY FOR MEASUREMENTS IN CONCRETE**

Ghantous, R.M. (1, P); Weiss, J. W. (1); Reese, S. R. (2); (1) Oregon State University, Civil and construction engineering Department. (2) Oregon State University, Nuclear and Science Engineering Department. (P) Presenting Author.

Neutron radiography is used to study concrete due to the large thermal scattering and absorption cross sections with hydrogen (i.e., water). Primarily two types of studies have been performed: (1) investigations on the reaction of water with cement (i.e., hydration), (2) investigations on the transport properties (i.e., the movement of water through the porous network). Transport of water in concrete occurs primarily through five mechanisms: wicking, permeation, diffusion, absorption and drying. Many studies have focused on the determining the rate of fluid transport in concrete, particularly near the concrete surface. The depth of water penetration from a concrete surface can be determined with a high accuracy using neutron radiography unlike the other techniques that are destructive and rely on visual inspection. This presentation will provide examples of the use of neutron radiography in examining drying, specifically as it relates to studying 3D printed samples exposed to air causing drying directly after printing. In addition, this presentation will describe how neutron radiography is used to assess the reaction of cement (i.e., curing). Measurements associated with curing have enabled the degree of hydration, and porosity, to be quantified as a function of distance from the drying surface. Finally, examples of fluid absorption are presented. Neutron radiography is a powerful technique for studying concrete.

Log 433. **3D-PRINTED MICROFLUIDIC EXTRACTION CELLS FOR FIELD-DEPLOYABLE NUCLEAR FORENSICS**

Glennon, K.J.(1,P); Valdovinos, H.F.(1); Parsons-Davis, T.(1); Shusterman, J.A.(1); Gharibyan, N.(1). (1) Lawrence Livermore National Laboratory, (P) Presenting Author.

Developing field-deployable chemistry systems can significantly improve time-critical analyses for the characterization of post-detonation nuclear debris. Compact (5 x 5 x 1 cm) microfluidic modules have been designed with CAD software and produced using a high-resolution 3D printer as a field-deployable chemistry platform. A porous PTFE membrane is soaked with a tributyl phosphate (TBP) solution and is placed between two modules; separation is performed as a liquid-liquid extraction across this membrane, where the forward and back-extractions occur within one system. The modules may be stacked into a tower performing multiple separations until the desired decontamination factors have been reached. The single-module system has been used to separate Pu from U with (98.9 ± 2.0)% and (97.6 ± 2.7)% yield for Pu and U, respectively. These extractions are complete in under an hour and use less than 800 uL of solution total. The system has also been used to selectively extract U from solutions containing fission products and dissolved soil with U yields > 98%. The microfluidic system has been coupled with a custom 3D printed alpha chamber to perform Pu isotopic analysis in the field. The system has been benchmarked against a solution of CRM 138 Pu with a Pu/U mass ratio of 1E-4 followed by an online isotopic measurement of the Pu by alpha spectrometry; the measured Pu isotopics were consistent with certificate values for CRM 138 Pu.

Log 434. EVALUATION OF TWO AG1 - X8 COLUMNS FOR RAPID SEPARATION OF MULTIPLE RADIONUCLIDES

Staci Herman, Leah M. Arrigo, Ean Arnold, Morgan M. Haney, Lindsay Irwin, Bethany Lawler, Chelsie Beck

A sequential separation involving two anion exchange columns was developed to isolate and radiometrically analyze several activation products including isotopes of Co, Cd, Ir, Zn, Fe, and Ga. The developed method was tested using stable tracers in a sample of irradiated uranium with $1x10^{12}$ fissions. The first separation involved a decreasing HCl gradient to separate fractions containing Co, Cd, Zn, and Ir with elemental recoveries being 98 ± 4 %, 100.2 ± 0.8 %, 100 ± 1 %, and 39 ± 5 %, respectively. From the first column, a fraction containing Fe and Ga was also collected. Separation of these two elements was achieved using the second anion exchange with ascorbic acid being used to provide redox control to enhance the ion exchange. The separated Fe and Ga fractions contained elemental recoveries of 95 ± 3 % and 85 ± 2%, respectively. Using the two sequential anion exchanges columns, six elements were successfully separated from one another with high recoveries and from major interferences in a mixed activation/ fission product sample.

Log 435. SEQUENTIAL SEPARATION SCHEME FOR REFRACTORY ACTIVATION PRODUCTS

Staci Herman, Evan Warzecha, Riane Stene, Hilary Emerson, Morgan Haney, Lori Metz, and Chelsie Beck

Multiple activation products can be separated from a mixed activation and fission product sample through a sequential separation scheme. The 17 different activation products that will be separated are Au, Be, Cr, Cu, Fe, Ga, Mn, Ni, Pd, Po, Pt, Sc, Ta, Ti, V, W, and Zn. A TEVA-TRU double stack is used first to separate Ta and W from the rest of the activation and fission products. A cation exchange column is used next to separate Au, Pd, Pt and Po. Finally, an actinide column is used with an increasing HNO_3 gradient to form a Ni, Cr/ Cu, Be/ Mn, and Ti/ V fractions. A sequential separation for multiple activation products maximizes the number of isotopes that can be separated when sample is limited.

Log 436. **COMPARISON OF MIXED-MODAL RESIN TO SOLVENT EXTRACTION FOR USE IN NUCLEAR FORENSICS**

Matthew Risenhuber, Staci Herman, Evan Warzecha, Hilary Emerson, Chelsie Beck, Brienne Seiner.

The actinide lanthanide separation (ALSEP) process is a well-known solvent extraction system that uses two active extractants, N,N,N',N'-tetraoctyldiglycolamide (TODGA), and an acidic extractant 2-ethylhexylphosphonic acid mono-2-ethylhexyl ester (HEH[EHP]), to remove and purify minor actinides from used nuclear fuel. The current work aims to understand the translatability of ALSEP to a multi-modal resin chromatography system using Eichrom Technologies commercial resins, DGA-n and LN2 resin, which hold the same extractants used in the ALSEP solvent extraction process. Through transition of solvent extraction to column chromatography, less organic radiological waste will be formed decreasing the inherent human and environmental risk. The solvent extraction method will be compared to both a stacked and mixed DGA-n and LN2 resin in a single column bed. Presented will be the results comparing the classic ALSEP process as well as the double stack cartridge and the mixed resin single column with DGA-n and LN2. These results will demonstrate the benefits and limitations of the ALSEP process at the radioanalytical scale using mixed resins as opposed to a solvent extraction system.

Log 437. **ELECTROCHEMISTRY TO IMPROVE SEPARATION PERFORMANCE AND ANALYSIS OF PURE ALPHA AND BETA NUCLIDES**

Martin Heule (1, P), Paul Dutheil (1). (P) Presenting Author.

The potential of electrochemical procedures both for separation and analysis purposes has not been fully exploited in deconstruction efforts that require increasing numbers of samples for analysis of pure alpha and beta emitters. The sample materials include a variety of metals, plastics as well as concrete. Sample preparations require digestion, chemical separation of the nuclides and deposition for measurement on a surface, e.g. for alpha spectrometry. However, these methods are very labor-intense and usually involve preparative chromatography or precipitation reactions. The contribution will highlight the advantages complementary to the use of traditional techniques. Electrochemistry allows both for inducing selective separation and for analytical monitoring of the quality of the separation processes at the same time. In contrast to established methods, there is a big potential for automatisation and adaptation of the process to the individual chemical matrix of the sample at hand. Techniques include cyclovoltammetry, fixed potential reduction, the modulation of oxidation states in solution, complex-based modification of reduction potentials for improved selectivity, but also include the possibilities of chronopotentiometric evaluation. The discussion will include properties of various working electrode materials such as modified glassy carbon as well as ionic liquids. While it is not realistic that electrochemical methods can fully replace the traditional processes, a special focus is given on methods of interfacing various process steps.

Log 438. **OBTAINING SPATIAL DATASETS OF ATMOSPHERICALLY PRODUCED COSMOGENIC ^{10}Be/^{7}Be TO FINGERPRINT STRATOSPHERIC DISTURBANCES**

Hidy, A. J. (1, P); Wharton, S. (1); Skinner, S. N. (2); Ehrmann, T. S. (1); Repasch, M. N. (1). (1) Lawrence Livermore National Laboratory. (2) University of Maryland. (P) Presenting Author.

Atmospheric 7Be and 10Be are produced mostly in the stratosphere from cosmic ray collisions with heavier nuclei. Once produced, they adsorb strongly to ambient aerosols. These aerosols accumulate 10Be and 7Be throughout their stratospheric residency until they are ultimately transported to the troposphere where they are quickly removed by scavenging or deposition. Because of the short half-life of 7Be (53 days) with respect to 10Be (1.4 Myr), 10Be/7Be in aerosols is highly sensitive to stratospheric residence time and is thus a powerful tracer of air mass transport and stratosphere-troposphere exchange. However, the low abundance of these isotopes has restricted in-situ atmospheric collections to aircraft campaigns capable of sampling large air volumes. Similarly, the logistics of counting numerous low-level 7Be samples from a single campaign has limited creation of the large spatial datasets needed for event-based applications. To overcome these limitations, we apply an accelerator mass spectrometry (AMS) capability recently developed at CAMS-LLNL. This technique allows quick analysis of 10Be and 7Be from a single AMS target, needs as little as 10,000 atoms of either isotope, and requires minimal sample preparation. In FY22 we plan to obtain several large spatial datasets of 10Be/7Be from high-altitude balloon missions and ground-based rain collectors to investigate mechanisms behind troposphere-stratosphere exchange processes including gravity waves induced by deep convection. Prepared by LLNL under Contract DE-AC52-07NA27344.

Log 439. **STIR BAR SORPTIVE EXTRACTION AND COMPREHENSIVE GCXGC HIGH-RESOLUTION MASS SPECTROMETRY FOR THE ANALYSIS OF ORGANO-IODIDES IN SURFACE AND GROUND WATERS**

Mannion, J.M. (1,P); Brant, H.(1); Gamble, S.(1); Labone, E.(1); Mannion, D.R.(1); Samperton, K.(1). (1) Savannah River National Laboratory. (P) Presenting Author.

The high mobility and long half-life of 129I make this species a particular environmental concern at contaminated locations such as the Savannah River Site (SRS), South Carolina. The complex biogeochemistry of iodine is influenced by various factors including redox potential, sediment organic matter concentrations, and microbial activity with resultant iodine speciation dictating mobility through aquatic environments. Based on environmental conditions a significant fraction of total iodine can be in an organic form within the hydrologic system. Understanding of specific chemical species within this organo-iodide fraction is currently limited. A high sensitivity analytical method was developed to interrogate the organo-iodide fraction of natural waters using stir bar sorptive extraction with comprehensive multidimensional gas chromatography (GCxGC) and high-resolution mass spectrometry (HRMS). Organic species within the aqueous sample are directly extracted onto a stir bar coated with a sorptive material then introduced to the GCxGC system via thermal desorption. Due to the unique mass defect of iodine high-resolution mass spectrometry can differentiate iodine-bearing ions from non-halogenated interferents. The comprehensive GCxGC provides improved separation over single dimensional gas chromatography and markedly improves sensitivity (approximately 1.5 orders of magnitude)

through chromatographic focusing. In combination, GCxGC and HRMS allow for limits of detection approaching the low picogram range for numerous organo-iodide species. Untargeted screening for organo-iodides within high complexity samples is possible with methods such as mass defect analysis. An automated data analysis pipeline using the R statistical programming environment is under development for interrogating these complex datasets.

Log 440. **UNIVERSITY OF NEVADA, LAS VEGAS: EDUCATIONAL OPPORTUNITIES FOR WOMEN IN NUCLEAR SCIENCE**
Johns, W. (P). (P) Presenting Author.

A Ph.D. from UNLV in the field of radiochemistry provides career opportunities in areas such as radiochemistry, nuclear chemistry, materials research, renewable energy studies, nuclear forensics, and environmental studies. UNLV is a Minority Serving Institution seeking to enhance education in the STEM disciplines for women and minorities. Since 2008, 30% of radiochemistry PhD graduates have been women. With current enrollment of 50% women, our program is positioned to continue making impactful gains in opportunities for women in the nuclear sciences. The program at UNLV is a student-driven, research intensive program that stresses fundamental aspects of radiochemistry science allowing students to gain hands-on experience in handling, manipulating and detecting unsealed radioactive material early in their career. The curriculum and research provide a comprehensive and interdisciplinary examination of topics and experiences necessary to produce graduates who are ready to secure employment. The program facilities contain a host of experimental equipment for use in research including radionuclide counting facilities, spectroscopy, microscopy and x-ray diffraction. It focuses on student development in the areas of technical competency, communication skills, analytical and critical thinking skills and expertise in the field of radiochemistry. Women graduates from the UNLV Radiochemistry PhD program have secured employment and forged successful careers in industry and national laboratories such as BNL, LLNL and LANL.

Log 441. PLUTONIUM ISOTOPIC MEASUREMENTS USING A SEGMENTED HIGH-PURITY GERMANIUM DETECTOR

Rim, J.H. (1,P); Porterfield, D.R.(1); Smith, M.(1). (1) Los Alamos National Laboratory. (P) Presenting Author.

A segmented High-Purity Germanium (HPGe) detector can enable identification, location, and quantification of a radioactive source by combining gamma-ray spectral data from a segmented germanium crystal with an imaging system. This technology was primarily tested and utilized in an emergency response area to locate and identify nuclear material [ref]. This presentation will discuss work performed to use a segmented HPGe detector for Pu sample identification and to further perform Pu isotopic analysis on multiple samples from a single measurement. The PHDS Co (Knoxville, TN, USA) GeGI HPGe detector system was used in the performance of this work. Two to three Pu samples were measured using a lead collimator in front of the GeGI HPGe detector. The localization of Pu items can be performed rapidly, within 15 minutes, using the in-grown Am-241 and low-energy Pu gamma emissions. The Pu isotopic abundance measurements required longer count durations due to low counting statistics for Pu-240 (160 keV) but Pu isotopic measurements can be performed individually on multiple samples once adequate counts are collected by the instrument. Localized Pu spectra are extracted from the instrument data file to perform isotopic analysis. The FRAM code was used for the Pu isotopic measurements and results were compared to the known values or mass spectrometry results. This document had been reviewed and assigned publication number: LA-UR-21-32277.

Log 442. IN-LINE NON DESTRUCTIVE ISOTOPICS MEASUREMENT FOR PLUTONIUM SAMPLES

Rim, J.H.(1,P); Tandon, L.(1); Winkler, R.(1). (1) Los Alamos National Laboratory. (P) Presenting Author.

A study was conducted to determine the accuracy of Pu isotopic abundances and Am-241 concentration measurements on large Pu metal samples in a glovebox environment using a High Purity Germanium (HPGe) Detector system. The sample masses were greater than 2 kg with a high background from the surroundings. The detector system is collimated and shielded using a lead/tungsten collimator to minimize background from the surroundings. A low energy gamma signal from Am-241 was filtered from the detector using a sheet of cadmium/copper to minimize detector dead time caused by samples. A typical count duration ranged from 30 to 45 minutes based on gamma signals measured from the sample. The collimator and filter setup was effective in decreasing unwanted low energy gamma Am-241 emission from reaching the detector crystal. However, it was also significantly cutting down 160 keV Pu-240 gamma emission signal and increasing the overall uncertainty of the Pu isotopic measurement. The non-destructive measurement results are compared to the destructive chemistry results to assess accuracy of the measurements. Additionally, a Monte Carlo simulation was performed to identify ideal counting geometry for the type of Pu samples analyzed by the detector system. This document had been reviewed and assigned publication number: LA-UR-21-32317

Log 443. **SEQUENTIAL SEPARATION AND MEASUREMENT OF RADIOCESIUM AND PLUTONIUM IN NORTH PACIFIC OCEAN SEAWATERS**

Kaizer, J. (1,P); Aoyama, M.(2,3); Bujdoš, M.(1); Pánik, J.(1); Povinec, P.P.(1); Sýkora, I.(1); Tateda, Y.(4); Terrassi, F.(5). (1) Comenius University. (2) Fukushima University. (3) University of Tsukuba. (4) Central Research Institute of Electric Power Industry. (5) 2nd University of Naples. (P) Presenting Author.

Long-lived anthropogenic radionuclides, and their distribution in the oceans have been studied since scientist had discovered their potential for better understanding of various oceanographic processes. The scientific interest in radiocesium and plutonium present in the marine environment has significantly risen in the past years as consequence of the Fukushima accident and is expected to be ongoing due to many unresolved questions. Hence, continuous development of new and verification of old analytical methods should be on the top of the list of the community, working on the topic. The aim of this study was to process and analyze several seawater samples, collected in the different time frames (2011-2015) from the North Pacific Ocean offshore Fukushima, in order to determine their radiocesium activity concentration, radiocesium activity ratio and plutonium atom ratio. For this purpose, we exploited a method based on the sequential scavenging of the investigated radionuclides and gamma spectrometry and accelerator mass spectrometry (AMS) for their determination . Some samples from the set, smaller in volume, were not processed but rather directly measured for radiocesium content by gamma spectrometry. Because of the complexity of the applied procedures, several problems emerged during the analyses, which we tried to address with the use of additional methods (e.g., measurements of uranium by ICPMS). Possible reasons for these issues and their solutions are discussed in the paper as well.

Log 445. **RADIOXENON SIGNATURES OF MOLTEN SALT REACTORS**

Mitchell, M. (1,P); Sobel, P. (1), Kazaroff, K. (1), Biegalski, S.R. (1) (1) Georgia Institute of Technology. (P) Presenting Author.

Developments of molten salt reactor technologies are making rapid progress across the globe. Many of these reactor designs are anticipated to release radionuclides through off-gas systems. Primary radionuclides anticipated to be released include isotopes of iodine, krypton, and xenon. This work examines the signature of the off-gas signal and then asseses detection technology to be utilized to monitor the emissions.

Log 446. **REDUCTION OF RADIOACTIVE INTERNAL CONTAMINATION AMONG THE MEDICAL PERSONNEL OF NUCLEAR MEDICINE FACILITES WITH THE USE OF RESPIRATORY TRACK PROTECTION MEASURES**

Brudecki, K. (1); Borkowska, E. (2); Gorzkiewicz, K. (1); Kostkiewicz, M. (2); Misiak, R. (1), Nalichowska, E.(1); Mróz, T. (3). (1) Institute of Nuclear Physics, Polish Academy of Sciences, Radzikowskiego 152, 31-342, Kraków, Poland. (2) Nuclear Medicine Department, John Paul II Hospital, Prądnicka 80, Kraków, Poland, (3) Institute of Physics, Jagiellonian University, Łojasiewicza 11, 30-348, Kraków, Poland National Laboratories. (3) Georgia Institute of Technology. (P) Presenting Author.

Internal radioactive contamination occurs when a radioactive substance enters the human body in an undesirable and unplanned way. In Poland, the largest professional group that may be affected by internal contaminations problem is the medical personnel of nuclear medicine facilities. Every year, they perform hundreds of thousands of diagnostic and treatment procedures, mainly with utilization of F-18 (positron emission tomography imaging), T-99mc (single photon emission tomography imaging) and I-131 (diagnosis and treatment of thyroid diseases). During only one procedure activities administered to patients may reach up to several GBq. Investigations conducted in period of 2015-2020 have confirmed the existence of the problem of internal radioactive contaminations among medical personnel working in nuclear medicine facilities. Conducted investigations allowed one to estimate internal radioactive contamination levels among medical personnel which reaches even several thousand of Bq in the case of Tc-99m and several hundred Bq in the case of I-131. What is more, studies have shown that inhalation is the main source of exposure. Nowadays, the basic radiation protection measures for medical personnel working with radiopharmaceuticals are lead coats and disposable gloves. Meanwhile, conducted research demonstrated that, utilization of upper respiratory tract protection systems may significantly reduce internal radioactive contamination (even 80%). Such a reduction will significantly improve both safety and working conditions of medical personnel.

Log 447. **A TABLETOP X-RAY TOMOGRAPHY INSTRUMENT FOR SPECTRAL IMAGING UTILIZING A TRANSITION EDGE SENSOR SPECTROMETER**

Nathan Nakamura (1,P), Bradley K. Alpert (1), Dan Becker (1), Douglas A. Bennett (1), Amber L. Dagel (2), Gabriella Dalton (2), W. Bertrand Doriese (1), Malcom Durkin (1,3), Joseph W. Fowler (1,3), Edward J. Garboczi (1), Johnathon D. Gard (1,3), James Zachariah Harris (2), Jozsef Imrek (1), Edward S. Jimenez (2), Kurt Larson (2), Zachary H. Levine (1), John A. B. Mates (1), Daniel McArthur (2), Kelsey M. Morgan (1,3), Galen C. O'Neil (1), Nathan J. Ortiz (1,3), Christine G. Pappas (1,3), Daniel R. Schmidt (1), Daniel S. Swetz (1), Paul Szypryt (1,3), Kyle R. Thompson (2), Joel N. Ullom (1), Leila Vale (1), Christopher Walker (2), Joel C. Weber (1,3), and Jason W. Wheeler (2). (1) National Institute of Standards and Technology. (2) Sandia National Laboratories. (3) University of Colorado, Boulder. (P) Presenting Author.

The ability to 3-dimensionally image and map the chemical state of nuclear materials can provide key information for nuclear safeguarding. To image with high spatial and spectral resolution, an instrument which provides both focused x-ray generation for tomographic imaging and broadband, high energy resolution x-ray detection is required. We have developed the x-ray tomography instrument TOMCAT (TOMographic Circuit Analysis Tool), which achieves nanometer-scale spatial resolution and precise, broadband x-ray detection via a transition edge sensor (TES) microcalorimeter spectrometer. TOMCAT utilizes the electron beam of a scanning electron microscope (SEM) to generate a highly focused x-ray spot from a metal target. The high energy resolution of the TES spectrometer then allows individual fluorescence lines from the target to be identified with high signal-to-noise and used for tomographic imaging. TOMCAT has recently demonstrated 3-dimensional image reconstructions with ~100 nm spatial resolution on an integrated circuit. While TOMCAT has been optimized for measurements on microelectronics, this new concept should be adaptable for imaging on nuclear materials and other samples. Furthermore, the ability to resolve individual fluorescence lines with high fidelity across a broad energy range using the TES spectrometer may enable spectral imaging with chemical state determination in the reconstructed volume. In addition to current TOMCAT performance, we will discuss paths towards high-speed spectral imaging involving multi-metal x-ray targets and next-generation TES arrays.

Log 448. **PRODUCTION AND CHARACTERIZATION OF NBL PLUTONIUM ISOTOPIC STANDARD CRM137A**

Parsons-Davis, T. (1, P); Henderson, R.(1); Holland, M.(2); Holliday, K.(1); King, C.(1); Mason, P.(2); Tourville, A.(2); Watters, R.(2); Williams, R.W.(1); Wimpenny, J.(1);   (1) Lawrence Livermore National Laboratory. (2) New Brunswick Laboratory Program Office. (P) Presenting Author

Lawrence Livermore National Laboratory (LLNL) and the NBL Program Office (NBL PO) are collaborating on production of purified sub-units of the aging plutonium isotopic reference materials formerly called NBS 936, 937 and 938, which will be distributed as certified reference materials (CRMs) CRM 136A, CRM 137A, and CRM 138A, respectively. Here we describe production and characterization of CRM 137A, the first of the series to be completed. An original unit of CRM 137 (formerly NBS 937) was purified in a new glovebox at LLNL via two-stage anion exchange chromatography to significantly reduce the quantities of U, Am, and Np in the source material. The purified plutonium was dispensed in nitric acid into individual units containing 1 mg Pu in 30 mL FEP bottles. Solution was gently evaporated to dryness and the units were packaged for distribution. The concentrations of Pu, U isotopes, 241Am, and 237Np in three analytical units were measured to provide informational values via isotope dilution (ID) multicollector–inductively coupled plasma–mass spectrometry (MC-ICP-MS). Results indicate the anion exchange procedure completely removed 241Am and U, while the Np concentration was reduced to 11.18 ± 0.15 ppm. High-precision measurements of the Pu isotopic composition have been made at LLNL, and units have been shipped to four other DOE laboratories for independent isotopic characterization. The NBL PO evaluation of isotopic data will be used to issue a certificate for CRM 137A as well as a new certificate for the CRM 137 source material. Prepared by LLNL under Contract DE-AC52-07NA27344. LLNL-ABS-830172

Log 449. **ADDITIVE MANUFACTURING OF SURROGATE DEBRIS REFERENCE MATERIALS AND MICROANALYTICAL STANDARDS**

Parsons-Davis, T. (1, P); Harris, S.L. (1); Sio, C.K.(2); Shusterman, J.(1); Lee, E.(1); Joshipura, I.D.(1); Goodell, J.J.(1); Shih, A.(1); Dory, C.(1); Henderson, R.(1); Ramon, C.(1); Lindvall, R.(1); Wimpenny, J.(1); Kuntz, J.D.(1); Pascall, A.J.(1); Bandong, B.B.(1). (1) Lawrence Livermore National Laboratory. (2) University of Toronto. (P) Presenting Author

Reproducible realistic surrogate debris reference materials (SDRMs) are needed to test and validate analysis techniques for identifying, quantifying, and interpreting major, minor, and trace elemental and isotopic constituents of radioactive debris from a nuclear detonation. Solid-phase SDRMs and other standards with precise elemental and isotopic compositions in desired proportions are needed to fully assess analytical timelines, accuracy, and robustness, and to evaluate the utility of in-situ microanalytical techniques. This talk presents novel methods to fabricate solid-phase SDRMS and microanalytical standards using additive manufacturing techniques with potential for 3-dimensional compositional and spatial control with μm-scale resolution. To form custom feedstocks, target analytes are doped into sub-micron sized Stöber-type silica particles. Particle suspensions are formed into three-dimensional cm-scale solid samples (tens of mg to several g) via electrophoretic deposition (EPD), then green-body deposits are calcined and sintered at high temperature to form densified glassy reference materials. In preparation for EPD-fabricated actinide and fission product-doped glasses (see related presentation by Shusterman et al.), we studied incorporation of over 50 trace elements into Stöber particles, their retention during thermal treatment, and their spatial distribution in feedstock particles and EPD-prepared samples. Preliminary data indicate a more homogenous distribution of many trace elements in EPD-prepared samples relative to sintered feedstock particles. These results will be discussed, along with approaches to multi-material deposition under development to enable silicate reference materials with customizable bulk and trace elemental compositions and desired spatial distribution.

Log 450. **DEVELOPMENT AND TESTING OF SCHOTTKY CONTACTS ON SiC USING A PNEUMATIC PRINTING SYSTEM.**

Safranek, A.W.(1)(P); Kymissis, I.(2); Cao, L.R.(1). (1) The Ohio State University. (2) Columbia University. (P) Presenting Author.

Silicon carbide (SiC) is an indirect wide-bandgap (WBG) semiconductor of high interest due to its applications in radiation-hard detection and monitoring for increased autonomy in nuclear reactor systems. Because of its high thermal conductivity, high breakdown electric field, high carrier saturation, high drift velocity, and large displacement energy, silicon carbide is also a suitable material for replacing conventional radiation detectors (Si, Ge, CdTe, CdZnTe) in specialized applications. In this study, a 4H-SiC Schottky device was fabricated and tested. Silver Schottky contacts were deposited onto a bare wafer in a square pattern of side-lengths between 1 mm and 4 mm using a pneumatic printing system. The silver contacts were then annealed at 150 °C. The SiC wafer has a 50 μm n-type epitaxial layer with net dopant concentration of $1.5 \cdot 10^{14}$ cm^(-3). Electrical characterization and x-ray response data were obtained to assess performance as a radiation detector, relative to the standards of today. Capacitance-voltage curves are used to estimate the net doping concentration in the device, and current-voltage to measure reverse-bias leakage current. X-ray response data is used to estimate carrier lifetime. The dependence of the current response to incident x-ray intensity and energy is measured and fitted to a model. Measured properties support the band theory for the metal-semiconductor junction. Characterization data will be compared between different contact sizes and metals such as copper, and with an ZTO oxide layer. The performance of this device is compared to traditional silicon detectors in addition to Schottky devices fabricated by metal deposition to determine the suitability of this fabrication method.

Log 451. UNDERSTANDING TRACE METAL TAGGANT DISTRIBUTION IN OXIDE AND METALLIC URANIUM MATRICES

Said, M. (1, P); Marks, N.E.(1); Garza, E.V.(1); Ramon, C.E.(1). (1) Lawrence Livermore National Laboratory. (P) Presenting Author.

The purpose of intentional nuclear forensics is to facilitate rapid provenance assessment of nuclear materials through the deliberate incorporation of distinctive taggant materials. The most promising candidates include multi-isotopic taggants with perturbed isotopic distribution that could be easily incorporated and detected in a uranium matrix. Target materials include nuclear materials that are sometimes associated with theft and interdiction, including UO2 fuel pellets and uranium metal. In support of Intentional Forensics R&D, LANL has produced a set of depleted uranium metal samples that are tagged with varying amounts of transition metal elemental taggants. We are also preparing a set of UO2 materials tagged with isotopically perturbed taggants using an established synthetic sol-gel process to test incorporation and forensic recovery of trace levels of isotopically perturbed Mo, W, and Ni. Our initial work at LLNL has focused on developing effective techniques for these samples to 1) understand the spatial distribution and diffusion profiles of tagged materials using microbeam techniques; 2) understand the effects of specific taggant candidates on grain size and other basic properties of tagged material; and 3) evaluate rapid recovery strategies for detecting taggants in materials under investigation. Results of these studies will inform future taggant production and incorporation strategies within the intentional forensics venture.

Log 452. QUALITATIVE ASSESSMENT OF URANIUM ORE CONCENTRATES AND RELATED MATERIALS USING SCANNING ELECTRON MICROSCOPY

Said, M. (1, P); Marks, N.E.(1); Dai, Z.(1); Roberts, S.(1); Sharp, M.(1); Lindvall, R.E.(1). (1) Lawrence Livermore National Laboratory. (P) Presenting Author.

Lawrence Livermore National Laboratory maintains a database of uranium ore concentrate (UOC) analytical signatures as a tool for provenance assessment and nuclear forensic signature research. It includes data of thousands of samples from facilities in more than 30 countries as well as an archive of several hundred physical samples. Each sample has been carefully analyzed for uranium assay and isotopes, trace element concentration, chemical phase (XRD), as well as several stable and radiogenic isotopes. Scanning electron microscope (SEM) imaging of these powdered samples has been a useful approach for researching morphological signatures of UOC materials. In recent years, great strides have been made to understand the morphology of nuclear material in relation to process history through both qualitative and quantitative efforts. However, these studies are mostly constrained to uranium oxide materials produced under tightly controlled conditions at the bench scale, while UOC materials produced at the plant scale tend to have a more complex morphological range. This work aims to provide qualitative morphological assessments for a suite of real-world uranium ore concentrates using the previously published lexicon of Tamasi et al., (2016). This work will inform UOC morphological characterization of commercially produced materials and will form the basis for future machine learning efforts for UOC materials.

Log 453. **LEVEL AND DISTRIBUTION OF Pu ISOTOPES AND U-236 IN LAGOON AND OFFSHORE WATERS OF RUNIT ISLAND ENEWETAK ATOLL**

Shaw, A.C.(1,P); Hamilton, T.F.(1); Martinelli, R.E.(1); Ahrens, N.L.(1,2); Tumey, S.J.(1); Brown, T.(1). (1) Lawrence Livermore National Laboratory. (2) Colorado State University. (P) Presenting Author.

Enewetak Atoll is a former U.S. atmospheric nuclear weapons test site, active from 1948 to 1958, and located in the Republic of the Marshall Islands. Local fallout deposition resulting from 43 nuclear detonations on and around the atoll led to widespread radioactive contamination of islands and the local marine environment. TRU contamination primarily from Pu isotopes and Am 241 entering the marine environment by direct deposition or from physical disturbance of island soils during test site preparations formed a reservoir and continuous source-term for remobilization and redistribution of Pu 239 and 240, in addition to Am 241 into the water column. The contemporary analyses shown here confirm that persistently higher levels of about 100 to 150 fold for Pu 239 and 240 remain in surface waters of Enewetak Lagoon relative to open ocean surface waters. We also report results from a unique study showing the distribution of Pu contained in a deep water ocean profile collected off Runit Island.

