American Chemical Society

Division of Nuclear Chemistry and Technology

ABSTRACTS

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March 23-27, 2003
NUCL

DIVISION OF NUCLEAR CHEMISTRY & TECHNOLOGY

S. C. Srivastava, Program Chair

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SUNDAY MORNING

Section A
Convention Center -- Room 390

Synchrotron-Based Analytical Techniques for Nuclear and Environmental Sciences
Cosponsored with Division of Analytical Chemistry, and Division of Geochemistry

Inorganic Surface-Solution Interactions

S. Kelly and J. G. Catalano, Presiding
M. C. Duff, Organizer

8:30 — Introductory Remarks.

8:40 — 1. XAFS study of americium sorbed on groundwater colloids. Claude André Degueldre¹, Donald T. Reed², Arthur J. Kropf², and Andreas Laube³. (1) LWV - NES, Paul Scherrrer Institute, CH-5232 Villigen - PSI, Switzerland, Fax: +41-56-3102205, claude.degueldre@psi.ch, (2) Chemical Technology Division, Argonne National Laboratory, (3) Paul Scherrrer Institute, Nuclear Energy and Safety Division

The sorption of americium, as Am(III), on groundwater colloids obtained from a marl aquifer was studied in the 2x10⁻² M sodium bicarbonate groundwater and 2x10⁻² M sodium chloride bicarbonate-free solutions. At the in-situ groundwater pH of 8.6, the americium was strongly sorbed onto the colloids. XAFS analyses were performed on these sorbed Am species to establish the oxidation state and its near-neighbour bonding. These XAFS data, obtained at 400 mg l⁻¹ colloid concentrations and total Am concentration of 1.53x10⁻⁵ M (dissolved and onto colloids), indicated that Am remains trivalent, that surface complexes are formed with the colloids without surface precipitation. This conclusion is based on the absence of Am-Am interaction in the second or third shells. The surface complexes generated by the Am(III) sorbed on active sites were described on the basis of the XAFS data. They include the presence of about seven water molecules around the ternary surface complexes of this trivalent actinide.

9:00 — 2. Actinyl: From solution to model mineral surfaces. Christophe Den Auwer¹, Eric Simoni², Romuald Drot², Marie Christine Charbonnel¹, and Philippe Moisy¹. (1) Marcoule, CEA, Bagnols sur Cèze 30207, France, Fax: +4 66 79 63 25, (2) IPN Orsay

Evaluation of the actinide interactions with the geosphere is essential for the planning of their long term disposal. Consequently, structural and electronic investigation of the actinide-solution, surface interactions have always been a point of major interest. In a structural point of view, merging the molecular level description with the macroscopic data as sorption curves should lead to the understanding of the sorption or solvation mechanisms. In this work, the uranyl U(VI)O₂²⁺ cation has been mainly chosen because of its ubiquity in the geosphere and the most electrochemically stable behavior of this oxidation state in aqueous media. Model mineral surfaces as TiO₂ or SrTiO₃ have been selected for the simplicity of the crystallographic plane description. From the solution to the surface, this is really a site by site analysis that has been attempted here by probing the actinide coordination sphere using X-ray Absorption Spectroscopy.

9:20 — 3. EXAFS studies of the interaction of Pu(VI)O₂²⁺ with goethite. Richard E. Wilson¹, Dawn A. Shaughnessy², Corwin H. Booth², Yung-Jin Hu³, Philip A. Wilk³, and Heino Nitsche³. (1) Chemical Sciences Division, Lawrence Berkeley National Laboratory, and Department of Chemistry, Univ. of California, Berkeley, MS 70A-1150, 1 Cyclotron Rd, Berkeley, CA 94720, Fax: 510-486-7444, (2) The Glenn T. Seaborg Center, Chemical Sciences Division, Lawrence Berkeley National Laboratory, (3) Nuclear Sciences Division, Lawrence Berkeley National Laboratory, and Department of Chemistry, Univ. of California, Berkeley

The understanding of the migration and speciation of actinides in the geosphere is critical for the assessment and remediation of radionuclide contaminated sites. Sites contaminated with plutonium can pose both a chemical and radiological risk to surrounding populations and ecosystems. The molecular level interactions of
plutonium with common soil components are necessary for a better chemical understanding of these systems. The sorption of Pu(VI)O₂²⁺ on the iron oxide goethite was studied. Batch sorption experiments were performed at variable pH and at atmospheric carbonate concentrations. An extended x-ray absorption fine structure (EXAFS) spectroscopy study was conducted to identify the structure and oxidation state of the plutonium sorbed to the goethite surface at different pH and plutonium loading concentrations. EXAFS and sorption results are presented.


Disposition of High-level Waste (HLW) material produced from spent nuclear fuel reprocessing is a prime mission for the U.S. Dept. of Energy. Treatment designs at the Savannah River Site (Aiken, SC) include the use of monosodium titanate (MST) sorbent material to concentrate radiostrontium (Sr) and actinides from liquid HLW. We seek a greater understanding of the affinity of Sr and actinides in Na⁺-rich alkaline solutions for MST. We used XAFS spectroscopic techniques to characterize interactions between actinides and Sr with MST. Sorbed Sr exhibits specific adsorption as partially-hydrated species. Sorbed U(VI) exhibits specific adsorption as monomeric and dimeric U(VI) complexes. Plutonium [added as Pu(IV)] exists as colloidal species whereas Np [added as Np(V)] specifically adsorbs as polymeric species. Actinide sorption is site specific-occurring on distorted and perfect Ti octahedra. Differences in speciation and sorption mechanisms may account for difficulties associated with predicting actinide loading and removal kinetics using MST.

10:00 — 5. Pu(VI) reduction by Fe and Al metal in the presence of alpha radiolysis by products in brine solutions. Mei Ding¹, Patricia Paviet-Hartmann², Steven D. Conradson³, Phill Palmer⁴, James L. Conca², and Thomas Hartmann². (1) Isotope and Nuclear Chemistry Group, C-INC, Los Alamos National laboratory, Los Alamos, NM 87545, mding@lanl.gov, (2) Carlsbad Operations, EES-12, Los Alamos National Laboratory, (3) Materials Science and Technology Division, Los Alamos National Laboratory, (4) Isotope and Nuclear Chemistry Group, C-INC, Los Alamos National Laboratory

To confirm assumptions and refine parameters that may be used in the Performance Assessment of nuclear repository situated in deep geological salt formation, X-ray absorption fine structure (XAFS) spectroscopy was used to determine the oxidation state and chemical speciation of the reaction products between Pu(VI) and metallic Fe and Al in brine solutions in the presence of hypochlorite, produced by alpha radiolysis of TRU waste, which may oxidize Pu(III, IV) to Pu(V, VI). All our experiments show that Pu(VI) was reduced to Pu(IV) in four weeks. However, the spectra are neither identical to PuO₂ nor to Pu(IV) colloids formed by hydrolysis. Our results also show that Fe-reduced samples display broad but symmetric nearest neighbor O shells and small, narrow Pu shells, whereas Al-reduced samples exhibit complex nearest neighbor O shells and broad Pu distributions. The addition of NaOCl reduces the number of oxygen in the first coordination sphere around Pu(IV) in the Fe-reduced samples.

10:20 — Intermission.

10:40 — 6. Grazing incidence XAFS measurements of metal-cation sorption onto mineral surfaces. Melissa A. Denecke¹, Joerg Rothe², Kathy Dardenne², and Patric Lindqvist-Reis². (1) INE, Forschungszentrum Karlsruhe, P.O. Box 3640, Karlsruhe D-76021, Germany, Fax: +49-7247-823927, denecke@ine.fzk.de, (2) Forschungszentrum Karlsruhe

GIXAFS measurements of Hf(IV) sorbed onto both the (001) basal plane of mica and a surface oxidized silicon wafer and of U(VI) sorbed onto the (110) surface of alpha-Al₂O₃ are studied. EXAFS metrical parameters for Hf(IV) on both the mica (001) basal plane and on the oxidized silicon surface show that a mononuclear surface species forms. The cations are bound to silanol / aluminol groups present as defects on the mica surface in a fashion equivalent to binding to clay (hk0) edge sites. The polarization...
dependency of the XANES/EXAFS for the uranyl sorbed alpha-Al₂O₃ shows that the linear uranyl cations exhibit a preferred orientation on the (110) surface. Comparison of the XANES spectral trends with previously reported single crystal data and fits to the EXAFS allows identification of the reactive sites as the AlO₆ octahedral edges on the alpha-Al₂O₃ surface.

11:00 — 7. Sorption mechanisms of radionuclides on clay-mineral surfaces as determined by polarized X-ray absorption spectroscopy. **Rainer Daehn¹, Andre Scheidegger¹, Alain Manceau², and Daniel Grolimund³.** (1) Laboratory for Waste Management, Paul Scherrer Institute, Villigen 5232, Switzerland, Fax: +41563104438, rainer.daehn@psi.ch, (2) Lawrence Berkeley National Laboratory, Advanced Light Source, (3) Swiss Light Source, Paul Scherrer Institute

Sorption of heavy metal ions on mineral surfaces strongly affects the fate and mobility of heavy metals and radionuclides in the geosphere. Therefore an atomic/molecular level understanding on the surface reactivity of clay particles with respect to metal uptake is of fundamental importance for maintaining environmental quality and for assessing the long-term stability of waste repositories. On clay particles several uptake modes of metal ions have been proposed: Sorption on edge sites, sorption on interlayer sites and the formation of lamellar nucleation phases such as neoformed layer silicates and mixed layered double hydroxides. In this study powder and polarized EXAFS were used to investigate the uptake mechanisms of safety relevant radionuclides onto clay minerals important in the nuclear waste management.

11:20 — 8. Metal sorption onto hydrated iron oxides in anoxic environments: Role of Fe(II) ions, reaction time, and temperature. **Paras Trivedi¹, Donald Sparks¹, James A. Dyer², Noel Scrivner², and Kaumudi Pandya³.** (1) Department of Plant and Soil Sciences, University of Delaware, 152 Townsend Hall, Newark, DE 19717, Fax: 302-831-0605, paras@udel.edu, (2) DuPont Engineering Technology, (3) National Synchrotron Light Source, Brookhaven National Laboratory

The fate of metal contaminants including Pb and Cr in anoxic environments such as, ground water and subsurface waste storages, is not completely understood. In anoxic environments, both Fe(II) and Fe(III) species exist and interact with the metal contaminants. In this research, combined results of macroscopic studies and XAS studies suggest these metal ions chemisorb either directly onto the ferrihydrite and/or form ternary complexes with Fe(II) ions on the ferrihydrite surface. Because the local structure of Pb ions did not change up to 60 d of contact time, the slow sorption kinetics are attributed to intraparticle diffusion. In contrast, the changes in these sorption complexes, at elevated temperatures, result from the rearrangement of the Fe(II)-ferrihydrite complexes to form a stable Fe(II)-Fe(III) mixed oxide. Overall, these studies underline the need to include the interactions of metal contaminants with Fe(II) ions for assessing their mobility and bioavailability in anoxic aquatic environments.

11:40 — 9. Phosphate speciation on calcite and gibbsite surfaces: A combined X-ray absorption spectroscopy and solid-state NMR investigation. **Stefan Hunger,** and Donald L. Sparks, Department of Plant and Soil Sciences, University of Delaware, 149 Townsend Hall, Newark, DE 19717, Fax: 3028310605, hunger@udel.edu

Phosphate sorption in neutral to acid agricultural soils is controlled by Al and Fe (oxy)hydroxides, in more alkaline soils by reactions with Mg and Ca minerals. The current understanding is that phosphate forms surface complexes on metal hydroxide surfaces, and calcium phosphate or apatite surface precipitates on calcite. In limed agricultural soils, phosphate can react with both kinds of solid phases (calcium carbonate and Fe/Al-(oxy)hydroxide coatings). Furthermore, an elevated concentration of Ca cations exists in the soil solution in equilibrium with the calcium solids, which will influence phosphate sorption as well. Results are presented from an XAS and solid-state P-NMR investigation of the speciation of phosphate in ternary model systems containing both calcium carbonate and gibbsite or gibbsite and free calcium. These findings are combined with macroscopic sorption and dissolution data in order to better understand the reactions that determine mobility and bioavailability of phosphate in soils.
Section B
Convention Center -- Room 391

Environmental Restoration and Green Chemistry in Nuclear Technology
Cosponsored with Committee on Environmental Improvement

A. A. Ekechukwu, Organizer, Presiding
D. L. Hjeresen, Organizer

8:30 — Introductory Remarks.