Log 454. **SIMULTANEOUS MEASUREMENT OF Sr, Zr, AND Mo ISOTOPES IN SiC STARDUST GRAINS USING RIMS**

Shulaker, D.Z. (1,P); Ong, W.J.(1); Trappitsch, R.(2); Savina, M.R.(1). (1) Nuclear and Chemical Sciences Division, Lawrence Livermore National Laboratory. (2) Department of Physics, Brandeis University. (P) Presenting Author.

Micrometer-sized stardust particles (also known as presolar grains), which can be found in meteorites, record the isotopic composition of nucleosynthesis processes that occurred under different stellar conditions. Because isotopic systems in unadulterated stardust represent ground truth values as inputs for nucleosynthesis models, studying isotopic systems in stardust are important for understanding the formation of p- and s-process nuclei. However, stardust is difficult to analyze since each grain is micrometer-sized and its trace elements are thus atom limited. We present data from over 50 SiC presolar grains using newly developed resonance ionization mass spectrometry (RIMS) methods for collecting 16 isotopes of 3 elements (Sr, Zr, Mo) simultaneously on the LION instrument at LLNL. After removing atoms from the sample surface with a desorption laser, we first pulse lasers tuned to resonance ionization wavelengths that are specific for ionizing Sr and Mo neutral atoms. 200 ns after ionizing Sr and Mo, Zr ionization lasers are pulsed and ionize neutral Zr atoms in the same cloud of neutral atoms. Because Zr is accelerated through the time-of-flight mass spectrometer 200 ns after Sr and Mo, Zr and Mo isobaric interferences can be discriminated. Ultimately, this method allows for elemental analysis, regardless of isobaric interferences, of micron-sized particles for isotopes without performing chemical separation. This method is applicable to analyzing nuclear material with application to nuclear forensics and safeguards in future. This work performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344. LLNL-ABS-830294.

Log 455. **ELECTROCHEMICAL ENRICHMENT OF ULTRATRACE ELEMENTS FOR NEUTRON ACTIVATION ANALYSIS**

Stary, T. (1,P); Welch, J.M. (1); Sterba, J.H. (1); Mokina, V. (2); Kirchweger, V. (1). (1) Center for Labelling and Isotope Production, TRIGA Center Atominstitut, TU Wien, Vienna, Austria. (2) HEPHY - Institute of High Energy Physics, Vienna. (P) Presenting Author.

Determination of very-low thorium concentrations (activity) in high-purity copper shielding has become pivotal for the CRESST (Cryogenic Rare Event Search with Superconductive Thermometers) experiment, since precise quantification of thorium activity is essential for accurate simulation of background events masking the rare, sought-after, dark matter events. To date, it has only been possible to verify that the amount of Th-232 in said copper is below the detection limits of the methods applied. Therefore, we will report our efforts to develop a method to efficiently quantify ultratrace amounts of thorium in copper by electrochemical enrichment and subsequent neutron activation analysis (NAA). Initial experiments inspired by industrial production (electrorefining) of high-purity copper showed that rather than being enriched in the electrolyte (nitric acid), as might be expected, thorium was readily (and remarkably selectively) enriched on the copper cathode. To take advantage of this effect, we have systematically varied the electrolysis duration and temperature, operating current and voltage, electrode materials and specific surface area and thorium concentration in the electrolyte. We have found that due to different deposition rates of copper and thorium, the latter may be significantly enriched relative to the former on a graphite cathode, which may, in turn be directly analyzed for thorium content by NAA. This method should allow detection of ultratrace amounts of thorium in copper and deliver the desired data for the CRESST experiment.

Log 456. **LASER INDUCED FLUORESCENCE AND IMAGE ANALYSIS FOR MONITORING ENVIRONMENTAL CONTAMINATION IN BIOTA**

Truax, K. (1,P); Dulai, H. (1); Misra, A. (1); Kuhne, W. (2); and Fulkey, P. (1). (1) University of Hawaii at Manoa. (2) Savannah River National Laboratory. (P) Presenting Author.

The ability to detect and characterize the spatial distribution of contaminants and radionuclides related to nuclear forensics is of ongoing interest. A widespread tool for monitoring comes in the form of vegetation that bioaccumulates compounds such as metals and can serve as indicators of their recent and historic releases. Large scale sampling of biota, however, can be time- and labor-intensive, leaving room for a non-invasive in-situ technique for monitoring. Laser induced fluorescence (LIF) is quickly gaining ground as an effective way to excite organic molecules, such as chlorophyll, and measure the wavelengths of short lifetime fluorescence to observe the health of plants and algae. The technique presented here utilizes LIF images collected of moss to identify the presence of metals at the nmol/cm2 levels, specifically of Cu, Zn, Pb, and their mixture. The effect of environmental factors, such as length of photoperiod, drought, and nutrient levels is also explored and seems to result in minimal interference with LIF image changes produced by metal contamination. While the method has been successful in differentiating and identifying individual metals at environmentally relevant levels in the laboratory, continuing work seeks to make the technique chlorophyll specific and applicable in any light condition for a multitude of vegetation types, as well as field portable.

Log 457. IMPROVING HIGH THROUGHPUT GAMMA SPECTROMETRY ANALYSIS BY IDENTIFYING PROBLEMATIC SPECTRA USING MACHINE LEARNING

Truax, K. (1,P); Dulai, H. (1); Garces, M. (1); Bowyer, T. (2); Friese, J. (2); and Metz, L. (2). (1) University of Hawaii at Manoa. (2) Pacific Northwest National Laboratory. (P) Presenting Author.

Gamma-ray spectrometry has long been used to identify radionuclides and monitor their activity in environmental, industrial, and nuclear non-proliferation surveillance. The application of high-purity germanium detectors is now widespread thanks to their energy resolution which allows for identification of radionuclides in a complex matrix. Multiple spectrum analysis software packages have also advanced resulting in excellent performance. However, long-term monitoring efforts with thousands of spectra produced each year introduce challenges to high-throughput analysis. The first of these comes from the radionuclide source itself where changing radionuclide content and variable activities may present a problem in identifying peaks of interest. Peak fitting on top of variable backgrounds and Compton edges combined with physical environment and measurement condition changes also introduce variability that affect the quality of spectra and hence the reproducibility of their evaluation. We present here on efforts employing machine learning techniques to identify problematic spectra where automated spectrum analyses fail. The spectra consist of two components: the signal coming from the radionuclide source and the noise resulting from the physical environmental and detector health parameters. First steps identify problematic spectra and their relationship with environmental and detector specific parameters. Second, we evaluate how changes in the source itself affect spectra and peak fitting, for example over high/variable backgrounds and Compton Edges. Currently, analysis is conducted with previously documented spectra, but future goals including adapting methods for real-time quality control.

Log 459. **RAPID ASSESSMENT OF URANIUM ACTIVITY AND MASS FOR NUCLEAR FORENSICS**

Valdovinos, H.F. (1,P); Glennon, K.J.(1); Parsons-Davis, T.(1); Shusterman, J.A.(1); Gharibyan, N.(1). (1) Lawrence Livermore National Laboratory. (P) Presenting Author.

Rapid assessment of short-lived uranium isotope activity and mass in the event of an unplanned nuclear explosion can assist in the subsequent forensic analysis. A deployable instrument for measurements in the field is one approach to provide rapid analyses. This work presents a flow chemistry system programed in LabVIEW that integrates three functionalities: 1) selective extraction of U from fission products (FP) via a supported liquid membrane module; 2) UV-Visible absorbance spectrophotometry to measure the total U concentration in the strip flow; and 3) U-237 activity concentration in the strip solution using a portable CdTe gamma spectrometer coupled to a 3D-printed flow cell. The system was validated using standard U/HEU solutions and its functionality was demonstrated using a solution of depleted-U spiked with U-237 and mixed-FP. U concentrations of 100-3,000 ppm were directly quantified inline using a 100 mm optical flow cell and U concentrations of 0.1-10 ppm were quantified after inline mixing of the strip solution with a flow of colorimetric reagent. The mass concentration measurement takes approximately half an hour while gamma spectrometry varies depending on activity of the sample, with 1 kBq/mL of U-237 requiring about an hour of acquisition time. This platform provides a potentially fieldable approach for quantifying uranium isotope activity and masses associated with post-nuclear explosion materials. Auspices: U.S. DOE Contract DE-AC52-07NA27344; LLNL's LDRD project 19-ERD-016; U.S. DOE's NNSA Office of DNN R and D.

Log 460. **RADIOLABELING OF ELASTOMERS**

Sahovic, N. (1); Welch, J.M. (1P); Sterba, J.H. (1). (1) Center for Labelling and Isotope Production, TRIGA Center Atominstitut, TU Wien, Vienna, Austria. (P) Presenting Author.

The breakdown and wear of organic elastomers under mechanical stress or in the environment are complex and poorly understood processes. Radiolabeling of such materials should offer significant insight into these processes. However, most commercial elastomers are sophisticated, heterogeneous, multi-component and often proprietary mixtures. Therefore, radiolabeling of the individual components of such mixtures followed by effective curing to a finished polymer with properties comparable to commercial analogs would be intensely laborious and prone to many physical, chemical and technical difficulties. A less rigorous, but potentially simpler, approach is to label commercially available materials by chemical incorporation of a radiotracer (doping). If the amounts of inactive carrier are minimized, it should be possible to dope elastomeric materials with isotopes of sufficient half-life to allow for radiometric studies of polymer degradation without affecting their initial, macroscopic physical properties. To this end, we have conducted initial investigations into doping elastomeric materials with ruthenium by gas-phase exposure to in situ generated ruthenium tetroxide. Ruthenium combines multiple radioisotopes (Ru-97, Ru-103 and Ru-105) accessible by thermal neutron activation with a volatile oxide known for reactivity toward organic materials. Doping of several elastomeric substances will be detailed and initial investigations into the properties of these samples will be reported.

Log 461. **STRONTIUM AND CESIUM ABSORPTION CAPACITY OF NATURAL ZEOLITIC MATERIALS**
Grill, V. (1); Welch, J.M. (2,P); Foster, M. (2); Sterba, J.H. (2); Streli, C. (3). (1) Radiation Protection and Radiochemistry, AGES, Vienna, Austria. (2) Center for Labelling and Isotope Production, TRIGA Center Atominstitut, TU Wien, Vienna, Austria. (3) Atominstitut, TU Wien, Vienna, Austria. (P) Presenting Author.

The properties of natural zeolites combined with their relatively low cost lead to application in diverse areas ranging from agriculture to acoustic technologies and beyond. Specifically, the ion-exchange and adsorptive properties of clinoptilolite-rich materials are potentially useful for bulk decontamination of large amounts of aqueous low-level radioactive wastes. The principal long-lived contaminants in older low-level waste from nuclear power applications are radio-cesium (predominantly Cs-137) and radio-strontium (Sr-90). Therefore, the (saturation) capacity of clinoptilolite-rich zeolite products (LithoFill and LithoGran) were determined by contact with various concentrations of aqueous strontium (0.05 – 0.5 M) and cesium (0.1 – 1.0 M) under rigorously controlled conditions using Cs-134 and Sr-85 as gamma-emitting indicators. These studies suggest strontium saturation capacities ranging from 30 – 80 mg/g and cesium capacities from 140 – 170 mg/g for the materials studied. The results will be presented in the context of material properties and application potential.

Log 462. **ANALYSIS OF ELEMENTAL CONTAMINANTS AND IMPURITIES IN PLUTONIUM METAL USING MICRO-X-RAY FLUORESCENCE**
Worley, C.G. (1,P); McIntosh, K.G. (1). (1) Los Alamos National Laboratory. (P) Presenting Author.

Analysis of elemental impurities in plutonium metal is important to ensure the metal will have desired material properties. Unexpected issues occurred during processing and handling two plutonium metal batches. Elemental impurity spatial distributions were examined in the two plutonium metals using micro-X-ray fluorescence (MXRF). Optical microscopy was also used to study microscopic features that correlated with areas of interest seen in the MXRF elemental maps. Areas on the samples containing a locally high concentration of zinc and copper were detected with MXRF, but the plutonium was not expected to contain these elements. The surface of the samples were determined to be contaminated with brass particles because zinc and copper were co-located in these areas, the copper/zinc peak intensity ratios correlated with brass, and shiny deposits were seen with the microscope in the same Zn and Cu-containing locations. After discussions with the sample submitter, it was determined that the samples were inadvertently contaminated with brass particles while processing the metal batches. Other features were seen on the samples using optical microscopy, and MXRF indicated they contained one or more of the following elements: Fe, Cr, Ni, Ca, Ti, Zn, Ce, and W. These results demonstrated how MXRF was important in troubleshooting unexpected problems that occurred during plutonium processing.

Log 463. METHODS OF UTILIZING RADIOXENON STATIONS AND LABORATORIES FOR NUCLEAR EXPLOSION MONITORING

Michael Foxe (P), Michael Mayer, Jennifer Mendez, Johnathan Slack, Theodore Bowyer, Matt Cooper, Ian Cameron, James Hayes, Rose Perea. (P) Presenting Author.

Radioxenon laboratories are an integral part of the International Monitoring System (IMS). As radioxenon samples are collected around the world at the IMS stations, a subset of those samples are sent to radionuclide laboratories around the world for re-analysis. PNNL operates the U.S. Noble Gas Laboratory (US-NGL), which was certified in December of 2016. There is also an opportunity to utilize the radioxenon laboratories in a field system comparison experiment. In this presentation, we detail current and potential future methods of utilizing the radioxenon laboratories. For a laboratory system to be used for verification of continuously operating systems, it is important to routinely validate the energy and efficiency calibration of the laboratory system. We discuss methods of further leveraging radioxenon stations and laboratories in both a verification and collaborative nature for nuclear explosion monitoring. Additionally, we present the recent efforts to increase the throughput and discuss the impact oof throughput on utilization methods.

Log 464. IMPACT OF A SILICON BETA CELL ON RADIOXENON SOURCE IDENTIFICATION

Michael Foxe (P), Michael Mayer, Johnathan Slack, Eric Becker, Alex Couture, Thomas Hallen, Mike Ripplinger, James Hayes. (P) Presenting Author.

Beta-gamma detectors are utilized extensively for the detection of radioxenon, but the beta detection is primarily performed with a plastic scintillator cell. Two areas of improvement for plastic scintillators are the sample carry-over ("memory effect") and energy resolution. While the scintillator can be coated to remove the memory effect, the energy resolution must be improved with a different detector material. Silicon is the current leading candidate for the future beta cell material due to the much-improved energy resolution compared to plastic scintillators (factor of ~3x). PNNL is developing a silicon beta cell for use as a potential modular replacement within Xenon International (a next-generation radioxenon detection system currently undergoing acceptance testing for potential inclusion in the International Monitoring System). With improved resolution, the radioxenon monitor should more easily discriminate the isotopes or radioxenon and thus the source of radioxenon. We will discuss the development of the silicon beta cell and the potential impact of a silicon beta cell in improving the radioxenon background measurements and source identification.

Log 465. **ANALYSIS UPDATES UTILIZED FOR A SILICON BETA CELL FOR RADIOXENON DETECTION**
Johnathan Slack (P), Michael Foxe, Michael Mayer. (P) Presenting Author.

Beta-gamma detector systems for radioxenon measurements often use a scintillating plastic gas cell for the beta portion of the detector. Silicon can be substituted in for the plastic for an improvement in the conversion electron peak energy resolutions at the expense of an increase in sensitivity to sources of noise. In this work, a beta cell constructed of four monolithic silicon chips arranged in an aluminum, gas-tight, cylinder was tested. Multiple improvements were made to overall performance through a combination of procedural changes and additional analysis/calibration techniques. A backscatter analysis was performed for this configuration to account for coincidence events between the silicon chips and determine the interference between isotope regions of interest. Using a dual isotope quality control source of Ba-133 with Cs-137 provided additional performance metrics for gain and stability monitoring. To improve activity and concentration analysis a new calibration routine, the Metastable Interference Terms (MITS) algorithm, was also applied. We will discuss the impact of electron backscatter on the radioxenon detector sensitivity and how the signals from a silicon beta cell can be used to optimize the isotopic discrimination.

Log 466. **SOURCE CHARACTERIZATION OF DD NEUTRON GENERATOR**
Mukhopadhyay, S. (1, P); Nelson, W. (1); Irvin, V. (1); Hertel, N.E. (1). (1) Georgia Institute of Technology. (P) Presenting Author.

An n-Gen DD neutron generator is used for various experiments ranging from irradiating cell cultures to approximate the LET spectrum in space radiation environments, testing different neutron sensitive detectors, and for neutron activation analysis just to name a few of its uses at the Georgia Tech Radiological Science and Engineering Laboratory. The source strength characterization of this neutron source is currently under investigation. We are using different direct and indirect measurements to fully understand the generator performance and the associated neutron field. Absorbed dose and dose equivalent has been measured in the neutron field of the generator with REM500 Neutron Survey Meter which uses a tissue equivalent (TE) proportional counter. Absorbed dose rates have also been measured through the application of the Bragg-Gray principle and a tissue-equivalent (TE) ion chamber (IC). Neutron activation of both gold and indium foil has enabled us to determine the source strength in terms of neutrons emitted per second. The neutron generator has been modelled using Monte Carlo simulations to estimate the activation levels in these foils for 1 neutron per second. Comparison of the measured activity levels with those of the unit source activity levels, yield a first-order approximation to the absolute source strength of the DD neutron generator.

Log 467. **Radioxenon detections during the SPALAX-NG CTBTO qualification**

Topin, S. (1, P); Achim, P. (1); Gross, P.(1); Generoso, S.(1); Douysset, G.(1); Delaune, O.(1);Philippe, T.(1); Ungar, K.(2); Bean, M.(2); Hoffman, I.(2). (1) CEA/DAM Ile de France (2) Health Canada. (P) Presenting Author.

The SPALAX NG has recently been qualified for deployment in the CTBTO monitoring network. This system has been operated for several years by CEA/DAM. The SPALAX NG is a major evolution compared to today's SPALAX with detection limits of 0.2 mBq.scm in 8 hours for all the relevant radioisotope of xenon. This presentation provides the feedback on the use of the system in an operational context and the study of several relevant detections observed during the qualification periods in France and Canada.

Log 468. **IDENTIFICATION, ISOLATION, AND ANALYSIS OF FUKUSHIMA-DERIVED MICROPARTICLES**

Macsik, Z. (1); Hudston, L.A. (1); Wurth, K.N. (1); Meininger, D. (1); Jesinghaus, C. (1,2), Tenner, T.J. (1); Naes B.E. (1); Shozugawa, K. (3); Steiner, R.E. (1); Steinhauser, G. (1,2,P). (1) Los Alamos National Laboratory. (2) Leibniz University Hannover. (3) The University of Tokyo. (P) Presenting Author.

In the course of the Fukushima nuclear accident major amounts of radionuclides were released in volatile form to the atmosphere and dissolved form to the Pacific Ocean. However, minor amounts of particulates were released in the form of cesium-rich microparticles. In this study, we used autoradiography to identify unusually large microparticples (diameter approx. 20 um) on blades of grass that were collected in the vicinity of Fukushima Daiichi NPP soon after the accident (2011). SEM revealed a spherical shape of the particles and EDX the basic composition. Low-level gamma spectrometry indicated detectable but comparably low activities of Cs-137. This may be the first observation of this type of particles.

Log 469. DETERMINATION OF DETECTION EFFICIENCY FOR VOLUMINOUS GAMMA-RAY SOURCES AT COLLIMATION GEOMETRY

Kang, M.Y. (1,P); Sun, G.M. (2); Choi, H.D. (3). (1) Korea Atomic Energy Research Institute, Radiation Safety Management Division. (2) Korea Atomic Energy Research Institute, HANARO Utilization Division. (3) Seoul National University. (P) Presenting Author.

For the various types of gamma-ray sources considering the geometrical effect and attenuation, determination of the full energy absorption peak efficiency (detection efficiency) is one of the important processes of the gamma-ray activation analysis. In this study, we extended and verified the performance of the EXVol(Efficiency calculator for eXtended Voluminous source) code, which is a detection efficiency calculation code using the effective solid angle method. The previous EXVol only determined the detection efficiency of the source-detector coaxial structure for the whole volume source. After expansion of the performance, EXVol can calculate both coaxial and asymmetric structure. In addition, the introduction of a collimator made it possible to reduce the radiation intensity of a high radiation source. And it is possible to determine the precise detection efficiency according to the energy of a gamma ray at a specific position of the volume source. To verify the performance of the EXVol, a high resolution gamma spectroscopy system was constructed and measurement and analysis were performed. Measurements were performed on coaxial, asymmetric and collimated structures with standard point source, standard 1 L liquid volume source and HPGe detector. The measured results were compared with the calculation results of EXVol. The relative deviation of the measurement and calculation in the coaxial and asymmetric structures was 10%, and that of the collimation structure was 20%. Results can be available in analysis of environmental radiation samples, the applied research field using gamma-ray measurement and non-destructive assay and radioactivity determination of high-level radiation and large volume structures.

Log 470. **THERMOPHYSICAL PROPERTIES OF BINARY CL COMPOSITIONS FOR NEXT GENERATION MOLTEN SALT REACTORS**

Lonergan, J. (1,P);Swinhart, M. (3);Sudowe, R. (3); Guo, X. (2); Clark, R. (1);McNamara, B. (1); Paviet, P. (1); (1) Pacific Northwest National Laboratory. (2) Washington State University. (3) Colorado State University. (P) Presenting Author.

Molten salt reactors (MSRs) have received growing interest due to a desire for high temperature reactors that are not constrained by the limitations of solid fuels and conventional coolants. The proposed salt systems found in the literature vary widely and basic properties are missing. In particular, thermophysical properties that are critical for design of an efficient and reliable heat exchange fluid are not well documented or understood. This study has focused on the binary composition KCl-NaCl, to systematically study the phase diagram from pure KCl to NaCl, as a baseline to understand the effects of composition on heat capacity, melting temperature, and enthalpy. Heat capacity was measured from 25 to 900oC for all five compositions. A 17% change in heat capacity was measured for pure KCl (0.964 J/g*K) to pure NaCl (1.162 J/g*K) at 900oC. For all salts, heat capacity increased until melting, at which point the measured values exhibited a small decrease in heat capacity with increasing temperature. Numerous challenges were overcome to test the molten salts safely and accurately due to their volatile and corrosive nature. Custom nickel, aluminum, and boron nitride crucibles were designed and fabricated. Metal crucibles were hermetically sealed utilizing laser welding to test properties safely and accurately through differential scanning calorimetry and high temperature drop calorimetry. This talk will detail the development process, characterization techniques, and the effects of composition on the thermal properties of the binary molten salt systems.

Log 471. **EVALUATING THE QQQ-ICP-MS FOR UNDERSTANDING ACTINIDE REACTIVITY**

French, A.D (1) Melby, K. (1) Arnquist, I.J (1) Eiden, G (1) Beck, C (1) Seiner, B (1) Harouaka, K. (1) Pacific Northwest National Laboratory. (P) Presenting Author.

The inline separation capability of tandem mass spectrometry (MS/MS) uses two mass filters and a collision/reaction cell to separate interference ions from the analyte(s) of interest through gas-phase ion-molecule interactions. Traditionally, separation of the analyte from matrix interferants is achieved off-line through complex and time-consuming chemical separations, typically via ion exchange column chemistry. For actinide analyses in particular, decreasing sample preparation time is necessary to reduce human exposure to hazardous samples and reduce the production of waste. The aim is to employ QQQICP-MS to explore actinide reactivity that may prove useful for sample analysis and for which there is limited literature available using the latest technology. In this talk, we present preliminary data showing the reactivity of actinides with several reaction/collision gases using the Agilent 8900 QQQ-ICP-MS. These investigations allow for the evaluation of the gas phase interactions and reactivity of the actinides, providing insight into their fundamental chemistries. Reaction gases are identified that may be leveraged for isobaric and polyatomic interference removal in samples that are relevant to nuclear forensics and geochemical dating.

Log 472. AN IN-POOL MOLTEN SALT TESTING FACILITY AT THE PULSTAR REACTOR

Hawari, A.I. (1,P); Liu, M.(1); Lassell, S.A.(1). (1) North Carolina State University. (P) Presenting Author.

An in-pool facility for molten salt irradiation experiments (MSIE) is being established at the 1-MWth PULSTAR reactor of North Carolina State University. MSIE will allow temperature controlled irradiations and will be located in a vertical beam tube in the pool of the PULSTAR. This enables the irradiation of fueled/unfueled samples of molten salt (e.g., FLiBe) with a volume reaching a few cubic centimeters. Helium gas will flow through the samples to sweep potential fission products and impurities that may result from the irradiation process. Two types of spectroscopy techniques are being investigated to understand the production and release of such products from irradiated salts. This includes in-situ laser induced breakdown spectroscopy (LIBS) to be performed near the flow exit, and gamma-ray spectrometry (GRS), which will performed upstream. In this case, LIBS should be beneficial in elemental assay, and GRS will further assay particular isotopes. Using this approach, it is anticipated that release-to-birth ratios of the produced elements will be established, which may be utilized in the validation of computational simulations that seek to estimate elemental and impurity release from molten salts. At this stage, MSIE design, using the COMSOL multi-physics platform and the MCNP6 neutronics code, has been performed establishing operational properties and illustrating the detectability of various released radionuclides.

Log 473. PRODUCTION OF XENON-135 FROM ISOTOPICALLY ENRICHED XENON-134 AND XENON-136 TARGETS

Horkley, J.J (P)(1); Brookhart, J.L.(1); Cardenas, E.S.(1); Carney, K.P (1); Hines, C.(2); Houghton, T.P(1); Robinson, T. A.(1); Watrous, M. G.(1). (1) Idaho National Laboratory. (2) Washington State University. (P) Presenting Author.

Atmospheric radioxenon analyses conducted under the Comprehensive Test Ban Treaty Organization (CTBTO) necessitates the collection and analysis of air samples to quantify 135Xe, an indicator for nuclear explosions and reactor operations. Analytical instrument calibration for these analyses requires high quality, monoisotopic reference standards that are typically short lived, and therefore must be produced and shipped promptly to monitoring stations worldwide. This work describes the preparation of 135Xe by neutron capture (n, gamma) on monoisotopic 134Xe and high energy photon irradiation (gamma, n) on monoisotopic 136Xe. The targets are prepared by electromagnetic isotope separation of natural xenon. Results from each process are compared for 135Xe yield and purity.

Log 474. ANALYSIS OF A MAXIMUM HYPOTHETICAL ACCIDENT FOR A MOLTEN SALT RESEARCH REACTOR

Bekker, JP (1)(P); Scherr, J.B. (2); Haas, D.A. (1). The Unviersity of Texas at Austin (2) Abilene Christian University. (P) Presenting Author.

The Nuclear Energy eXperimental Testing Research Alliance (NEXTRA) consists of four universities (Abilene Christian University, The University of Texas at Austin, Texas A&M University, and Georgia Tech) and is funded by Natura Resources, LLC to design, license, and build a Molten Salt Research Reactor (MSRR) on the Abilene Christian University campus. As part of the licensing process, it is critical to understand the consequence of a bounding accident scenario known as a Maximum Hypothetical Accident (MHA). Simulations of dose consequence start from abundantly conservative (worst possible case) instantaneous release of all noble gases and halogens from the reactor loop and off-gas system straight into the atmosphere, before applying more realistic assumptions such as leakage rates from the reactor enclosure or plate-out of certain nuclides onto the metal surfaces in the reactor loop. Results from these analyses will be used to influence system design and set operation requirements for the MSRR. Data from experiments at the new molten salt loop at MIT's reactor and operation of the MSRR at low power will be used to refine MHA radionuclide release values in support of higher power operations of the MSRR.

Log 475. SOL-GEL PARTICLE SYNTHESIS AS A BOTTOM-UP METHOD FOR PRODUCTION OF SURROGATE NUCLEAR EXPLOSIVE DEBRIS

Justin T. Cooper (1,P), George L. Diehl(2), Robert Lusk(1), Kevin Carney(1), Thomas V. Holschuh(1), David Chichester(1), Mathew S. Snow(1). (1) Idaho National Laboratory. (2) University of Utah. (P) Presenting Author.

Development and production of surrogate nuclear explosive debris is of interest to the nuclear nonproliferation community for training first responders and validating nuclear forensic techniques. Surrogates should have a size, color, elemental composition, and radionuclide content that resembles what first responders might encounter in the field following a nuclear detonation. However, the ability to tailor each of these parameters accurately is difficult due to the wide range of fallout types and the various types of detonation scenarios that could occur. Nuclear fallout can contain materials from the surrounding environment; thus, fallout elemental composition can vary widely. Validation of laboratory-level nuclear forensics techniques require surrogates that can be tailored to represent the wide range of fallout types and detonation scenarios. Sol-gel techniques have shown to be suitable to meet these challenges. The sol-gel process consists of forming a metal oxide material, often silica, through polymerization of a metal-alkoxy precursor. This bottom-up synthesis approach yields a large degree of tunability of both the size, shape and composition of the final material. In this work, we demonstrate the ability to produce particles with controllable size, shape and color. We also use scanning electron microscopy with energy dispersive x-ray spectroscopy data to characterize the ability to load the sol-gel surrogate debris particles with secondary elemental components such as iron, aluminum, and calcium, toward approximating the elemental composition of debris from various detonation environments. Finally, we demonstrate the ability to quantitatively encapsulate specific radionuclide content within the sol-gel particles, confirmed via gamma-ray spectrometry.

Log 477. **PARTICLE THERAPY INTERNATIONAL MASTERCLASS: THE FIRST ITALIAN EXPERIENCE**

Groppi, F. (1,2,P); Capua M. (3,4); Manenti, S. (1,2); Tucci, R. (4,5); Cagnetta, MF (2,6); Colucci, M. (1,2); (1) LASA Lab., Physics Dept. of Milano University. (2) INFN-MI, Italy. (3) Physics Dept. of Calabria University, Italy. (4) INFN-CS, Italy. (5) Liceo Scientifico E. Fermi, Cosenza, Italy. (6) Liceo Scientifico Donatelli-Pascal, Milano , Italy. (P) Presenting Author.

For the first time, Italy participated in the International Particle Therapy Masterclass, a training event promoted by the International Particle Physics Outreach Group, with the purpose to bring students of the last year of High School closer to physics applied to medicine, showing them the importance of basic research and its impact on society, in particular, on human health. The event was organized as follow. The morning was dedicated to seminaries devoted to explanation of artificial radionuclides production for theranostics application in Nuclear Medicine; to the radiotherapy carried out with different external beams, with an in-depth discussion and debate about the strengths and weaknesses that each technique presents; to a virtual visit to the Italian National Center for Oncological Adrotherapy in Pavia (CNAO). In the afternoon, the students participated to a hand-on session with the use of MatRad s/w, developed and used by German Cancer Research Center DKFZ in Heidelberg. The students realized a real treatment plan to treat tumors of specific organs with different particle beams (photons, protons, carbon ions). Finally, they presented their results in an international virtual meeting organized at CERN with the students of the other participant institutes around the world and in the presence of experts. In this contribution, this rich and stimulating experience which made "researchers" for one day twenty-two students selected from different schools in Lombardy and Calabria Italian regions, will be presented.

Log 478. **DEVELOPMENT OF GALLIUM OXIDE BASED ULTRAWIDE BANDGAP SEMICONDUCTOR DETECTOR FOR ALPHA SPECTROSCOPY**

Heckert, C.J. (1,P); Taylor N.R.(1,2); Dhara, S. (1); Rajan, S.(1); Cao, L.R. (1); Blue, T. E. (1). (1) The Ohio State University. (2) Oak Ridge National Laboratory. (P) Presenting Author.

A set of semiconductor radiation detectors intended for real-time alpha spectroscopy of molten actinide salt has been fabricated at The Ohio State University's Nanotech cleanroom facility using epitaxial gallium oxide grown at a doping concentration on the order 1015 cm-3 by Novel Crystal Technologies. The detectors are designed to include a 22 nm thick $BaTiO_3$ electron blocking layer sputter coated onto an 11-micron thick epitaxial layer of gallium oxide grown on bulk gallium oxide wafers. This is capped with a 100 nm gold contact forming a Schottky barrier between the gold and gallium oxide while the $BaTiO_3$ prevents electrons tunneling past the Schottky barrier. This contact is protected by a 30 nm Pt outer layer with metal contacts annealed to form an Ohmic contact on the reverse face. Alpha spectra are collected under exposure to an Am-241 source in vacuum through a charge sensitive preamplifier and digitizer setup then passed to a PC. The detectors are evaluated using C-V and I-V electrical characterization to establish the formation of Schottky diodes and the maximum applied reverse bias prior to breakdown. Previously fabricated gallium oxide Schottky detectors displayed an effective resolution of 11%, potentially degraded by imperfections present in the epitaxial layer and surface leakage current.

More recent detectors fabricated using less highly doped gallium oxide give spectra with improved resolution. It is thought that resolution of these detectors will increase with further improvements to the crystal growth techniques and applied passivation methods.

Log 479. EFFECT OF TAILING CORRECTION ON MINOR ISOTOPIC ABUNDANCES: LARGE ENOUGH TO MAKE THE MAJOR ISOTOPE ABUNDANCE PASS OR FAIL IN A PROFICIENCY TEST CAMPAIGN?
K.J. Mathew (P); C.F. Ottenfeld; N. Butterfield; S. Levesque. (P) Presenting Author.

Total evaporation is the preferred measurement technique for major isotope ratio measurements using thermal ionization mass spectrometry instruments. For the major isotope abundances, this technique yields superior quality (better precision and accuracy) data due to the nature of this analysis technique. However, minor isotope abundance measurements using this technique have the known disadvantage of being systematically biased higher due to peak tailing from the major isotopes. As isotopic abundances are correlated systems (sum of all isotopic abundances must be 100%), we investigate whether the tailing corrections at the minor isotopes are large enough to shift the major isotope abundance values outside the ITV-2010 values, which are routinely used as pass/fail criteria in proficiency test programs. Measured data in a plutonium sample, a uranium sample, and a uranium separate (from decay of Pu isotopes) is investigated. The difficulty performing the tailing corrections on a forensic sample of plutonium material for chronometric purposes is evaluated as the isotopic abundances produced as a result of the Pu isotope decay is distinct from isotopic standards available for instrument calibration and estimation of the magnitude of the tailing corrections. LA-UR-21-32416

Log 480. UNCERTAINTY BUDGETS FOR MINOR ISOTOPE RATIOS OF PLUTONIUM: AN EVALUATION BASED ON TRACEABLE STANDARDS FROM NBL
K.J. Mathew; C.F. Ottenfeld; N. Butterfield (P). (P) Presenting Author.

Certified reference material (CRM) isotopic standards are used for calibration of the mass spectrometer instruments, for measurement control, and for validating analytical methods used by safeguards laboratories. We evaluated the isotopic data on plutonium CRMs 136, 137, 138, and 126-A to estimate the contributing factors to the uncertainty budgets. All data utilized in the evaluation here were obtained using the total evaporation methodology, which is considered the superior analytical technique for the $240Pu/239Pu$ major ratio measurements due to the excellent precision and accuracy obtained through the use of this technique. The minor isotope ratios were corrected for abundance sensitivity (peak tailing effects) using the manufacturer specified abundance sensitivity values and using observed systematic biases using replicate analysis performed within the same turret. For the minor isotope ratios, the per turret correction yielded overall uncertainties that are a factor of 2 to 3 smaller than the correction based on an assumed correction factor. LA-UR-21-32410

Log 481. METHOD DEVELOPMENT IN SUPPORT OF RECERTIFICATION OF PLUTONIUM CRMS 136, 137, and 138

K.J. Mathew (1); P. Mason (2); C.F. Ottenfeld (1,P); R. Keller (1). (P) Presenting Author.

The Standard Reference Material (SRM) standards (originally from the National Bureau of Standards --NBS) 946, 947 and 948, currently referred to as CRMs 136, 137, 138, were first produced in the 1960's. Since then, significant improvements have been made in Thermal Ionization Mass Spectrometry (TIMS) instrumentation and measurement techniques that allow characterization of isotope ratio data with uncertainties 5 to 10 times lower than those stated in the SRM 946, 947, and 948 certificates. Amounts typically used for our separations procedure are about 20 micrograms of plutonium. We performed a scaled-up separation experiment using 20 milligrams, in an effort to check the separation efficiency using the larger amounts. Results from this separation experiment demonstrated that Pu decay products (241Am, and 234U, 235U, 236U isotopes produced from the decay of 241Pu, 238Pu, 239Pu, and 240Pu) can be separated with equal efficiency using the same columns we use for separation of the 20 microgram quantities. We will discuss the results of the scaled up separations that we performed, which were funded through NBL as part of the office of science. LA-UR-32414

Log 482. VERIFICATION PERFORMANCE OF THE WORLD'S FIRST RADIOXENON ARRAY

Ringbom, A. (1,P); Aldener, M.; Elmgren, K.; Fritioff, T.; Kastlander, J.; Karlkvist, L.; Olsson, H.; (1) Swedish Defence Research Agency. (P) Presenting Author.

The world's first radioxenon array was installed in Sweden in May 2021. The array consists of five so called SAUNA CUBE units measuring atmospheric radioxenon, placed with 200 – 500 km inter-distance. The basic idea, supported by simulations, is that a combination of several less sensitive radioxenon measurement systems can outperform the verification performance of a single state-of-the-art ultra-sensitive system. Data from eight months of operation will be presented together with an analysis of detection-, location and source characterization parameters obtained by the array. The results will be compared with a state-of-the art SAUNA III system belonging to the CTBT International Monitoring System (IMS) that was operated simultaneously

Log 483. ORIGIN OF RADIOCESIUM IN WILD BOARS FROM BAVARIA

Stäger, F. (1,P); Zok, D. (1); Schiller, A.K. (1); Steinhauser, G. (1). (1) Leibniz University Hannover. (P) Presenting Author.

The beginning of atmospheric nuclear explosions marked the onset of the contamination of nearly the entire northern hemisphere with anthropogenic radionuclides. The Chernobyl nuclear accident (1986) contaminated wide parts of Europe with volatile fission products. Therefore, ^{137}Cs has become an ubiquitous environmental radionuclide in Europe. Its levels are easily measureable in various environmental samples. While the effective half-life of radiocesium led to a permanent decrease of contaminations levels, the contamination levels in wild boar tissue do not follow this trend. This phenomenon is still not fully understood and has been termed "wild boar paradox". The persistently high contamination of wild boars may correlate with their consumption of deer truffles. These underground mushrooms accumulate Cs efficiently and are a food source for wild boars. To investigate the source of the radiocesium in the animals and in deer truffles, we analyzed their ^{135}Cs/^{137}Cs isotope ratio by ICP-QQQ-MS. By applying this ratio, it is possible to distinguish between the sources of the radiocesium. It varies strongly between nuclear weapons fallout and releases from reactors. To solve this paradox, wild boar and deer truffle samples were collected across Bavaria. After a chemical separation of cesium and measurement of the ^{135}Cs/^{137}Cs ratio, all samples indicated mixing between reactor and nuclear weapons fallout, partly heavy on the side of reactors release. Using the ratio of ^{135}Cs/^{137}Cs, we now have the tools to understand and predict the contamination levels in wild boars.

Log 484.   **RADIOLOGICAL DISPERSAL DEVICE (RDD) PARTICLE GENERATION FOR POST-DISPERSAL FORENSIC METHOD DEVELOPMENT - THE FIRST STEP**

Totland, M. (1,P); Helal, A.(2); Dimayuga, I.(1); Chaudhuri, A.(1); Carrie, J.(1); Maach, S. (2). (1) Canadian Nuclear laboratories. (2) Canadian Explosives Research Laboratory. (P) Presenting Author.

Canadian Nuclear Laboratories (CNL) has a long history of research and development including radioactive source and medical isotope production, reactor and nuclear fuel development and the associated characterization methods for radioactive and nuclear materials. CNL has been leveraging its knowledge base and specialized facilities to support the enhancement of Canadian nuclear forensics capabilities in partnership with Government of Canada departments. Recently this has expanded to the study of radiological dispersal devices (RDD, the so called "dirty bomb") which uses a conventional explosive to disperse radioactive material. Post-dispersal forensics of a RDD aims to connect people, places and events, for both law enforcement and broader security purposes, through the examination of resultant radiological and explosive residue and particles. CNL recently worked with the Canadian Explosives Research Laboratory (CERL) to generate post-dispersal particles for use in subsequent characterization method development. The initial tests conducted at CERL utilized non-radioactive surrogate sources fabricated at CNL (filled with stable cesium chloride) and the common industrial explosive ANFO. This presentation will detail the fabrication of the explosive device, sample collection and subsequent analytical plan for characterization. Future plans for work in this area will also be covered. This work is funded under Atomic Energy of Canada Limited's Federal Nuclear Science and Technology Work Plan.

Log 485. STABLE CHLORINE ISOTOPE MEASUREMENTS OF ASTROMATERIALS USING THE MODIFIED MIDDLETON SOURCE OF AN ACCELERATOR MASS SPECTROMETER

Tyler Anderson (1,P); Alan Hidy(2); Jeremy W. Boyce(1); Francis M. McCubbin(1); Scott Tumey(2); Jordyn-Marie Dudley(3); Nikole C. Haney(3); Gérard Bardoux(4); Magali Bonifacie(4). (1) NASA-Johnson Space Center, Mail Code XI, Houston, TX 77058, USA. (2) Center for Accelerator Mass Spectrometry, Lawrence Livermore National Laboratory, Livermore, CA 94550. (3) Jacobs, NASA-Johnson Space Center, Mail Code XI3, Houston, TX 77058, USA. (4) Université de Paris, Institut de Physique du Globe de Paris, CNRS, F-75005 Paris, France. (P) Presenting Author.

The volatile element Cl can be lost during planetary formation and evolution, leading to fractionation of its two stable isotopes 35Cl and 37Cl. Chlorine isotope variations (reported as d37Cl in parts per thousand (per mil) relative to Standard Mean Ocean Chloride, SMOC) are documented to exceed 80 per mil between different lunar rock samples and have been variably interpreted as the fingerprint of degassing during accretion, magma ocean, or volcanic portions of lunar history. The large intersample and intrasample variations observed by bulk isotope ratio mass spectrometry (IRMS) and in-situ secondary ion mass spectrometry (SIMS) methods are difficult to interpret in part because of a paucity of bulk Cl isotope measurements. This lack of high-precision bulk data is due to the relative rarity of IRMS laboratories capable of making these high precision measurements on small samples of precious planetary materials such as those returned by human or robotic exploration. We present a new method for performing high precision d37Cl measurements using the modified Middleton ion source of an accelerator mass spectrometer. For samples with as little as 1 microgram Cl–equivalent to 2-4 mm3 of a typical lunar rock sample–the average cathode accuracy is ~1 per mil. Cathode reproducibility is typically ~1 per mil (2 sigma) for samples with at least 10 micrograms of Cl, increasing to ~3-6 per mil for aliquots with ~1-2 micrograms Cl, similar to SIMS results and sufficient to study astromaterials from the Moon, Mars, or 4 Vesta, which have tens of per mil variations.

Log 486. CHARACTERIZATION OF THE NEW PROMPT GAMMA NEUTRON ACTIVATION ANALYSIS FACILITY AT THE DALAT RESEARCH REACTOR

Nguyen Canh Hai1, Nguyen Nhi Dien1, Vuong Huu Tan2, Tran Tuan Anh1, Pham Ngoc Son1, Phan Bao Quoc Hieu1, Nguyen Canh Hai Tran1, Ho Huu Thang1, Tuong Thi Huong1. 1) Dalat Nuclear Research Institute, 01 Nguyen Tu Luc Street, Dalat, Vietnam 2) Vietnam Atomic Energy Society. (P) Presenting Author.

A new Prompt Gamma Neutron Activation Analysis (PGNAA) facility has been installed at horizontal channel No. 4 of the Dalat research reactor. The facility comprises a collimator with neutron beam filters, a beam shutter, a sample chamber with holder table, a beam stopper, a biological shielding and a gamma HPGe spectrometer. The materials used in the construction were lead, lithium florin, cadmium, Teflon, and iron. The characteristics of the new PGNAA facility, including the neutron flux, gamma dose and cadmium ratio at the sample position, absolute efficiency, background gamma-ray spectrum, as well as analytical sensitivies and detection limits, were determined and indicated to be much better than those of the old facility. Keywords: PGNAA facility, DRR, characteristics of the PGNAA facility.

Log 487.   **ULTRA-HIGH-RESOLUTION CRYOGENIC X-RAY SPECTROMETER FOR ADVANCED ELECTRON MICROSCOPE ACTINIDE MATERIAL MAPPING**

Carpenter, M. H. (1,P); Mark P. Croce, M. P. (1); McNeel, D. G. (1); Schrieber, K. A. (1); Ullom, J. N. (2); Bennett, D. A. (2); Mates, J. A. B. (2); Gard, J. D. (3); Imrek, J. (3); Becker, D. T. (3); Wessels, A. L. (3); Morgan, K. M. (3). (1) Los Alamos National Laboratory. (2) National Institute of Standards and Technology, Boulder. (3) University of Colorado, Boulder. (P) Presenting Author.

The Safeguards Science and Technology group (NEN-1) at Los Alamos National Laboratory has commissioned a next-generation microcalorimeter x-ray spectrometer for advanced nanoscale elemental and chemical species mapping in the scanning electron microscope (SEM). SEMs are widely used in nanoscale microanalysis of materials in many fields including nuclear forensics. Energy-dispersive spectroscopy (EDS) of the characteristic x-rays excited by the electron beam provides spatially-resolved elemental concentration information down to sub-micron resolution scales. Standard commercial silicon drift detectors (SDDs) used for EDS have typical energy resolution on the order of 100-150 eV for x-rays between 0.3-20 keV covering the full periodic table. Low-temperature microcalorimeters based on Transition Edge Sensors (TESs) achieve 10-20X better energy resolution than standard EDS detectors while measuring the full spectral band with comparable efficiency to EDS. TES detectors can better resolve overlapping lines from different elements in a sample, especially complex heterogeneous environmental samples, and achieve lower detection thresholds for trace elements while improving measurement uncertainty of major components. Additionally, this energy resolution enables analysis of minor satellite emission lines that demonstrate chemical sensitivity, yielding information about oxidation state and bound ligand species in certain scenarios. Here we describe the design and operational parameters of this integrated SEM-TES system, and discuss example measurements of actinide-bearing samples.

Log 488. **MEMBRANE ADSORBERS WITH COVALENTLY TETHERED DIGLYCOLAMIDE LIGANDS FOR ACTINIUM-225 PURIFICATION**

Sibley, M.M. (1,P); Sepesy, M.R.(1); Scott, J.V.(1); Kozar, T.J.(1); Ford, A.G.(1); Yen, T.(1); Johnson, A.B.(1); Duval, C.E.(1). (1) Case Western Reserve University. (P) Presenting Author.

Actinium-225 is an alpha-emitting isotope that shows promise for targeted alpha therapy, an emerging cancer treatment. It is produced by irradiating a thorium target then, after production, microgram quantities of Ac-225 must be purified from residual thorium and byproducts (e.g. lanthanides). Current purifications rely on DGA resin (diglycolamide ligands physisorbed in an acrylic resin). Transport of ions to binding sites in resin-packed columns is governed by diffusion. Thus they must be operated at low flowrates (< 1 mL/min) which leads to long purification times (> 6 hours). Membrane adsorbers are an alternative to resins in which ion transport is governed by convection; therefore they have the potential to perform the same separation in < 1 minute. In contrast to liquid-supported membranes, membrane adsorbers contain covalently bound ligands, preventing ligands from leaching at high flowrates. This contribution describes our efforts to 1) synthesize new, acid-stable membrane materials via electrospinning and 2) synthesize and covalently attach DGA ligands to those membranes. Membranes are synthesized by electrospinning poly(styrene-co-chloromethyl styrene) fiber mats and are characterized for surface area, permeability and porosity. DGA derivatives are synthesized with reactive end groups (amines or alkenes) to bind to membranes. In initial experiments, lanthanum (a surrogate for actinium) is used to determine the binding capacity and kinetics of the DGA-functionalized membranes. The results of this research are paving the way for faster separations in radiochemistry.

Log 489. **ADDITIVE MANUFACTURING OF ACTINIDE- AND FISSION PRODUCT-DOPED SILICA FOR PRODUCTION OF SURROGATE DEBRIS REFERENCE MATERIALS**

Shusterman, J. (1,P); Parsons-Davis, T. (1); Harris, S. (1); Joshipura, I. (1); Henderson, R. (1); Goodell, J. (1); Lee, E. (1); Kuntz, J. (1); Pascall, A. (1); Bandong, B. (1). (1) Lawrence Livermore National Laboratory. (P) Presenting Author.

Surrogate debris reference materials (SDRMs) are necessary for the verification and validation of laboratory methodologies employed in the analysis of post-detonation nuclear debris. Ideal solid SDRMs will incorporate relevant actinides, fission products, and activation species with matrix elements in realistic ratios and forms, be highly reproducible from piece to piece, and control for desired spatial heterogeneities (see related presentation by Parsons-Davis et al.),. Towards developing robust SDRMs, additive manufacturing, in particular electrophoretic deposition (EPD)of radionuclide-doped silica particles has been employed to fabricate tenth-of-a-gram scale glassy samples. The degree of incorporation of each element, homogeneity within the piece, and reproducibility across sample sets are assessed in the development of the SDRMs. The first use of EPD with fission products and actinides was conducted for SDRM development and the results of these studies will be presented. *This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344. This work was supported by Laboratory Directed Research and Development Funding 20-ERD-021.

Log 490.   **THE IMPACT OF SILVER ON THE ANALYSIS OF URANIUM BY KINETIC PHOSPHORESCENCE ANALYSIS**

Arnold, Ean (P) (1); Uhnak, Nic (1); Metz, Lori (1); Beck, Chelsie (1). (1) Pacific Northwest National Laboratory. (P) Presenting Author.

The analysis of uranium by Kinetic Phosphorescence Analysis (KPA) is negatively impacted by the presence of silver (Ag). KPA uses a laser to excite uranyl ions causing phosphorescence of those exited ions over time. An external calibration curve is generated to quantify the uranium in an unknown and generally complex sample. However, there are known interferences such as chloride that quench the phosphorescence intensity. The purpose of this work is to understand the effects of Ag on uranium analysis by KPA, specifically to determine the concentration of Ag that begins to quench the phosphorescent signal.   Results demonstrated that silver concentrations greater than 0.1 ppm Ag impacted quantification regardless of the concentration of uranium present in the sample.

Log 491. **XENON INTERNATIONAL TESTING OPERATIONS**

James C. Hayes (1,P), Mark E. Panisko (1), Matthew W. Cooper (1), Warren W. Harper (1), Michael F. Mayer (1), James H. Ely (1). (1) Pacific Northwest National Laboratory. (P) Presenting Author.

This presentation will discuss the Xenon International data observed during several phases of testing during the last 3 years. Xenon International was tested alongside of 2 separate International Monitoring Systems and has completed testing at Teledyne Brown Engineering (TBE) in Knoxville, TN. This presentation will review the Xenon International data with data collected on IMS stations that have different collection and processing times and will review the unusual detects of radioxenon isotopes never before observed in an IMS radioxenon system. We will also review data collected on Xenon International using the new PNNL developed MITER analysis code that is designed to get good resolution between Xe-133m and Xe-131m. PNNL will present on the several levels of testing performed with Xenon International and evaluate time and detector resolution.

Log 492. CORRELATING EFFECTS OF ROCK DAMAGE FROM UNDERGROUND CHEMICAL EXPLOSIVES WITH NATURAL RADON GAS BACKGROUNDS

Christine Johnson (1, P), Justin Lowrey (1), Xiao Luo (1), Hunter Knox (1), James Knox (1), Benjamin Roberts (1), Mark Rockhold (1), Khiloni Shah (1, 2), Christopher Strickland (1). (1) Pacific Northwest National Laboratory. (2) The University of Texas at Austin. (P) Presenting Author.

Rock damage from explosions in the subsurface directly impacts material flow and transport properties of bulk geology through the generation of fractures and the compression of porous material. In particular, understanding how rock damage from underground nuclear explosions immediately impacts the mobility of the associated radioactive materials is critical to improving predictive modeling of such nuclear explosions. As part of a large effort to investigate rock damage patterns, Pacific Northwest National Laboratory and Sandia National Laboratories have conducted a series of chemical explosive experiments at Blue Canyon Dome in Socorro, New Mexico employing a number of traditional and novel measurement and sensing techniques. We present here a segment of this research focused on assessing the evolution of natural radon gas in response to three chemical explosives in the testbed. The Blue Canyon Dome testbed consists of a central ground zero borehole where the explosions are detonated surrounded by eight concentric wells completed with gas sampling zones and a variety of other sensors. Results of pre- and post-explosion radon gas measurements are analyzed in the context of other site characterization techniques, including electrical resistivity tomography to directly image rock damage and gas flow patterns under experimental conditions.

Log 493. ASSESSING Xe-133 GAS TRANSPORT IN SHALLOW ALLUVIUM

Christine Johnson (1, P), Michael Mayer (1), Justin Lowrey (1), Khiloni Shah (1, 2), Dustin Clelland (1), Jim Fast (1), Brad Fritz (1), Xiao Luo (1), Justin McIntyre (1), Mark Rockhold (1), Signe White (1). (1) Pacific Northwest National Laboratory. (2) The University of Texas at Austin. (P) Presenting Author.

We present an overview of a small-scale tracer migration experiment that was carried out in July 2018 in Area 2 of the Nevada National Security Site (NNSS). This experiment involved the injection of 133Xe into the deepest zone (about 7 m depth) of a multi-completion shallow borehole. Sampling was then conducted on higher regions of the injection borehole and in a second borehole located approximately 17 m away from the injection site. A simple system for measurement of the 133Xe activity in whole air using a CsI(Tl) detector was utilized to analyze samples onsite immediately after collection. This allowed for the entire tracer experiment to be conducted at the injection location, without the need to ship samples off-site for analysis. In addition to the experiment results, we give an assessment of the tracer gas movement along with insights gained from numerical modeling. We show the results of predictive simulations that were performed prior to the experiment made using a system that was numerically modeled as a simple, homogenous media case with varying permeabilities. Later, a more complex numerical model of the system was developed in an attempt to fit simulated gas arrival times and concentrations with experimental data despite a lack of detailed site characterization data.

Log 494. **VARIABILITY OF ARGON EMANATION RATES AND RELEVANCE TO PREDICTIONS OF NATURAL ARGON GAS BACKGROUND IN THE SUBSURFACE**

Justin Lowrey (1), Christine Johnson (1, P). (1) Pacific Northwest National Laboratory. (P) Presenting Author.

Radioactive isotopes of argon, particularly Ar-37 and Ar-39, are important activation products generated from neutrons bombarding natural geology in underground nuclear explosions. While the production of these isotopes is conceptually straightforward and can be predicted using information about the elemental composition of local geology and the relevant interaction cross-sections, substantial uncertainty remains in predicting how much of the isotope remains trapped in the geology and how much is released and accessible to gas sampling techniques. This discrepancy between the amount of activated argon that is generated and the amount that can actually be collected through whole air gas sampling is termed the emanation fraction. Past efforts at Pacific Northwest National Laboratory (PNNL) have led to the direct measurement of the emanation fraction of Ar-37 and Ar-39 from five rocks and two powders. Here we examine the variability of the argon emanation measurements in the context of results from previous argon background sampling campaigns to determine whether the improved emanation fraction measurements can improve predictions of background levels of radioargon. Additionally, the underlying mechanisms responsible for emanation variability among rock types is discussed, and an argument is presented for treating emanation as time-dependent process better described as an emanation rate than simply a single characteristic fraction.

Log 495. **AN OVERVIEW OF SIMULATED RADIONUCLIDE RELEASE SIGNATURES FROM A TYPICAL MODERN MEDICAL ISOTOPE PRODUCTION FACILITY**

Justin Lowrey (1), Christine Johnson (1, P), Jacob Inman (1), Judah Friese (1), Harry Miley (1). (1) Pacific Northwest National Laboratory. (P) Presenting Author.

Medical isotope facilities are one of the primary contributors to the global radioxenon background. Past work has made predictions of the expected emissions from medical isotope facilities, particularly those operating in the early 2000s. However, significant changes have occurred since these previous studies, including improvements to modeling software, the closure of the Chalk River facility, and the conversion of most medical isotope production to the use of low-enriched uranium (LEU) targets. In an effort to update predictions of the impact of medical isotope emissions on the global radioxenon background, an updated simulation framework has been developed to explore releases from typical modern medical isotope production facilities. A range of operating conditions were simulated to examine the impact of operational variabilities on emissions and the range of radioisotopic signatures that may be observable.

Log 496. DIFFERENTIAL ADVECTIVE TRANSPORT OF VARIOUS NOBLE GASES THROUGH CONSOLIDATED ROCK CORES

Tara Scarborough (1, 2), Justin Lowrey (1), Mark Rockhold (1), Tom Wietsma (1), Christine Johnson (1, P), Derek Haas (2), Joseph Lapka (2). (1) Pacific Northwest National Laboratory. (2) The University of Texas at Austin. (P) Presenting Author.

Within nuclear explosion monitoring applications, the mobility of noble gases in the environment has traditionally been regarded very simply. For example, in past numerical simulations concerning radioactive materials from underground nuclear explosions, noble gas transport through porous rock was described by straightforward mass-dependent diffusion and non-reactive advective flow. As radioactive noble gases like Xe-133 and Ar-37 serve as key indicators of underground nuclear explosion activity, the potential for these gases to be impacted differentially by more complex interactions in the environment must be better understood. Thus, in recent years, a greater amount of research effort has been dedicated to understanding reactive, multi-phase flow of noble gases through the subsurface. Numerical model validation against data from controlled, gas flow experiments is key. Here we present an overview of a straightforward experimental system developed at the Pacific Northwest National Laboratory to study pressure-driven noble gas flow through bulk rock cores. The system utilizes a tri-axial core holder to confine up to 8-inch cores; a xenon, argon, krypton, and neon mix of noble gases in nitrogen is driven through the core at variable pressure from one end and measured by a mass spectrometer at the opposite end. We also present an initial data set of differential transport rates through unsaturated sandstone core material at various gas flow rates, core confining pressures and system temperatures.

Log 498. IN-SITU IDENTIFICATION AND QUANTIFICATION OF NORM

Zickefoose, J.K. (1,2,P); Sullivan, D.A.(1); Persson, H.(1); Ilie, G.(1); Dang, E(1). (1) Mirion Technologies. (2) University of Connecticut. (P) Presenting Author.

The identification and quantification of naturally occurring radioactive materials (NORM) is an important aspect of radioprotection in a number of industries such as oil and gas, heavy metals mining, coal burning, and water treatment. In general, NORM is detected through gamma ray spectroscopy where, depending on the nuclide, gamma lines from 45 keV up to 2615 keV are measured and the presence of certain lines indicates the presence of a given nuclide. Ideally these measurements are conducted in shielded environments where the interfering contributions to the gamma spectrum from the environment may be removed. However, in many cases shielded laboratory measurements are not possible and in-situ measurements must be conducted in the field where shielding is reduced or even eliminated. In these cases, the analysis may become complex, and uncertainties may become enlarged. Several options exist for both laboratory and in-situ field measurements ranging from HPGe detectors to various scintillators to room temperature semiconductors. The performance of coaxial, planar, and well type HPGe detectors will be considered including an HPGe detector optimized for in-situ measurements. Furthermore, the performance of NaI(Tl), SrI2(Eu), LaBr3(Ce), LaBr3(Ce+Sr), CeBr scintillators and CZT will be considered. The benefits of each detector type in both shielded and unshielded environments will be compared and discussed along with the challenges encountered when measuring NORM.

Log 499. SOURCE PREPARATION AND SPECTRAL ANALYSIS FOR MASSIC ACTIVITY BY DECAY ENERGY SPECTROMETERY USING TRANSITION EDGE SENSORS

Fitzgerald, R. (1,P), Bergeron, D.E.(1), Essex R.(1), Nour, S.(1); Shaw, G.(1), Verkouteren, R.M.(1). (1) National Institute of Standards and Technology. (P) Presenting Author.

Cryogenic decay energy spectrometry (DES) is a potentially powerful method for activity quantification in applications including nuclear safeguards, forensics, dating, and measuring precise nuclear decay data that improve other methods such as liquid scintillation. Ideally DES would be linked to massic activity, which requires liquid drop mass metrology at the few mg level. Here we present quantitative source preparation and spectral analysis methods to enable that link. Quantitative source preparation consists of gravimetric dispensing of alpha-emitting radionuclide solutions onto nanoporous gold substrates, which are then folded into a 4 pi absorber geometry. Due to the limits on overall absorber mass, the dispensed volumes are limited to the few microliter (milligram) level. Mass uncertainties of 1 % are adequate for many applications, but in some metrology applications, 0.1 % is desired. We present a microdrop method validated by liquid scintillation counting of Am-241 to be accurate at approximately the 1 % level. Quantitative activity analysis is pursued using Geant4 Monte Carlo modeling of DES spectra. We present simulation results for Am-241 and for mixed-nuclide sources such as Pu-239 & Pu-240, which demonstrate the dependence of full-energy peak efficiency on source geometry due to alpha and photon escapes. Potential additional applications include standardization of radionuclides in the presence of impurities (e.g. Ac-227 in Ac-225) and as a complimentary method to mass spectrometry for isobars (e.g. Pu-238 & U-238).

Log 500. SEPARATION OF URANIUM FROM THORIUM IN SUPPORT OF PARTICLE REFERENCE MATERIAL PRODUCTION

Chelsie L. Beck (1,P) Ean S. Arnold (1), Evan J. Warzecha(1), Katherine Koh(1), Riane E. Stene(1), Timothy R. Pope(1), Neil Henson(1), Stephan Vogt(1), Matthew S. Wellons (2), Christopher A. Barrett(1). (1) Pacific Northwest National Laboratory, Richland, WA (2) Savannah River National Laboratory, Aiken, SC. (P) Presenting Author.

Trace analysis of particulate material collected on environmental swipe samples continues to be one of the cornerstones of the IAEA's process of verifying member state compliance as part of the Non-Proliferation Treaty (NPT). The ability to measure the "age" of individual particles through radiochronometry provides valuable information about when a sample was manufactured or produced which can also provide insights into a nuclear facility's operational history. Consequently, the 230Th/234U radiochronometer is a reliable indicator of nuclear compliance and verification with disarmament accords and fissile material cut-off treaties. However, reference materials with a suitable particle form-factor and well-documented purification dates are continually needed. One mechanism to address this need is to use legacy uranium inventories. However, these materials have unknown amounts of 230Th. Therefore, the U and Th must be quantitatively separated to both reset the age of the material, as well as generate new model ages. This work used UTEVA resin, with and without the addition of oxalic acid, for the separation and inductively coupled plasma mass spectrometry to measure the Th and U. Separations using oxalic acid caused unacceptable levels of Th background on the mass spectrometer. The removal of oxalic acid reduced the Th background and improved uranium recovery for the same volume of eluent. High recoveries and adequate separation factors have been demonstrated with this method.

Log 501. A CONCEPTUAL MOLTEN SALT MASS MEASUREMENT TECHNIQUE BASED ON RADIOACTIVE TRACER DILUTION AND GAMMA SPECTROSCOPY

Cao, G. (1, P); Li, S. (1); Cao, R. (2); Kandlakunta, P. (2). (1) Idaho National Laboratory. (2) Ohio State University. (P) Presenting Author.