8:40 — 10. Bench-scale testing of remedial technologies for NAPLs containing PCBs and chlorinated benzenes. Bryan R. Maurer, Cummings/Riter Consultants, 339 Haymaker Road, Parkway Bldg, Suite 201, Monroeville, PA 15146, Fax: 412-373-5242, bmaurer@cummingsriter.com

Bench-scale tests of potential remedial technologies were performed on NAPLs and NAPL-saturated soils containing PCBs and chlorinated benzenes from a CERCLA site in western Pennsylvania. Two destruction technologies were tested in a laboratory setting to determine if the technologies are capable of remediating the specific contaminants and concentrations found in site media. Solubility and viscosity tests using various solvents and at various temperatures were also performed to determine the viability of solvent washing and/or electrical resistivity heating. The bench-scale oxidation tests indicate a full-scale efficiency of approximately 5 pounds hydrogen peroxide per pound of organic contaminant, depending on the silt content of the impacted soils. The biological treatment tests indicated destruction of up to 89 percent of the organic contaminants within two weeks. Solvent washing with isopropanol indicated a transfer of up to 88 percent of the organic contaminants from the impacted soil to the solvent.

9:00 — 11. Field test of limestone as a treatment medium for groundwater at the Savannah River Site. Miles E. Denham, Frank C. Sappington, Gerald C. Blount, and Amy A. Ekechukwu, Savannah River Technology Center, Westinghouse Savannah River Company, Building 773-42A, Savannah River Site, Aiken, SC 29808, Fax: 803-725-7673, miles.denham@srs.gov

Groundwater downgradient from the F- and H-Area Seepage Basins at the Savannah River Site is contaminated with acids, metals, radionuclides, and tritium originally released as part of low-level waste streams from the Separations Areas processing facilities. To stem the flux of tritium discharged from the basin area, a series of wells was installed to extract the groundwater and re-inject it upgradient of the seepage basins. Tritium is captured in an extraction-re-injection cycle that allows it to decay. To meet regulatory criteria for re-injection, metals and radionuclides must be treated and removed. One method under consideration for this removal is in-situ contact with limestone. Equilibration of the groundwater with limestone raises the pH to approximately 8. At this pH, metals and radionuclides can be removed by direct precipitation, co-precipitation, and enhanced adsorption. A pilot test was performed to provide data required to assess the applicability of limestone in groundwater treatment. This demonstration unit consisted of several columns of limestone with different total column lengths and thus different residence times. Ground water from the basins was passed through each of the columns and analyzed for metals, radionuclides, ions, pH, and alkalinity. Results of this field test will be discussed.


Most strategies for remediating metals and radionuclides in groundwater focus on solid and aqueous phases. Yet the gas phase can also be an important component of remediation. Contaminants that can exist in volatile forms can be sparged from groundwater with air. Likewise, carbon dioxide manipulation can help stabilize contaminants, slowing their migration to exposure points. Finally, gas phase reagents can be used to stabilize inorganic contaminants.

Research at the Savannah River Site has shown that gas phase manipulation can be an effective remediation tool. Dissolved mercury was successfully removed from contaminated
groundwater by reduction with stannous chloride followed by air sparging. Similarly, calculations suggest that air sparging of diiodine may be effective for remediating iodine-129. Studies also suggest that some contaminants can be co-precipitated with calcium carbonate by reducing carbon dioxide partial pressure with air sparging after acidic groundwater reacts with limestone. Another approach, currently under study, is injection of phosphate in a gas phase to promote microbial stabilization of inorganic contaminants.

9:40 — Intermission.

10:00 —13. Field measurement of Cs-137 content in soil. Kenneth J. Hofstetter, Donna M. Beals, and Linda S. Nichols, Savannah River Technology Center, Westinghouse Savannah River Company, Building 735-A, Aiken, SC 29808, Fax: 803-725-4478, kenneth.hofstetter@srs.gov

A rapid technique has been developed to determine the Cs-137 content in contaminated soil using in-field gamma-ray spectrometry. Spectra from a 5 cm x 5 cm NaI(Tl) detector coupled to a portable multi-channel analyzer are recorded in the field, with and without shielding, to determine the Cs-137 content in a 2000 cm² area. The field surveys take less than 10 minutes at each location. The spectral data are analyzed and compared to model predictions based on postulated source distributions. An experimental method has also been developed using a down-hole NaI(Tl) probe for determining the vertical radionuclide distribution. The results of the calculated radioisotope content compare favorably to sample results taken at the same locations. The minimum detectable activity for Cs-137 is approximately 1 pCi/gm by this field technique. A discussion of the model development and comparison of the experimental and calculated results will be given.


The Ribbon NAPL Sampler (RNS) is a direct sampling device that provides continuous depth discrete mapping of Non Aqueous Phase Liquids (NAPLS) in a subsurface borehole. This technique uses the Flexible Liner Underground Technologies (FLUTE) membrane to deploy a hydrophobic absorbent ribbon. The system is placed into the borehole and the ribbon absorbs the NAPL that is in contact with it. A dye, sensitive only to NAPL, is impregnated in the ribbon and changes color when the contaminants are contacted. The presence and depth discrete location of DNAPL is indicated by physical color changes on the ribbon. The Ribbon NAPL Sampler can be deployed with direct push methods or drilling methods in both the vadose and saturated zones. The RNS has been deployed at numerous DNAPL sites, provides confirmation of the presence of NAPL and negates the necessity of bringing large volumes of soil core to the surface for analysis.

10:40 —15. Time-dependent retention of dichlorophenol and its polymerization products on soils. Monica Palomo, and Alok Bhandari, Department of Civil Engineering, Kansas State University, 2118 Fiedler Hall, Manhattan, KS 66506-5000, Fax: 785 532 7717, mpa7777@ksu.edu

Horseradish peroxidase mediated oxidative polymerization has been proposed as a means to clean up phenol contaminated soils and groundwater. This study evaluated the retention and binding of 2,4-dichlorophenol (DCP) and its polymerization products generated in the presence of peroxidase on two soils. DCP and its polymerization products were equilibrated for soil-solute contact times ranging from 0.083 to 84 days at initial aqueous concentrations ranging from 5 to 500 µM. ¹⁴C-DCP was used to improve detection limits and track the contaminant in soil and solution. Extractability of the parent DCP and its polymerization products was evaluated by subjecting the soils to water and methanol extractions. ¹⁴C activities associated with the humic acid, fulvic acid and humin/mineral fractions were also quantified. Retention and binding behavior of DCP and its polymers was expressed in terms of sorption isotherms, the Extractability Index (EI), and time-dependant distributions of the contaminant among various soil components.
SUNDAY AFTERNOON

Section A
Convention Center -- Room 390

Synchrotron-Based Analytical Techniques for Nuclear and Environmental Sciences
Cosponsored with Division of Analytical Chemistry, and Division of Geochemistry

Overviews, New Developments, and Microbial Interactions

K. R. Czerwinski, Presiding
M. C. Duff, Organizer, Presiding

1:00 — Introductory Remarks.


The new Environmental Remediation Sciences Division in the Office of Biological and Environmental Research at the Department of Energy has a strong interest in the application of state-of-the-art experimental techniques that use synchrotron radiation. The Division’s Natural and Accelerated Bioremediation Research (NABIR) program supports several research projects that use x-ray spectroscopy and micro-imaging to study interactions of microbes with heavy metals and radionuclides in the laboratory and in field samples. The Division’s Environmental Management Science Program (EMSP) has provided funding to infrastructure projects to enable research on radioactive samples at synchrotrons and supports several projects using x-ray techniques to study radioactive wastes and contaminated environmental samples. Examples of successful applications of synchrotron techniques in projects in these programs will be described. Plans for future support of synchrotron resources for environmental remediation research will be discussed.

1:35 — 17. Overview of applications of synchrotron-based techniques in environmental and nuclear science. Gordon E. Brown Jr., Department of Geological & Environmental Sciences, Stanford University, Building 320 Room 118, Stanford, CA 94305, gordon@pangea.Stanford.EDU

The availability of synchrotron radiation (SR) to the scientific community has revolutionized the way X-ray science is done in many disciplines, including environmental and nuclear sciences. The key reason is that SR provides continuum vacuum ultraviolet and X-ray radiation 5-10 orders of magnitude brighter than that from sealed or rotating anode X-ray tubes. This increase in brightness has resulted in many studies at the molecular scale that could not have been done using conventional X-ray sources and in a number of new discoveries about chemical and biological processes that determine the behavior and fate of trace elements, plant nutrients, and environmental pollutants in complex natural systems. This presentation will highlight some of these recent studies, including (1) sorption of heavy metals/radionuclides at mineral-water interfaces; (2) speciation of Cr, U, and Pu in contaminated soils and sediments at DOE sites; and (3) interaction of heavy metal(loid)s with biofilm-coated mineral surfaces.

2:05 — 18. Use of microXAS to study the influence of the inherent heterogeneity of nuclear-waste repository materials on radionuclide uptake. Andre Scheidegger¹, Daniel Grolimund², and Rainer Daehn¹. (1) Laboratory for Waste Management, Paul Scherrer Institute, Villigen 5232, Switzerland, Fax: +41 56 310 3171, Andre.Scheidegger@psi.ch, (2) Swiss Light Source, Paul Scherrer Institute

The research at the Laboratory for Waste Management concentrates on the understanding of safety relevant mechanisms and processes that govern the release of radionuclides from waste matrices. For this reason, detailed sorption studies of radionuclides in waste repository systems are
conducted. The studies are combined with X-ray Absorption Spectroscopy (XAS) measurements to understand the uptake mechanisms on a molecular level. In this study micro X-ray fluorescence (XRF) and micro XAS were used to gain spatially resolved information on the speciation and structural coordination environment of Co solidified in cement, a complex heterogeneous mixture with discrete particles typically in the size range of a few to a few hundred micrometers. Another example addresses the corrosion of iron waste canisters used in waste management. Under anoxic conditions zero-valent iron can react with water to produce a thin green rust layer that is highly reactive towards reduction processes of radionuclides.

2:25 — Intermission.


An elliptical polarization undulator (EPU) based soft x-ray user facility for molecular environmental science research (MES) is now available at the Advanced Light Source. The variable angle plane grating monochromator serves two branchlines. One branchline is optimized for scanning transmission x-ray microscopy (STXM) over the energy range of 180 to 1500 eV. High quality images from the STXM were obtained within days of “first light”. The other branchline serves specialized surface science end stations on a rotating platform, one capable of operating at up to 20 Torr, allowing experiments in the presence of water vapor or other high-pressure gas from 75 to 2000 eV. These end stations provide opportunities for the use of x-ray photoelectron, x-ray emission, and x-ray absorption fine structure spectroscopies for MES investigations. The characteristics, performance, and first results from the new beamline will be presented.

3:05 — 20. XAS and fundamental plutonium interaction processes with environmental microorganisms. Heino Nitsche1, Petra J. Panak2, Corwin H. Booth3, and David K. Shuh3. (1) University of California Berkeley, Department of Chemistry, and Nuclear Science Division, Lawrence Berkeley National Laboratory, MS 70R0319, Berkeley, CA 94720-8169, Fax: 510-486-7444, h_nitsche@lbl.gov, (2) Institute for Nuclear Waste Management, Forschungszentrum Karlsruhe, (3) Chemical Sciences Division, The Glenn T. Seaborg Center, Lawrence Berkeley National Laboratory

Microorganisms can interact with actinides by both direct interaction (biosorption, bioaccumulation, oxidation and reduction reactions) and indirect interaction (change of pH and redox potential). They may play an important role in the immobilization and mobilization of actinides in aquifers and subsurface environments. This talk will present examples of the interaction of aerobic soil bacteria with plutonium. Details of the nature of the bacterial functional groups involved in the interfacial actinide interaction process will be reported. Based on synchrotron X-ray absorption spectroscopy (XANES and EXAFS) measurements, and time-resolved laser-induced fluorescence spectroscopy (TRLFS) studies on the homologous uranium system, molecular-level mechanistic details of the different interaction processes will be discussed.