For safeguards purposes, in molten salt systems such as molten salt reactors or spent nuclear fuels pyroprocessing facilities, it is necessary to know the molten salt mass to determine the nuclear material inventory. However, the irregular or complicated shape of molten salt containers and inherent molten salt characteristics such as density variabilities make accurate mass measurements very difficult. To address this safeguards challenge, an innovative, conceptual molten salt mass measurement technique based on radioactive tracer dilution and gamma spectroscopy was proposed. The main features of the technique include: (1) a small amount of radioactive tracer salt—of known radioactivity—is added into a molten salt container, of which the mass of the molten salt is unknown, (2) a salt sample is taken from the molten salt container after the tracer salt is uniformly mixed with the molten salt of unknown mass, and (3) the radioactivity of the salt sample is measured by gamma spectroscopy and the molten salt mass is then calculated. The technical details of the conceptual molten salt mass measurement technique are provided using spent nuclear fuels pyroprocessing facilities and Na-22 tracer as

examples. The factors that may potentially affect the accuracy and repeatability of molten salt mass measurement associated with the technique are also discussed.

Log 502. EXAMINATION OF USING BORON-DOPED DIAMOND FOR SPECTROELECTROCHEMICAL ANALYSIS IN HARSH ENVIRONMENTS

Rakos, Jason (1,P); Patenaude, Hannah K. (1); Olney, Renee (1); Damjanovic, Nastasija (1); Rusinek, Cory A. (1). (1) University of Nevada, Las Vegas. (P) Presenting Author.

Spectroelectrochemistry (SEC) is an interesting technique that enables the determination of several fundamental redox and thermodynamic properties of a given electrochemical reaction. Furthermore, SEC offers enhanced selectivity in harsh environments (HE). While several electrode materials have been used for SEC, boron-doped diamond (BDD) has optimal characteristics for complex media such as high chemical and mechanical stability. BDD also has several beneficial electrochemical characteristics such as a wide potential window and low capacitive background current. This allows for a better signal-to-noise ratio that lowers the detection limit. In this study, we present a free-standing BDD electrode mini-grid for SEC measurements in HE. First, Ferri/Ferrocyanide was analyzed using cyclic voltammetry (CV), choronocoulometry (CC) and chronoabsorptometry (CA) as a proof of concept to determine changes in electrochemical behavior, and several parameters were assessed such as diffusion coefficient. To assess the ability to collect measurements in complex media, studies in strong acids and strong bases were executed. The performance and resilience of the BDD mini-grid in chloride-based molten salts was also examined at varied temperature (450, 550 and 650 C). Topographically, scanning electron microscopy (SEM) was used to examine changes in the diamond structure before and after exposure. Overall, this work highlights the applicability of diamond electrodes and SEC for measurements in HE, paving the way for electroanalytical studies in media previously not possible.

Log 503. **EMISSION GHOST IMAGING**

Chen-Mayer, H.H. (1, P); Coakley, K.P. (1); Ravel, B. (1); Josell, D. (1); Klimov, N.N. (1); Robinson, S.M. (1); Hussey, D.S. (1). (1) National Institute of Standard and Technology. (P) Presenting Author.

Ghost imaging (GI), an emerging alternative to conventional imaging, has seen greater utility with "computational GI", a concept we are exploring for imaging neutron induced gamma-ray emission. The incident beam is coded with predetermined pseudo random patterns or "speckles" from which an approximate orthogonal basis can be derived. The images of the speckle patterns are first measured as prior knowledge. Subsequent measurements of the sample only require signals from a "bucket detector" for each speckle pattern. This eliminates the need for position sensitive detector or physical collimation, both of which are difficult to achieve for gamma rays. We simulated GI of prompt gamma emission from a 2D distribution comprised of H, C, Ca, and N under noise-less conditions. While waiting to perform neutron-gamma measurement at NIST, we carried out an analogous experiment in which the neutron beam was replaced by a synchrotron x-ray beam and gamma-ray emission with x-ray fluorescence (XRF). A total of 442 unique speckle patterns were created using a "barcode" composed of randomly spaced gold-filled lines. A 7.5 mm (H) x 2.5 mm (V) test sample of 0.3 mm diameter Cu-Ag wire was scanned vertically across the 8 mm (H) x 1 mm (V) beam in five increments. For each increment, the Cu-K-a,b XRF bucket signals excited by the same 442 patterns were used to reconstruct a 1D GI at 0.165 mm resolution. These 1D GIs were stacked together to form a 2D reconstruction. The data processing method can be readily applied to our future neutron-gamma experiment.

Log 505. **GAMMA-RAY SPECTROMETRY ANALYSIS METHODS FOR RADIOISOTOPE DISSOLUTION AND MIXING FOR NUCLEAR FORENSICS APPLICATIONS**

Holschuh, T.V. (1,P); Ocampo Giraldo, L.A. (1); Snow, M.S. (1); Chichester, D.L. (1). (1) Idaho National Laboratory. (P) Presenting Author.

The production of surrogate nuclear explosive debris involves the dissolution of irradiated fissile material, elemental isolation, and recombination based on required fission product ratios. Gamma-ray spectrometry is used at each step in the production process to assess the radioactive fission product inventory. A high-purity germanium detector is used to measure each sample, and the detector efficiencies are determined prior to sample measurements using a calibration source and Monte Carlo N-Particle simulations to correct for geometric differences between the calibration source and dissolved material. Through radiochemical techniques, elements are isolated and then recombined into samples with specific fission product ratios. The concentration of each isolated element to be mixed is calculated from the activity calculations provided through gamma-ray spectrometry. However, impurities or imperfect chemical separations cause the recombined mixture to possess larger or smaller fission product ratios. A matrix-based approach can be taken based on the fission product activities in each separated solution along with each respective solution's volume to calculate the proper mixture volumes to use to obtain intended volatile-to-refractory fission product ratios, or R-values.

Log 507. **DISSOLUTION OF URANIUM HEXAFLUORIDE AND PRECIPITATION OF URANIUM FROM 1-METHYL-1-PROPYLPIPERIDINIUM BIS(TRIFLUOROMETHYLSULFONYL)IMIDE**

Higgins, C.J. (1,P); Lubke, K.I.(1); Poineau, F.(1); Czerwinski, K.R.(1); Hatchett, D.W.(1); (1) University of Nevada, Las Vegas. (P) Presenting Author.

The current method for deconversion of the USA's 700,000 metric tons of depleted uranium hexafluoride (DUF6) waste involves the mixing of volatilized DUF6, steam, H2, and N2 to make uranium oxides and HF gas. The dissolution of solid UF6 at depressed temperature into room temperature ionic liquid, 1-methyl-1-propylpiperidinium bis(trifluoromethylsulfonyl)imide ([MPPi][TFSI]) and recovery of uranium is the focus of the current studies. The spontaneous dissolution occurs with the oxidation of TFSI and reduction of UF6. XAFS and UV-Vis data of the solution indicates the U(V) oxidation state. Over time a solid precipitate crashes out of solution. TGA analysis indicates that 58% of the mass is lost at temperatures exceeding 1148 oC. PXRD shows the product after TGA is UO2. The mass losses from the TGA data are consistent with precipitation of [MPPi]2[UF6] salt. An alternative route for recovery of uranium is the addition of deionized water to the solution of UF6 in [MPPi][TFSI] producing immediate precipitation. PXRD and TGA data shows the precipitate to be hydrated UF4, which is easily converted to uranium oxides with heating. Work has been done to maximize recovery of the uranium from the [MPPi][TFSI] without loss to the aqueous phase.

Log 508. TRANSFER PARAMETERS OF RADIONUCLIDE INTO FARM ANIMAL PRODUCTS IN SEMIPALATINSK TEST SITE CONDITION (MAIN RESULTS AND PROSPECTS)

Baigazinov, Zh.A. (1,2), Lukashenko, S.N.(3), Kovacs T. (2) (1) Institute of radiation safety and ecology, National Nuclear Center of Republic of Kazakhstan, Kurchatov, Kazakhstan (2) Institute of Radiochemistry and Radioecology, University of Pannonia, Veszprem, Hungary. (3) Russian Institute of Radiology and Agroecology, Obninsk, Russia. (P) Presenting Author.

Starting from 2007 on the Semipalatinsk Test Site (STS) diverse natural experiments investigating the parameters of the radionuclide transfer into agricultural products in order to assess the opportunities for agricultural usage of the STS territory. These investigations employ a special experimental farm located on one of the highly contaminated sites of the test site. The natural experiments are set during summer time. Typical for these region agricultural animals (cattle, small cattle, horses, pigs, wild boars, hens, broilers) are farmed and investigated. Different groups of animals and birds were fed with contaminated forage, soil and water; in some of the experiments, the animals were watered with a solution of known amounts of radionuclides. The forage and soil were prepared at diverse contaminated testing sites of the STS with a different character of radionuclide contamination. Main parameters which were studied are the dynamics of accumulation and excretion of radionuclides into the organs; distribution of radionuclide in the body and transfer coefficient (Tf, CR) data. Investigation showed that the use of agricultural commodities produced at the STS lands outside the contaminated testing grounds remains absolutely radiologically safe. This conclusion is based on the relatively low migration capabilities of the radionuclides in the system "soil – vegetation – livestock products". This fact is a peculiar feature of the STS contamination. Among other important factors are the localized character and relatively small areas of the radionuclide contamination.

Log 509. **IMPLEMENTATION OF LOW-TEMPERATURE DETECTORS FOR NUCLEAR MATERIALS ANALYSIS**

D.T. Becker (1, P), B.K. Alpert(2), D.A. Bennett(2), D.E. Bergeron(2), M. H. Carpenter(3), M. Croce(3), R.M. Essex(2), R.J. Fitzgerald(2), J.W. Fowler(1,2), J. D. Gard(1), G.C. Hilton(2), A.S. Hoover(3), J. Imrek(1), K.E. Koehler(3), J.A.B. Mates(2), D.J. Mercer(3), K.M. Morgan(1), N.J. Nakamura(2), S. Nour(2), G.C. O'Neil(2), N.J. Ortiz(2), C.D. Reintsema(2), D.R. Schmidt(2), K.A. Schreiber(3), G.A. Shaw(2), D.S. Swetz(2), P. Szypryt(2), J.N. Ullom(1,2), L.R. Vale(2), R.M. Verkouteren(2), D.T. Vo(3), S.L. Weidenbenner(3), A. Wessels(1), D. Yan(2), (1) University of Colorado, Boulder, Colorado, USA, (2) National Institute of Standards and Technology, Boulder, Colorado, USA, (3) Los Alamos National Laboratory, Los Alamos, New Mexico, USA. (P) Presenting Author.

Ultra-low temperature microcalorimeters based on superconducting detectors and readout have long been recognized as having exquisite energy resolution for single photons and massive particles, achieving resolving powers from 1000-5000 for a wide energy range from 1keV to 10 MeV. Practical application to Nuclear Materials Analysis has been hampered by low collection area and cumbersome ultra-low temperature refrigeration technology. Advancement in the production and readout of large arrays of detectors addresses the first issue. The availability of reliable commercial cryogen-free ultra-low temperature refrigeration systems addresses the second issue. We will describe the working principles of these detectors and how we have met the implementation challenges of these technologies, focusing on three real-world systems. First, the portable SOFIA gamma-ray spectrometer in operation at Los Alamos National Laboratory (LANL), which can resolve closely spaced actinide gamma-ray and x-rays lines in the 90 – 220 keV region, allowing quantitative measurements of actinide isotopic content. Second, a high-resolution x-ray spectrometer also in operation at LANL, which can determine spatially-resolved elemental concentration information with sub-micron spatial resolution, while allowing closely-spaced lines to be resolved. Finally, a new total Decay-Energy Spectrometry system currently under development at the National Institute of Standards and Technology will allow quantitative massic activity measurements of radionuclides for samples with no chemical separation with ~ 1 Bq activity.

Log 510. **A QUANTITATIVE COMPARISON OF MICROCALORIMETERS AND HIGH-PURITY GERMANIUM DETECTORS FOR NON-DESTRUCTIVE PLUTONIUM ISOTOPIC ANALYSIS**

D.T. Becker (1, P), D.A. Bennett(2), M. H. Carpenter(3), M. Croce(3), J.W. Fowler(1,2), J. D. Gard(1), G.C. Hilton(2), A.S. Hoover(3), J. Imrek(1), K.E. Koehler(3), J.A.B. Mates(2), D.J. Mercer(3), G.C. O'Neil(2), N.J. Ortiz(2), C.D. Reintsema(2), D.R. Schmidt(2), K.A. Schreiber(3), D.S. Swetz(2), J.N. Ullom(1,2), L.R. Vale(2), D.T. Vo(3), S.L. Weidenbenner(3), A. Wessels(1). (1) University of Colorado, Boulder, Colorado, USA, (2) National Institute of Standards and Technology, Boulder, Colorado, USA, (3) Los Alamos National Laboratory, Los Alamos, New Mexico, USA, (P) Presenting Author

Gamma-ray spectroscopy using low-temperature microcalorimeters has matured significantly in the last few years with the deployment of the SOFIA (Spectrometer Optimized for Facility Integrated Applications) instrument at Los Alamos National Laboratory. SOFIA contains 256 microcalorimeter pixels, and routinely achieves an energy resolution of 60 eV at 97 keV. A next-generation instrument is currently scheduled to be installed at Idaho National Laboratory in 2022. This enhanced instrument includes 384 pixels and has an architecture that supports scaling to even higher pixel counts. The goal of this program has been to take advantage of the excellent energy resolution provided by microcalorimeters to close the gap in accuracy between non-destructive and destructive assay, with applications in nuclear materials control and accounting. We will describe our recent efforts in this area, focusing on the determination of the relative abundances of different plutonium (Pu) isotopes in 13 standard Pu reference materials. We have acquired high statistics spectra of these materials with both our microcalorimeter array and with High Purity Ge (HPGe) detectors, and analyzed both data sets in a consistent way with a newly developed analysis code, SAPPY. We additionally analyze the HPGe data with a standard HPGe analysis code (FRAM) in order to connect to prior work. We report on the results of this comparison. We gratefully acknowledge the support of the U.S. Department of Energy through the NE and NEUP programs, as well as the Los Alamos National Laboratory G.T. Seaborg Institute.

Log 511. TAILORING THE MICROSTRUCTURE OF UO2 FUEL CANDIDATES FOR INTENTIONAL FORENSICS

Finkeldei, S.C. (1,P); Connor, T. (1); Hunt, R.D. (2). (1) The Unviersity of California, Irvine. (2) Oak Ridge National Laboratory. (P) Presenting Author.

Uranium dioxide is the most commonly used fuel in nuclear power plants, and hence, much focus has been placed to improve its efficiency and safety. The strategy to enhance UO2 fuel by increasing its thermal conductivity and ability to retain fission products could improve the operation of worldwide fleet of light water reactors. The development of suitable fabrication methods to synthesize UO2 feedstocks for high-performance UO2 fuel candidates will be discussed. A bottom-up approach has been used to tune the synthesis route that could yield in UO2 with controlled amounts of dopants in order to induce targeted UO2 grain growth. Enlarged UO2 grains in a nuclear fuel pellet are expected to slow down fission gas diffusion, which is considered to be a superior property for these high-performance fuel candidates. The unique fabrication processes for microspherical feedstock via the internal gelation approach have been further developed to enable the dopant uptake within the UO2 microspheres. Selected dopants in the parts per million range were added into UO3 spheres. Even though, certain amounts of the dopants volatized during the sintering process, structural analyses, including scanning electron microscopy revealed the formation of large, up to 80 μm, UO2 grains for the pellets prepared from doped microspherical feedstocks. Synthesis capabilities presented here that enable altering the UO2 microstructure and morphology along with their functional properties might be of interest to the nuclear forensics community and this presentation is seeking to initiate this conversation.

Log 512. SURVEY OF ADDITIVE MANUFACTURING SIGNATURES FOR THE PREVENTION OF NUCLEAR PROLIFERATION

Gladden, B.J.(1,P); Snarr, S.E.(1); Snarr, P.L.(1); Beaman, J.J. (1); Haas, D.A. (1). (P) Presenting Author.

Improvements in additive manufacturing technologies will enable multiple-material and advanced material capabilities, creating opportunities to improve and expand the nuclear fuel fabrication process. Additive manufacturing can allow fuel to have complex geometries and contain composites of materials that would otherwise be difficult or impossible to make with traditional manufacturing methods. This opportunity also has implications for nuclear proliferation. We present a survey of additive manufacturing technologies and relevant signatures that could be used to identify processes, materials, or part properties.

Log 513. ENERGY DEPENDENCE OF FISSION PRODUCT YIELDS FROM THE MAJOR ACTINIDES

Gooden, M.E. (1,P); Bredeweg, T.A.(1); Finch,S.(2); Ramirez, A. (3); Silano, J.(3); Tonchev, A. (3); Tornow, W. (2); Wilhelmy, J. (1). (1) Los Alamos National Laboratory;(2) Duke University/Triangle Universities Nuclear Laboratory; (3) Lawrence Livermore National Laboratory. (P) Presenting Author.

A joint collaboration between LANL-LLNL-TUNL has produced a large set of absolute differential cumulative fission product yields for the major actinides. This data has shown new energy dependence to a number of peak yield fission products. Measurements have been made in nearly 1 MeV steps from 0.5 – 14 MeV using quasi-monoenergetic neutrons from the Triangle Universities Nuclear Laboratory (TUNL) Tandem accelerator. This new energy dependent fission yield data is highly useful to reactor design, nuclear forensics, and the forth coming new fission data evaluation. Data will be presented on dozens fission product yields, whose half-lives vary from minutes to months and were measured using fission chambers, for absolute fission number of fissions, and off-line gamma-ray spectroscopy. We will also discuss current efforts on very short-lived fission products using a rapid transfer system and cyclic activation where we are measuring fission yields who half-lives are on the order of seconds.

Log 514. RADIONUCLIDES FOR THERANOSTICS APPLICATIONS: Mn-52 PRODUCTION BY DEUTERON BEAMS IRRADIATION

Groppi, F. (1,P); Bianchi, F. (1); Hassan G. (1,2); Manenti, S. (1); (1) LASA Lab., Physics Dept. of Milano University and INFN-MI. (2) College of Science, Physics Dept. Salahaddin University - Erbil, IRAQ. (P) Presenting Author.

Now a day the new frontier of Nuclear Medicine (NM) is the use of artificial radionuclides that present nuclear properties suitable for theranostic applications that combine therapy and diagnosis in the same radiopharmaceutical to go in the direction of personalized medicine. This term is applied also to nuclides with properties that can give complementary diagnostic information. A promizing nuclide that presents these characteristics is manganese. In particular manganese-52 is a radionuclide which decays with positron emission and electron capture, with a medium-long half-life (T1/2 = 5.591 d). with possible and promising use in NM as a radiotracer for PET diagnostic tests; the low energy of the positrons emitted (244.6 keV) and the short range in the tissues (0.63 mm) would allow to acquire diagnostic images of a quality similar to those obtained with radiotracers already in use, such as fluorine-18 (252 keV; 0.66 mm). Moreover, the stable isotope of manganese Mn^{2+} presents paramagnetic properties that make it suitable for use in MRI, opening the possibility of obtaining multi-modal PET / MEMRI images. Currently manganese is produced by irradiation with protons on chromium targets. We have studied the production of Mn-52 using natCr(d,x) nuclear reaction by deuteron beams irradiation, that could be more advantageous. New sets of excitation functions for this nuclear reaction and the co-produced contaminants were obtained and compared with the other sets present in literature and the results of simulation with EMPIRE 3.2.2 and TALYS codes.

Log 515. **PREPARATION OF URANIUM TETRAFLUORIDE MICROSPHERES USING AMMONIUM BIFLUORIDE**

Jang, H.(1,P); Louis-Jean, J.(1); Poineau, F.(1). (1) University of Nevada, Las Vegas. (P) Presenting Author.

The need to better understand the formation, reactivity and morphology of nuclear materials is an important factor for nuclear forensics and nonproliferation efforts. Of particular interest is the chemistry of uranium fluoride (UF4). This material is a key intermediate compound in the uranium fuel cycle, which is used as a precursor for UF6 and uranium metal. Historically, UF4 has been produced by the direct fluorination of UO2 powder with gaseous HF. Given that bifluoride salts are a strong fluorinating agent, the present work studies the reactivity of UO2 microspheres in indirect contact with ammonium bifluoride under autogenous pressure using an autoclave system. For this, a series of reactions were investigated at 373 K to 473 K for 1 to 30 days. This provides an alternative route for laboratory scale production for UF4 preparation. Chemical signatures and fingerprints aroused from this process were studied using scanning electron microscopy (SEM) and energy-dispersive x-ray spectroscopy (EDS). This was done in an effort to clearly establish any correlation between morphology and structural composition of uranium fluoride to a given synthetic route. The results of this study provide a thorough understanding of actinide fluoride chemistries and their implications in nuclear forensics and nonproliferation.

Log 516. **DETERMINATION OF Cf ISOTOPES IN MARK 18A TARGET MATERIAL**

Fenker, K. M. (1,P); DiPrete, D. P.(1); Eldridge, H. W. (2); Armstrong, C. R.(1). (1) Savannah River National Laboratory. (P) Presenting Author.

The Savannah River National Laboratory (SRNL) was tasked by the National Nuclear Security Administration (NNSA) to recover isotopes from irradiated Mark-18A targets, including 244Pu. The Savannah River Site (SRS) has sixty-five Mark-18A targets currently stored in L-Area Basin that will be transported to SRNL for processing. The Mark-18A targets include 242Pu that has been irradiated under high neutron flux in SRS's K-Reactor. In support of the analytical characterization of the Mark-18A process, the Nuclear Measurements Group (NMG) is developing a new method to determine the californium concentration in Mark 18A target material following the extraction of the plutonium. A protocol utilizing Eichrom's RE or DGA resin to clean up the matrix followed by LN resin to extract the tetravalent isotopes is being developed and tested. The LN extraction conditions will be tailored to purify the Cf from the many other tetravalent isotopes present in the Mark 18A target matrix. Radiological and non-radiological simulants will be tested to develop an extraction scheme that purifies the Cf from Cm, Am, and Eu, to remove gamma interferences. The results of those experiments will be presented. The new Cf extraction method will be a key component of the analytical characterization of the Mark 18A target material.

Log 517. EXTRACTION OF Pm-147 FROM SRS NUCLEAR MATERIAL MANAGEMENT PROGRAM SOLUTIONS

Fenker, K. M. (1,P); DiPrete, D. P.(1). (1) Savannah River National Laboratory. (P) Presenting Author.

The Savannah River Site (SRS) regularly receives, stores, and processes highly enriched spent nuclear fuel from foreign and domestic nuclear reactors. Spent fuel is processed at SRS's chemical separation facility, H-canyon, through a modified PUREX process. The raffinate, which contains fission products, is transferred to SRS's liquid waste treatment facilities for disposal. Many of those fission products are of scientific interest and are difficult to access elsewhere. One such isotope is 147Pm, which has no stable isotopes but is a prominent fission product. Recently, the Savannah River National Laboratory's Nuclear Measurements Group (NMG) was challenged to extract 147Pm from SRS spent fuel processing rather than dispose of it. An existing method, used to determine 147Pm and 151Sm in a variety of radiochemical matrices, was modified to purify significant quantities of 147Pm from the H-Canyon waste. SRS spent fuel raffinate is a complicated matrix, it has a high whole-body dose from 137Cs and high extremity dose rate from 90Sr and its daughter, 90Y. The first step in the separation protocol is to remove these high dose isotopes. Then, the trivalent elements (147Pm, 90Y, 241Am, 151Sm, 152, 154, 155Eu, and 144Ce) are extracted. Non-radiological simulants and mixtures of 241Am, 147Pm and 151Sm were tested to develop the extraction scheme. The developed scheme was tested on an aliquot of H-Canyon waste solution from the processing of spent fuel from the High Flux Isotope Reactor at Oak Ridge National Laboratory. Results from these experiments will be discussed.

Log 518. ASSESSMENT OF A CORROSION RESISTANT AMORPHOUS ALLOY COATING ON SS 316 EXPOSED TO MOLTEN LiCl-KCl EUTECTIC

Killinger, D. (1, P), Phongikaroon, S. (1) Virginia Commonwealth University. (P) Presenting Author.

Amorphous alloys have gained interest as a pretreatment method for preventing the corrosion of metallic and alloy containing components in molten salt environments. In this study, SS 316 coupons with an amorphous alloy coating were exposed to a molten salt environment (LiCl-KCl eutectic, 500 C) to assess their performance in preventing attack on the substrate as well as corrosion of the amorphous alloy layer. Electrochemical techniques were employed during the exposure to monitor cell potential and obtain Nyquist and Tafel plots to estimate corrosion rate and evolution of the corrosion mechanism. Material characterization techniques were used to characterize the major constituents of the amorphous alloy and its morphology to ultimately compare them to findings after exposure to the salt. Mass spectrometry was performed on salt samples obtained during the experiment to assess for dissolution of alloy components into the salt.

Log 519.   EFFECT OF MOLTEN SALT COMPOSITION ON THE MORPHOLOGY OF ELECTRODEPOSITED URANIUM CRYSTALS

Killinger, D. (1, P), Phongikaroon, S. (1) Virginia Commonwealth University. (P) Presenting Author.

A benefit of salt fueled molten salt reactors over salt cooled molten salt reactors is the possibility to perform online salt conditioning and fuel reprocessing. The U(III)/U(0) redox couple has a potential to be used as a method to control the uranium levels in fuel salt. Extensive work has already been performed in chloride surrogate salts that are modeled after the electrorefining step of pyroprocessing technology; however, work in other chlorides and fluorides is lacking and the dependence of electrocrystal morphology on the molten salt environment's composition is not well understood. In order to have control over the electrocrystals in a molten salt reactor, this knowledge must be known. In this study, we varied the compositions of host salts (LiCl-KCl, NaCl-CaCl2, LiF-NaK-KF, 600 C) and target analytes (UCl3 and CeCl3, 1-5 wt%) to investigate their effects on the morphology of the uranium electrocrystals. Electrochemical analysis techniques were employed to determine any changes due to salt composition (e.g. CV, EIS, OCP, CA, CP). Material characterization techniques were employed to study the morphologies resulting from the changes and assess any composition changes in the electrocrystals (if any).

Log 520.   A COMPARISON OF ALTERNATIVE ISO-11929 COMPLIANT DETECTION LIMIT CALCULATIONS FOR THE CASE OF LARGE SYSTEMATIC UNCERTAINTIES

Kirkpatrick, J.M. (1, P). (1) Mirion Technologies (Canberra), Inc. (P) Presenting Author.

The ISO 11929 formulation of the detection limit for radiation measurements extends the long-established Currie method to include the systematic effects of uncertainties in the instrument calibration. However, the ISO formula is known to fail for cases in which those uncertainties are too large, making it unsuitable in certain applications where large calibration uncertainties are common. This occurs due to the use of a Gaussian approximation for both the random counting process and the systematic calibration uncertainties. The most recent (2019 - 2020) revision of the standard allows the use of alternative calculation methods to evaluate the detection limit in such circumstances. Here the numerical results of two such alternative approaches are compared. The first is an approximation to the ISO detection limit that combines the Gaussian limit of counting statistics with a log-normal representation of the calibration uncertainties, which was recently implemented in Mirion's NDA 2000 software to support a non-destructive waste assay system requiring ISO-compliance for a European site. The second method is a first-principles numerical calculation that accurately represents the Poisson nature of counting statistics and accommodates a variety of realistic (positive definite) distributions to model the calibration uncertainty. Several different choices of calibration uncertainty models are demonstrated. Results are calculated with both methods over a range of calibration uncertainty values, and at different background levels. The relative merits of both approaches are discussed

Log 521. **EXTREMELY HIGH RESOLUTION GAMMA RAY SPECTROMETER FOR THE ANALYSIS OF NUCLEAR FUEL AND REACTOR MATERIALS**

Schreiber, K.A. (1,P), Croce, M.P. (1), McNeel, D.G. (1), Carpenter, M.C. (1), Wessels, A. (3), Ortiz, N. J. (3), Ullom, J. N. (2,3), Gard, J.D. (3), Bennett, D. A., (2), Mates, J.A.B. (3), Imrek, J. (2), Becker, D.T. (2), Swetz, D.S. (2), Schmidt, D.R., (2). (1) Los Alamos National Laboratory, (2) National Institute of Standards and Technology, Boulder (3) University of Colorado Boulder. (P) Presenting Author.

Nuclear energy depends on the development of new reactor technologies which are increasingly safe, secure, and economical. In support of this mission, the Materials and Fuels Complex (MFC) Analytical Laboratory at Idaho National Laboratory provides sophisticated analysis of nuclear materials. An important component is nondestructive assay of fresh and spent fuel, reactor components, radioactive waste, and other materials from the nuclear fuel cycle. This assay includes gamma ray spectroscopy, which measures isotopic composition. For an accurate measurement, a gamma ray spectrum with high resolution and high counting statistics is desirable. However, such spectra can be difficult to obtain from the materials to be studied, which often consist of very dilute samples. Here we present a new gamma ray spectrometer, developed at Los Alamos National Laboratory in collaboration with the National Institute of Standards and Technology and University of Colorado, to be installed in the MFC in 2022. The spectrometer uses a microcalorimeter array, a gamma ray detector with unparalleled energy resolution. Microcalorimeters are sensors which operate below 100 milliKelvin and respond sensitively to the absorption of a photon. This gives them energy resolution on the order of 60-80 eV FWHM in the 50-250 keV range. The detector is mounted within a dilution refrigerator, and can measure continuously, permitting multi-day measurements with thousands of counts per second. This gamma ray spectrometer will permit precise, high statistics measurements on small amounts of novel nuclear fuel cycle materials. We will discuss the development of this detector and present spectra demonstrating its capabilities. LA-UR-21-32401

Log 522. **ACTINIDE CHEMICAL SPECIATION FROM ULTRA-HIGH-RESOLUTION CRYOGENIC X-RAY EMISSIONS SPECTROSCOPY**

Daniel G. McNeel (1,P); Matthew H. Carpenter (1); Mark P. Croce (1); Katherine A. Schreiber (1); Joel Ullom (2); Douglas Bennett (2); John A. B. Mates (2); Johnathon Gard (3); Jozsef Imrek (3); Daniel Becker (3); Abigail Wessels (3); Kelsey Morgan (3). (1) Los Alamos National Laboratory, (2) National Institute of Standards and Technology, Boulder, (3) University of Colorado, Boulder, (P) Presenting Author.

Ultra-high-resolution x-ray emission spectroscopy (XES) of actinide compounds can reveal chemical information from minor spectral features. This information may be able to identify the creation process or the source of the material, making this a potentially valuable tool to chemically analyze samples non-destructively. To be able to ascertain the chemical state of unknown compounds first requires understanding the ways that chemistry affects the observed spectra. We have studied 16 uranium containing chemical compounds using ultra-high-resolution XES over an energy range of 200 eV to 9 keV to determine which uranium M lines are sensitive to chemical speciation. The measurement campaign spanned different oxidation states, ligands, and contaminants in the form of standard soil samples mixed with pure compounds. The goal of these measurements is to develop a theoretical framework for analyzing unknown samples from our observations of these spectra. Ultra-high-resolution XES has been achieved using an array of 128 cryogenic Transition Edge Sensors (TESs), each having an energy resolution of approximately 6 eV FWHM at 3 keV. These measurements represent a database of known compounds that may be used to validate a theoretical framework for calculating the M line shifts in other compounds or actinides. We will discuss the chemically relevant features in the spectra and how these may be used in future analysis.

Log 523. RADIOCHEMICAL SEPARATION OF POLONIUM-210 FROM HIGH PURITY COPPER - METHODOLOGY AND DETECTION LIMITS

Mroz, T.(1,P); Brudecki, K.(2); Wojcik, M.(1); Zuzel, G.(1); (1) Jagiellonian University, Institute of Physics. (2) Institute of Physics Polish Academy of Sciences. (P) Presenting Author.