Acknowledgement: This work was supported by the Natural and Accelerated Bioremediation Research Program (NABIR), Office of Biological and Environmental Research (OBER), of the U.S. Department of Energy under Contract No. DE-AC03-76SF00098.

3:30 — 21. EXAFS study of uranyl adsorption to bacteria. Jeffrey A. Warner1, Tracy E. Letain2, Corwin H. Booth1, David K. Shuh1, and Heino Nitsche3. (1) The Glenn T. Seaborg Center, Chemical Sciences Division, Lawrence Berkeley National Laboratory, 1 Cyclotron Rd., Berkeley, CA 94720, Fax: 510-486-5596, jawarner@lbl.gov, (2) Earth Sciences Division, Lawrence Berkeley National Laboratory, (3) Nuclear Sciences Division, Lawrence Berkeley National Laboratory
Metal complexation reactions by bacteria can have a large effect on metal contaminant distributions in the environment. Uranium complexation by bacteria is of particular interest because of its possible importance in uranium transport and waste remediation. There have been a number of studies showing that bacteria can adsorb significant amounts of uranium from solution. Here, we present results on the identity of binding sites of uranium on a bacterial surface using temperature-dependant Extended X-ray Absorption Fine Structure (EXAFS) Spectroscopy. EXAFS was carried out at the uranium L3 edge for a series of uranyl phosphate and acetate model compounds and uranyl adsorbed onto bacteria. Uranyl adsorption onto bacteria was performed as a function of pH and uranium loading. No evidence was seen of either uranium reduction or precipitation under the conditions used. EXAFS fitting results will be discussed in detail.

3:50 —22: Chromium(VI) is reduced by sulfur nucleophiles and bacteria: Mechanistic studies using XANES spectroscopy. Murthy Vairavamurthy, Department of Energy Sciences & Technology, Brookhaven National Laboratory, Building 815, Upton, NY 11786, Fax: 631-344-5526, vmurthy@bnl.gov

The reduction of environmental Cr(VI) to Cr(III) constitutes a valuable mechanism of detoxification which can be accomplished by both abiotic and biotic processes. We studied the mechanism of abiotic Cr(VI) reduction by hydrogen sulfide and several thiols, which play a crucial role in reducing Cr(VI) in anaerobic environments. The biochemical pathway was examined using a highly resistant Bacillus sp. XANES spectroscopy performed at the NSLS revealed changes in the speciation of both chromium and sulfur. Our results suggest that intracellular sulfur plays a direct role in the bacterial transformation of environmental chromate.

Section B
Convention Center -- Room 391

Environmental Restoration and Green Chemistry in Nuclear Technology
Green Chemistry
Cosponsored with Committee on Environmental Improvement

D. L. Hjeresen, Presiding
A. A. Ekechukwu and D. L. Hjeresen, Organizers

1:30 — Introductory Remarks.

1:40 —23. Granulation technology for the minimization of radioactive contaminated waste. Donald Quintana¹, Fernando Roybal¹, Susan S. Ramsey², Jay Stimmel², and Kevin B. Ramsey³. (1) ESA-AET, Los Alamos National Laboratory, P.O. Box 1663/MS J576, Los Alamos, NM 87545, Fax: 505-667-0600, (2) NMT-7, Los Alamos National Laboratory, (3) NMT-15, Los Alamos National Laboratory

A granulation system will be deployed to volumetrically reduce transuranic solid waste generated at the Plutonium Facility at Los Alamos National Laboratory. Volume reduction through granulation technology can reduce the cost of waste packaging and disposal operations by at least 50 percent. Waste materials such as plastic bottles, tygon tubing, plastic bags, crucibles, and glass have been reduced up to 80 percent in volume through granulation. Analysis of the application of granulation technology to transuranic waste matrices indicates that not only is significant volume reduction possible for waste generated from this point on, but also that reprocessing of existing waste inventories will also be economic. Thus, granulation can have a very significant impact on the total volume of transuranic waste destined for the Waste Isolation Pilot Plant.

2:00 —24. CerOx process for on-site organic waste destruction: A nonthermal alternative to incineration. Norvell Nelson, and Stuart L. Nolan, CerOx Corporation, 2602 Airpark Drive, Santa Maria, CA 93455, Fax: 805-925-8218, njnelson@cerox.com
The CerOx process employs electrochemically generated Ce(IV) ions to oxidize organic materials in an electrolyte solution. In the course of the reaction, the reactive oxidant Ce(IV) ion is reduced to a Ce(III) ion. The Ce(III) is then recycled back through the electrochemical cell where it is oxidized back to the active Ce(IV) ion. As an electrochemical process, the CerOx process is readily scaled to match the waste volume produced. This non-thermal process is a technically and commercially viable alternative to incineration. Further, the process is performed in a series of tanks which makes it exempt from RCRA regulation. The compact size and self-contained operation of the process lends it to on-site operation for destruction of organic waste materials, including mixed waste materials, that could only be disposed by off-site incineration after transportation through our neighborhoods. The chemistry of the process will be described along with details of its operation on familiar laboratory waste streams.

2:20 —25. Expansion of Green is Clean Program at Nuclear Materials Technology Division facilities. Robert L. Dodge¹, Kathleen M. Gruetzmacher¹, Edward L. Horst¹, Steve Meyers², Susan S. Ramsey¹, and Richard M. Salazar¹. (1) NMT-7, Los Alamos National Laboratory, MS E501, Los Alamos, NM 87545, Fax: 505-667-9201, dodge@lanl.gov, (2) Eberline Services, Los Alamos National Laboratory

The operations within the radiological control areas of NMT Division generate approximately 262 m³ of LLW per year from the routine disposal of laboratory room trash. It is estimated that as much as 50% of LLW boxes are free of radioactive contamination and are suitable for inclusion in the Green Is Clean (GIC) waste program. The GIC program minimizes the generation of “suspect” low-level waste (LLW) by providing a standardized method of waste segregation that utilizes the concept of acceptable knowledge (AK) to free release waste from a radiological control area. Preliminary results of high sensitivity non-destructive assay (NDA) indicate greater than 40% of these items are free of radioactive contamination. Thus, the expansion of the GIC program to include laboratory room trash that is considered to be “suspect” LLW can significantly reduce the total volume of LLW generated at Los Alamos.

2:40 —26. Molecularly imprinted polymers for the specific rebinding of macrocyclic metal complexes via noncovalent interactions. Xiaobin Zuo, Donnati Mosha, Mansour M. Hassan, and Daryle H. Busch, Department of Chemistry, University of Kansas, 1251 Wescoe Hall Dr, Lawrence, KS 66045, Fax: (785)8645747, xiaobinzuo@hotmail.com

The specific binding of templated macroporous polymers is of much interest for separations science and enzyme-like catalysis. While covalently bound imprints work best, non-covalently imprinted polymers are attractive because of the easy removal of the imprint and rapid rebinding. We explore hydrogen bonding, electrostatic interactions, and minor metal-ligand interactions in new non-covalently imprinted polymers designed for the specific recognition of macrocyclic metal complexes. The imprinting of all $N,N',N''$-tetra(2-carbamoyethyl)cyclam-nickel(II) salts involve multiple hydrogen bonding interactions when combined during copolymerization with the functional monomer acrylamide or with the cross-linking monomer $N,N'$-ethylenebisacrylamide. Parallel experiments using the vinylsulfonate salt of the same macrocyclic complex add electrostatic interactions, and use of good coordinating groups as functional polymers (e.g. vinyl pyridines) adds the minor ligand component. Selectivity is being explored for a range of metal ions. The polymers are characterized by the extent of rebinding, selectivity, and morphological determinations.

3:00 — Intermission.

3:20 —27. Preorganized malonamide ligands: New ligands and materials for f-block ion binding. James E. Hutchison¹, Robert D. Gilbertson², Bevin W. Parks¹, B. P. Hay³, and Brian M. Rapko⁴. (1) Department of Chemistry and Materials Science Institute, University of Oregon, Eugene, OR 97403, hutch@oregon.uoregon.edu, (2) Department of Chemistry, University of Oregon, (3) Pacific Northwest National Laboratories, (4) Radiochemical Processing Group, Pacific Northwest National Laboratory
Application of green chemistry in nuclear technology is made possible by new advances in materials (such as selective, high affinity ligands) for more efficient sensors, separations and sequestration. Rational design of a new ligand architecture for f-block ion binding led to the discovery of a bicyclic malonamide ligand \((3,9\text-diaza-3,9\text-dialkylbicyclo[4.4.0]\text-decane-2,10-dione, 1)\), that exhibits dramatically enhanced binding affinities (ten million-fold greater) compared with typical acyclic malonamides (JACS 2002, 124, 5644). An efficient and convenient synthesis leading to a family of derivatives of the bicyclic malonamide 1 (with \(R=\text{alkyl or omega-substituted alkyl}\)) will be presented. The coordination chemistry with lanthanide (trivalent Eu, Nd, Gd) and actinide (uranyl) metal ions has been explored by single crystal x-ray structural studies to determine how the new architecture affects the stoichiometry and ligand geometry within the complexes. Incorporation of the ligand substructure into solid-phase materials (e.g. polymers) will also be described.

3:40 — Panel Discussion.

4:40 — Concluding Remarks.

MONDAY MORNING

Section A
Convention Center -- Room 390

Glenn T. Seaborg Award Symposium
L. G. Sobotka, Organizer, Presiding
W. Reviol, Organizer

8:30 — Introductory Remarks.

8:45 —28. Award Address Interplay of gamma-ray spectroscopy and reactions in nuclear-structure studies in the past 35 years. Demetrios G. Sarantites, Department of Chemistry, Washington University in St. Louis, Campus Box 1134, One Brookings Drive, St. Louis, MO 63130-4899, Fax: 314-935-6184, dgs@wuchem.wustl.edu

A short survey will be given of the developments in the use of nuclear reactions as an aid to nuclear structure studies via high resolution gamma-ray spectroscopy. Some recent results on superdeformation in the mass 80 and 40 regions will be presented. Emphasis will be given on transition quadrupole moments of the superdeformed structures in these mass regions. An outlook for structure studies of translead nuclei will be given. Finally a perspective will be given for nuclear structure studies in light mass nuclei with the oncoming GRETA array augmented by auxiliary detector systems.

9:15 —29. The impact of nuclear chemistry on nuclear medicine. Edward J. Hoffman, Dept. of Molecular and Medical Pharmacology, The David Geffen School of Medicine, 10833 Le Conte Ave., B2-096 CHS, Los Angeles, CA 90095-6948, Fax: 310-825-4517, ehoffman@mednet.ucla.edu

Methods used in Nuclear Chemistry form the basis of modern Nuclear Medicine. Isotopes are produced at accelerators and reactors, and techniques for fast chemical separation are used for short-lived isotopes. PET bears striking resemblance to crystal balls used to study multiplicity of gamma emission in nuclear reactions. PET is now a dominant imaging modality. Recently, new designs in photomultipliers, solid state detectors and detector materials for nuclear chemistry/physics have paved the way for new miniature instrumentation, that can be brought into the operating room to guide the surgeon to locate cancers and assure the removal of the lesion, or detectors that can be threaded up the coronary or carotid artery to identify plaque that can potentially cause heart attacks or strokes.


X-ray microbeams (MBs) are parallel arrays of ultra-thin (<100 nm) planar slices of synchrotron x rays. Single-exposure MBs are tolerated by normal brain of young and adult rats and ducks at 10-fold higher in-beam doses than broad beams. Furthermore, unidirectional MBs preferentially
ablate malignant intracranial rat brain tumors and subcutaneous murine mammary carcinoma tumors. The main hypothesis for the mechanisms underlying the MB effects is that both are mediated, at least in part, by the tissue's vasculature. Experimental results of the MBs' sparing of the central nervous system tissues and the skin will be presented together with the MBs' preferential tumoricidal results and preliminary mechanistic results. The method's potential for clinical use, Microbeam Radiation therapy (MRT), is also discussed.

10:15 — Intermission.