Construction of large scale, underground located detectors for detection of dark matter, neutrinos or neutrino-less double beta decay require very careful material selection to ensure lowest possible backgrounds. In case of direct dark matter detection one of the most important background sources are neutrons from (a,n) reactions, which generate signal indistinguishable from the interactions of dark matter particles. Usually shielding and support structures of these detectors are fabricated from industrial or electroformed high purity copper in which, microbecquerel activities of long lived nuclides like 238U and 232Th are measured by means of mass spectrometry, while 226Ra can be analyzed with low-backgorund gamma-ray spectrometers. Because production process of copper can cause disequilibrium in decay chains, it is necessary to determine also activity of 210Pb, what can be done by alpha-spectrometry counting of its decay product, 210Po. Polonium activity in the bulk material can be measured by deconvolution of spectra collected with large area alpha spectrometers, however, sensitivity of this method is insufficient in many situations, therefore it was necessary to develop a radiochemical procedure for separation of 210Po from copper. Proposed method and its optimisation in terms of sensitivity will be discussed. Separation of 210Po form bulk material by ion-exchange chromatography and counting with dedicated, low-background spectrometer usually allow us to achieve detection limits at level of around 10 mBq/kg, however by taking special care of factors like for example sample preparation or ion-exchange resin selection, it is possible to reduce blank count rate by a factor of about 2.

Log 524. THERMODYNAMIC AND TRANSPORT PROPERTIES OF MOLTEN PuCl₃-NaCl MIXTURES: ATOMISTIC MODELING

Manh-Thuong Nguyen (1,P); Roger Rousseau (1); Vassiliki-Alexandra Glezakou(1). (1) Pacific Northwest National Laboratory, Richland, WA 99354, USA. (P) Presenting Author.

Actinide molten salts are integral part of highly important nuclear applications like Gen-IV reactors. Due to inherent difficulties associated with experiment at high temperatures, the radioreactive nature of actinides, and complexity of multi-component systems, molecularly understanding their properties is challenging. Atomistic modeling, which in principles can provide an alternative approach, also have significant issues. Because of the complexity of electronic structure due to the 5f orbitals, it difficult to obtain information on properties that require extensive statistical sampling such as transport. To tackle this problem, we adopted a machine-learning approach to study PuCl3-NaCl mixtures. Machine-learning interatomic potentials, trained on density functional theory potential energy surfaces, allow us to obtain long molecular dynamics trajectories (ns) for large systems (~1000 atoms) at a considerably low computing cost, thereby efficiently gaining information about their bonding structures (e.g., bond distance, coordination environment), thermodynamics (specific heat), and dynamics (mass and heat transport). Comparison with available experimental data will be discussed.

Log 525. **ADDRESSING FUTURE CHALLENGES IN RADIOCHEMISTRY WITH HIGH PERFORMANCE COMPUTING**

Penchoff, D.A. (P) (1,2); Peterson, C.C (3), Valeev, E. (4); Benny, P. (5) Bosilca, G. (1); Harrison, R.J. (6). (1) Innovative Computing Laboratory, The University of Tennessee. (2) Department of Nuclear Engineering, The University of Tennessee. (3) Office of Advanced Research Computing, University of California Los Angeles. (4) Department of Chemistry, Virginia Polytechnic Institute. (5) Isotope Science and Engineering, Oak Ridge National Laboratory. (6) Institute for Advanced Computational Science, Stony Brook University. (P) Presenting Author.

Advances in High-Performance Computing (HPC) have accelerated discoveries in many areas of science. However, even with significant recent developments in supercomputing resources, the ability to model rare earth elements (REEs) and actinides needed in radiochemical applications requires solutions to an imbalanced hardware-software ecosystem. This demands multidisciplinary synergistic approaches to develop robust and efficient solutions. This presentation will highlight efforts at the intersection of scientific computing and applied computational modeling of REEs and actinides. A brief evolution of HPC, availability of artificial intelligence applications, and progress and challenges in modeling REEs and actinides as we enter the Exascale era of supercomputing will be presented. Applications relevant to separations of REEs and actinides will be discussed with focus on separating actinium-225 from lanthanum-140. Actinium-225 is a promising radiotherapeutic which often needs to be purified from accelerator production to remove lanthanum-140. Furthermore, the complexation of actinium-225 to be administered to patients must have a well defined coordination environment with inherent stability. A recent study evaluating binding preferences and effects in selective separations for purification of actinium-225 will be highlighted.

Log 526. UNCERTAINTY PROPAGATION USING CORRELATIONS IN GAMMA SPECTROMETRY AND THE EFFECT ON EFFICIENCY CALIBRATION AND ACTIVITY DETERMINATION

Persson, H. (1, P), Anderson, T. (1), Kirkpatrick J.M. (1), Phillips, K. (1) Mirion Technologies. (P) Presenting Author.

Accurate and defendable uncertainty propagation from input quantities to final radionuclide activities is important for assaying gamma-emitting radionuclides in many applications, including environmental studies and nuclear non-proliferation. In high precision gamma spectroscopy, the uncertainties are often dominated by the efficiency calibration, which is made by fitting or interpolating a set of measured or calculated efficiency reference points. If these data are correlated, it will increase the uncertainty when the fit is evaluated at a particular energy as well as the correlation between two efficiencies calculated at different energies. These correlations need to be properly accounted for when the efficiency calibration is used to calculate radionuclide activities and uncertainties, when more than one gamma ray energy is used. During this presentation we will review a method for including correlations for efficiency calibration, weighted mean activity calculation, and interference corrected activities for radionuclides with overlapping peaks. For nuclide activity interference correction we will also present how uncertainties in the gamma ray emission intensities are included in the calculation. We will show how various correlation assumptions for the efficiency calibration will affect the uncertainty of the radionuclide activity and how using multiple uncorrelated calibration sources can reduce the uncertainty of the radionuclide activities. The method will be applied to a complex gamma spectrum containing typical fission radionuclides.

Log 527. AN AUTOMATED METHOD FOR RESOLVING UNIDENTIFIED PEAKS IN GAMMA SPECTROMETRY

Persson, H. (1, P), Phillips, K. (1), Tischenbach, E (1) Mirion Technologies. (P) Presenting Author.

Gamma spectra are typically analyzed by locating peaks and calculating their area, perform background subtraction, applying full energy peak efficiency and performing a nuclide identification using a user adjustable nuclide library. In a complete gamma spectrometry analysis, every peak that has been found in a measured spectrum needs to be identified. It is common that the nuclide identification step does not identify all found peaks. In addition to full energy peaks from radionuclides in the sample, many spectra also include single and double escape peaks, random and coincidence sum peaks, lead x-ray peaks and an annihilation peak. Manually identifying all peaks in a complex gamma spectrum can be a time-consuming task. A new prototype algorithm has been developed that helps the user to resolve these unidentified peaks. The algorithm uses the count rates in the largest peaks to identify random sum peaks. To identify coincidence sum peaks, it uses the full energy peak efficiency calibration and known nuclide and atomic decay data. The Pb X-rays from a lead shield and the annihilation peak at 511 keV are labelled accordingly. Single and double escape peaks are labeled based on the energy difference from the full energy peak and finally it uses a master nuclide library to look for low intensity emissions from already identified radionuclides and identifies candidate radionuclides that was missed during the analysis. In this presentation I will discuss how it can be used to improve challenging spectral analysis.

Log 528. **ANALYSIS OF ACTIVATED ELEMENTS IN MODERN URBAN ENVIRONMENTS**

Phelps, J. (1,P); Baciak, J. (1); (1) The University of Florida. (P) Presenting Author.

Over the past several decades urban environments have transitioned from the traditional concrete jungle, to the modern spectacles of today; filled with eye-catching lights, colors and the attraction electronic devices provide. The addition of modern electronics into the structure of urban environment provides new methods for nuclear forensics by analysis of activated electronic debris following a nuclear detonation in these environments. Analysis of this post detonation debris can provide insight into characteristics of the detonated device. Utilizing a 100 kW Argonaut reactor available at The University of Florida, the activation characteristics of LED's and other display board electronics to neutron and gamma fluences associated with post detonation events were determined through use of neutron activation analysis. LED samples from a variety of manufactures in a range of conducting materials were irradiated in the University's reactor and the samples activated spectra were measured using HPGe detectors. Analysis of these spectra show that despite a variety of makes and manufacturers these LEDs all had some long lived isotopes in common with each other. These isotopes can help determine characteristics of the detonated nuclear device by giving information on the spectrum of the device through examination of the ratios of activated elements or from the slope of the activation of these elements as the distance from the detonation point increases.

Log 529. **DICER: A NEW INSTRUMENT FOR NUCLEAR DATA FOR NUCLEAR SECURITY**

Stamatopoulos, A. (1,P); Koehler, P. (1); A. Couture (1); B. DiGiovine(1); Matyskin, A. (2), Mocko, V(2); G. Rusev (2); Ullmann, J. (1); Vermeulen, C. (2). (1) Physics Division, Los Alamos National Laboratory, 87545, NM, USA. (2) Chemistry Division, Los Alamos National Laboratory, 87545, NM, USA. (P) Presenting Author.

Radiative neutron capture data targeted to nuclear security purposes are essential, however, challenging to acquire for short lived radionuclides. A technique that has been recently developed at the Los Alamos Neutron Science Center (LANSCE), can provide accurate data on several radionuclides relevant to nuclear criticality safety, radiochemical diagnostics, astrophysics, nuclear forensics and nuclear security, by measuring the transmission of neutrons through radioactive samples. To apply this technique a new instrument was developed and commissioned: the Device for Indirect Neutron Capture Experiments on Radionuclides (DICER). DICER has been in commissioning phase the last two years and already data relevant to nuclear security and radiochemical diagnostics were acquired on stable nuclei such as 191,193Ir and 147,149Sm. DICER performed its first measurement on a radioactive sample recently, by irradiating liquid 88Zr samples that were produced in collaboration with the Isotope Production Facility (IPF) at LANSCE. 88Zr was recently reported to have the second largest neutron capture cross section at the thermal point and DICER will be able to provide a justification as to what is the physics mechanism that causes that. DICER has currently about 50 radionuclides within its reach. Future plans include the upgrade of the instrument to enhance its capability to perform measurements on other radioactive samples such as 88Y and 171Tm. A description of the new apparatus, preliminary data on a few stable isotopes and 88Zr as well as future upgrades will be presented.

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g 530. **NEW HORIZONS IN MICRO PARTICLE FORENSICS: ACTINIDE IMAGING AND DETECTION OF Pu-238 AND Am-242M IN HOT-PARTICLES**

Bosco, H. (1); Hamann,L (1); Kneip,N (2); Raiwa,M (1), vanEerten,D(1); Weiss,M(1); Wendt,K(2); Walther,C (1,P) 1 Institute of Radioecology and Radiation Protection, Leibniz University Hannover. 2 Institute of Physics, Johannes Gutenberg-University Mainz. (P) Presenting Author.

Micrometer-sized pollutant particles are of highest concern in environmental and life sciences, cosmochemistry and forensics. From their composition, detailed information on origin and potential risks to human health or environment are obtained. We combine secondary-ion mass spectrometry with resonant laser ionization to selectively image elemental and isotopic composition of individual particles at below 100 nm spatial resolution. Using newly developed laser ionization shemes, isobaric interferences particularly U-238/Pu-238 and Pu-241/Am-241 are suppressed by ca. five orders of magnitude. In contrast to most mass spectrometric techniques, only negligible mass is consumed leaving the particle intact for further studies. Single particles are identified by combining flotation and SEM, isolated by use of a micromanipulator and glued to tungsten needles, avoiding any chemical pretreatment. Identification of actinide elements and their isotopes on Chernobyl hot-particles, including 242mAm at ultra-trace levels, proved the performance. Beyond that, the technique is applicable to almost all elements and opens up new scientific applications.

Log 531. **AUGMENTED COOPERATION IN EDUCATION AND TRAINING IN NUCLEAR AND RADIOCHEMISTRY**

Walther, C. (1,P). (1) Leibniz University Hannover /Institute of Radioecology and Radiation Protection, 30419 Hannover, Germany. (P) Presenting Author.

In order to maintain European nuclear operations, expertise in nuclear and radiochemistry (NRC) is of strategic relevance for amongst others the energy sector, medical treatment and diagnosis, material sciences and dating. The A-CINCH project is the latest of four CINCH-based projects aiming on cooperation in education in nuclear chemistry and radiochemistry. In the first three projects, minimum requirements for bachelor, master and postgraduate programs to achieve approved NRC curricula were defined, a number of theoretical and practical courses were developed using hands-on and e-learning approaches and platforms. MEET-CINCH counteracted the massive lack of NRC expertise by building teaching packages for high schools and a MOOC on NRC for the general public to attract young persons to the NRC field. New E&T approaches based on remote teaching, remote controlled RoboLab experiments, interactive screen experiments and the flipped classroom concept were developed and implemented. ECVET course modules are offered in an e-Shop adapted to the needs of end-users. A-CINCH makes use of state of the art virtual and augmented reality tools. It implements a innovative Virtual Laboratory. The wide mix of e-learning and presence teaching in nuclear and radiochemistry education aims to increase the number of students and trainees in the field. To address the new target groups directly and efficiently attract the attention, new didactical tools, suitable for today's youth, are used. These newly developed courses and all previous CINCH results are accessible via the CINCH Hub platform.

Log 532. **PRELIMINARY RESULTS OF THE UNIVERSITY OF TEXAS NUCLEAR ENGINEERING TEACHING LABORATORY PROMPT GAMMA ACTIVATION ANALYSIS SYSTEM REDESIGN**

Beauvais, Z.S. (1,P); Charlton, W.S. (1); Andrews, M.J. (1); Payne, W.M. (1). (1) The University of Texas at Austin. (P) Presenting Author.

The Nuclear Engineering Teaching Laboratory (NETL) at the University of Texas is equipped with a 1.1 MW TRIGA reactor with an installed cold neutron source. The cold neutron source has typically been used to perform cold neutron prompt gamma activation analysis (PGAA). The PGAA system at NETL was recently redesigned and reconstructed to implement a Compton suppression system, a sample chamber with seamless gamma ray shielding, and enhanced neutron shielding. The system was redesigned to fully eliminate hydrogenous shielding and structural materials with an aim toward improving the overall hydrogen measurement sensitivity of the system. The redesign was accomplished through extensive Monte Carlo radiation transport simulation to optimize detector dimensions, detector configuration, shielding material selection and shielding configuration. Performance baseline measurements were completed using a NIST SRM 2454 standard with hydrogen content of 0.0211 wt. % and comparison to background measurements. These initial measurements show significant improvements in background and continuum reduction compared to the previously implemented system design and improved sensitivity for hydrogen concentration measurement. This paper presents the system design and preliminary performance results.

Log 533. **PLUTONIUM BEHAVIOR IN IRON-RICH SILICATE IMMISCIBILITY REGIONS OF NUCLEAR FALLOUT GLASS**

Genda, T.P. (1,2, P); Matzel, J. E. (2); Dai, Z. R. (2); Balboni, K. (2); Knight, K. B. (1); Hoseman, P. (1). (1) The University of California, Berkeley. (2) Lawrence Livermore National Laboratory. (P) Presenting Author.

Particulate fallout formed in nuclear explosion fireballs are the result of an array of complex high temperature processes. Recently discovered two-phase microstructure "amoeboids" with a range of morphologies are preserved within the rims of some aerodynamic fallout glass from an historic U.S. nuclear test, and have the potential to constrain fireball temperature and oxygen behavior. Their two-phase compositions and textural evidence suggest amoeboids result from liquid immiscibility processes, but their exact formation mechanism is unknown. Two possible formation mechanisms include: (1) an agglomeration and emulsification of two distinct liquids of immiscible composition, and (2) liquid-liquid phase separation (LLPS) from a homogenous condensate. We have investigated the LLPS hypothesis using a qualitative phase field method (PFM) model of liquid phase separation and demonstrate that amoeboid morphologies are consistent with LLPS processes. We estimated plutonium concentrations in the glass using nano-scale secondary ionization mass spectrometry (nanoSIMS), including the first ever Pu measurements within silicate liquid immiscibility textures. Pu concentrations range from 1-10 ppm and a positive correlation exists between Pu, Fe, and Ca in regions of the glass without immiscible textures. However, the Pu/Fe correlation is reversed in regions with immiscible textures. These results are inconsistent with expected theoretical two-liquid partitioning coefficients, suggesting complex non-equilibrium melt conditions and/or evidence for the emulsification hypothesis of amoeboid formation. This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52- 07NA27344 and was supported by the LLNL-LDRD Program under Project No. 20-SI-006.

Log 534. MICROMOLAR LEVEL SENSING AND QUANTITATIVE RECOVERY OF LANTHANIDES FROM ALKALINE SOLUTIONS WITH SIMPLE O-SULFONAMIDOPHENOL DERIVATIVES

Adedoyin, O.W. (1); Gonzalez, C.(1); Morozov, A.N.(1); Mebel, A.M.(1); Chakraborty, I.(1); Raptis, R.G.(1); Kavallieratos, K. (1,P) (1) Florida International University. (P) Presenting Author.

Sensing of f-elements; actinides (An), but also lanthanides (Ln), which are often fission products, is a challenge both within the nuclear fuel cycle, but also for applications to radiopharamaceuticals and nuclear forensics. As part of our effort to improve alkaline tank waste processing at DOE's Savannah River Site, we have introduced lipophilic sulfonamide ligands that extract Ln(III) from highly alkaline solutions with recoveries >90% in chlorinated solvents and >50% in dodecane/octanol after a single loading-stripping cycle. Herein, we are expanding into Ln(III) optical and fluorescence sensing with two o-sulfonamidophenols, bearing a nitrophenol group (1) and a dansyl group (2) for optical and fluorescence sensing, respectively. Ligand 1 gave µM level selective sensing for Lu(III) in alkaline media, and in the presence of high concentration backgrounds of HLW metals, including Na(I), K(I), Cs(I), and Sr(II). Titrations of 1 with Lu(III) in the presence of 2.5 eq. of diisopropylethylamine in MeCN gave 1:1 complexation, with clear isosbestic points, a visible color change, and binding in the µM range. A linear response range of 0–4.93 µM for Lu(III) with a limit of detection as low as 0.207 µM was determined in high metal backgrounds (Na/Lu>5000). DFT calculations corroborated the spectral changes, while X-ray showed a unique Lu(III) trimeric cluster in the solid state. For 2, fluorescence titrations with Ln(III) in alkaline solutions gave 3-fold turn-on fluorescence. Sm(III) recovery of 92.2(±13.5)% from pH 13.0 into DCM was observed (single loading/stripping cycle).

Log 536. IDENTIFYING AND QUANTIFYING FORENSIC TAGGANTS IN URANIUM METAL SAMPLES

Bartlett, J.H. (1); Boland, K.S. (1); Chamberlin, R.M. (1,P); Emberley, W.C. (1); Erickson, K.A. (1); Kral, G.A. (1); Rearick, M.S. (1) Los Alamos National Laboratory. (P) Presenting Author

Intentional nuclear forensics approaches are designed to enable practitioners to rapidly and confidently assess the provenance of nuclear materials found outside of regulatory control. With careful taggant design, nuclear forensics laboratories will be able to "read" an encoded signature in various materials in a minimally destructive way, supporting rapid incident response and augmenting investigations. Beyond the analytical chemistry and materials characterization methods that would be employed in nuclear forensics investigations, fuel fabrication and safeguards verification activities will also drive the need for rapid, reliable methods of detecting and identifying the presence of taggants in a fuel. This is especially true in early stages of adoption, when a mixture of tagged and untagged fuels would be in circulation. For elemental quantification, routine methods such as inductively-coupled plasma spectroscopies are sufficient to quantify taggants in a metal or ceramic sample, although the elevated concentrations of novel elements may require special procedures. Non-destructive screening methods that are sensitive to the identity and spatial distribution of taggants in a fuel matrix would also be of high value to the forensics community, allowing sample flow to be optimized and early provenance information to be returned. Results of initial laboratory characterization studies of uranium metals and oxides (or their surrogates) that have been tagged with elemental mixtures will be presented.

Log 537. **KEY STRATEGIES AND SCIENTIFIC QUESTIONS FOR AN INTENTIONAL FORENSICS APPROACH TO NUCLEAR MATERIAL PROVENANCE ASSESSMENT**

Chamberlin, R.M. (1,P); Marks, N.E.(2); Shields, A.E.(3); Wellons, M.S.(4) (1) Los Alamos National Laboratory (2) Lawrence Livermore National Laboratory (3) Oak Ridge National Laboratory (4) Savannah River National Laboratory. (P) Presenting Author

Intentional Forensics is the deliberate introduction of benign and persistent material signatures into nuclear fuel fabrication and processing. Its purpose is to reduce the lag time between the recovery of a material outside of regulatory control and the identification of its original provenance. An integrated, multi-laboratory project has been initiated to develop a scientific and technical basis that would enable adoption of this forward-looking approach to nuclear forensics. Key research questions are: What are the best strategies for intentionally tagging various nuclear materials, and where in the fuel cycle should they be introduced? How can we design taggants that provide the desired nuclear forensics outcomes while also remaining benign under reactor irradiation? How can we rapidly measure and confidently assess the information encoded in tagged nuclear material, even after it has been processed? A preliminary definition of taggants for this project is the use of mixtures of heteroelements, which ideally contain significant perturbations from their naturally occurring isotopic ratios, incorporated into the bulk or on the surface of metallic or ceramic nuclear fuels. This presentation will give an overview of the challenges in developing a taggant selection scheme that integrates probative value, manufacturability, reactor safety, and persistence in the fuel cycle. Initial work on potential solutions will be discussed in more depth elsewhere in this session.

Log 538. **NEUTRON RADIOGRAPHY AT LANSCE: INTERROGATION AND CHARACTERIZATION OF MATERIALS FOR NEXT GENERATION NUCLEAR REACTOR DEGSINGS**

Long, A.M. (1); Balke, T. (1); Jackson, J.M. (1); Luther, E. (1); Mehta, V. (1); Monnreal, M. (1); Parker, S.S. (1) ; Shivprasad, A.P. (1); Trellue, H. (1); Tremsin, A. (2); Vogel, S.C. (1). (1) Los Alamos National Laboratory; (2) University of California, Berkeley. (P) Presenting Author.

Neutron radiography is an ideal probe for integrating and characterizing potential nuclear fuel and moderator materials in next generation nuclear reactor designs. Due to the nature of interaction with the nucleus, neutrons have complex attenuation functions that result in contrast mechanisms and material penetrabilities that are not only well-suited investigating materials that contain high-Z isotopes (such as actinides in nuclear fuels), but also characterizing hydrogen distributions in various hydride materials considered in future moderator designs. Additionally, with a high intensity short-pulsed neutron source, Energy-Resolved Neutron Imaging can be utilized to map out specific isotopes based on neutron resonance absorptions with in a given material. Presented will be efforts in developing these capabilities on FP5, along with initial results from notable measurements ranging from thermophysical property characterization of uranium based molten salts, to H-concentration mapping in hydrides, to isotope mapping in fresh and irradiated fuel samples for post-irradiation examination (PIE).

Log 539. **CHEMICAL AND STRUCTURAL CHARACTERIZATION OF PARTICULATE FALLOUT ISOLATED FROM AIR FILTERS**

Balboni, E., Dai Z., Matzel J., Kerlin M., Knight, K.

Nuclear particulate fallout is the radioactive byproduct of a nuclear event formed by the mixture of proximate environmental materials with vaporized bomb debris. The debris can be transported into the atmosphere during cloud rise, raining out locally and globally constituting a radiation hazard. Questions remain on how entrainment of environmental material in the fireball affects fallout formation processes and radionuclide incorporation during cooling. Here we analyzed a selection of archived air filters collected from aircrafts in the aftermath of ground interacting nuclear tests. Particles were isolated and analyzed using scanning electron microscopy, transmission electron microscopy and NanoSIMS (Secondary Ionization Mass Spectrometry) to determine the particulates chemistry, structure, and particle size distribution. Our results demonstrate that the particles isolated from the filters are mainly composed of Fe and Si oxides, are characterized by variable minor/trace element composition, and display complex internal structures. The characterization of these fallout particulate samples can provide information on particulate nucleation and particle growth from the vapor phase and help determine how radionuclides are incorporated in macro-sized debris such as aerodynamic fallout. This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52- 07NA27344 and was supported by the LLNL-LDRD Program under Project No. 20-SI-006.

Log 540. SOLID-PHASE ISOTOPE HARVESTING OF ^{88}Zr

Jake Bence (P,1,2), Samridhi Satija (3), Katharina Domnanich (3), John Despotopulos (4), Kelly Kmak (4), Paige Abel (3), Hannah Clause (3), Scott Essenmacher (3), Chloe Kleinfeldt (3), Wesley Walker (3), Colton Kalman (3), Chirag Vyas (3,5), Nicholas Scielzo (4), Tashi Parsons-Davis (4), Gregory Severin (3,5), Jennifer Shusterman (1,2,4). (1) Hunter College of the City University of New York (2) Graduate Center of the City University of New York (3) Department of Chemistry, Michigan State University (4) Lawrence Livermore National Laboratory (5) National Superconducting Cyclotron Laboratory. (P) Presenting Author.

During routine operation of the Facility for Rare Isotope Beams (FRIB), high-purity radioactive ion beams will be produced for nuclear science research experiments. During the beam purification process, byproduct radionuclides will accumulate along the beamline. There is interest in collecting, purifying, and using these radionuclides in a process called isotope harvesting. Preliminary isotope harvesting efforts have been ongoing at the National Superconducting Cyclotron Laboratory (NSCL), such as the aqueous collection of 24Na, 47Ca/47Sc, 48V, 62Zn, 67Cu, and 88Zr. In this work, the viability of isotope harvesting using solid collection of 88Zr, an isotope relevant to stockpile stewardship, was examined by performing a proof-of-concept experiment at the NSCL where 88Zr beam was stopped in a series of collectors comprised of Al, Cu, W, and Au. 88Zr was radiochemically recovered from each of these collectors with yields exceeding 80% and decontamination factors on the order of 105 by using a combination of solvent extraction and ion-exchange chromatography. Results of the harvesting experiment and the radiochemical recovery of 88Zr and its decay product 88Y from Cu and Al collectors will be discussed. For elements of interest that readily hydrolyze in near-neutral pH aqueous conditions, such as Zr, harvesting through solid-phase collection has been shown to result in higher recovery yields compared to aqueous harvesting.

Log 541. OPTICALLY-READABLE ALD-DEPOSITED SURFACE MARKINGS FOR INTENTIONAL FORENSICS

Condon, N.J. (1, P); Laudadio, E.D. (1); Jamison, L. (1); Mouche, P.A. (1); Elam, J.W. (1); Mane, A.U. (1); Yacout, A.M. (1). (1) Argonne National Laboratory. (P) Presenting Author.

The incorporation of taggants directly into nuclear fuels has many advantages for intentional forensics, but a challenge for this method is the difficulty in "reading" the tag without destructive analysis at a remote site. In this work, we are addressing this difficulty by preparing optically-readable surface markings for onsite identification of nuclear materials using conventional visible, infrared, or UV cameras. We deposit surface coatings using atomic layer deposition (ALD) which allows for atomic level control of material thicknesses. The thickness, smoothness, index of refraction, and other characteristics of these films will determine how they reflect light, so tuning these parameters can be used to control how they appear using various imaging techniques, leading to an optical signature. In addition, 2-D patterning of these films can be used to add additional, easily-read information to the tag. The result will be robust, unique tags that can be easily read out using inexpensive instrumentation in the field.

Log 542. DIGITAL NEUTRON IMAGING OF TRANSIENT IRRADIATED NUCLEAR FUELS

Craft, A.E. (1,P); Schulthess, J.L. (1); Papaioannou, G.C. (1); Chuirazzi, W.C. (1); Kane, J.J. (1); Cordes, N.L. (1). (1) Idaho National Laboratory. (P) Presenting Author.

Neutron radiography and tomography are some of the first methods used to examine nuclear fuels after irradiation. Nondestructive neutron imaging techniques provide valuable information about the internal geometric condition of irradiated nuclear fuels that is used to plan subsequent destructive examinations and sometimes provides early indication of unanticipated features or material behaviors. Traditionally, transfer method neutron radiography using film has been the only available technique for nondestructively visualizing highly radioactive samples. Recently, however, modern digital neutron imaging capabilities have been developed to examine irradiated samples. This paper presents results of digital neutron tomography of transient-irradiated nuclear fuels using a modern digital camera-scintillator type imaging system.

Log 543. Multi-detector Neutron Depth Profiling for Probing Li distribution in Li-Ion Batteries

Safranek, A.W. (1); Wood D. (2); Co A. (2); Kauffman A. (3); Cao L.R. (1)(3); Downing, G. (4)(P); (1) Department of Nuclear Engineering, The Ohio State University. (2) Department of Chemistry, The Ohio State University. (3) Nuclear Reactor Laboratory, The Ohio State University. (4) AwareAbility Technologies, LLC. (P) Presenting Author.

Small research reactors play an important role in education and they assist university researchers working with industry toward a cleaner environment. A prime example is the application of nuclear analytical techniques in the study of lithium-ion batteries (LIBs), an energy carrier widely in use today. The Ohio State University Nuclear Reactor Laboratory (NRL) operates a 500 kW research reactor with in-core and neutron beam research facilities. One of the instruments, Neutron Depth Profiling (NDP), was recently utilized to demonstrate that the reactor facility can fully compete with National Laboratory Reactors investigating lithium transport and its fixation in (LIBs) structures while working in partnership with academic and industrial labs to enhance the performance of these devices. This NDP facility is unique in the world having 8 detectors that simultaneously view the sample under neutron irradiation. And, although the reactor operates with a thermal power of only 500 KW, by utilizing those detectors in equivalent geometric view of the sample, the NDP instrument functions at the equivalent to one at a 4 MW reactor. No other NDP facility in world utilizes this great of a solid angle. In this recent study, using a data acquisition time of an hour or less, NDP was used to examine LIB solid-state sample areas of 10 mm^2 or less to nondestructively acquire Li depth profile spectra and compare the near-surface loss of lithium of as-manufactured battery components to those components after charged-discharged cycled unto failure.

Log 544. USING LASER ABLATION TO INFORM NUCLEAR FIREBALL CHEMISTRY: HOW OXYGEN AVAILABILITY AFFECTS URANIUM SPECIATION

Burton, M.A. (1,P); Auner, A.W.(1); Crowhurst, J.C.(1); Boone, P.S.(1); Finney, L.A.(2); Weisz, D.G.(1); Koroglu, B.(1); Jovanovic, I.(2); Radousky, H.B.(1); Knight, K.B.(1). (1) Lawrence Livermore National Lab (2) University of Michigan. (P) Presenting Author.

Following a nuclear event, fallout particulates are formed from and affected by gas-phase chemical reactions and micro-physical processes involving hot nuclear material and the environment. Current models, based on empirical data from historic nuclear tests, aim to predict the chemical speciation of a nuclear explosion but reflect testing performed across a limited set of environments. To more broadly predict the behavior of actinides, such as uranium, and the chemical behaviors that dictate their incorporation into debris, new experimental inputs are needed to determine key system sensitivities such as local environment conditions. Benchtop laser experiments were performed to provide new insights on the interaction of uranium and oxygen as hot metal rapidly cools (from several thousand Kelvins). Results, collected using a novel approach of particle collection and in-situ analysis, show the nature of formed particulates is sensitive to the available oxygen, varying from a highly oxidized amorphous form (am-UOx where $3 \leq x \leq 4$) to crystalline UO_2.

Log 545. BULK URANIUM METAL TAGGING FOR INTENTIONAL FORENSICS

Hackenberg, R.E. (1) ; Luitjohan, K.E.(1, P); Imhoff, S.D.(1); O'Brien, L.B.(1). (1) Los Alamos National Laboratory. (P) Presenting Author.

One strategy to provide a unique taggant signature in bulk metal uranium includes adding natural isotopes of various elements during casting, one of the first processing steps. Each element on the periodic table was screened for practical suitability through three filters that reduced the possible taggants to 43 elements. Of those 43, 16 were selected. In this study, those 16 potential taggant elements were probed for detectability and behavior in three distinct stages of experimentation. The elemental additions were first alloyed with uranium in unique groupings of four species to probe initial detectability. Further mixing and dilution castings were performed to observe both species interaction as well as detectability under more dilute conditions. Finally, a reprocessing study was used to determine the persistence of the alloying additions after repeated melting and casting. Microscopy and chemical assessment techniques were utilized to track the effects of taggant species. Chemical taggant detection in all three stages of experimentation was generally good while microscopy revealed the second phases largely, but not always, contained the expected tagging elements.