10:30 — 31. Neutron-rich radioactive beams at the HRIBF. J.R. Beene, Physics Division, Oak Ridge National Laboratory, P.O. Box 2008, Bldg. 6000, MS6368, Oak Ridge, TN 37831-6368, Fax: 865-574-1268, beenejr@ornl.gov

The world's first post-accelerated beams of heavy neutron-rich radioactive nuclei are now available at the Holifield Radioactive Ion Beam Facility (HRIBF). We have used these unique beams to carry out a number of experiments ranging from Coulomb excitation of first excited 2+ states, to transfer reactions and sub-barrier fusion. I will discuss some key features of the beam production systems we have developed and selected experimental results. Finally, I will mention some of the residual benefits that remain at HRIBF from the decade or so that Demetrios Sarantites was a regular user of our facility.

Oak Ridge National Laboratory is managed by UT-Battelle, LLC, for the U.S. Department of Energy under contract DE-AC05-00OR22725

11:00 — 32. Probing liquid-gas phase changes of light-to-medium mass nuclei. Y. Ma, and Joseph B. Natowitz, Cyclotron Institute, TAMU, 3366 TAMU, College Station, TX 77843, Fax: 979-845-1899, Natowitz@comp.tamu.edu, Natowitz@comp.tamu.edu

Light particle probes of the evolution of temperature and size indicate significant expansion of disassembling hot nuclei produced in heavy ion collisions. Apparent densities of ~ 0.4 times normal density are reached at higher excitation energies. The mass dependence of nuclear caloric curves suggests that light nuclei present the most favorable venue for observation of the critical point in a liquid-gas phase change of nucleonic matter. The extent to which the data for lighter systems manifest critical behavior is discussed.

11:30 — 33. High-resolution in-beam particle-gamma coincidence spectroscopy. Dirk Rudolph, Department of Physics, Lund University, Box 118, S-22100 Lund, Lund, Sweden, Fax: 46-46-2224015, Dirk.Rudolph@kosufy.lu.se

During recent years, the nuclear decay mode of discrete-energy prompt proton- and alpha-particle emission has been established in nuclei near $^{56}\text{Ni}$. Different from the ground-state proton emitters, the prompt particle emission competes with gamma rays instead of beta decay. The new decay mode proceeds from superdeformed initial states to spherical daughter states, which implies a drastic rearrangement of the nuclear mean field. Hence, it may be viewed as self-regulated two-dimensional quantum tunneling process, which is unique in Nature. I will report on recent results obtained with an unprecedented set-up aiming at combined high-resolution in-beam particle-gamma-gamma coincidence spectroscopy. They include the first observation of 'fine structure' for the new decay mode and involve states with competing alpha, proton, and gamma radioactivity. Future options and plans will also be outlined.

MONDAY AFTERNOON

Section A
Convention Center -- Room 390

Glenn T. Seaborg Award Symposium

W. Reviol, Presiding
L. G. Sobotka and W. Reviol, Organizers

1:30 — 34. From Spinspectrometer to Greta. I.Y. Lee, Nuclear Science Division, Lawrence Berkeley National Lab, One Cyclotron Rd., MS 88R0192, Berkeley, CA 94720-8101, Fax: 510-486-7983, iylee@lbl.gov
Study of gamma-ray emission from nuclei is one of the most powerful methods for the understanding of a broad range of nuclear properties. New discoveries of nuclear phenomena have come from major advances in gamma-ray detector technology. Spinspectrometer is the first 4-pi gamma ray detector array with a large number of NaI detectors, which represents a major increase in efficiency, and resolution in gamma-ray multiplicity and total energy. It was followed by arrays of Compton suppressed Ge detector, which give a much better energy resolution. Recent developments in highly segmented Ge detector, digital electronics, and computing power will enable us to construct an array entirely with Ge detector and to operate it as a gamma-ray energy track array (GRETA). I will review the design and the achievements of the existing arrays, and will discuss the concept, R&D status and scientific opportunities of GRETA.

2:00 — 35. Structure of neutron-rich Ti nuclei and shell closures above $^{48}$Ca. Robert V Janssens, Physics Division, Argonne National Laboratory, Bldg. 203, 9700 S. Cass Ave., Argonne, IL 60439, Fax: 630-252-6210, janssens@phy.Anl.gov

The level structure of neutron-rich Ti isotopes has been explored for the first time by combining beta-decay measurements from fragmentation products with prompt gamma-ray spectroscopy following deep-inelastic reactions. The latter technique was also instrumental in tracing $^{52-54}$Ti to high spin. The data provide new tests of effective interactions for full pf-shell calculations in neutron-rich nuclei above $^{48}$Ca. The data indicate the presence of a significant subshell gap at N=32 and comparisons between theory and experiment suggest an additional shell closure at N=34 in Ca and Ti isotopes.

*This work was supported in part by the U. S. Department of Energy, Nuclear Physics Division, under Contract No. W31-109-ENG-38.

3:00 — Intermission.

3:15 — 37 Ten years of Gammasphere physics. Augusto O. Macchiavelli, Nuclear Science Division, Lawrence Berkeley National Laboratory, MS 88R0192, Berkeley, CA 94720-8101, Fax: 510-486-7983, aom@lbl.gov

GAMMASPHERE, a gamma-ray spectrometer of unparalleled performance, is a National Facility funded by DOE. With its unique characteristics - high energy resolution, large efficiency and granularity - enhanced by a suite of auxiliary devices, the array had a major impact in the study of nuclear structure. As we approach 10 years of successful operation, that started with its Early Implementation Phase in 1993, we will review the rich and broad physics program carried out so far, including applications in nuclear structure, nuclear astrophysics and fundamental interactions.

3:45 — 38. Structure of exotic nuclei. W. Nazarewicz, Department of Physics and Astronomy, University of Tennessee, Knoxville, TN 37830, Fax: 865-574-4745, witek@utk.edu

There are many theoretical challenges related to nuclei far from stability. In many respects, weakly
bound exotic nuclei are much more difficult to treat theoretically than well-bound systems. The major theoretical difficulty and challenge is the treatment of the particle continuum. The residual-interaction coupling to the continuum can influence nuclear binding, effective interaction, and core polarization. It can give rise to a new class of collective phenomena. Continuum can also dramatically influence shell structure, many-body correlations (such as pairing) and can impact the appearance of cluster structures. Consequently, many cherished approaches of nuclear theory such as the conventional shell model must be modified in order to properly take into account unbound states. The main objective of this presentation is to discuss various challenges in theoretical nuclear structure. A review of the recently developed Gamow Shell Model will be presented. This work was supported by the U.S. DOE under Contract Nos. DE-FG02-96ER40963 (University of Tennessee), and DE-AC05-00OR22725 (Oak Ridge National Laboratory).

Section B
Convention Center -- Room 391

Analytical Chemistry in Nuclear Technology
Developments in Nuclear Chemistry and Cosponsored with Division of Analytical Chemistry

Counting Methods
D. E. Hobart, Organizer, Presiding
C. J. Coleman, D. T. Hobbs, and J. C. Griffin, Organizers

1:30 — Introductory Remarks.

1:35 — 39. Radionuclide-selective sensors for water monitoring: In situ sensor system for analysis of Tc-99 in Hanford groundwater. Oleg Egorov, Matt O'Hara, Jay W. Grate, Michael Knopf, and John Hartman, Environmental Molecular Sciences Laboratory, Pacific Northwest National Laboratory, P7-22, 902 Battelle Blvd, PO Box 999, Richland, WA 99352, oleg.egorov@pnl.gov

Production of nuclear weapons materials and storage of nuclear wastes at the U.S. Department of Energy (DOE) Hanford site has lead to the radioactive contamination of soil and groundwater. Monitoring of the radionuclide contaminants such as 99Tc in groundwater represents an important element in support of the remediation and long-term stewardship activities. Direct, isotope-specific determination (sensing) of pure beta-emitting 99Tc in aqueous matrix represents a substantial challenge due to short radiation range and energy overlap or interference problems. The research in our laboratories has been directed at addressing the fundamental challenges of monitoring of low levels of non-gamma-emitting radionuclides (e.g. Tc-99) in groundwater. We will present results on the development and testing of the reagentless Tc(VII)-selective radiometric sensor concept for in-situ detection of Tc in groundwater. The concept is based on the use of a dual-function composite sensor column designed to incorporate radionuclide selective uptake and scintillating properties. Sensor element, scintillation detection, sample delivery, data acquisition and instrument control components were integrated into a single functional unit compatible with the 3.5-inch well geometry.


Fission products from irradiation of U-235 and Pu-239 in Savannah River Site (SRS) reactors are currently being immobilized into a durable glass for final disposal. The products are in interim storage as caustic slurries in million gallons tanks along with other products of processing at SRS. Prior to immobilization, the concentrations of the fission products have to be measured or estimated in the waste. At Savannah River Technology Center appropriate samples of the slurries are dissolved remotely and the solutions analyzed by a variety of techniques. Concentrations of ~70 fission products have been measured. Maximum concentrations of others can be estimated using a fission yield scaling factor. This factor is calculated from those products whose chemical properties are such that their concentrations are directly proportional to their fission yields from either U-235 or Pu-239...
depending on the source of the waste. Results for a waste from Hanford will also be presented.

2:15 — 41. Direct measurement of actinides in liquids with PIPS diodes. R. Shane Addleman\(^1\), Oleg B. Egorov\(^2\), Matt O’Hara\(^3\), and Jay Grate\(^3\). (1) Environmental Technology Division, Pacific Northwest National Laboratory, P.O. Box 999, Richland, WA 99352, shane.addleman@pnl.gov, (2) Environmental Molecular Sciences Laboratory, Pacific Northwest National Laboratory

The determination of actinides in nuclear wastes, process solutions and various other liquid matrices is required in support of environmental restoration and waste processing activities. We will show that passivated implanted planar silicon (PIPS) diodes can provide rapid direct actinide assay of high level solutions without human exposure or waste generation issues. For low activity samples we will demonstrate direct isotopic assay of liquids for actinides with polymer membrane modified PIPS diodes. This synergistic combination of a chemically selective polymer film that concentrates ions of interest upon the diode surface and the spectral energy resolution available from the PIPS detector enables excellent isotopic selectivity and sensitivity. With thin films we have obtained high resolution alpha energy spectra directly from solution. Further, by using plasticizers that are also strong complexants, we have been able to selectively concentrate the actinides upon the diode surface resulting in a significant increase in sensitivity relative to direct analysis.

2:35 — 42. Determination of trace radionuclides in depleted uranium recovered from irradiated target element reprocessing. Donna M. Beals, Steven P. LaMont, James R. Cadieux, Chuck R. Shick, and Greg Hall, Savannah River Technology Center, Westinghouse Savannah River Company, Building 735-A, Aiken, SC 29808, Fax: 803-725-4478, donnabeals@msn.com

Savannah River Site depleted uranium (DU) required the radioactivity level of \(^{241}\text{Am},^{226}\text{Ra},^{137}\text{Cs},^{129}\text{I},^{99}\text{Tc}\) and \(^{90}\text{Sr}\) to be determined prior to off-site waste shipment. Methods were developed to measure these radionuclides with detection limits on the order of a few tens of pCi/g of DU. After chemical separation, \(^{241}\text{Am},^{226}\text{Ra},^{137}\text{Cs}\) and \(^{129}\text{I}\) were determined by gamma spectroscopy in an ultra low-level counting facility. Strontium-90 was measured by gas flow proportional counting, \(^{99}\text{Tc}\) by liquid scintillation counting, and \(^{237}\text{Np}\) and \(^{238}\text{Pu}\) by alpha spectrometry. The remaining Pu isotopes were determined by high sensitivity thermal ionization mass spectrometry. Technetium-99 was the only radioisotope detected significantly above the detection limit. The uranium isotopic abundances were determined by alpha spectrometry; selected samples were analyzed by mass spectrometry in order to determine the \(^{236}\text{U}\) separately from the \(^{235}\text{U}\).

2:55 — Intermission.

3:10 — 43. Determination of waste-acceptance radionuclides in DWPF sludge batch two N. E. Bibler, J. R. Harbour, T. L. Fellinger, and D. P. Diprete, Savannah River Technology Center, Westinghouse Savannah River Co, Aiken, SC 29808, Fax: 803-725-4704, ned.bibler@srs.gov, john.harbour@srs.gov

The Defense Waste Processing Facility (DWPF) at Savannah River Site is currently processing HLW Sludge Batch 2 for geologic disposal. The Waste Acceptance Product Specification 1.2 for disposal requires that radionuclides that have half lives greater than 10 years and contribute more than 0.05 \% of the radioactivity at any time from production through the 1100-year period between 2015 and 3115 be reported. A sample of Sludge Batch 2 was extensively analyzed at Savannah River Technology Center and concentrations of 40 radionuclides were determined or estimated. Calculations of radioactive decay and daughter production for these radionuclides were then preformed for every 100 years out to the year 3115. As a result twenty-nine radionuclides were identified as reportable. The radionuclides include neutron activation products, \(^{235}\text{U}\) fission products, and actinides. Examples are \(^{59}\text{Ni},^{63}\text{Ni},^{79}\text{Se},^{99}\text{Tc},^{121}\text{Sn},^{233}\text{U},^{237}\text{Np},^{239}\text{Pu},^{239}\text{Am},^{246}\text{Cm}\). Details will be discussed.

3:30 — 44. Equipment and process development for fabricating plutonium primary analytical chemistry standards. Gerald Coriz, NMT-11: Actinide Fuel Cycle Technology Group, Los Alamos National Laboratory, PO Box 1663, MS
The focus of this talk will be a discussion of the techniques and equipment used in the fabrication of High-Purity Plutonium Primary Analytical Standards. The goal of the Plutonium Standards Fabrication Project was to fabricate 1200 nominal 1 gram samples of high purity plutonium (Pu) metal, sealed in evacuated glass ampoules. Equipment developed for this process included a hydraulic press and pump, microprocessor controller and data recorder, tool-steel extrusion die and resistance band heater, and a rebar cutter and glass sealer. Process steps included casting, machining, wire extrusion, wire cutting, encapsulation, and delivery. The plutonium metal samples will be characterized at the Los Alamos National Laboratory (LANL) and New Brunswick Laboratory (NBL) followed by a statistical evaluation of the analytical chemistry data by NBL. NBL will then issue the metal samples to the nuclear community as a plutonium metal certified-reference material for plutonium assay. These high purity 1 gram Pu metal samples will be used as primary analytical standards across the Department of Energy (DOE) complex and the world.

3:50 —45. Counting techniques applied to fabrication of nondestructive assay standards for the National TRU Waste Program. Sandra L. Mecklenburg 1, Amy S. Wong 2, and Denise L. Thronas 2. (1) Actinide Analytical Chemistry, Los Alamos National Laboratory, P.O. Box 1663, MS G740, Los Alamos, NM 87545, Fax: 505-665-4737, meck@lanl.gov, (2) NMT-9, Los Alamos National Laboratory

The National TRU Waste Program requires traceable nuclear material standards for certification of non-destructive assay instrumentation used to quantify nuclear material in DOE-generated waste before shipment for final disposition at the Waste Isolation Pilot Plant (WIPP). Over 170 standards containing various isotopes of plutonium, americium, or uranium dispersed in an inert matrix have been fabricated and certified. A number of counting techniques were employed in characterization of the feedstock nuclear materials, as well as in characterization of the completed standards. Radiochemical analyses of americium, neptunium, uranium, californium, and curium content of starting materials were performed using gross alpha counting, alpha spectroscopy, and gamma-ray spectroscopy. Homogeneity of nuclear material-matrix blends was measured before encapsulation by gamma-ray methods. Holdup measurements were performed on fabrication tools used to more accurately quantify the nuclear material content of each standard. Finally, gamma-ray measurements were performed on completed standards to verify their uniformity and content.

4:10 —46. Microdosimetry studies of plutonium oxides. Lav Tandon 1, Alice K. Slemmons 1, John Blackadar 1, Jay A. LaVerne 2, John D. Robertson 3, Jeffrey Schwarz 2, Anthony J. Sanchez 2, and David E. Hobart 1. (1) Chemistry Division, Los Alamos National Laboratory, Mail Stop G740, Los Alamos, NM 87545, Fax: 505-665-4737, tandon@lanl.gov, aslemmons@lanl.gov, (2) Notre Dame Radiation Laboratory, University of Notre Dame, (3) Department of Chemistry, University of Missouri, (4) Department of Chemistry, University of Kentucky

Important issues surrounding long-term storage of plutonium are corrosion and gas generation that lead to ultimate failure of the stainless steel containers. The chemical reactions used to model these radiolytic effects depend upon the dose (energy) escaping from these materials. An important variable in calculating and modeling these effects is the physical particle size of the plutonium oxide. The average alpha particle energy and dose emitted by varying particle size fractions of PuO2 has been studied experimentally in the present work and will be presented. Alpha spectrometry, ionization chamber counting, and gamma spectroscopy are being used to measure the alpha energy, total energy, and isotopic composition of the materials. The present results are novel because the dose fraction actually emitted by the various particle sizes has previously only been predicted, based on spherical models, rather than actually determined experimentally. Comparisons of experimental data with these physical models will be discussed.
SYNCHROTRON-BASED ANALYTICAL TECHNIQUES FOR NUCLEAR AND ENVIRONMENTAL SCIENCES CHARACTERIZATION

COSPONSORED WITH DIVISION OF ANALYTICAL CHEMISTRY, AND DIVISION OF GEOCHEMISTRY

D. K. SHUH AND M. A. DENECKE, PRESIDING
M. C. DUFF, ORGANIZER

1:00 — Introductory Remarks.

1:05—47. X-ray spectroscopic and diffraction study of the distribution and speciation of uranium in contaminated sediments from the DOE’s Hanford site. Jeffrey G. Catalano1, M. Zachara2, S. M. Heald3, and Gordon E. Brown Jr.1. (1) Department of Geological and Environmental Sciences, Stanford University, Bldg. 320, Room 118, Stanford, CA 94305-2115, Fax: 650-725-2199, catalano@pangea.stanford.edu, (2) Environmental Molecular Science Laboratory, Pacific Northwest National Laboratory, (3) Environmental Molecular Sciences Laboratory, Pacific Northwest National Laboratory

Extensive uranium contamination of the subsurface at the Department of Energy’s Hanford site has occurred through the leakage of significant quantities of high-level nuclear waste. An understanding of the speciation and distribution of uranium in the vadose zone is needed to predict the future migration of uranium into groundwater. We have applied synchrotron-based x-ray spectroscopic and diffraction techniques to characterize the speciation of uranium in contaminated sediments from the Hanford site. X-ray fluorescence microprobe ( -XRF) studies show uranium distributed heterogeneously, occurring in discrete particles <5 m in diameter. X-ray absorption fine structure (XAFS) spectroscopic studies demonstrate that >95% of the uranium in all samples occurs as U(VI), and that the primary uranium species in most locations is an uranophane-group mineral, most likely boltwoodite. These studies suggest that boltwoodite is the primary phase controlling the geochemistry of U at the Hanford site. X-ray microdiffraction studies of these samples will also be presented

1:25—48. X-ray microtomography study of metal distribution in sediments related to pore structure modification by mineral dissolution and neophase formation under extremely alkaline conditions. Soh-joung Yoon1, Keith W. Jones1, Antonio Lanzirotti2, Huan E. Feng3, Wooyong Um4, R. Jeffrey Serne5, K. G. Karthikeyan5, and William F. Bleam6. (1) Environmental Sciences Department, Brookhaven National Laboratory, Building 901A, Brookhaven National Laboratory, Upton, NY 11973, Fax: 631-344-5271, syoon@bnl.gov, (2) University of Chicago, (3) Earth and Environmental Studies, Montclair State University, (4) Applied Geology and Geochemistry, Pacific Northwest National Laboratory, (5) Department of Biological Systems Engineering, University of Wisconsin, Madison, (6) Soil Science, University of Wisconsin-Madison

REDOX tank waste streams at the Hanford DOE sites are extremely alkaline and silicate minerals in surrounding sediments would dissolve and secondary solid phases would form as the alkaline solution with a high aluminum concentration infiltrates. These processes may modify the pore structure of the sediments, subsequently affecting the solute transport. In this study, simulated tank waste solutions comprising high levels of hydroxide, aluminum, sodium, and nitrate are flowed through miniature sediment columns with mineral phases commonly occurring in Hanford sediments under a saturated flow condition. The pore structure modifications are examined by x-ray absorption microtomography study and the distributions of metal contaminants in the columns are observed by x-ray fluorescence microtomography study at the National Synchrotron Light Source. We also determine whether these metal distributions are related with silicate neophase distributions using germanate ions as tracers for dissolved silicate ions by x-ray fluorescence study.


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microdiffraction studies of these samples will also be presented

1:05—47. X-ray spectroscopic and diffraction study of the distribution and speciation of uranium in contaminated sediments from the DOE’s Hanford site. Jeffrey G. Catalano1, John M. Zachara2, S. M. Heald3, and Gordon E. Brown Jr.1. (1) Department of Geological and Environmental Sciences, Stanford University, Bldg. 320, Room 118, Stanford, CA 94305-2115, Fax: 650-725-2199, catalano@pangea.stanford.edu, (2) Environmental Molecular Science Laboratory, Pacific Northwest National Laboratory, (3) Environmental Molecular Sciences Laboratory, Pacific Northwest National Laboratory

Extensive uranium contamination of the subsurface at the Department of Energy’s Hanford site has occurred through the leakage of significant quantities of high-level nuclear waste. An understanding of the speciation and distribution of uranium in the vadose zone is needed to predict the future migration of uranium into groundwater. We have applied synchrotron-based x-ray spectroscopic and diffraction techniques to characterize the speciation of uranium in contaminated sediments from the Hanford site. X-ray fluorescence microprobe ( -XRF) studies show uranium distributed heterogeneously, occurring in discrete particles <5 m in diameter. X-ray absorption fine structure (XAFS) spectroscopic studies demonstrate that >95% of the uranium in all samples occurs as U(VI), and that the primary uranium species in most locations is an uranophane-group mineral, most likely boltwoodite. These studies suggest that boltwoodite is the primary phase controlling the geochemistry of U at the Hanford site. X-ray microdiffraction studies of these samples will also be presented

1:25—48. X-ray microtomography study of metal distribution in sediments related to pore structure modification by mineral dissolution and neophase formation under extremely alkaline conditions. Soh-joung Yoon1, Keith W. Jones1, Antonio Lanzirotti2, Huan E. Feng3, Wooyong Um4, R. Jeffrey Serne5, K. G. Karthikeyan5, and William F. Bleam6. (1) Environmental Sciences Department, Brookhaven National Laboratory, Building 901A, Brookhaven National Laboratory, Upton, NY 11973, Fax: 631-344-5271, syoon@bnl.gov, (2) University of Chicago, (3) Earth and Environmental Studies, Montclair State University, (4) Applied Geology and Geochemistry, Pacific Northwest National Laboratory, (5) Department of Biological Systems Engineering, University of Wisconsin, Madison, (6) Soil Science, University of Wisconsin-Madison

REDOX tank waste streams at the Hanford DOE sites are extremely alkaline and silicate minerals in surrounding sediments would dissolve and secondary solid phases would form as the alkaline solution with a high aluminum concentration infiltrates. These processes may modify the pore structure of the sediments, subsequently affecting the solute transport. In this study, simulated tank waste solutions comprising high levels of hydroxide, aluminum, sodium, and nitrate are flowed through miniature sediment columns with mineral phases commonly occurring in Hanford sediments under a saturated flow condition. The pore structure modifications are examined by x-ray absorption microtomography study and the distributions of metal contaminants in the columns are observed by x-ray fluorescence microtomography study at the National Synchrotron Light Source. We also determine whether these metal distributions are related with silicate neophase distributions using germanate ions as tracers for dissolved silicate ions by x-ray fluorescence study.

The rapid microbial reduction of soluble U(VI) to U(IV) as insoluble uraninite is an attractive remediation strategy for U(VI)-contaminated groundwaters. However, the factors affecting U(VI) reduction have not been fully explored. Recently, Ca$^{2+}$ in solutions with U(VI) has been shown to inhibit the reduction of U(VI), purportedly through the formation of a Ca-UO$_2$-CO$_3$ complex. These complexes have been largely overlooked, because, most equilibrium speciation models predict the dominant uranyl species in calcareous groundwater to be uranyl carbonates. X-ray absorption fine-structure (XAFS) measurements of U(VI) in solutions of Ca$^{2+}$ and CO$_3$$^{2-}$, with and without acetate, indicate that Ca-UO$_2$-CO$_3$ complexation dominates. XAFS measurements of several solutions with various U:Ca concentration ratios show that the number of Ca atoms complexed to UO$_2$ (CO$_3$)$_3$$^{4-}$ decreases as the ratio of U(VI) to Ca decreases from approximately 1:100 to 1:20. At higher U:Ca ratios the dominant species is Ca$_3$(UO$_2$)(CO$_3$)$_3$$^{2+}$.