Log 546. DEVELOPMENT OF A GOLD NANOROD RADIOTRACER

Dos Santos, A.K.G.(1); Brandao, L.E. (2); Lima, I (1,P).1). (1) Federal University of Rio de Janeiro. (2) Nuclear Engineering Institute. (P) Presenting Author.

In the highly competitive environment of the petrochemical industry, efficiency has become an important condition in the production cost of petroleum derivatives. Radiotracer methodology is one of the most used technical tools for the evaluation of different units in an oil prospecting/processing plant /gas. For a radiotracer to be considered ideal for evaluating these units, prior knowledge of their physicochemical characteristics is necessary. The purpose of this work is the production of a new gold nanorod radiotracer to be applied in flow measurement in pipelines. The production of this radiotracer begins with the dissolution of metallic gold in aqua regia to form chloroauric acid. From the acid produced, with the addition of a strong reducing agent we produce the spherical gold nanoparticles and in the next step with the aid of growth drivers we will finally produce the gold nanorods. To monitor the sizes of the formed nanoparticles we will use UV-visible spectrophotometry analysis and to evaluate the shape of the produced nanorods we will use transmission electron microscopy. We are currently in the stage of morphological classification of the gold nanoparticle so that we can carry out a comparative study of the qualities of the nanosphere and the gold nanorod as a radiotracer for flow measurement in pipelines.

Log 547. EVALUATION OF A DEPLOYABLE HPGE DETECTION SYSTEM FOR USE IN HIGH ACTIVITY URANIUM OR PLUTONIUM PROCESS APPLICATIONS

Sullivan, D. F. (1,P); Ilie, G. (1); Ralet, D. (2); (1) Mirion Technologies Meriden. (2) Mirion Technologies Lingolsheim. (P) Presenting Author.

The capacity to perform accurate gamma spectroscopic measurements of Special Nuclear Materials (SNM) at any stage in the uranium fuel cycle requires a system that maintains a delicate balance of detector performance. Superior resolution is required so one may determine the isotopic vector of the target sample. Meanwhile the system's dynamic range of count rate must match the source emission activity such that an accurate quantification can be made of any radionuclides present in the sample. Typically, the application will dictate which performance characteristics are most important but, in some cases, both good resolution, and high throughput are required. One additional resource recently developed at Mirion Technologies is a deployable high purity germanium (HPGe) gamma spectrometer, the MicroGe. In this work, the system was used to measure both uranium and plutonium standards to evaluated how its performance compared to portable alternatives. Additionally, the maximum throughput was optimized and characterized to evaluate its application to the numerous high count rate environments. Finally, a detection efficiency characterization was made of the detector. A series of point source measurements was used to create a benchmarked simulation of the detector. An ISOCS™ mathematic efficiency characterization was created and tested.

Log 548. GAINING INSIGHT AND CERTAINTY ON UNCERTAINTY WITH THE ISOCS UNCERTAINTY ESTIMATOR TOOL

Ilie, G. (1); Sullivan, D. F. (1,P); Persson, H. (1); (1) Mirion Technologies Meriden. (P) Presenting Author.

The Mirion Technologies In-situ Object Counting System (ISOCS(TM)) and Laboratory Sourceless Calibration Software (LabSOCS(TM)) are used to calculate efficiencies for gamma spectroscopy detectors used for in situ measurements and laboratory geometries respectively. One component of these packages, the ISOCS Uncertainty Estimator (IUE), employs a probabilistic estimation methodology to evaluate the impact that each variable, in each measurement geometry, will have on the detection efficiency. IUE can also be used in a sensitivity mode to evaluate which aspects are most important towards further reducing the uncertainty in the efficiency. In this work, IUE will be applied to geometries common in a laboratory to illustrate the insights that can be gained from the uncertainty analysis. These will be compared to a large collection of measurements of the same laboratory samples containing a multi-nuclide, multi-energy line source matrix. By fully understanding how each component of the measurement geometry impacts not only how robust the measurement uncertainty is known but will equip the user to make decisions on how best to plan, perform and evaluate their gamma spectroscopy measurements with improved productivity, accuracy, and confidence.

Log 549. THERMODYNAMIC MODELING TO FACILITATE SAFE HANDLING OF CESIUM CHLORIDE SEALED SOURCES

Weber, C.F. (1,P); Abrecht, D.G. (1). (1) Oak Ridge National Laboratory. (P) Presenting Author.

For more than 50 years, radioactive 137Cs has been a major source material for radioactive sealed sources, usually constructed as CsCl salt loaded into double-walled stainless-steel capsules. A complication develops as 137Cs decays to 137Ba, since this process creates a strongly reducing environment inside the capsule. A potential hazard exists if the capsule is breached and air ingress induces rapid exothermic oxidation, and this mechanism is suspected as responsible for the well-known contamination incident at the Harbor View facility in Washington state. The current study has performed numerous thermodynamic evaluations to assess the internal state of capsules after several decades of decay, and to describe the potential oxidation if the capsule contents were to suddenly be exposed to air. Results suggest that reduction of impurities such as Cu, Fe, Pb, and Cr to metal will occur. If these impurities are lacking, it is possible that even Ba metal will form. In most cases, rapid oxidation can occur and the exothermic reactions are sufficient to vaporize portions of the contents, which would include remaining 137Cs.

Log 550. **THERMOPHYSICAL PROPERTIES OF LIQUID CHLORIDES FROM 600 – 1600 K**

Stephen Scott Parker (1,P), A. Long (1), C. Lhermitte (2), S. Vogel (1), M. Monreal (2), J.M. Jackson (1) (1) Los Alamos National Laboratory: Materials Science and Technology Division (2) Los Alamos National Laboratory: Chemistry Division. (P) Presenting Author.

Melt point, enthalpy of fusion, heat capacity, and volumetric expansion of single- and multi-component liquid chlorides {NaCl, KCl, LiCl, MgCl2, CaCl2, UCl3} were measured experimentally. These properties and materials are relevant in applications such as heat transfer, liquid nuclear fuel, and pyrochemical processing. A novel method for density measurement by neutron radiography was shown to produce high-quality data, consistent with reference literature where available, and allowed measurement of some materials for the first time. This method is especially useful in the characterization of sealed sample crucibles, given the ability of neutrons to penetrate the containment and surrounding furnace material. Measurement of the heat capacity of these ionic liquids was achieved by differential scanning calorimetry, and the results of this study are a first for most compositions examined. All results are presented within the context of a comprehensive review of the available published data. The purpose of this review is to integrate measurements of the thermophysical properties of liquid chlorides into empirical descriptions of the relationships between composition, temperature, and thermophysical properties. Models for the prediction of the density and heat capacity of mixtures of liquid chlorides are proposed and demonstrated within a case study of the {NaCl + x mol% UCl3} system

Log 551. **PROGRESS OF NEUTRON ACTIVATION ANALYSIS IN KOREA**

Sun, Gwang-Min;Korea Atomic Energy Research Institute, HANARO Utilization Division, Presenting Author.

Neutron activation analysis activities in Korea are being conducted at the HANARO research reactor of the Korea Atomic Energy Research Institute. Therefore, it is inevitably affected greatly by the operation status of HANARO research reactor, which has only been operated intermittently since 2014 in accordance with the earthquake reinforcement work and stricter regulations in the aftermath of the 2011 East Japan Earthquake. Fortunately, it is gradually stabilizing with the start of operation for three cycles (three months) in the second half of 2021. It is true that neutron activation analysis has significantly reduced its usefulness due to the limitation of its accessibility and the development of other instrumental analysis methods. This situation not only raises questions about the sustainability of neutron activation analysis in any country, but also raises a problem in fostering a new generation. It is not easy for students to be interested in neutron activation analysis, and it is not easy to make a decision to conduct research or engage in this field by taking the hassle out of it. The situation is even worse in Korea, where the operation of the research reactor has been problematic for nearly eight years. Existing research groups in Korea are getting older and older, and students have lost the opportunity to use them and discover their usefulness during their degree. How to show them new possibilities will be the most important for the sustainability and development of neutron activation analysis in Korea in the future. Currently, the fields most effective in Korea are studies of cultural heritages, geology, and development of standard materials. The Korea Atomic Energy Research Institute is strengthening cooperation with the Korea institute of Geoscience and mineral

resources and the associated department of universities. In the field of standard material development, cooperation with the Korea Institute of Standards and Science is being carried out. In this presentation, the latest trends and trends in neutron activation analysis in Korea are presented along with examples. In addition, a future vision in Korea is presented.

Log 552. UPDATED CUMULATIVE FISSION YIELDS FROM 14 MeV NEUTRON FISSION ON A PLUTONIUM-239 TARGET

Uhnak, N.E (P); Haney, M. M.; Pierson, B.; Greenwood, L.; Friese, J.; Metz, L. Pacific Northwest National Laboratory. (P) Presenting Author.

Nuclear data provides the foundation for the computational modeling of nuclear systems, whether that be for nuclear forensics, nuclear power, or medical isotope production. The bulk of the nuclear data, particularly those from 14 MeV fission, are based on data collected from the National Laboratory system in the 60s through the 80's. These measurements relied on the state of the science at the time, however modern technologies and analysis methods have greatly improved the analysis capabilities for radioanalytical measurements.   In this work we investigated the irradiation of a 0.2301 g 239Pu target using a 14 MeV D-T generator, analyzing the fission and activation products using standard gamma ray spectroscopy. Analysis of the target was conducted over a large time scale ranging from 30 minutes to greater than 100 days after the end of bombardment. From this data we determined the cumulative fission yields of many fission products, improving uncertainties on many, as well detection of several activation products such as 240Am.

Log 553. DEVELOPMENT OF NOBLE METAL SELECTIVE EXTRACTION RESINS FOR USE IN NUCLEAR FORENSICS APPLICATIONS

Uhnak, N. E.(P); Risenhuber, M.; Lawler, B.; Arnold, E.; Beck, C.; Seiner, B. Pacific Northwest National Laboratory. (P) Presenting Author.

Extraction chromatography represents one of the best tools for the separation of metal analytes, particularly at small scales like those used in nuclear forensics or medical isotopic separations. Though there are many commercial extraction resins available, many of these resins have limitations that preclude their implementation due to their chemical applicability and lack of flexibility specifically for sequential separation schemes. We have synthesized a series of extraction resins, based on first principles, aiming at the selective separation of select noble metals (Ag, Au, Ir, Pd, Pt, Ru) from mixed activation and fission product samples. These resins have been characterized, their extraction capabilities determined for a significant number of metals in both HCl and HNO3 matrices and have been used in separations for complex mixed analyte solutions. The results from these investigations will be discussed focusing on the noble metal selectivity in comparison to commercial resins.

Log 554. **ACTIVATION ANALYSIS OF PRECAST CONCRETE IRRADIATED DURING PROTON FLASH-RADIOTHERAPY**

Sriharsha Mangu, Henry Spitz (P) , Michael Lamba, SE Glover. University of Cincinnati. (P) Presenting Author.

Proton and concomitant neutron activation products generated in a concrete core sample after exposure to a Flash radiotherapy proton beam were measured to determine whether the residual activation products would present a potential source of occupational radiation exposure during operation and maintenance of the facility. A 1 cm diameter, 24 cm long concrete cylinder was cut into 5 slabs along its length and placed at the isocenter of a proton accelerator. Slabs of high-density polyethylene 10-cm thick were placed above and below the concrete to simulate a clinical beam passing through a patient.   An absorbed dose of 8 Gy was delivered to a tissue-equivalent depth of 5 cm in a 7 x 7 cm field by 250 MeV protons in 700 ms. High resolution gamma spectrometry measured activation products with increasing depth in the concrete after the exposure to the proton beam. A second concrete core sample containing pairs of gold wires inserted into the first three slabs was irradiated to evaluate thermal neutron abundance. Long-lived radioisotopes observed in the first four concrete slabs were annihilation radiation (511 keV), 7Be (477 keV), 22Na (1275 keV), and 52Mn (744 and 935 keV). Approximately 65% of the neutron flux was due to thermal neutrons.   Simulations of proton interactions using MCNP6 demonstrated the significance of (p,n) interactions in generating products of neutron activations. This study suggests that although the FLASH proton radiotherapy beam can generate long-lived proton and neutron activation products in concrete structural materials, the intensity of that radiation is not likely a significant source of occupational radiation exposure.

Log 555. **PRODUCTION OF ACTIVATION SPECIES AT NCERC AND INVESTIGATION OF THE ^{9}Be(n,γ)^{10}Be INTEGRAL CROSS SECTION**

Goodell, J.J. (1,P); Church, J.A.(1); Keith, C.C.(2); Bredeweg, T.A.(2); Pierson, B.(3); Zhao, P.(1); Harward, N.K.(1); Bandong, B.B.(1). (1) Lawrence Livermore National Laboratory. (2) Los Alamos National Laboratory. (3) Pacific Northwest National Laboratory. (P) Presenting Author.

The National Criticality Experiments Research Center (NCERC) provides a unique capability to irradiate materials in different neutron spectral environments not available at typical nuclear science user facilities. One such capability is the Flattop critical assembly, which has a natural uranium reflector and interchangeable enriched uranium/plutonium cores. The characteristics of Flattop, including a transverse sample bore, allow for different neutron spectra within the same critical assembly. In this work, we investigate the production of activation species from various structural materials irradiated in the Flattop critical assembly with fission energy neutrons. Stainless steel (and its elemental components), beryllium, and gold were irradiated at multiple positions across the assembly using both cores in separate measurement campaigns. We present progress on efforts to model the Flattop critical assembly, validate and assess the accuracy of the model, and use the derived neutron spectra to calculate the 9Be(n,gamma)10Be integral cross section. Modelling efforts were conducted using MCNP radiation transport code, while validation and assessment efforts employed the STAYSL-PNNL neutron spectrum adjustment code and the FISPACT-II inventory code. Integral cross section values were determined based on the total neutron fluence and the 10Be/9Be ratio as measured by accelerator mass spectrometry. LLNL-ABS-830208. This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344.

Log 556. ADDING CAPPING LAYERS TO NDP SAMPLES BROADENS AND ATTENUATES SPECTRA

Weaver, J.L. (1); Downing, R.G. (1). (1) Material Measurement Laboratory, National Institute of Standards and Technology.

Protective capping layers are used in neutron depth profiling (NDP) experiments for ambient and/or vacuum sensitive samples. The layer(s), often thin polyimide or metal films, can prevent a sample from changing while being loaded into and/or run in a NDP chamber. They can also function as a filter, minimizing overlap between NDP measured reaction product profiles. In this study the consequences of placing variable thicknesses of polyimide films on Li containing NDP samples are investigated. Li is a component of several industrially important materials (batteries, glasses, and ceramics), and can be measured by NDP through the $6Li(n,4He)3H$ neutron (n) absorption reaction. The addition of the polymer film was found to a) reduce the number of neutrons reaching the sample, and b) increase energy broadening in the NDP spectra. The beam attenuation is neutron wavelength and capping layer thickness specific and occurs through neutron scattering and absorption by the polymer. Increases in spectral broadening were also relative to capping layer thickness and caused by the reaction product(s) undergoing additional scattering events while passing through the layer. The Li concentration scale of NDP profiles (y-axis) is influenced by the attenuation of the neutron beam and increased scattering. The NDP depth scale (x-axis) is impacted by the increased reaction product scattering. Taken together, these two effects decrease the certainty to which the amount and position of Li within a sample can be determined by NDP. Suggestion on how to measure and mitigate these issues is presented.

Log 557. APPLICATION OF ZERO RESISTANCE AMMETER TO REAL TIME MEASUREMENT OF REDOX CONTROL IN MOLTEN CHLORIDE SALTS

Gonzalez, M. (1,P); Hamilton, E.(1); Simpson, M.F.(1). (1) The University of Utah, Department of Materials Science and Engineering. (P) Presenting Author.

A new in-situ redox control measurement device has been demonstrated based on a zero resistance ammeter (ZRA). In the test system, molten $NaCl-CaCl_2$ was dosed with small amounts of $NiCl_2$ to cause the redox potential to spike. A metallic zirconium rod was used as the working electrode (WE) and immersed in the molten salt. The zirconium WE was shorted to the stainless steel beaker (counter electrode (CE)) used to contain the salt through a potentiostat operating in ZRA mode. The potentiostat simultaneously measured electric current between the WE and CE and the potential of the WE versus a Ag/AgCl reference electrode. With each dosing of $NiCl_2$, the current spiked indicating oxidation of the Zr metal rod and the Zr metal rod OCP spiked before returning to its initial value or lower. In a molten salt system such as an electrorefiner or molten salt reactor where Zr metal can be an effective redox buffer, this device can be used to continuously monitor the rate at which redox buffering is needed. This is useful for measuring rate of ingress of corrosive materials into the molten salt such as air and water. In a molten salt reactor, it is hypothesized that the device can measure the rate of fission which is suspected to cause simultaneous increase in the salt's redox potential.

Log 558. **RADON-PROGENY BLIND TRACE ACTINIDE IN AIR ALPHA SPECTROSCOPY WITH TMFD SENSORS**

Hemesath, M. (1,2); Boyle, N. (1,3,P); Archambault, B. (1,4) Lorier, T. (5); DiPrete, D. (5); Taleyarkhan, R. P. (1). (1) Purdue University. (2) Niowave, Inc. at Lansing. (3) US Intelligence Comm. Post.Doc Fellow -ORISE. (4) Pacific Northwest National Laboratory. (5) Savannah River National Laboratory. (P) Presenting Author.

This article describes research outcomes pertaining to ascertaining for the presence of ultra-trace level actinides in air from their alpha emission signatures, while remaining blind to the relatively large (1,000× higher activity) alpha emissions from Rn-progeny (Po-214 and Po-218). Apparatus and techniques were developed to collect and characterize alpha-emitting nuclides of Rn-progeny and actinides in air on a polycarbonate 3 micron pore size continuous air monitor (CAM) filter. A wet-chemistry approach was developed and validated for successfully separating the Rn-progeny alpha emitting isotopes of Po-214 and Po-218, while extracting the actinides (U, Pu, Am) in a fluid mixture that is suitable for conduct of alpha spectroscopy with a centrifugally tensioned metastable fluid detector (CTMFD). The resulting A-TMFD technology was compared against the state-of-art "Alpha-SentryTM" CAM system commonly utilized world-wide. Results indicate that the A-TMFD technology can potentially offer complementary and superior performance in multiple performance categories, and ~18× improvement in the time to detect (e.g., at 0.02 Derived Air Concentration (DAC) within ~3 h, vs ~70 h for Alpha-SentryTM) for actinides of interest while also remaining ~100% blind to ~103× higher Rn-progeny background – with the added potential for offering few keV scale energy resolution without resorting to peak shape fitting, vs ~300-400 keV for existing CAM systems which utilize PIPS detectors.

Log 559. **HYBRID MASS-ALPHA SPECTROMETRY -TRACE LEVEL UNDER 10 keV ACTINIDES IN FLUID IDENTIFICATION WITH TMFD SENSOR TECHNOLOGY**

Harabagiu, C. (1); Boyle, N. (1,2); Archambault, B. (1,3). DiPrete, D. (4); Taleyarkhan, R. P. (1). (1) Purdue University. (2) US Intelligence Comm. Post.Doc Fellow -ORISE. (3) Pacific Northwest National Laboratory. (4) Savannah River National Laboratory. (P) Presenting Author.

A novel and rapid hybid (mass-alpha) spectroscopic technique was developed for detecting trace (< 0.001 Bq/mL) level alpha emitting radionuclides in fluids with ~10 keV alpha energy resolution. The Centrifugally Tensioned Metastable Fluid Detector (CTMFD) sensor technology was adapted to decipher trace level Pu-239 and Pu-240 content in mixtures of these two isotopes ranging in content from 1:0 to 0:1 in relative proportions with gamma-beta rejection, and ~100% (4 pi) sensitivity validated for accuracy (within +/-5% with NIST standards). Pu-239 and Pu-240 isotopes emit closely spaced (< 10 keV) alpha particles and constitute a known challenge to decipher without resort to microcalorimetry or mass spectrometry. A relatively rapid (< 1h) sampling protocol was developed to create mixtures of these isotopes for CTMFD based examination and to derive the mixture's characteristic response function, viz., alpha decay detection rate over a range of tensioned metastable state negative pressure spanning the detection thresholds for the two isotopes. An accompanying methodology and algorithm were developed to analyze-deconvolute the Pu isotopes bearing mixture's response curves, and to determine the composition of each isotope within the mixture. Results revealed this technique identified the Pu-239:Pu-240 isotopic activity ratio combinations within < +/- 12% for each of the samples ranging from 1:0, 5:1, 1:1, to 0:1 ratios – enabling alpha spectroscopy within few hours of counting, for an arbitrary Pu-239:Pu-240 (< 0.001Bq/mL) mixture, using a ~16 mL CTMFD.

Log 560. **A COMPARISON OF DEHBA AND TBP AS EXTRACTING AGENTS FOR TETRA- AND HEXAVALENT ACTINIDES IN THE CHALMERS GOUPED ACTINIDE EXTRACTION PROCESS (CHALMEX)**

Authen, T. L. (P, 1), Tekikachew, B. E. (1), Foreman, M. R. StJ. (1), Wilden, A. (2), Ekberg, C. (1). (1) Division of Nuclear Chemistry and Industrial Materials Recycling, Department of Chemistry and Chemical Engineering, Chalmers University of Technology, Kemivägen 4, 412 96 Gothenburg, Sweden; (2) Forschungszentrum Jülich GmbH, Institut für Energie- und Klimaforschung, Nukleare Entsorgung und Reaktorsicherheit (IEK-6), 52428 Jülich, Germany. (P) Presenting Author.

The Chalmers Grouped ActiNide EXtraction (CHALMEX) process is a solvent extraction process for the homogeneous recycling of spent nuclear fuel. The current version of the process combines the well-known extractant tri-n-butyl phosphate (TBP) with the European reference molecule for An/Ln separation, CyMe4-BTBP, in the heavy diluent phenyl trifluoromethyl sulfone (FS-13). This combination of extractants provide targeted and direct extraction of tetra- and hexavalent major actinides and tri- and pentavalent minor actinides respectively. However, the use of TBP can be problematic for several reasons, such as troublesome degradation products causing crud formation, decreased extraction yield and the possibility of explosive red oil reactions. Earlier work on the replacement of TBP with DEHBA has been continued for the CHALMEX FS-13 process and is presented here. Factors ranging from physical properties such as density and surface tension, to radiolytic stability and extraction properties under various conditions have been investigated and compared with already established data for the TBP-system. Our findings suggest that DEHBA can be a suitable extracting agent for use with the FS-13 solvent. It extracts tetra- and hexavalent actinides with comparable separation factors compared to the TBP molecule. A disadvantage for the use of DEHBA is its low density: combined with our FS-13 diluent, the density difference between the solvent and the metal loaded aqueous phase is much lower than for the TBP-FS-13 solvent. This could cause phase separation issues, and further hydrodynamic studies are suggested.

Log 561. **ASSESSING AIR POLLUTION WITH SPANISH MOSS AS AN BIOINDICATOR IN THE LOW COUNTRY OF SAVANNAH RIVER BASIN**

Hall, C. (1); Nangeelil, K.(1); Lassel, S.(2); Frey, W.(3);Sun, Z.J.(1). (1) Unviersity of Nevada Las Vegas. (2)North Carolina State University. (3) Unviersity of California Davis (P) Hall, C. (P) Presenting Author.

Spanish moss (Tillandsia usneoides) is epiphyte that grows upon large trees in tropical and subtropical climates. It is a common plant in the low country of the savannah river basin. It absorbs nutrients and water through its leaves from the air and rainfall, which makes it conceivable as a bio-indicator of local air pollution. In this study, Spanish moss samples were collected along the savannah river, near all the industrial sites and superfund sites from Aiken, SC to Savannah, GA, in order to seek the relationship of heavy metals the epiphyte and the air pollution levels in the low country of Savannah River Basin. All the samples were dried and grinded into fine powder, and then studied systematically by instrumental neutron activation analysis (INAA). The preliminary results indicate that: (1) INAA can determine the level of elements in Spanish moss with high accuracy and extreme sensitivity. It is an appropriate tool for conducting multi-element analysis in biological samples; (2) The concentrations of heavy metals in the samples has clearly correlations with the pollution sources in the Savannah River Area, which proves that Spanish moss can serve as an efficient bio-indicator of air pollution.

Log 562. **DETERMINING TRACE ELEMENTS IN COTTON SEEDS WITH INSTRUMENTAL NEUTRON ACTIVATION ANALYSIS (INAA)**

Nangeelil, K.(1,P); Lassel, S.(2);Sun, Z.J.(1). (1) Unviersity of Nevada Las Vegas. (2) North Carolina State University. (P) Presenting Author.

Cotton assimilates numerous trace elements from the soil during its growth, including the heavy metals. Some trace elements are accumulated and enriched in cotton seeds. Therefore, cotton seeds can potentially serve as a biological indicator of heavy metal contamination in local soil. In this study, several cotton seeds and corresponding local soil samples from the Midlands Region of South Carolina were studied by instrumental neutron activation analysis (INAA). The results demonstrate INAA is capable to determine the level of elements in soil and plants with high accuracy and extreme sensitivity. It is promising to apply INAA for multi-element analysis of agricultural products.

Log 563. **FORGERY IDENTIFICATION OF HETIAN JADE WITH INSTRUMENTAL NUCLEAR ACTIVATION ANALYSIS (INAA)**

Dimpfl, P.(1); Mamtimin, M(2); Sun, Z.J.(1,P). (1) Unviersity of Nevada Las Vegas. (2) Halliburton. (P) Presenting Author.

Instrumental neutron activation analysis (INAA) is non-destructive and sensitive in quantifying the elements of interest and have the potential to determine the provenance of the geological samples. In this study, INAA was applied to several jade-related objects from the Xinjiang province of China. Samples include authentic Hetian jade, counterfeit samples purchased at the local market, and natural stones from the same geographic location. The study indicates that INAA can be applied successfully to qualitatively identify similarities and differences in the elemental composition of the mineral samples, and quickly distinguish the forgery from the genuine Hetian jade.

Log 564. **ALPHA/BETA GATED GAMMA-GAMMA SPECTROSCOPY OF MIXED FISSION PRODUCTS FOR TRACE ANALYSIS**

B. Pierson (1,P), B. Archambault (1), A. Hagen (1), L. Greenwood (1), M. Cantaloub (1), S. Herman (1), M. Haney (1), J. Estrada (1), J. Bowen (1), N. Uhnak (1). (1) Pacific Northwest National Laboratory. (P) Presenting Author.

A custom-built liquid scintillation detector combining gamma/gamma coincidence detection with alpha and beta coincident and anti-coincident analysis capabilities was evaluated using a mixed fission product sample. The selectivity offered by gamma/gamma coincidence and background modulation afforded by alpha and beta gating were also demonstrated for ^{136}Cs. By using optimized beta thresholds for several isotopes including ^{91}Y, ^{144}Ce, ^{147}Nd, and ^{153}Sm, the minimum detectable activity (MDA) was reduced by a factor of 1-2 relative to traditional gamma counting. For isotopes including ^{89}Sr, ^{95m}Nb, ^{115}Cd, and ^{137}Cs the use of beta anti-coincidence counting resulted in a reduction of MDA by factors of 3-5; an even greater reduction of MDA by factors of 5-7 was observed for ^{235}U with alpha coincidence counting. The combination of time-correlated radiation signatures and resultant improved detection sensitivity for fission products and actinides is of great interest to a variety of applications including nuclear fission physics, environmental monitoring for nuclear waste and reactor accident releases, and nuclear forensics. Quantification of several hard-to-detect fission products directly from mixed fission product samples has been demonstrated here using a novel detection system.

Log 566. **PVT SCINTILLATOR CHARACTERIZATION AND FAST NEUTRON COMPUTED TOMOGRAPHY AT IDAHO NATIONAL LABORATORY'S NEUTRON RADIOGRAPHY REACTOR**

Oksuz, I.(1, P); Chuirazzi, W.(2); Cao, L.(1); Cherepy, N.(3); Craft, A. (2). (1) The Ohio State University. (2) Idaho National Laboratory. (3) Lawrence Livermore National Laboratory. (P) Presenting Author.

Fast neutron imaging offers a powerful, non-invasive investigation technique to inspect thick, dense samples while causing less activation than thermal neutron imaging. However, lower interaction cross-sections of fast neutrons with materials pose a challenge in designing an efficient neutron imager that can produce the many projections needed for fast neutron computed tomography (nCT) that would allow for visualization of small 3-D features. In this study, two PVT scintillators, fabricated by Lawrence Livermore National Laboratory, with thicknesses of 3 mm and 10 mm and doped with 2%Flrpic (Ir-Complex) were characterized in terms of relative light output utilizing a lens-based automated imaging setup comprising of a CMOS camera, a light-tight box, a rotation stage, at Idaho National Laboratory's (INL) Neutron Radiography Reactor (NRAD) North Radiography Station (NRS) beamline, which supplies an estimated fast neutron flux of ~2.7×10^7 n·cm-2·s-1 (>1MeV). Fast nCT was also performed utilizing an imaging phantom that is a 5 cm cube made of HDPE covered with 2 mm thick aluminum sheet, with four through holes (1.25cm dia.) each filled with aluminum nuts, paraffin wax, cement, and stainless-steel screws. A total of 720 projections of the phantom, from 0° to 360° at 0.5° increments, were acquired with a total acquisition time of 60 hours. Characterization results showed that the 10 mm thick scintillator provided ~8x higher light output compared to the 3 mm scintillator. The volume-rendered images of the phantom clearly discerned each of the four materials filling the holes.

Log 567. **ELECTROCHEMICAL TRANSIENT TECHNIQUE FOR THE MEASUREMENT OF HIGH CONCENTRATION OF METAL IONS IN LiCl-KCl MOLTEN SALT**

Choi, S. (1); Bae, S.-E.(1, P); Park, T.-H.(1); Kim, J.-Y.(1); Ji-Hye Kim, J.-H.(1). (1) Korea Atomic Energy Research Institute. (P) Presenting Author.

For a real-time monitoring of nuclear materials in pyroprocess, repeating chronoamperometry (RCA) is proposed in this work. The RCA technique was tested in LiCl-KCl melt containing various metal ions such as uranium trichloride, and magenssium bichloride, and lanthanium trichloride. The magnesium was selected as a surrogate for plutonium. In the multi-component system, the current response and passed charge of electrodeposition and dissolution reactions were monitored as a function of the concentration of the metal ions. The results showed that the current and passed charge increased linearly with the concentration of the metal ions in LiCl-KCl melt when the concentration of metal ions extended up to ~10 wt%. The RCA technique was successfully applied to determine unknown concentration of the metal ions in LiCl-KCl melt, indicating that the RCA is one of useful methods for the real time monitoring of the metal ion concentration in a pyroprocess.

Log 568. **APPLYING INAA METHOD TO ASSESS THE NUTRITIONAL COMPOSITION OF EDIBLE BIRD'S NEST IN VIETNAM**

Truong, TS. (1,2); Hoang, S.M.T. (3,P). (1) Ho Chi Minh City University of Education. (2) Nuclear Training Center - VINATOM. (3) Institute of Applied Technology - Thu Dau Mot University. (P) Presenting Author.