2:25 — Intermission.

2:45 — 51. Aqueous colloids from corrosion of metallic uranium fuel: A SAXS and TEM study. Jeffery A. Fortner$^1$, C. J. Mertz$^1$, Margaret M. Goldberg$^1$, and Sönke Seifert$^2$. (1) Chemical Technology Division, Argonne National Lab, 9700 South Cass Avenue, Argonne, IL 60439, fortner@cmt.anl.gov; (2) Chemistry Division, Argonne National Laboratory

Metallic uranium fuel from the Hanford N- Reactor represents a substantial portion of the DOE-owned spent fuel inventory. While it is widely observed that the aqueous corrosion of metallic uranium fuels is generally rapid, little attention has been paid to the generation of uranium-containing colloids arising from this corrosion. We describe measurements on aqueous colloids formed during contact with solution chemistry conditions ranging from deionized water to simulated repository groundwater (silicate/carbonate saturated). Colloids were analyzed by inductively coupled plasma mass spectrometry, transmission electron microscopy, photon correlation spectroscopy, and synchrotron small-angle x-ray scattering. We find that initially, stable suspensions of small (1-10 nm diameter), spherical uranium oxides are generated and aggregate to approximately 100-200 nm colloids. In silicate solutions, acicular uranium silicate colloids are eventually formed in small quantities, as are uranium-bearing smectite clay colloids. Low-silicate/high carbonate solutions are found to generate both colloidal uranium carbonate and uranium-bearing calcite.
3:05 —52. XANES study of iron and uranium oxidation states in Nevada Test Site nuclear melt glasses. Erik J. Nelson¹, Timothy P. Rose², Patrick G. Allen¹, and Frederick J. Ryerson³. (1) Seaborg Institute for Transactinium Science, Lawrence Livermore National Laboratory, P.O. Box 808 L-231, Livermore, CA 94551, Fax: 925-422-3160, nelson87@llnl.gov, (2) Analytical and Nuclear Chemistry Division, Lawrence Livermore National Laboratory, (3) Geosciences and Environmental Technologies Division, Lawrence Livermore National Laboratory

The redox state of multivalent elements in nuclear melt glasses is an important factor in determining the solubility and transport of uranium and transuranics from these glasses in local groundwater. Synchrotron-based x-ray absorption near-edge spectroscopy (XANES) measurements are used to determine the oxidation states of iron and uranium in three nuclear melt glass samples from the Nevada Test Site (NTS). Comparison of the U LIII and Fe K edge XANES data to standards of known oxidation state indicate that uranium and iron are in mixed valence states (U⁵⁺/U⁶⁺ and Fe²⁺/Fe³⁺). The proportions of Fe²⁺ and Fe³⁺ in the glasses are quantified by fitting the Fe K pre-edge feature. The melt glass samples contain between 33 and 53 at% Fe²⁺, implying moderately reducing conditions. The uranium and iron edge XANES results coupled with published redox studies of synthetic glasses suggests that plutonium is present as Pu⁴⁺ in the melt glasses.

Specimens of uranyl oxyhydroxide alteration phases derived from CSNF were examined by synchrotron x-ray absorption spectroscopy (XAS) to better determine neptunium uptake in these phases. Neptunium fluorescence was not observed and we firmly establish upper limits for Np incorporation that are significantly below previously reported concentrations obtained using electron energy loss spectroscopy (EELS). We attribute the discrepancy to a peculiar plural-scattering event that creates a spurious EELS peak at the Np-M5 energy.

3:45 —54. EXAFS characterization of uranium speciation in an amorphous metallic matrix
Virginia L. Curran, Nuclear Engineering Department, Massachusetts Institute of Technology, 290 Ferry St, Everett, MA 02149, Fax: 617-253-7300, ginipig@mit.edu, Ken R. Czerwinski, Department of Nuclear Engineering, Massachusetts Institute of Technology, and Patrick G. Allen, G. T. Seaborg Institute for Transactinium Science, Lawrence Livermore National Lab

Three metallic slag samples recovered from a site several decades after their inadvertent creation were analyzed using x-ray absorption spectroscopy techniques to determine the speciation of any uranium present. The samples contained highly enriched uranium in concentrations up to 2 wt%. Elemental composition of various sections of each sample was determined using ICP-AES and ICP-MS. High lead content limited the analysis of some of the specimens to short k ranges. Analysis of XANES and EXAFS spectra showed the uranium was a mixture of uranyl and a less oxidized species bonded with aluminum, with phase composition ranging from 33% to 50% uranyl. Results of the analysis were used to aid in remediation of the original contaminated site.
56. **Novel solid and liquid dosimetry systems.** Lav Tandon, Alice K. Slemmons, Wolfgang Runde, Velma M. Montoya, Lee J. Trujillo, Stephanie E. Field, and Apara R. Dave, Chemistry Division, Los Alamos National Laboratory, Mail Stop G740, Los Alamos, NM 87545, Fax: 505-665-4737, tandon@lanl.gov, vmontoya@lanl.gov

To address the issues surrounding long-term storage of highly radioactive plutonium-bearing materials, researchers at Los Alamos National Laboratory are studying the radiolytically induced corrosion and gas generation in stainless steel storage containers. The chemical reactions used to model these radiolytic effects depend upon the nature and energy of the radiation released from these materials. Because the stored materials may have significant amounts of adsorbed water layers on the particles, it is important to estimate the dose received by the physi- and chemisorbed water layers. Thus, novel chemical dosimeters are being used to determine the total amount of chemical species generated that can potentially lead to chemical reactions external to the particle. Other solid and liquid chemical dosimeters have been studied for its application as non-destructive detection of radioactive material. Irradiation experiments conducted with alpha, beta, gamma, and neutron sources will be presented and discussed.

57. **Temperature-dependent heat-source effects in cooling nuclear reactors.** Kal Renganathan Sharma, Department of Chemical and Biotechnology, Anna University - Sakthi Engineering College, Principal, Mathur Post, Oragadam, Kancheepuram, Chennai 602105, India, Fax: 2526019, jyoti_kalpika@yahoo.com

Nuclear power plant uses heavy water as coolant or liquid metals. The rate of heat removal has to equal or be greater that that generated during the fission reactions. The transient heat conduction using the Cattaneo and Vernotte (1958) expression for non-Fourier heat conduction is used to study the temperature dependent heat source. The governing equation is written as;

\[ U_{xx} = U_{tt} + Ut (1 - Us^*) - Us^*u \]
where \( u = \frac{T}{T_1}, \ \text{Us}^* = u''/S \) where \( S \) is the storage coefficient with units of \( \text{W/cu.m/K} \), \( (\rho \ C_p/\tau_r) \) where \( \tau_r \) is the relaxation time. The damping term can be removed from the governing equation by assuming the solution of the form \( u = \exp(-nt)W \). When \( n = (1 - \text{Us}^*)/2 \) the governing equation reduces to

\[
W_{xx} = W_{tt} - \frac{(1 + (\text{Us}^*))^2}{4}
\]

Assuming the transformation

\[
\eta = t^2 - X^2
\]

the equation is transformed to a Bessel equation:

\[
\eta^2 W_{\eta\eta} + \eta W_{\eta} - \eta/8 (1 + (\text{Us}^*))^2 = 0
\]

Comparing this equation with the generalized form of the Bessel equation:

\[
W = C_1 I_0 (\sqrt{1/2(1 + (\text{Us}^*))^2}) (t^2 - X^2)
\]

or \( u = I_0 (\sqrt{1/2(1 + (\text{Us}^*))^2}) (t^2 - X^2) \)

\[
/I_0(\sqrt{1 + (\text{Us}^*))^2}/2
\]

The heat removal is hence a critical phenomena in nuclear power plants.

**TUESDAY MORNING**

**Section A**

Convention Center -- Room 390

**Development of High-LET Therapeutic Radiopharmaceuticals**

**Bismuth-213 Radiopharmaceuticals**

_Cosponsored with Division of Inorganic Chemistry, and Division of Medicinal Chemistry_

S. Mirzadeh, _Presiding_

M. W. Brechbiel and D. S. Wilbur, _Organizers_

**8:00. — Introductory Remarks**

**8:05 —58. High-LET radiopharmaceuticals: Practical or pretender? Robert Atcher,**

Bioscience Div, Los Alamos National Laboratory, MS E529, Los Alamos, NM 87545, Fax: 505-665-5052, ratcher@lanl.gov

The potential for high LET emitters has been known for decades. The combination of physical attributes equips these radionuclides with superior cytotoxic properties. In comparison to low LET emitters, specifically beta emitters, their entry into clinical practice has been extremely slow. In this talk, I will review some of the advantages of high LET emitters and discuss some of the impediments to their wider use.

**8:35 —59. Production of actinium-225 for alpha-particle-mediated radioimmunotherapy. Rose A. Boll1, Dairin W. Malkemus2, and Saed Mirzadeh2. (1) Department of Chemistry, University of Tennessee, Knoxville, TN 37996-1600, (2) Nuclear Science and Technology Division, Oak Ridge National Laboratory**

Within the past five years, the investigation of targeted cancer therapy using \( \alpha \)-emitters has developed considerably and recent clinical trials have generated promising results. Bi-213, a member of the \(^{229}\text{Th}\) chain has been used with success in \( \alpha \)-radioimmunotherapy. This paper describes our seven years experience producing \(^{225}\text{Ac}\) (parent of \(^{213}\text{Bi}\)) in partial support of the ongoing clinical trials. The initial purification of \(^{229}\text{Th}\) from waste material began at ORNL in 1995, and since then the separation process has been refined and expanded to a current production level of 500 mCi of \(^{225}\text{Ac}\) in 2002. The chemical processes involved in the initial clean up of \(^{229}\text{Th}\) from \(^{233}\text{U}\) waste material and the routine separation of \(^{225}\text{Ra}\) and \(^{225}\text{Ac}\) from \(^{229}\text{Th}\) will be discussed. The semi-remote process involves a series of ion-exchange columns ranging in size form 1000 to 0.25 mL. The final purification of \(^{225}\text{Ac}\) and the required quality control will also be presented.

**8:55 —60. Alternative routes for the production of \(^{229}\text{Th}, \ ^{225}\text{Ra}, \text{ and } \ ^{225}\text{Ac}. Marc A. Garland,**

Charles W. Alexander, and Saed Mirzadeh, Nuclear Science and Technology Division, Oak Ridge National Laboratory, 1 Bethel Valley Road MS
Increasing interest in clinical applications of bismuth-213 coupled with limited availability of thorium-229 necessitate investigation of alternative routes of production. Currently, $^{233}\text{U}$ is the only viable source for $^{229}\text{Th}$, however, demand for $^{225}\text{Ac}$ may soon exceed the levels of $^{229}\text{Th}$ present in the $^{233}\text{U}$ stockpile. Considering the rather low annual production rate of $^{229}\text{Th}$ from $^{233}\text{U}$ and difficulties associated with $^{233}\text{U}$ safeguards, routine processing of $^{233}\text{U}$ is unfeasible. Alternative routes for production of $^{229}\text{Th}$, $^{225}\text{Ra}$ and $^{225}\text{Ac}$ include both reactor and accelerator approaches. $^{229}\text{Th}$ can be produced by neutron transmutation of $^{226}\text{Ra}$ targets. The calculated yield of $^{229}\text{Th}$ is ~0.02 mCi per mg of $^{226}\text{Ra}$ for 100-day irradiation in HFIR. $^{228}\text{Th}$ and $^{227}\text{Ac}$ contaminants are co-produced in quantities much greater than $^{229}\text{Th}$. However, even from a mixture of $^{228}\text{Th}$ and $^{229}\text{Th}$, high purity $^{225}\text{Ac}$ can be obtained by initially extracting Ra from Th and then extracting $^{225}\text{Ac}$ from the Ra.

9:15 —61. Recoil separation of $^{221}\text{Fr}$ and $^{213}\text{Bi}$ from $^{225}\text{Ac}$. Ann Marie Doneksi Craig¹, Daniel J. Marsh², George K. Schweitzer³, and Saed Mirzadeh³. (1) Department of Chemistry, University of Tennessee, Knoxville, TN 37996, acraig2@utk.edu, (2) Oak Ridge National Laboratory, (3) Nuclear Science and Technology Division, Oak Ridge National Laboratory

A novel method for separation of $^{221}\text{Fr}$($^{213}\text{Bi}$) from $^{225}\text{Ac}$ is based on collection of daughter recoiled atoms in an electrostatic field. The recoil energy during decay of $^{225}\text{Ac}$ imparts ~100 keV to the $^{221}\text{Fr}$ nucleus, sufficient to lift a $^{221}\text{Fr}$ atom from the surface. To collect the recoil atoms in an electrostatic field, a very thin source of $^{225}\text{Ac}$ is essential since the range of the recoil atoms is very short (µm). An $^{225}\text{Ac}$ source prepared by conventional electrodeposition on a Pt foil electrode (~1 cm²) has been successful. The electrodeposition process has high efficiency with >90% of $^{225}\text{Ac}$ being plated within 60 minutes under an EMF of 12 V in 0.01M HNO₃ electrolyte. The process appears to be sensitive to some unknown variable, while occasionally efficiency drops. The recoil-separation of $^{221}\text{Fr}$ and $^{213}\text{Bi}$ with 75% efficiency was achieved using a catcher electrode placed ~5 mm away from the $^{225}\text{Ac}$ source and applying 750 V (He) or 3000 V (air) across the electrodes.


A novel radionuclide generator concept has been developed and demonstrated for the production of $^{212}\text{Bi}$ from $^{224}\text{Ra}$/$^{212}\text{Pb}$, $^{213}\text{Bi}$ from $^{225}\text{Ac}$, and $^{90}\text{Y}$ from $^{90}\text{Sr}$. The new concept, called a Multicolumn Selectivity Inversion Generator (MSIG), involves the selective retention of the desired daughter radionuclide from a solution of the parent (or parents) by a primary separation column followed by stripping the daughter radionuclide from the primary column, and without feed adjustment, passing the daughter through a secondary separation column that retains the parents while the daughter elutes unrestrained. The new generator system minimizes the effects of radiation damage to the column material, requires only very small chromatographic columns, and achieves extraordinarily high decontamination factors. These features permit the rapid and reliable production of nuclides of high chemical and radionuclidic purity.

9:55 —63. A multicolumn selectivity inversion generator for the production of high-purity actinium-225 for use in therapeutic nuclear medicine. Andrew H. Bond¹, E. Philip Horwitz¹, Daniel R. McAlister¹, and Kenneth R. Givens². (1) PG Research Foundation, 8205 S. Cass Ave., Ste. 111, Darien, IL 60561, Fax: 630-963-6432, abond@pgrf.com, (2) MedActinium, Inc

The widespread success of $^{213}\text{Bi}$ in radioimmunotherapy depends on the efficient production of the $^{225}\text{Ac}$ parent. To optimize $^{225}\text{Ac}$ recovery and purification, a multicolumn selectivity inversion generator (MSIG) was developed in which $^{225}\text{Ac}$ and its $^{225}\text{Ra}$ parent are extracted from a sulfate solution of $^{229}\text{Th}$ by a cation-exchange primary separation column. The $^{225}\text{Ac}$ and $^{225}\text{Ra}$ are stripped with HNO₃ and passed through a guard column that retains only adventitious $^{229}\text{Th}$. The guard column eluate is directed without chemical adjustment to a third column where $^{225}\text{Ac}$ is separated from $^{225}\text{Ra}$. The $^{225}\text{Ac}$ is recovered in 0.1
M HCl, which is readily incorporated into the MSIG developed for $^{213}$Bi. The $^{225}$Ac purification process affords nearly quantitative recovery of $^{229}$Th, permits the efficient utilization of $^{225}$Ra, and enables the reliable production of $^{225}$Ac of high radionuclidic and chemical purity. A discussion of the $^{225}$Ac flowsheet and experimental testing using radioisotopic analogs will be presented.

10:15 — Intermission.

10:30 — 64. Mesoporous sol-gels for use in clinical-scale $^{225}$Ac/$^{213}$Bi generators. John W. Clavier, Saed Mirzadeh, David Depaoli, and Michael Z-C. Hu, Nuclear Science and Technology Division, Oak Ridge National Laboratory, 210 East K Street, Elizabethton, TN 37643, john.w.clavier@gsk.com

Commercial cation exchange resins (BioRad AG or MP50) are currently used in $^{225}$Ac-213Bi biomedical generators. The PVC-backbone resins deteriorate in the high radiation fields of >20 mCi of $^{225}$Ac used in clinical-scale generators, consequently shortening the generator lifetime. We envisioned improvement in radiation stability by substituting an inorganic sol-gel for the resin backbone, and have synthesized several mesoporous sol-gels resins incorporating covalently tethered bifunctional sulfonic acid ligands as the metal binding sites. The resins were characterized by XPS, BET and SEM, and their performance was evaluated in comparison with commercial resins. The synthesized sol-gels offered rapid, quantitative uptake of $^{225}$Ac, and high (>70%), and selective elution of $^{213}$Bi.

10:50 — 65. Ac-225/Bi-213 generator chemistries compatible with automation. Oleg Egorov$^1$, Matt O’Hara$^1$, Sergei I. Sinkov$^1$, and Kenneth R. Givens$^2$. (1) Pacific Northwest National Laboratory, PO Box 999, Richland, WA 99352, oleg.egorov@pnl.gov, (2) MedActinium Inc, 313 Woodland Trace Drive, Knoxville, TN 37922, kgivens@medactinium.com

The short-lived (half-life 45.6 minutes) alpha-emitter Bi-213 is a highly promising radionuclide for the radioimmunotherapy of cancer. Bismuth-213 is generated from the decay of Ac-225, which has a half-life of 10 days. Development of viable Bi-213-based treatments requires the availability of reliable and safe generator systems that can produce clinical levels of high purity Bi-213 over extended periods of time. Moreover, the generator must be compatible for integration with automated dose preparation kits. This presentation will provide a brief overview of the existing Ac-225/Bi-213 generator chemistries and systems. Furthermore, we will present our recent results on the development and initial testing of an improved tandem column Ac-225/Bi-213 generator system. The primary column is composed of a large bed of the extraction chromatographic sorbent. To minimize the radiation dose density, the Ac-225 source is uniformly distributed through the sorbent bed. A small secondary column enables selective capture and concentration of the Bi-213 species directly from the primary column eluent, without the need for dilution. High purity Bi-213 product is obtained by eluting the secondary column with a sodium acetate buffer solution that is compatible with the subsequent labeling and purification steps.

11:10 — 66. An automated radionuclide separation system for nuclear medical applications. Andrew H. Bond, E. Philip Horwitz, and Daniel R. McAlister, PG Research Foundation, 8205 S. Cass Ave., Ste. 111, Darien, IL 60561, Fax: 630-963-6432, abond@pgrf.com

The purification of radionuclides for medical uses is challenging as chemical purity is needed to minimize the impact of interferents on radioimmunoconjugation reactions and radionuclidic purity is required to prevent patient exposure to potentially hazardous radiation. The production of therapeutically useful radionuclides (e.g., $^{212/213}$Bi, $^{90}$Y, $^{186/188}$Re) presents unique challenges, because the comparatively high LET of such radionuclides can cause degradation of the support materials responsible for product purity. An instrument compatible with Multicolumn Selectivity Inversion Generator technologies was developed that permits the rapid purification of clinically useful quantities of radionuclides. The modular device comprises controller software and a low pressure chromatography system with disposable chromatographic columns. This instrument is ideal for use in radionuclide generators as separations can be performed remotely in <5-10 min with decontamination factors of >10$^6$. Instrument performance during the automated purification of
212Bi, 213Bi, and other therapeutically useful radionuclides will be discussed.

11:30 — 67. Improved method for labeling DOTA analogs with 213Bi, 225Ac, and lanthanides. **Tuomo K Nikula**, 1 Christos Apostolidis1, Ramon Carlos-Marquez1, Philippe Jehenson1, Helmut Mäcke2, Simona Martin1, and Ali Ouadi1. (1) Nuclear Chemistry, European Commission, JRC, Institute for Transuranium Elements, B.O.Box 2340, 76125 Karlsruhe, Germany, Fax: +49-7247-951924, nikula@itu.fzk.de, (2) Radiobiological Chemistry, University Hospital Basel, Institute for Nuclear Medicine

Although radio-labelled DOTA-analogues have a good in vivo stability, their slow reaction kinetics at room temperature has limited their use mainly to peptides. This has been a critical issue, especially with short half-life isotopes such as bismuth-213. We have overcome this problem with a new buffer system that takes into account the protonation of amines in the structure of cyclam.

A DOTA-Substance P analogue was radio-labelled with bismuth-213 (obtained from 225Ac/213Bi-generator as BiI5-ion) at pH 9 and at the room temperature. After 10 minutes incubation the labelling efficiency was over 90%. Actinium-225 shows considerable lower labelling efficiency than bismuth-213, but still an increased yield at pH 9 compared to pH 5.5. The method was also tested with different lanthanides and yttrium at nmol level resulting in practically complete chelation with the DOTA-chelate in less than 10 minutes.


The bismuth-213 (213Bi) therapeutic kit developed by MedActinium is the first to address the logistical challenges of widespread alpha particle immunotherapeutic clinical deployment; the kit integrates the multi-functional requirements of chemical separation, purification, monoclonal antibody conjugation, sterilization, and automation. The 213Bi therapeutic kit contains actinium-225 (225Ac) from which 213Bi is selectively extracted using a newly developed generator technology. The 213Bi is then conjugated with the monoclonal antibody using an automated, temperature–controlled internal mixing operation. The final step transfers the 213Bi-labeled monoclonal antibody solution to a syringe for dose adjustment prior to administration. The 213Bi therapeutic kit design is fully automated to provide a sterile environment for the safe and reliable production of multiple on-site doses for up to 30 days, until the 225Ac is expended.

12:10 — Discussion.

Section B
Convention Center -- Room 391

Analytical Chemistry in Nuclear Technology
Cosponsored with Division of Analytical Chemistry

Developments in Waste-Tank Separation and High-Level Sludge Analytical Methods

S. M. Serkiz, Presiding
C. J. Coleman, D. T. Hobbs, J. C. Griffin, and D. E. Hobart, Organizers

8:30 — Introductory Remarks.

8:35 — 69. Thermodynamic studies of actinide complexation at elevated temperature. **Sue B. Clark**, and Alena Paulenova, Department of Chemistry, Washington State University, POBox 4630, Pullman, WA 99164-4630, Fax: 509-3358867, s_clark@mail.wsu.edu

Organic ligands used in separation processes or generated in radioactive waste can have a significant impact on chemical behavior and solubility of actinides (An), and little is currently known about the impact of temperature on these processes. A thermodynamic model describing actinide speciation at high temperature is under development for the processing and vitrification of high-level waste. In support of this model development, complexation equilibria for Anz+(aq) + Ln-(aq)→AnL(z-n)+(aq) have been studied over the temperature interval from 10-750°C, pH 5 - 6.
Apparent stability constants with EDTA, NTA, acetate, oxalate, gluconate and citrate have been measured using solvent extraction at ionic strength 0.1 M NaClO4. Values for the apparent stability constants have been extrapolated to zero ionic strength using SIT parameters. Complexation enthalpies and entropies were calculated from temperature dependences of stability constants. The impact of temperature will be discussed in the context of effective cationic charge for the actinide ions.

8:55 —70. Evaluation of silver-impregnated activated carbons for I-129 and Tc-99 from an acidic groundwater. Steven M. Serkiz¹, K.M. Kanzleiter¹, Erin E. Cumbie², and Tanju Karanfil³. (1) Savannah River Technology Center, Westinghouse Savannah River Company, 773A rm B121, Aiken, SC 29808, steven.serkiz@srs.gov, (2) Environmental Engineering and Science Dept, Clemson University, (3) Department of Environmental Engineering and Science, Clemson University

Iodine-129 and Technetium-99 are long-lived nuclides and because they are predominately anionic species their removal from aqueous waste streams is often difficult. Data will be presented on the removal of I-129 and Tc-99 from contaminated groundwater at the Department of Energy’s Savannah River Site (SRS) using commercially available anion exchange resin, activated carbon, and several silver impregnated activated carbons (SIACs) in column breakthrough experiments. A range of SIAC materials, including Ag(0) and AgCl impregnated materials, were tested to examine the effects of silver content and silver surface speciation on anion removal. Because the low disposal limits for I-129 bearing waste, the spent sorbent leaching properties were evaluated using standard leaching protocols. The results from this study show that the SIACs and the commercially available anion exchange resin were effective in removing I-129 and Tc-99 from the feed water to low levels. The secondary waste leaching properties of the SIACs, however, appear superior to the organic-based anion resin tested in this work.