Edible bird's nest (EBN) is a resource and a noble food that has excellent uses in nourishing the body, clearing heat, increasing resistance, anti-aging and increasing longevity. The nutritional composition of EBN is determined in the form of organic compounds as Serine, Glutamine acid, Glycine, Lysine, Valine and a few elements such as Zn, Fe, Mn, Se ... In this study, the INAA method was applied to analyze about 18 elements in EBN in Khanh Hoa City (Vietnam) with the highest element concentration being Ca (18720 ppm), followed by Na (16290 ppm), Cl (14150 ppm), Fe (111.6 ppm). These are nutritional elements necessary for humans. Relatively high concentrations of Na and Cl indicate a high salt concentration in the analyzed sample, which may be due to salt vapors of the sea, food, etc. Se and Hg elements result in relatively low concentrations, 0.2789 ppm, and 0.2676 ppm, respectively. Elements that give reliable analytical values with low errors such as Sm (1.2%), Na (0.7%), Cl (4.4%), Ba (5.2%), Fe (5%), ... Elements with a high error such as Eu (43.2%), Cr (39.9%), Mn (63.5%), Th (50.2%) due to the analytical content approximate the detection limit or possibly due to the influence in experiments lead to errors. Regarding toxic elements, NAA analyzed the Hg content about 0.25 ppm and Se about 0.28-0.42 ppm, within the UL limit of IOM and WHO. At this level, Se is considered a nutritional element, essential for cell function, and Hg is not toxic.

Log 569. **APPLYING PRIMO PROGRAM TO SIMULATE ELEKTA LINAC FOR STANDARD DOSE CALCULATIONS AND TREATMENT PLANNING**

Dang, M.P. (1,2); Pham, N.V.H (3); Hoang, S.M.T. (4,P). (1) Cho Ray Hospital. (2) Nuclear Training Center - VINATOM. (3) Institute for Nuclear Science and Technology – VINATOM. (4) Institute of Applied Technology - Thu Dau Mot University. (P) Presenting Author.

Radiation therapy is one of the most popular methods in cancer treatment; with modern radiotherapy techniques on new generation accelerators, asymmetric projection fields are used quite commonly, creating significant advantages in technical manipulation and radiation dose distribution for patients. The purpose of this study was to quantify the differences in dose distributions computed by Monte Carlo simulations against experimental measurements from an Elekta Synergy linear accelerator. This study was conducted to apply for the PRIMO program, a PENELOPE-based Monte Carlo code, in simulating radiotherapy linear accelerators, checking the simulation accuracy on the water phantom to proceed to the simulation directly on the model human phantom created from CT images. Experimental depth doses and profiles were obtained using a pair of CC13 IBA ionization chambers at an SSD as 100 cm in an IBA Blue phantom. The simulation results of the depth and the horizontal dose distributions correspond entirely to the results obtained from the experiment with a discrepancy of less than 2%, helping to simulate the dose calculation in planning. Using PRIMO, the Monte Carlo simulation showed good agreement compared to experimental measurements for both depth dose and dose profiles for the evaluated fields sizes, primarily used in commonly radiotherapy treatments.

Log 570. **COMPARISON OF THE ENVIRONMENTAL IMPACTS OF NUCLEAR AND COAL-FIRED POWER PLANTS BY EMISSIONS OF RADIONUCLIDES**
Wieczorek, J. (1,2,P); Steinhauser, G. (1); Blenke, T. (1); Zok, D. (1,3,P); Stäger, F. (1); (1) Leibniz University Hannover (2) Faculty of Chemistry of the University of Gdansk. (P) Presenting Author.

The search for clean and sustainable energy sources has led to increased scrutiny for the emissions of noxious emissions from the production of electric power. In our present study, we compare the atmospheric emissions and liquid effluents of radionuclides from coal-fired and nuclear power plants, respectively. Two German power plants in Lower Saxony were chosen to exemplify these emissions. Water and soil samples were collected in the present winter period. In our comparison, two types of possible radionuclide groups were selected. On the one hand, naturally occurring radionuclides such as Po-210 and U-235/U-238, on the other hand, typical fission products such as Cs-134 and Cs-137. Especially, we are focusing on radionuclides, which were partly understudied in risk assessments and emissions. After a specific sample preparation, alpha, gamma, and mass spectrometric measurements were performed and will be discussed in detail.

Log 571. **HEALTH RISK ASSESSMENT OF *Cannabis Sativa* L. PRODUCTS BY Po-210 AND Pb-210**
Wieczorek, J. (1,P); Borylo, A.(1); Kaczor, M.(1); Skwarzec, B.(1,); . (1) Faculty of Chemistry of the University of Gdansk. (P) Presenting Author.

The consumption of tobacco involves a well-studied increased health risk due to inhalation of not only noxious chemical compounds but also radionuclides from the natural decay chains. The direct incorporation of harmful substances mostly effects the lung as well as surrounding tissues and increases the risk for cancer significantly. Since Cannabis Sativa L. products have now become legal in Poland, these may warrant the same degree of scrutiny as tobacco products. For our study, we selected the radionuclides Po-210 and Pb-210 from the uranium decay chain for a detailed analytical investigation. Activity concentrations of these nuclides were investigated in 44 hemps, 20 hashish, and 8 hemp tea samples, as well as in three selected types of cannabis plants (Fenola, Fedora, and Futura). Analyses were performed using alpha spectrometry. In the case of hemp, the activity concentration of Po-210 ranged from 0.6 up to 34.7 mBq/g and, for Pb-210, from 0.2 up to 2.3 mBq/g, respectively. In hashish, the radionuclide activity concentrations ranged from 2.5 up to 164 mBq/g for Po-210 and from 0.5 up to 45 mBq/g for Pb-210. Additionally, effective doses were calculated and will be presented.

Log 572. **PRODUCTION AND SEPARATION OF Be-7 FOR ION-SOURCE PREPARATION AT THE NATIONAL SUPERCONDUCTING CYCLOTRON LABORATORY**

Satija, S. (1,2,P); Domnanich, K.A.(1,2); Chaple, I.F.(3); Shefali, F.(3); Cingoranelli, S.J.(3); Severin, G.W.(1,2); Lapi, S.E.(3); Sumithrarachchi, C.(2); Bollen, G.(2). (1) Michigan State University. (2) National Superconducting Cyclotron Laboartory. (3) The University of Alabama at Birmingham. (P) Presenting Author.

The stand-alone operation of the ReA (Re-accelerator) at the National Superconducting Cyclotron Laboratory is being facilitated by the use of offline sources after the coupled-cyclotron facility ceased operation. To aid this ReA experimental research program, approximately 5 mCi 7Be was required. This isotope would be placed in the Batch-mode Ion Source (BMIS), which consists of an oven, that provides beams of stable or long-lived radioactive isotopes coupled to an ion-source system with arrangements for beam transport. Owing to the commercial unavailability of this rare isotope Be-7 in sufficient quantity for ReA experiments, a production route through $10B(p,\alpha)7Be$ reaction was explored at the University of Alabama Birmingham (UAB) Cyclotron facility and subsequently, a separation methodology had been developed. Seven irradiations in total were carried out at the UAB Cyclotron with proton beam energy of 17.5 MeV with natural and 10B enriched boron powder targets that led to the production of 7Be. This was followed by dissolution of boron powder in concentrated HNO_3 followed by separation of 7Be from the bulk boron in HNO_3 medium employing the use of ion-exchange resin. Greater than 90% 7Be separation efficiency was achieved and the purified 7Be fractions were reconstituted in a small volume of 0.1 M HCl and deposited onto tantalum foils for introduction into the BMIS for 7Be beam delivery experiments. Through this experiment, production and delivery of 7Be was carried out to support the ongoing ReA experimental program of NSCL and a radiochemical methodology for separating 7Be from the boron target was established.

Log 573. **NEUTRON ACTIVATION ANALYSIS APPLICATIONS WITH A MEDICAL ISOTOPE CYCLOTRON**

Duke, M.J.M. Medical Isotope and Cyclotron Facility, University of Alberta.

During the past 25 years there has been a significant decline in the number of operating research reactors world-wide due to the decommissioning and permanent shutdown of facilities. With this decline there has been concomitant loss of activation analysis and radionuclide production capabilities, and of training, teaching and outreach opportunities. In comparison, the past 10-15 years has seen a substantial increase in the commissioning of cyclotrons world-wide, particularly medical cyclotrons in the 10-25 MeV range. In this regard the University of Alberta (UofA) decommissioned its SLOWPOKE reactor in 2017 but commissioned a TR-24 cyclotron in 2013. The primary purpose of the UofA TR-24 is to produce medical radionuclides via (p,xn) nuclear reactions. While neutrons generated in such reactions are typically considered problematic from a radiation safety perspective they may also serve as a neutron source for activation analysis. At the UofA cyclotron facility we have been exploring and developing the use of these 'parasitic' neutrons for NAA by: a) performing 1h-4h single and multiple sample batch irradiations in the cyclotron vault (utilizing activation products with intermediate half-lives, e.g., ^{56}Mn,^{24}Na,^{152m}Eu,^{165}Dy) and b) utilizing a re-purposed pneumatic sample irradiation controller to transfer samples in and out of the cyclotron vault during cyclotron operation thus permitting the use of shorter-lived radionuclides for FNAA and INAA. Plans are underway to significantly increase the neutron flux available for NAA by employing a Be metal target on one of the cyclotron beam lines. The utility of a TR-24 cyclotron for NAA and teaching will be presented.

Log 574. **HUMAN TISSUE SUBSTITUTES SUITABLE FOR A CLINICAL POLYENERGETIC PHOTON SPECTRUM BETWEEN 40 keV TO 120 keV**

Jimmy Stringer (P), Henry Spitz.   University of Cincinnati, College of Engineering & Applied Science, Cincinnati, OH. (P) Presenting Author.

The conventional process involved in designing and fabricating tissue substitute materials typically incorporates small quantities of additives into a base material to obtain a product that exhibits the desired effective Z, electron density, and mass attenuation coefficient (m/r) relevant to the discrete photon energy being measured. The acceptability of the tissue substitute can be determined by comparing the measured value of m/r, determined at the discrete photon energy, with the published value in ICRU Report #44. However, the typical x-ray energy spectrum produced in diagnostic radiology includes photons ranging from approximately 40 keV up to 120 keV. Depending upon the type of tissue in the x-ray beam, photon interactions are most likely a combination of photoelectric absorptions and Compton scattering events and will modify the intensity and energy distribution of the X-ray beam as it penetrates the tissue. Thus, formulation of a tissue substitute material suitable for computerized tomography (CT) is more challenging because the polychromatic continuum does not produce the same number of photons for all energies. Values of m/r, a critical performance metric, change significantly across this continuum as Compton rather than photoelectric interactions become more likely. Furthermore, clinical CT detectors measure the total number of photons that reach the detector rather than discrete photon energy fluence. Formulations for adipose, muscle, bone, and other tissues have been developed and tested that reflect the probability a photon of specific energy being generated in the polychromatic CT spectrum and its likelihood of interacting in the specific tissue of interest.

Log 576. **CRYOGENIC TRACER IRRADIATION FACILITY**

Hudson, C.C. (1,P); Lester, R. (1); Tipping, T.N. (1); Haas, D.A. (1). (1) The Unviersity of Texas at Austin. (P) Presenting Author.

We report on a cryogenic gas irradiation facility in the 1.1 MW TRIGA reactor at The University of Texas at Austin. The system was designed to produce radioactive xenon and argon for environmental studies, and it can be applied to produce other gaseous radiotracers. In this presentation, we report on the system design, transfer tests using nitrogen and xenon, and initial irradiations. The system design includes modeling accident scenarios to ensure operation does not risk damage to the reactor and minimizes risk of release of radioactive material. The SCALE code was used to model a 20-day irradiation (assuming 8-hour reactor operation followed by 16 hours of downtime per day) of one liter of Xe-126 and the predicted activity is 0.6 Ci of Xe-127 at irradiation end. Installation of the system in the reactor is underway and results of initial irradiations are expected to be included in the presentation and paper.

Log 577. **A PORTABLE, LOW-POWER ELECTROSTATIC PRECIPITION-BASED AEROSOL MONITOR FOR NUCLEAR EXPLOSION MONITORING**

Keillor, M.E. (1); Hallen, T.W. (1); Moore, M.E.(1); Kasparek, D.M.(1); Ortiz, A. (1). (1) Pacific Northwest National Laboratory. (P) Presenting Author.

A low-power aerosol collector with on-board measurement for gamma-emitting radionuclides is currently being developed at PNNL. An electrostatic precipitator with low airflow restriction is central to the design. The device uses a 3" wide roll of collection media, which can be advanced at a continuous rate, or in a stop-and-go approach to collection followed by measurement. Radiation measurement is accomplished with two radiation detectors, providing for the potential application of coincidence techniques. Detectors currently in-hand for testing in the system include Mirion sodium iodide (NaI(Tl)) and cerium bromide (CeBr) scintillators, and an H3D cadmium zinc telluride (CZT) detector module. The status of this development will be presented. The potential for valuable contribution to explosion monitoring with portable aerosol monitors processing lower air volumes at distances of less than ~1000 km from the source will also be considered.

Log 578. THE POTENTIAL APPLICATION OF THE TWO-FILTER RADON MONITOR DESIGN TO MONITORING FOR THE SHORT-LIVED FISSION PRODUCT KR-88

Keillor, M.E. (1); Burnett, J.L.(1); Ely, J.H.(1); Kasparek, D.M.(1); Moore, M.E.(1). (1) Pacific Northwest National Laboratory. (P) Presenting Author.

A series of developments in radon monitors aimed at studying radon levels over remote ocean areas led to the development of the Two-Filter Radon Detector design reported by Whittlestone in 1998. This style of radon monitor can achieve detection limits on the order of 1 mBq/m3 for Rn-222 based on collection and measurement of the daughter isotopes, with a relatively simple and robust system design. This work will primarily focus on the potential detection limits for the short-lived fission product Kr-88 based on collection and measurement of Rb-88; monitoring for the 2.84-hour half-life fission product would be most effective during the first ~10-12 hours after a nuclear explosion. Another potential application is measurement of the concentration of Ar-42 in the atmosphere; Ar-42 is a likely background contributor in underground physics experiments using liquid argon. Calculations similar to those for Kr-88 will be presented to consider whether the Two-Filter design has the potential to provide a useful measurement of the atmospheric concentration of Ar-42 in argon. Such a measurement would be accomplished by processing purified argon sourced from the atmosphere. The dominant source of Ar-42 in the atmosphere is believed to be from cosmogenic production in the upper atmosphere, while a smaller contribution is attributed to historic atmospheric nuclear testing. In a Two-Filter system, the K-42 daughter of Ar-42 would be collected onto the 2nd filter; the 12.4 hour half-life of K-42 would allow for direct measurement or transfer to an underground laboratory for low-background measurement.

Log 579. RADIOKRYPTON AND RADIOXENON DIFFUSION IN SILICATE AND SODIUM CHLORIDE MEDIA

Lapka, J.L. (1,P), Haas, D.A. (1),Lowrey, J.D. (2) The Unviersity of Texas at Austin. (2) Pacific Northwest National Laboratory (2). (P) Presenting Author.

The detection of volatile fission products such as krypton and xenon plays an important role in the monitoring of underground nuclear explosions. Different types of geological media impact the rates of transport of these gases and thus affect their ability to be detected by atmospheric monitoring stations. In this work the gaseous diffusion of neutron-activated krypton and xenon were investigated in silicate and sodium chloride media at multiple pressures. Measurements of radionuclide transport were conducted by introducing each gas into the bottom of a 316 stainless steel two-bulb gaseous diffusion apparatus which had been previously flushed with nitrogen. The porous media bridge between each bulb was filled with either Ottawa density sand or granular sodium chloride which were both sieved to obtain matching particle size distributions. The transport of each radionuclide was monitored by a collimated lanthanum bromide detector at each bulb over the course of several days until equilibrium distribution was achieved. Diffusion rates and relaxation times for each nuclide in different media and pressures are discussed and compared to data obtained in prior two-bulb diffusion studies.

Log 580. **INFLUENCE OF UNINTENDED CONVECTION ON DIFFUSION COEFFICIENT MEASUREMENT IN HIGH-TEMPERATURE MOLTEN SALT ELECTROCHEMICAL CELL**

Ohk, Seung-min(1,P); Park, Jaeyeong(1). (1) Ulsan National Institute of Science and Technology. (P) Presenting Author.

The diffusion coefficients in a high-temperature molten salt system have primarily been measured by cyclic voltammetry (CV) and chronopotentiometry (CP). However, there are several-fold differences among the obtained values depending on the used experimental parameters such as scan rate in CV and applied current density in CP. A reason for the discrepancy would be the inadvertent convection caused by thermal gradient and cell vibration disturbing the ideal theoretical diffusion. The rotating electrode can minimize the problem by producing forced convection, overwhelming the unintended convection. Herein, the influence of the undesired convection from thermal gradient on the diffusion boundary layer thickness and diffusion coefficient in the high-temperature molten salt system is evaluated for both static and rotating electrode cells by a computational COMSOL Multiphysics 5.6. The rotating speed of the electrode is controlled to cover from laminar to the turbulent regime. Also, the diffusion coefficients of Mg in LiCl-KCl-MgCl2 salts at 773 K were experimentally obtained by CV, CP, and a rotating cylindrical electrode with various operating parameters (e.g., scan rate, applied current density, and rotating speed), and the obtained values are compared quantitatively to each other and literature.

Log 581. **CHARACTERIZATION OF THE NEUTRON PULSE TIME PROFILE FROM A DEUTERIUM-TRITIUM NEUTRON GENERATOR**

Searfus, O. (1,P); Graham, C. (1); Jinia, A (1); Clarke, S. (1); Pozzi, S (1); Jovanovic, I. (1). (1) The University of Michigan Department of Nuclear Engineering and Radiological Sciences. (P) Presenting Author.

Precise knowledge of the temporal output profile of a pulsed neutron generator has the potential to increase the sensitivity of neutron active interrogation techniques, allowing for the simultaneous measurement of the characteristic decay time profile and gamma ray energy spectrum of an induced isotopic species during the neutron pulse. In most neutron generators, it is not sufficient to assume that the output pulse is a simple square wave in time. The true temporal profile is often difficult to measure, as both scattered primary radiation from the generator and secondary radiation induced by the generator can significantly distort the direct, primary signal. In this work, the direct, primary temporal profile of a pulsed deuterium-tritium neutron generator was isolated from scattered and induced contaminants with multiple detector types using the neutron generator electronic pulse as a time reference. The first is a "gamma-blind" He-4 fast neutron scintillation detector, while the second is a variety of pulse-shape discrimination-capable organic scintillation detectors. Both of these neutron detector types allow for fast timing, and their signals carry information on the incident neutron energy, so that the time profiles of high-energy depositions (corresponding to primary neutrons incident directly from the generator) and low-energy depositions (corresponding to scattered and secondary radiation in addition to primary direct radiation) can be separated. This spectro-temporal analysis will improve the sensitivity of many prompt neutron active interrogation techniques.

Log 582. **NICKEL-63 SEPARATION AND ANALYSIS IN STAINLESS STEEL**

Zayas, N.(1,P); Zigmond, J.(1); Larson, N.(1); Reese, R.(1); Schoendaller, K.(1); Allen, M.(1). (1) Sandia National Laboratories. (P) Presenting Author.

The Sandia National Laboratories Radiation Protection Sample Diagnostics (RPSD) laboratory validated a method to separate and quantify nickel-63 for activated stainless steel samples that contain a significant amount of cobalt-60. The validation study used a stainless steel standard reference material, along with subsamples of the activated stainless steel, presented as metal shavings, to develop the method. The samples were prepared through digestion with a nitric and hydrochloric acid mixture in a 1:3 molar ratio. Eichrom TRU resin was used to remove iron from the samples before performing two individual nickel separations using Eichrom Ni resin cartridges. Cobalt was added before each nickel separation to aid with cobalt-60 removal as a stable carrier. The inherent nickel content of the stainless steel was used to determine the chemical yield of the nickel after separation. Nickel analysis was performed using Inductively Coupled Plasma – Optical Emission Spectrometry (ICP-OES). Nickel-63 analysis was completed by Liquid Scintillation Counting (LSC) using a nickel-63 specific quench curve. The method met the Multi-Agency Radiological Laboratory Analytical Protocols (MARLAP) validation criteria requirements, resulting in an average nickel-63 recovery of $96.9 \pm 0.03\%$ for 21 validation samples. During method development, High Purity Germanium gamma spectrometry (HPGe) and ICP-OES aided in determining activity of cobalt-60 and iron, nickel, and cobalt content in the discrete fractions of samples and waste.

Log 583. **THE LOS ALAMOS LOW ACTIVITY INVESTIGATION ROOM (LAIR) AT THE EVENT VERIFICATION and IDENTIFICATION LABORATORY (EVIL)**

M. Boswell (1), D. Cox (1), D. Dry (1), S.R. Elliott (1), S. Lamont (1), R. Massarczyk (1), S. Meijer (1), W. Meijer (1). (1) Los Alamos National Laboratories. (P) Presenting Author.

The Los Alamos National Laboratory Nuclear & Radiochemistry Countroom has a distinguished history of nuclear forensics dating back to the 40's. Over the years the Countroom has evolved to serve many programs, processing over 55,000 measurements a year. In this talk, I will discuss our new facility for specialized applications like nuclear forensics. I will primarily focus on our new low-background laboratory being constructed in a historic Manhattan-project era tunnel in the Los Alamos canyon. The tunnel at TA-41 was built between June 1948 until May of 1949. The tunnel is lined with concrete and extends approximately 250 feet into the north side of Los Alamos Canyon, providing roughly 300 ft of rock overburden. Such a facility is a prime location for a low-background laboratory, and currently home to 3 HPGe detectors, with a plan to expand that to include 2 more detectors along with several low-background beta chambers. I will present data from our current detector operating in the facility, as well as discuss our future plans for additional detectors in the facility, and discuss the challenges in setting up such a facility in an historic location.

Log 584. **METHOD DEVELOPMENT FOR ICP-MS MEASUREMENTS OF U AND Np IN Pu METAL**

Bartlett, J.H. (P), Aragon, S.M. (P) Presenting Author.

Inductively-coupled plasma mass spectrometry (ICP-MS) is a technique most notable for its ability to analyze for nearly any element on the periodic table, with the main caveat that the element of interest must be able to be ionized by the plasma gas (Ar). In practical applications, many elements can be quantified by ICP-MS with just a single sample preparation and analysis run. ICP-MS has been well established over several decades for our laboratory to provide analytical support for Pu metal processing. Recently, interest has arisen in expanding the suite of analytes routinely measured by ICP-MS to include U and Np in Pu metal matrix. As a result, we have developed a column chromatography scheme to facilitate these measurements, using commercially-available UTEVA and TEVA resin cartridges for the ion-exchange reductive extraction of U and Np from Pu. Method validation data was benchmarked against gold-standard techniques throughout the development process; namely, the isotope dilution method using thermal ionization mass spectrometry (TIMS-IDMS) for U, and gamma or alpha counting for Np. The chemistry scheme will be presented along with the results and analysis of the validation data.

Log 585. **SYNCHROTRON X-RAY FLUORESCENCE AND TRANSMISSION IMAGING FOR NUCLEAR SAFEGUARDS**

Ditter, A. (1,P); Ward, J. (2); Miller, M. (2); Coles, R.(3); Bowerman, B.(3); Schoonen, M.(3); Thieme, J. (3); Duffin, A. (2);, Shuh, D. (1); (1) Lawrence Berkeley National Laboratory, (2) Pacific Northwest National Laboratory, (3) National Synchrotron Light Source II. (P) Presenting Author.

The current method for preparing particles for nuclear safeguards analysis involves utilizing fission tracks to locate particles of interest, then isolating and removing these particles for electron microscopy and/or thermal ionization mass spectrometry. This process very time consuming and involves a great deal of manual labor. Our collaboration between Lawrence Berkeley National Lab (LBNL), Pacific Northwest National Lab, and Brookhaven National Lab addresses these problems by synchrotron radiation to perform much of this analysis without ever removing those particles from the specimen. This talk will focus on recent experiments performed at the Advanced Light Source at LBNL in two areas: tender micro-XRF, specifically pushing this technique to measure lighter element fluorescence, and a rapid method of uranium particle location through full-field x-ray microscopy.

Log 586. ^{236}U/^{238}U MEASUREMENTS IN URANIUM ORE CONCENTRATES VIA ACCELERATOR MASS SPECTROMETRY

Dorais, C. (1,P); Tumey, S.J. (1); Marks, N.E. (1); Brennecka, G.A. (1). (1) Lawrence Livermore National Laboratory. (P) Presenting Author.

Uranium ore concentrate (UOC), the refined uranium product that is subsequently enriched for use in reactors and weapons, is one of the nuclear materials most frequently targeted for theft and trafficking. When UOC outside of regulatory control is recovered, nuclear forensic analyses are conducted to assist in provenance assessment. Isotope ratios of the major and minor U isotopes (234U, 235U, and 238U) are commonly measured in UOC samples; however, 236U is not often measured because of its low abundance. Although generally considered an anthropogenic isotope, 236U is also produced naturally at ultra-trace levels via neutron capture on 235U. Variation in the natural 236U/238U ratio, which has been shown to range from 10- 14 to 10-10, is thought to be controlled by factors that affect the production or moderation of neutrons in the ore body such as ore grade, deposit age, the amount of certain trace elements present, and water content. However, because 236U is present at such low concentrations in UOCs, Accelerator Mass Spectrometry (AMS) is required to make these measurements. In this presentation, details of the new 236U/238U measurement setup at LLNL's Center for Accelerator Mass Spectrometry will be presented, along with preliminary results from a suite of UOC samples.

Log 587. TIME-BASED DETECTION OF PARTICLES IN LASER ABLATION MULTICOLLECTOR ICPMS
Duffin, A.M. (1,P); VanGundy, R.A (1). (1) Pacific Northwest National Laboratory. (P) Presenting Author.

Mass spectrometry detection of nanoparticulate materials is challenging in that the ion counts from individual particles can be overwhelmed by background. We have developed a time-based ion signal detection scheme, exchanging intensity-based detection for temporal-based detection. This development allows for rapid identification and quantification of nanoparticulate signals. We demonstrate this rapid transient detection scheme with direct laser ablation analysis of filter material, showing clear distinction between particle signals and matrix material background signals.

Log 588. **RADIOCHEMICAL SEPARATION OF 48V FROM ALPHA-IRRADIATED NATSC FOIL**

Essenmacher, S.D. (1,2,P); Satija, S.(1,2); Kleinfeldt, C.R.(1,2); Domnanich, K.A.(1,2); Vyas, C.K.(2); Severin, G.M.(1,2). (1) Michigan State University. (2) Facility for Rare Isotope Beams. (P) Presenting Author.

Most direct neutron reaction cross section measurements to date have been performed with stable or long-lived nuclei. Performing direct measurements on relatively short-lived radionuclides would enhance the accuracy of theoretical calculations and improve stellar nucleosynthesis models and validations in nuclear applications. Obtaining high precision (10 b) cross section measurements of the (n, γ) and (n, p) reactions with the relatively short-lived radionuclide 48V (t1/2 = 15.97 d) is important for the Stockpile Stewardship program. A crucial requirement for a 48V target in these studies is the 48V radioisotopic purity; specifically, the ratio of 48V/49V atoms must be at least 1.0E+4. One possible method to obtain 48V in high isotopic purity is via natSc(α, n)48V as 48V/49V = 1.0E+3. Although this is lower than the desired ratio of 1.0E+4, the cross section data for natSc(α, γ)49V is based solely on theoretical calculations. To determine the actual 49V production rate, natSc foils were irradiated with alpha particles at the University of Notre Dame. The irradiated foils were used to determine the optimal radiochemical separation method for V and Sc. The results suggest two separation methods can be utilized to completely separate trace vanadium from bulk Sc target material with total 48V recoveries of 96(5)% and 97.9(7)%. Fractions from the optimal separations that contain 48V/49V were used to produce samples for liquid scintillation counting to determine the actual 48V/49V ratio and the viability of this production method to produce radioisotopically pure 48V.

Log 589. **RADIONUCLIDE SIGNATURES FROM COMPLEX RELEASES OF VENTED FISSION PRODUCTS**

Gordon, E.M. (1,P); Adhikari, P.(1); Lowrey, J.(2); Johnson, C.(2); Ely, J.(2);Haas, D.A.(1). (1) The Unviersity of Texas at Austin. (2) Pacific Northwest National Laboratory. (P) Presenting Author.

Following an underground nuclear explosion, fission products may be vented to the surface and transported through the atmosphere. Traditional requirements for nuclear explosion monitoring systems have focused on simple release scenarios. A more rigorous evaluation of radionuclide inventory releases will provide better requirements for measurement systems and improve analysis of detections.

The goal of this work was to compare isotopic signatures reaching monitoring stations under different venting scenarios. First, a radionuclide inventory of fission products was developed using the SCALE code system based on the fission of U-235. The release of fission products to the surface was modeled in two components: prompt and delayed releases. Both the prompt and delayed components were varied to produce 63 total scenarios (example: 0.1 percent gas vent prompt release and no delayed release). Dilution factors for the resulting plume reaching a selected subset of IMS monitoring stations were found using HYSPLIT, and the simulation was repeated over a period of 366 days. Detection frequency and ratios for multi-isotope detections are presented for the various release scenarios.

Log 590. Np-237 FISSION PRODUCT YIELDS INDUCED FROM THE GODIVA IV CRITICAL ASSEMBLY

Harke, J.T. (1,P); Tamashiro, A.S.(2); Burcher, S.P.(1), Padgett, S.W.(1), Zhao, P.(1), Pierson, B.(3); Gharibyan, N.(1); Goda, J.(4); Greenwood, L.(3); Hayes, D.(4), Hutchinson, J.(4); Harward, N.(1); Roberts, K.(1); Slavik, G.(1); Yap-chiongco, P.(1); Walker, J.(4); Bredeweg, T.A.(4). (1) Lawrence Livermore National Laboratory. (2) Oregon State University. (3) Pacific Northwest National Laboratory. (4) Los Alamos National Laboratory. (P) Presenting Author.

The accurate understanding of fission product yields is key for nuclear reactor fuel cycles, reactor decay heat, and nuclear waste inventory. We have performed gamma-ray spectroscopy on fission fragments produced via a prompt fission neutron spectrum irradiation using the Godiva IV critical assembly. During this experiment, Np237 samples were irradiated using the prompt burst mode operation of Godiva. Within 1 hour of irradiation, gamma-ray spectroscopy was commenced, and event-by-event list mode data was collected for up to 7 days post-irradiation. Several dozen fission product yields were identified and measured by using the unique gamma-rays emitted for the beta-decaying fission products. This measurement is part of a series of measurements that is part of a national effort to create a modern fission product yield database. It has been over 25 years since any coordinated major effort has occurred. The Np237 fission product yield measurements are part of a 10-year measurement campaign using the unique capabilities of the Godiva IV critical assembly. An update on this research and results to date will be presented.

Log 591. ABUNDANCE SENSITIVITY IN ^{236}U/^{238}U MEASUREMENTS: OPTIMIZATION OF CONDITIONS

Krogstad, E. (1) (P), McHugh, K.(1), Shen, S.(1), Conte, E.,(1), (1) PNW Ntl. Lab. (P) Presenting Author.

Measurement of very low 236U/238U ratios in environmental samples relies on optimization of conditions that limit precise and accurate results. These include ion optics, inelastic scattering, counting statistics, for plasma instruments the degree of hydride generation and, for all, sample concentrations. Modern sector field mass spectrometers across a wide range of instruments have ion optics that can produce peak shapes sufficient for low (e.g., <1E-06) 236/238 measurements. However, this ratio in environmental samples is commonly much less than 1E-6, with 1E-11 being thought to represent natural backgrounds. What can be done about the range 1E-07 to 1E-11 without the need to resort to accelerator mass spectrometry? A practical limit of ion counter detection, including detector noise, reflects one bounding condition. Sample utilization efficiency represents another, with both TIMS and MC-ICP-MS hindered by ionization and transmission limitations. For an ion beam of 1E10 cps on 238U+ and 10 cps on 236U+, an 236U/238U of 1E-09, challenges arise due to the need to have such a high 238 signal. Correction for 235UH+ is also a significant problem for plasma instruments. Peak tailing of the 238U beam to 1E-08 at mass 237 requires very good vacuums and likely a filter to reject low energy ions. To achieve 1E09 or better requires: 1) very high sensitivity and high 238U ion beams, 2) low counter noise, 3) well-characterized hydride (plasma), 4) very good operating vacuums (E-09 mbar or better), and probably 5) a map of scattered beam intensities at several half-mass conditions.

Log 592. **MAPPING URANIUM ISOTOPIC CONTENT AT THE NANOSCALE IN A NON-UNIFORM FUEL PELLET USING EXTREME ULTRAVIOLET LASER ABLATION MASS SPECTROMETRY**

Rush, L.A. (1,P); Cliff, J.B.(2); Reilly, D.D.(2); Duffin, A.M.(2); Menoni, C.S.(1). (1) Colorado State University. (2) Pacific Northwest National Laboratory. (P) Presenting Author.

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Extreme ultraviolet (EUV) lasers possess unique properties for ablation and ionization at the nanoscale (≤100 nm) due to their short wavelength, high absorptivity in most materials (i.e., 10's of nanometers deep), and efficient single photoionization in the laser-created plasmas. When coupled with a mass spectrometer, an EUV laser can be used to probe chemical information in three dimensions with nanoscale spatial resolution. We have previously built an EUV laser ablation and ionization time-of-flight mass spectrometer (EUV TOF) that operates at a wavelength of 46.9 nm (26.4 eV photon energy) and can achieve ~80 nm lateral and ~20 nm depth resolution when mapping the chemical content in organic and inorganic solids. Here, we present results from a recent study that extends EUV TOF's high spatial resolution capabilities to the analysis of an isotopically non-uniform uranium fuel pellet that was made by blending two isotopically distinct feedstock materials. We show that EUV TOF can map $^{235}U/^{238}U$ heterogeneity using 100 nm pixels, revealing non-uniformity that was not clearly identified at the microscale or with bulk analytical techniques. For comparison, nanoscale secondary ionization mass spectrometry (NanoSIMS) maps a similar distribution of U non-uniformity on a similar subsample at the same spatial scale. These results position EUV TOF as a promising technique for performing isotopic analyses at the nanoscale, finding applications not only in nuclear forensics but also geology and biology.

Log 593. **NEW APPROACH TO PRECISELY MEASURE GAMMA-RAY INTENSITIES FOR LONG-LIVED FISSION PRODUCTS**

N.D. Scielzo (1,P), K. Kolos1, D. E. M. Hoff1, M. Bencomo1, M.T. Burkey1, J.A. Clark2, J.C. Hardy3, V.E. Iacob3, D. Melconian3, E.B. Norman4, W.-J. Ong1, R. Orford4, D. Santiago-Gonzalez2, G. Savard2, M.A. Stoyer1, A. Tonchev1 (1) Lawrence Livermore National Laboratory, (2) Argonne National Laboratory, (3) Texas A&M University, (4) Lawrence Berkeley National Laboratory. (P) Presenting Author.

We have recently demonstrated a new experimental approach to precisely determine the gamma-ray intensities following the beta decay of long-lived fission products. For national-security applications, such as stockpile stewardship and nuclear forensics, one of the most straightforward and reliable ways to determine the number of fissions that occurred in a chain reaction is done via detection of the emitted gamma rays. The focus of this talk is on recent measurements to improve the nuclear-decay data for the fission products 95Zr, 144Ce, and 147Nd. For these isotopes, and many other fission products, the gamma-ray intensities are desired to high precision for these national-security applications. Our approach consists of implanting fission-product samples into a thin carbon foil using low-energy mass-separated ion beams from the CARIBU facility and then performing beta counting using a custom-made 4-pi gas proportional counter in coincidence with gamma-ray spectroscopy using the precisely-calibrated HPGe detector at Texas A&M University. Recent results for 95Zr, 144Ce, and 147Nd will be presented and future plans will be discussed. This work was supported under Contract DE-AC52-07NA27344 (LLNL), Office of Nuclear Physics Contract DE-AC02-06CH11357 (ANL), and DE-FG03-93ER40773 (Texas A&M).

Log 594. **U-238 FISSION PRODUCT MEASUREMENT USING THE LĀPAKI γ-γ ARRAY AT OREGON STATE UNIVERSITY**

Tamashiro, A.S. (1,2,P); Harke, J.T. (2); Palmer, C.J. (1); Menn, S. (1); Reese, S. (1); Minc, L. (1) . (1) Oregon State University. (2) Lawrence Livermore National Laboratory. (P) Presenting Author.

Fission product yields play an important role for nuclear reactor fuel cycle, nuclear reactor decay heat, and nuclear reactor waste inventory. We will be performing gamma-ray spectroscopy on beta-decaying fission products following a prompt fission neutron spectrum irradiation. Previous work has been performed using the Godiva-IV critical assembly to determine fission product yields starting 1 hour after irradiation and data was recorded for 7 days. In order to observe shorter-lived fission products, a new rabbit system and gamma-ray counting setup is being developed by Lawrence Livermore National Laboratory (LLNL) and Oregon State University (OSU) for use in a campaign of measurements on U-238 at the OSU TRIGA reactor. The counting setup will consist of eight BGO Compton-suppressed HPGe clover detectors connected to a new VME-based digital data acquisition system. Data will be recorded in list-mode to analyze time-dependant behavior of the gamma-spectra. The clover detectors have relative efficiency of 150% (to 3x3 in. NaI(Tl) crystal at 1.33 MeV) and the solid angle coverage of eight detectors is approximately 18% for the array. The U-238 samples will be delivered in front of the detectors directly from the reactor core via the rabbit facility. A study of different pure polyethylene sample materials from various suppliers at the OSU TRIGA reactor and the development of a new rabbit. Preliminary results from this measurement will be discussed.

Log 595. **TRACING SOURCES AND DISPERSAL OF ACTINIDES IN THE ENVIRONMENT USING BIOMONITORS**

Widom, E. (1,P); Kuentz, D.C.(1). (1) Miami University. (P) Presenting Author.

We have been investigating the utility of tree bark and lichen (± moss) as tracers of the source and geographic dispersal of actinides in the environment associated with diverse nuclear industry and munitions activities. Sample preparation and isotopic analysis of these biological materials is performed by ashing, microwave digestion, and chemical separation/purification by ion exchange chromatography, followed by thermal ionization mass spectrometry and/or multi-collector ICP-MS methods. Results from several case studies will be compared and contrasted, including those from the former Fernald Feed Materials Production Center, which produced uranium metal and intermediate compounds sourced primarily, but not exclusively, from natural uranium ore; the Miamisburg Mound Laboratories site, a former nuclear weapons research facility; the Portsmouth Gaseous Diffusion Plant, which previously produced highly enriched (weapons-grade) and low-enriched uranium; and the Pohakuloa Training Area, the site of a firing range at which depleted uranium spotting rounds were previously utilized for testing purposes. Collectively, our results demonstrate that tree bark and lichen are reliable long-term repositories of actinides, and that they retain information about the nature, extent and source of airborne environmental actinide contamination. Furthermore, analysis of these biomonitors can reveal the presence of ultra-trace levels of radionuclides in the environment, thus serving as highly sensitive monitors of any anthropogenic activities that might release actinides to the environment.

Log 596. **PULSE SHAPE DISCRIMINATION USING A COMPACT ASIC-BASED DATA ACQUISITION SYSTEM**

Wu, T. C (1,P); Li, V. A. (2); Sutanto, F. (2); Dazeley, S. (2); Jovanovic, I. (1). (1): University of Michigan Department of Nuclear Engineering and Radiological Sciences, Ann Arbor, MI, USA; (2) Lawrence Livermore National Laboratory, Livermore, CA, USA. (P) Presenting Author.

Neutron detector systems using silicon multiplier (SiPM) based detector systems require hundreds of channels, which necessitates the need for a high-channel-density and low-cost-per-channel data acquisition system. However, these systems lack full waveform digitization, and thus far pulse-shape discrimination (PSD) for discriminating different particle types has been difficult. This work demonstrates PSD on a channel of the TOFPET2 data acquisition system (PETsys Electronics), an ASIC-based data acquisition originally intended for positron emission tomography. A stilbene scintillator cube was placed on a pixel of a J-series SiPM with the TOFPET2 data acquisition and was exposed to gamma sources such as 137Cs, neutron sources such as DD and DT generators, as well as combined sources such as 252Cf. PSD was achieved by splitting the SiPM signal into two channels and integrating the charge for a different duration for each of the channels. For the stilbene cube exposed with a 252Cf source, a PSD figure of merit of 1.13 was achieved at 410 to 470 keVee with a short integration time of 131 ns and a long integration time of 421 ns. Gamma-only and neutron-only sources were used to verify that any PSD separation was indeed from different particle types: a pulse shape parameter of 0.30 was observed with gammas and 0.37 was observed for neutrons.

Log 597. **DECAY ENERGY SPECTROSCOPY FOR SAMARIUM CHRONOMETRY AND NUCLEAR MATERIAL ANALYSIS**

Kim, G.B. (1, P); Borg, L.E. (1); Boyd, S.T.P. (2); Cantor, R.H. (3); Despotopulos, J.D. (1); Drury, O.B. (1); Friedrich, S. (1); Gallant, A. (1); Hines, N.R. (1); Jacobs, A. (4); Jovanovic, I. (5); Kmak, K.N. (1); Kavner, A.R.L. (1,5); Kim, Y.H. (6); Kunz, P. (4); Kwiatkowski, A. (4); Kwon, D.H. (6); Lee, D. (1); Murb¨ock, T. (4); Scielzo, N.D. (1); Shollenberger, Q.R. (1); Sio, C.K.I. (1); Thomas, K.J. (1); Wooddy, T. (1); Walls, C. (4); Amelin, Y. (7); Parsons-Davis, T. (1); Brennecka, G.A. (1). (1) Lawrence Livermore National Laboratory, Livermore, CA 94550, USA. (2) University of New Mexico, Albuquerque, NM 87131, USA. (3) Star Cryoelectronics, Santa Fe, NM 87508,USA. (4) TRIUMF Laboratory, Vancouver, BC V6T 2A3, Canada. (5) University of Michigan, Ann Arbor, MI 48109, USA. (6) Center for Underground Physics, Institute for Basic Science, Daejeon 34047, Republic of Korea. (7) The Australian National University, Canberra, ACT 2601, Australia.

Decay energy spectroscopy (DES) is a technique that measures the total decay energies (Q values) of radioactive nuclides by directly embedding sources into cryogenic microcalorimeters. Each nuclide is identified from the high-resolution (few keV) decay-energy spectrum; the high resolving power of DES enables accurate analysis of the isotopic composition. Also, as the sources are fully embedded into detectors, DES provides a nearly 100% detection efficiency, which is ideally-suited for absolute activity measurements of nuclear materials. We employed the DES technique to re-measure the half-lives of ^{146}Sm and ^{147}Sm in order to improve the accuracy of Sm-Nd chronometers for early solar system chronology. The Sm sources are fully embedded into small Au foils and coupled to cryogenic magnetic microcalorimeters (MMCs). After the absolute decay counting is completed, the Sm sources will be chemically separated from the Au foils and quantified by thermal-ionization mass spectrometry to determine the number of Sm atoms embedded in each detector. Half-lives will be obtained by combining the activity results with the number of atoms determined from mass spectrometry. We will present experimental progresses on the ^{146}Sm and ^{147}Sm measurements. Additionally, DES results from uranium and plutonium isotopic analysis will be presented in the context of nuclear analytical applications.This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344. This work was funded by the Laboratory Directed Research and Development program of Lawrence Livermore National Laboratory (20-LW-024). This work was supported in part by the Department of Energy National Nuclear Security Administration, Consortium for Monitoring, Verification and Technology (DE-NE000863). The work at Institute for Basic Science is supported by Grant no. IBS-R016-A2.

Log 598. **SURFACE COATING ANALYSIS OFFERS INSIGHT INTO ALLEGED WWII GERMAN URANIUM CUBE PEDIGREE**

Robertson, B. (1,2,P); Fraga, C (1); McHugh, K. (1); Koeth, T.(3); Hiebert, M. (3); Palmer, C. (2); Schwantes, J. (1); (1) Pacific Northwest National Laboratory. (2) Oregon State University. (3) University of Maryland. (P) Presenting Author.

Efforts are underway to confirm the pedigree of three uranium metal cubes suspected to originate from Nazi Germany's nuclear program by measuring the casting dates using radiochronometry, comparing trace impurities with those from the two possible ore bodies under Nazi control in the 1940's, and analyzing cube surfaces for evidence of polystyrene or CN-based coatings used. Here, results of the latter are summarized. Briefly, surface samples from the cubes were placed in THF to extract polystyrene. These extractions were analyzed by GC-MS. In each of the three samples, significant quantities of polystyrene were detected. Two of the three samples also showed the presence of methyl methacrylate in addition to the polystyrene, suggesting these two samples may have originated from the same production facility.

Log 599. **QUANTIFYING THE POTENTIAL ARGON DETECTION CAPABILITIES FOR NUCLEAR EXPLOSION MONITORING**

Adhikari, P (1,P); Gordon, E.M. (1); Shah, K.A. (1); Eslinger, P.W. (2); Ely, J.H. (2); Miley, H.S. (2); Bowyer, T.W. (2); Haas, D.A. (1). (1) The University of Texas at Austin. (2) Pacific Northwest National Laboratory. (P) Presenting Author.

Current noble gas detection systems for nuclear explosion monitoring are based on the detection of four radioxenon isotopes - Xe-131m, -133, -133m, and -135. The data provided by radioxenon detection could be enhanced by other radionuclide signatures such as Ar-37. Activation of Ca-40 in rock by neutrons produces Ar-37, and monitoring for this additional nuclide could help distinguish detections of nuclear explosions from background sources such as medical isotope production. This work studies the capabilities of a hypothetical argon detection network. A 10 kt explosion was modeled using MCNP and SCALE to determine the inventory of Ar-37 created in a representative granite rock layer, assuming that either 0.1, 1, or 10% of the total inventory was released. The Ar-37 inventory was combined with atmospheric transport data from HYSPLIT compiled in a previous study, along with the detection limits of standard Ar-37 detection systems, to determine how many hypothetical monitoring stations would detect Ar-37 from the modeled explosion. This method was repeated for 365 HYSPLIT data sets to create a year's worth of hypothetical explosions, releases, and hypothetical Ar-37 detections. The study quantified the average number of detections per release, the number of stations detecting Ar-37, and the possibility of detecting Ar-37 in coincidence with xenon.

Log 600. **PULSED NEUTRON CHARACTERIZATION OF IRRADIATED FUELS AT LANSCE**

Vogel, Sven C. (1,P); Balke, Thilo (1,2); Bouman, Charles A. (2); Capriotti, Luca (3); Harp, Jason M. (4); Long, Alexander M. (1); Schaper, Danielle C. (1); Tremsin, Anton S. (5); Wohlberg, Brendt E. (1); Larson, Eric J. (1); Craft, Aaron E. (3); Gross, Brian J. (3); Carver, D. Travis (1); Angell, James R. (3); Mehta, Vedant K. (1). (1) Los Alamos National Laboratory. (2) Purdue University. (3) Idaho National Laboratory. (4) Oak Ridge National Laboratory. (5) University of California at Berkeley. (P) Presenting Author.

Neutrons offer bulk, non-destructive characterization of irradiated materials for which other bulk methods, e.g. X-ray diffraction or tomography, are not suitable due to the immense gamma background emitted from the samples. In particular, pulsed neutrons provide information from the ability to resolve the neutron energy or wavelength using their time-of-flight. This enables the potential to utilize neutron absorption resonance spectroscopy to characterize the spatial distribution of isotopes, so-called energy-resolved neutron imaging or neutron resonance imaging. Here, we report on characterization of an irradiated U-10Zr-1Pd fuel (6mm diameter, <2mm thick, 3R/hr dose rate) at LANSCE as well as our efforts to develop a cask enabling pulsed neutron characterization of entire irradiation capsules (<12mm diameter, <20cm length, 900R/hr dose rate), the so-call SHERMAN (Sample Handling Environment for Radioactive Material Analysis using Neutrons) cask.

Log 601. PREVENTING WHITE RESIDUE OR SOLIDIFIED Ca$_5$(PO$_4$)$_3$OH OBSERVED ON PLANCHETTES FOLLOWING ELECTRODEPOSITION WITH N,N-BIS(2-HYDROXYETHYL)-2-AMINOETHANESULFONIC ACID (BES) IN PLUTONIUM BIOASSAY.

Harris, M.N. (1,P); Debacker, K.B. (1); Hudston, L.A. (1); Zazueta, J.A.(1); Margiotta, C. (1); Zuniga, M.M. (1); Lamont, S.P. (1); Steiner R.E. (1). (1) Los Alamos National Laboratory.

A whitish residue coating on planchettes is periodically observed in plutonium bioassay following sample electrodeposition and is the result of unpredictable urine dynamics where significant levels of calcium, phosphoric acid, and/or phospho metabolites sometimes co-purity with the plutonium analyte following anion exchange. This coating covers the entire surface of the planchette and has been shown to attenuate plutonium isotopes during alpha counting resulting in poor recoveries of tracer and plutonium analyte. Studies of this white residue through X-ray fluorescence and X-ray diffraction have shown that the residue is high in calcium and phosphorous and has been identified as Ca5(PO4)3OH. During electrodeposition hydrogen evolution directly at the cathode increases the pH of the electrolytic solution and generates alkaline conditions, resulting in pH induced insolubility and precipitation of calcium phosphate directly onto the planchette surface. The precipitation of calcium phosphate can be prevented by the introduction of an appropriate buffer into the electrodeposition solution, which is capable of maintaining pH stability, thus preventing the formation of calcium insoluble HPO42- and PO43- species associated with high pH. Several buffers were tested for this purpose. It was discovered that a 150mM concentration of N,N-Bis(2-hydroxyethyl)-2-aminoethanesulfonic acid (BES, pKa 7.1) maintained proper pH control, was stable during electrodeposition, and was effective in preventing calcium phosphate precipitation onto the planchette. The use of this electrodeposition technique has greater implications within radiochemistry for sample types that have a tendency to co-purify insoluble components capable of precipitating onto planchettes if alkaline pH conditions are reached during electrodeposition.

Log 602. NON-RADIOACTIVE FISSION GAS ANALYSIS FROM HIGHLY IRRADIATED UO$_2$ FUEL

Liezers M. (1,P); Carman A.J.(1); Hilton C.D.(1); Reilly D.D.(1); Feldman J.D.(1); Springer K.W.(1) Eiden G.C.(1). (1) Pacific Northwest National Laboratory. (P) Presenting Author.

Stable Kr and Xe isotopes are produced at significant levels during the irradiation of nuclear fuel. The isotope ratio of these fission gases may retain information about sample irradiation history, but analyzing irradiated nuclear fuel usually falls inside the realms of operating equipment in hot cells and glove boxes, that can be both expensive and challenging. One solution to this problem is to make the sample very small and essentially non-radioactive, then the material can be analyzed on equipment in a non-rad setting. Size reduction also allows more detailed spatial measurements to be made across a sample. Results will be presented on the detection of fission Kr and Xe using a Noble Gas Mass Spectrometer from single irradiated UO2 particles down to a size of 1um (~10 pg UO2).

Log 603. **ICP-MS MEASUREMENTS OF U AND Np SEPARATED FROM Pu METAL**
Bartlett, J.H. and Aragon, S.M. (P). (P) Presenting Author.

Inductively-coupled plasma spectrometry (ICP-MS) is an analytical technique that has the capability to measure most elements at trace levels in different matrix solutions that have lower ionization potential the argon(Ar). The high sensitivity and large dynamic range of ICP-MS technique has the advantage over other element specific techniques such as atomic absorption or emission spectrometry. The Los Alamos National Laboratory (LANL) analytical chemistry group continues to process Pu metal samples for elemental analytes using ICP-MS techniques. Our team has recently incorporated a reductive extraction methnod for seperation of U and Np from Pu metal samples for anlaysis using ICP-MS. Results and data of ICP-MS measurements of U and Np in Pu metal using TEVA and UTEVA will be presented.

Log 604. **PNNL SUPPORT FOR NNSA'S Mo-99 CONVERSION PROGRAM**
Bowen, J.M.(1,P), Hayes, J.C.(1), Ritzmann, A.M.(1), Woods, V.T.(1), Fritz, B.G.(1), Arrigo, L.M.(1), Humble, P.H.(1), Gartman, B.N.(1), Reilly, D.D.(1). 1) Pacific Northwest National Laboratory. (P) Presenting Author.

The National Nuclear Security Administration's (NNSA) Office of Conversion (NA-231) has been providing assistance, including funding national laboratory support, for molybdenum-99 (99Mo) producers to convert from highly enriched uranium (HEU) targets to low enriched uranium (LEU) targets and encouraging domestic production. Pacific Northwest National Laboratory (PNNL) has been supporting this effort primarily through two activities: (1) emissions abatement for radioxenon and (2) iodine chemistry to improve capture of radioiodine. Support includes modeling traditional carbon delay beds, developing prototypes of a cooled abatement trap, optimizing iodine capture techniques, and characterizing iodine species under different radiochemical processing conditions.

Log 605. UNIQUE CAPABILITIES AND APPLICATIONS OF NEUTRON COUNTING MCP/TIMEPIX DETECTORS IN NEUTRON IMAGING AND DIFFRACTION EXPERIMENTS

A.S. Tremsin (1) in collaboration with many neutron imaging facilities. (1) The Unviersity of California at Berkeley. (P) Presenting Author.

In this talk we will discuss the capabilities of neutron counting detectors with Microchannel Plates combined with a Timepix readout for various applications in neutron imaging and diffraction. With such detectors implemented at spallation neutron sources it is now possible to measure simultaneously more than 250,000 neutron transmission spectra, each within a 55 μm pixel. Despite substantial limitations of this method (e.g. integration of materials characteristics along the direction of neutron beam propagation) this novel technique can be attractive for some studies where other techniques fail due to opacity of the materials or due to bulky sample environment equipment. We present the results of studies of microstructure and elemental composition within various polycrystalline and single crystal materials. These experiments enable mapping of residual strain, uniformity of texture and location of various crystalline defects as well as mapping the bulk elemental concentration, all non-destructively. Investigation of dynamic processes, such as water penetration into various porous materials, in-situ crystal growth and annealing of materials as well as neutron imaging of highly radioactive samples will also be presented.

Log 606. HYPERSPECTRAL NEUTRON CT WITH MATERIAL DECOMPOSITION

Balke, T. (1,2,P); Long, A. M. (2); Vogel, S. C. (2); Wohlberg, B. E. (2); Bouman, C. A. (1). (1) Purdue University. (2) Los Alamos National Laboratory. (P) Presenting Author.

Energy-resolved neutron imaging is an advanced neutron radiography technique capable of non-destructively extracting spatial isotopic information within a sample. In combination with uniquely characteristic isotopic neutron cross-section spectra, isotopic areal densities can be determined on a per-pixel basis, thus resulting in a set of areal density images for each isotope present in the sample. By performing measurements over several rotational views, an isotope-decomposed 3D computed tomography is possible. However, this objective is made difficult by having very low per-pixel neutron counts even after long scan times, significant background counts, and the non-linear effects of the finite neutron pulse width (moderator broadening). We demonstrate a model-based approach based on Poisson noise modeling that incorporates modeling of the background, moderator broadening and exposure variations. This approach allows for quantitatively accurate isotopic density estimates for both neutron time-of-flight radiography as well as tomography which is demonstrated in our experimental results collected for nuclear fuel samples on the FP-5 beamline at LANSCE.

Log 607. **RADIOCESIUM ANALYSES AT THE SAVANNAH RIVER SITE**

DiPrete, D.P. (1,P), DiPrete, C.C. (1), Jones, M. A.(1), Fenker, K. M.(1) (1) Savannah River National Laboratory. (P) Presenting Author.

Waste cleanup at the Savannah River Site has created an ongoing need to characterize the inventories of various radiological waste tank heels prior to tank closure. Each tank heel has unique chemical and radiological distributions, rendering the use of routine analyses inadequate. Since each tank is a completely new matrix with often-unforeseen interferences, method development must be performed for each tank's residue material. Due to its high neutron-induced uranium fission yield coupled with a 30 year half-life, Cs-137 is one of the most predominant beta-emitting isotopes in DOE radioactive waste inventories. While Cs-137 is simple to quantify via the intense 662 keV gamma ray associated with the Cs-137/Ba-137m decay, the high activity of this isotope in DOE waste streams can be problematic when quantifying other, significantly less abundant isotopes. Cs-135 has a high neutron-induced uranium fission yield, it is much less radioactive than Cs-137 due to its long (2.3E6 year) half-life. Cs-135 is also a pure beta emitter, requiring quantification by mass spectrometry. Trace elemental cesium and barium from reagent contamination become a concern. Ammonium molybdophosphate (AMP) is used extensively at SRNL as a Cs-removal agent in acidic solutions, but recovering extracted cesium from AMP is challenging. Crystalline silicotitanate (CST), calixarene compounds, namely calix[4]arene-bis-(t-octylbenzo-crown-6), and resorcinol formaldehyde provide attractive alternatives to AMP for analytical applications requiring the recovery of a cesium isotope as well as for reagent purification. Properties and applications of AMP and other specialized Cs-extractants for numerous radiochemical separation protocols for high activity residue will be discussed.

Log 608. **CHARACTERIZATION OF ACTINIDE PARTICLE EXPOSURE TO BIOTA USING SYNCHROTRON X-RAY BASED IMAGING TECHNIQUES**

Lind, O.C.(1,2,P); Brede, D.A.(1,2); Byrnes, I.B.(1,2); Garcia-Tenorio, R.(3); Janssens, K.(4); Nuyts, G.(4); Reinoso-Maset, E.(1,2); Rossbach, L.(1,2); Salbu, B.(1,2). (1) Norwegian University of Life Sciences. (2) Centre for Environmental Radioactivity (CoE CERAD). (3) University of Seville. (4) University of Antwerp. (P) Presenting Author.

Refractory radionuclides such as uranium (U) and plutonium (Pu) released from nuclear sources, including nuclear weapons tests and military or civilian nuclear accidents, are typically associated with radioactive particles, i.e., localized aggregates of radioactive atoms that give rise to an inhomogeneous distribution of radionuclides significantly different from that of the matrix background. The presence of such radioactive particles may have important radioecological and radioanalytical implications. Relatively large particles can carry a substantial amount of radioactivity and act as point sources of radiological concern, whereas submicron and nanometer sized particles are more mobile and potentially bioavailable. In the present work, a combination of radioanalytical methods and a suite of micro- and nanoanalytical X-ray techniques were employed to detect nanometer-micrometer sized actinide particles within biota following exposure in the environment or in laboratory-based toxicity tests as well as to characterize the exposure they represent to specific organs and cells. By utilizing these state-of-the art methods, the phenomenon of retention of actinide particles was demonstrated in snails living on U/Pu contaminated land in Palomares, Spain. Following biotest experiments involving U nanoparticle exposure, U biodistribution from whole organism to tissue level was determined for model organisms thus identifying potential target organs and tissues. By combining the high-resolution elemental biodistribution maps with spatially resolved biological endpoints such as in vivo redox status analysis and histology, U distribution could be linked with adverse effects.

Log 609. **COMPARISON OF NEUTRON ACTIVATION ANALYSIS AND PASSIVE COUNTING FOR ENVIRONMENTAL RADIOACTIVITY MEASUREMENTS**

Kaitschuck, N. (P);Bekker, JP.; Haas, D.A.; Landsberger.S. (P) Presenting Author.

Neutron activation analysis is not usually known to be used for environmental radioactivity measurements as compared to low-level passive gamma-ray counting. As part of the new project to design and build a 100-kW molten salt reactor, local background samples were taken as part of an environmental impact statement. The benefits and drawbacks of each method are explored for soil samples which are in secular equilibrium, including gamma-ray self attenuation, Compton suppression, sample size and counting time.

Log 610. RADIOCHRONOMETRY FOR NUCLEAR FORENSICS

Gaffney, A.M. (1,P). (1) Lawrence Livermore National Laboratory. (P) Presenting Author.

Radiochronometry is a powerful analytical tool for a nuclear forensic investigation because it can provide a predictive signature representing the amount of time that has passed since a nuclear material was last processed. In the uranium fuel cycle, as uranium is processed from ore to an oxide or metal form, it undergoes multiple purification steps and chemical phase transitions, any of which may partially or completely reset the uranium decay series clock of the material. For an investigator to confidently utilize a radiochronometric model age to reveal the history of a sample, several conditions must be met. First, the measurement must be sufficiently accurate and precise to meet the needs of the investigation. Improvements over the past decade in analytical methods and reference materials have enabled a robust analytical capability for many isotope systems used in radiochronometry. Second, the investigator must be able to confidently assume that the material was completely purified of decay products at some time in the past, and that the material has remained a closed system since that time, so that all decay product measured in the material now represents the complete record of ingrowth since the time that the material was last processed. The goal of our current research is to evaluate whether this assumption holds true for a range of uranium fuel cycle materials and quantify the extent to which model ages are reset, modified, or preserved throughout the fuel cycle. Prepared by LLNL under Contract DE-AC52-07NA27344, LLNL-ABS-831170.

Log 611. OPTIMIZING THE LANL CHROMATOGRAPHY CHEMISTRY FOR PURIFYING PROTACTINIUM FROM URANIUM-NIOBIUM METALS

Engel, J.R. (1, P); Denton, J.S. (1); Kayzar-Boggs, T.M. (1); Wende, A.M. (1), Cardon, A.R. (1), Edwards, M.E. (1). (1) Los Alamos National Lab. (P) Presenting Author.

Radiochronometry age-dating of interdicted nuclear materials is a cornerstone of nuclear forensics and supports nonproliferation missions. Generating reliable uranium (U) model ages requires separating trace amounts of the progeny isotopes – such as ^{231}Pa and ^{230}Th – from the bulk uranium matrix. While a number of separation chemistries exist for pure bulk-U materials, new inter-laboratory research between LANL, LLNL, and AWE seeks to extend ion-exchange chromatography procedures to complex U alloys. For example, weight-percent-level niobium (Nb) may be alloyed with U to give favorable neutron cross-sections in advanced reactor designs as well as provide excellent corrosion resistance. Unfortunately for forensics applications, the homologous chemical behavior of Nb and Pa hinders the separation of these elements resulting in challenges for high-precision mass spectrometry. In this contribution, we present optimized results from redesigning the LANL chromatography chemistry for purifying ^{231}Pa from uranium samples containing weight percent concentrations of Nb. We detail elution curves for maximizing the Pa/Nb separation factor from BioRad anion resin, AG 1-X8, using a range of ultra-dilute (0.001 – 0.05M) HF + 9M HCl solutions. Furthermore, we determine the carrying capacity of Nb on AG 1-X8, with implications for future resin-bed scaling based on experimental requirements. By separating the Pa and Nb fractions as much as possible in this initial anion step, we further purify Pa using subsequent silica-gel columns without bulk-Nb exceeding the column's retention capacity.

Log 612. **REAL-TIME RARE ISOTOPE HARVESTING, DETECTION AND IMAGING UTILIZING A HPGE NUCLEAR PHYSICS IMAGER**

Clause, H.K. (1,2); Domnanich, K.A. (1,2); Kleinfeldt, C. (1,2); Kalman, M. (1,2); Walker, W. (1,2); Vyas C. (1,2); Abel E.P. (1,2); Severin G.W. (1,2); Hull, E. L. (3); Kiser, M.R. (3); Longford, C.P.D. (3,P). Department of Chemistry, Michigan State University; (2) Facility for Rare Isotope Beams, Michigan State University; (3) PHDS Co. Knoxville.; (P) Presenting Author.

A double side HPGe strip detector (NP Imager) was utilized for real-time gamma spectroscopy, imaging and quantification of isotopes harvested from Kr-78 beam on a water target. A Kr-78 beam irradiation of a flowing water filled target system with aqueous & gaseous chemistry loops was monitored to detect, identify, and spatially quantify radioisotopes produced. The measurements were used to validate the mass transport model for Kr-76 and Kr-77 through the system. The NP-Imager germanium detector/imager provided HPGe gamma-ray spectroscopy and gamma-ray imaging at the same time to produce radiological-location maps of the distributions of isotopes before, during and after radiochemical production.
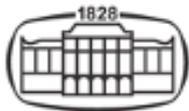
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