9:15 —71. On-line ICP-ES monitor and sampling system for measuring rhenium and technetium breakthrough from SuperLig 639 columns. W.D. King, and W.A. Spencer, Savannah River Technology Center, Westinhouse Savannah River Company, Aiken, SC 29808, william02.king@srs.gov

An on-line ICP-ES monitor and sampling system have been developed in support of the Hanford River Protection Project Waste Treatment Plant design. The technology was developed for potential plant application to monitor technetium removal from low-level radioactive waste solutions by ion exchange. During method development, the monitor was utilized to collect elemental composition data on a frequency of 15 minutes to support lab-scale anion exchange column testing of nonradioactive waste simulants containing rhenium as a surrogate for technetium.

High-resolution liquid-phase concentration profiles were generated for multiple metal during all phases of column operation (ranging from 5 M Na+ feed solutions at 25°C to water eluent solutions at 65°C). The fine time-scale data provided unique information on the presence of competitor species for resin adsorption sites and the timing of desorption for various species during column elution. This work showed that highly time-resolved analyses collected using the on-line ICP-ES system were essential in discerning many of the details of competitive ion sorption mechanisms for this system.

9:35 —72. New test apparatus for measuring particle kinetics and equilibrium isotherms based on a differential column approach. C.E. Duffey, W.D. King, and L.L. Hamm, Savannah River Technology Center, Westinhouse Savannah River Company, Aiken, SC 29808, Fax: 803-725-2756, cheryl.duffey@srs.gov, william02.king@srs.gov

An understanding of resin performance in a liquid chromatographic column requires that several underlying mass transport mechanisms be quantified. The development of a kinetics test apparatus based on a differential column concept will be presented. This setup allows for the estimation of particle pore diffusion coefficient,
particle liquid-film coefficient, and true equilibrium capacities (i.e., sorption isotherm) for a resin. Additionally, the design provides for increased flowrate control and is simple to fabricate. Experimental results for the removal of perrhenate ion (a chemical surrogate for pertechnetate) from a high alkaline, low-level radioactive waste stream simulant generated with the apparatus are shown. Impacts on mass transfer parameters from varying temperature, liquid-phase composition, and particle size are discussed. To demonstrate the composite impact of these mass transfer mechanisms, a column transport model using the experimentally determined parameters is compared to recently measured column behavior. Our experience indicates that this experimental setup provides consistent kinetics and equilibrium data key in the understanding and prediction of column performance.

9:55 — Intermission.

10:15 —73. Impact of precipitates on the removal of cesium from Hanford waste tank simulants. Steven M. Serkiz, Kimberly R. Powell, and A.L. Pishko, Savannah River Technology Center, Westinghouse Savannah River Company, 773A rm B121, Aiken, SC 29808, Fax: 803-725-1660, steven.serkiz@srs.gov, kim.powell@srs.gov

The Savannah River Technology Center has been investigating ion exchange methods for removing radio-cesium and technetium from alkaline wastes currently stored at the Hanford Site. In the current process flow sheet, including a precipitation and ultrafiltration step, the baseline technology for the Waste Treatment Plant removal of cesium is an elutable cesium-specific resin prepared by IBC Advanced Technologies. Recent work has shown the formation of a post-filtration precipitation and concerns have been raised on the impacts of these precipitates on cesium ion exchange. Data were collected on the development of post-precipitates from a range of alkaline waste simulants designed to represent the chemical composition of Hanford waste tanks. Digestion and elemental analysis, x-ray diffraction, scanning electron microscopy (SEM), and energy dispersive x-ray analysis (EDX) characterized these precipitates. Changes in resin performance were evaluated using batch contacts of the resin with stable cesium. Attempts were made to evaluate the relationship between the physicochemical properties of the precipitates formed and cesium sorption to the resin.


The Defense Waste Processing Facility (DWPF) at the Savannah River Site combines high-level radioactive defense waste with glass-forming chemicals at 1100 degrees C to make durable waste glass. The DWPF operates with a large tank that contains a 2-3 year supply of sludge feed (a macrobatch). Each macro-batch must be fully characterized to provide processing and waste regulation compliance information. The macro-batch feed that will be processed in 2003 presents a more challenging analytical matrix than previous feeds. Significant quantities of coal, sand, and sodium oxalate are present in the feed. These components must be measured because they can affect DWPF chemical processes and the large coal and sand particles can affect the DWPF tank sampling system. Development of analytical methods for this DWPF macro-batch sludge feed and application of these methods to the actual radioactive sludge will be discussed in this paper.


We have developed a method for measuring total carbon (TC) in simulated sludge containing insoluble organic particulates in the form of coal. The analysis is complicated by the presence of carbon compounds (such as coal) which are difficult to combust and by the difficulty in obtaining a
reproducible sample. Based on experiments performed on a variety of sludge matrices, the following were determined: Thermal oxidation at a minimum of 900 degrees centigrade is required to completely effect oxidation of the coal; persulfate oxidation and thermal oxidation below 900 degrees proved ineffective. Reproducible data can be obtained on undiluted sludge (well mixed immediately prior to analysis) or on diluted samples that have been micronized (crushed) prior to dilution. Reproducible data could not be obtained on diluted samples that had not been crushed prior to dilution. Results on simulated sludge samples as well as radioactive sludge will be presented.

11:15 —76. Rheological properties of DWPF sludge batch-two radioactive slurries. Terri L. Fellinger, Eric K. Hansen, and David C. Koopman, Savannah River Technology Center, Westinghouse Savannah River Company, Aiken, SC 29808, Fax: 803-725-4704, terri.fellinger@srs.gov

The Defense Waste Processing Facility (DWPF) at Savannah River Site is currently processing Sludge Batch 2 (a 500,000 gallon caustic slurry of HLW) for geological disposal. Rheological measurements on this batch are necessary to identify potential mixing and pumping problems during transfer of the batch to the DWPF and during DWPF processing. Sludge slurry samples were transferred to Savannah River Technology Center where rheological properties were measured remotely. Yield stresses and consistencies were obtained from flow curves (shear rate vs. shear stress). Results indicated that the consistency of the original slurry was such that it could be pumped to the DWPF, but was too high to be processed in the DWPF. Lowering the pH of the slurry decreased its consistency. However, additional acid was added to ensure that the final slurry (a mixture of waste and glass forming compounds) could be pumped to the melter. Rheological properties of other radioactive HLW will also be discussed.

TUESDAY AFTERNOON

Section A
Convention Center -- Room 390

Development of High-LET Therapeutic Radiopharmaceuticals
Cosponsored with Division of Inorganic Chemistry, and Division of Medicinal Chemistry

Bismuth-213 Radiopharmaceuticals

J. G. Jurcic and B. J. Allen, Presiding
M. W. Brechbiel and D. S. Wilbur, Organizers

1:30 —77. Alpha-particle immunotherapy with bismuth-213-labeled anti-CD33 monoclonal antibody HuM195 in myeloid leukemia. Joseph G. Jurcic¹, John M. Burke¹, Michael R. McDevitt², Chaitanya R. Divgi³, George Sgouros⁴, Ronald D. Finn², Steven M. Larson³, and David A. Scheinberg². (¹) Department of Medicine, Memorial Sloan-Kettering Cancer Center, 1275 York Avenue, Box 458, New York, NY 10021, Fax: 212-772-8441, jurcicj@mskcc.org, (²) Sloan-Kettering Institute, Memorial Sloan-Kettering Cancer Center, (³) Nuclear Medicine Service, Memorial Sloan-Kettering Cancer Center, (⁴) Department of Medical Physics, Memorial Sloan-Kettering Cancer Center

HuM195, a humanized anti-CD33 monoclonal antibody, targets myeloid leukemia cells. The feasibility, safety, and anti-leukemic activity of therapy with the α-emitting radioimmunoconjugate ²¹³Bi-HuM195 was shown in a phase I trial (Jurcic et al. Blood 2002). The physical properties of particles are ideally suited for treatment of small-volume disease. To determine the effects of particle immunotherapy against cytoreduced disease, we treated 11 patients with acute myeloid leukemia using the chemotherapeutic agent cytarabine followed by ²¹³Bi-HuM195 0.5-1 mCi/kg (30-90 mCi). Three patients, all treated at the 1 mCi/kg-dose level, responded. Two patients had complete remissions, and one achieved a partial remission. The median time from initiation of therapy to leukocyte recovery was 25 days (range, 17-49 days). One patient had dose-limiting...
myelosuppression lasting over 35 days. No other significant toxicities were seen. Sequential administration of chemotherapy and $^{213}\text{Bi}$-HuM195 is safe and can produce remissions in myeloid leukemia.

2:00 — 78. Alpha radioimmunotherapy of B-lineage non-Hodgkin’s lymphoma using $^{213}\text{Bi}$-labeled anti-CD19- and anti-CD20-CHX-A'-DTPA conjugates. Steffen Heeger¹, Gerhard Moldenhauer², Gerlinde Egerer², Horst Wesch³, Simona Martin⁴, Tuomo Nikula⁴, Christos Apostolidis⁴, Martin W. Brechbühl⁵, Anthony D. Ho⁴, and Rainer Haas⁶. (1) Department of Molecular Immunology, German Cancer Research Center, Im Neuenheimer Feld 280, 69120 Heidelberg, Germany, Fax: +49-6221-422572, S.Heeger@DKFZ.de, (2) Oncology and Haematology, University Hospital Heidelberg, (3) Department of Radiology, German Cancer Research Center, (4) Nuclear Chemistry, European Commission, Joint Research Centre, Institute for Transuranium Elements, (5) NCI, NIH, (6) Department of Hematology, Oncology and Clinical Immunology, University of Duesseldorf

The therapeutic potential of high LET alpha-emitting $^{213}\text{Bi}$ coupled to various antibody constructs was investigated for the use in B-lineage lymphoma. In preclinical experiments the strong cytotoxic potency of $^{213}\text{Bi}$-labeled anti-CD19 and anti-CD20 antibodies including recombinant constructs (diabodies, tetrabodies) for targeting lymphoma cells was demonstrated. Additionally, antibody conjugates showed favorable in vivo stability and biodistribution behavior. After 6 hours 55-70 % of the injected activity was still detectable in blood circulation and 32-45 % after 24 hours, respectively. Beside the induced myelosuppression, no other toxic side effects occurred. We are currently conducting a phase I dose escalation trial to determine safety and feasibility as well as pharmacology and dosimetry. So far, nine patients received activities of the $^{213}\text{Bi}$-CHX-A'-anti-CD20 radioconjugate ranging from 385 up to 1640 MBq. Acute toxic side effects were not observed except two cases of mild leukopenia (grade 1 CTC). Two patients responded to alpha-radioimmunotherapy. Our data indicate that $^{213}\text{Bi}$-labelled antibody conjugates represent promising tools for an improved therapy of refractory B-cell malignancies.

2:30 — 79. In vitro evaluation of $^{213}\text{Bi}$-rituximab versus external gamma irradiation for the treatment of B-CLL patients: Relative biological efficacy with respect to apoptosis induction and chromosomal damage by the micronucleus assay. Katia Vandenbulcke¹, Filip De Vos², Fritz Offner³, Jan Philippe⁴, Christos Apostolidis⁵, Roger Molinet⁵, Tuomo K Nikula⁵, Klaus Bacher⁶, Virginie de Gelder⁶, Anne Vral⁶, Christophe Lahorte¹, Hubert Thierens⁶, Rudi A. Dierckx², and Guido Slegers¹. (1) Department of Radiotherapy, University of Gent, Gent, Belgium, Katia.vandenbulcke@rug.ac.be, (2) Department of Nuclear Medicine, Gent University Hospital, (3) Department of Hematology, University of Gent, (4) Department of Clinical Chemistry, University of Gent, (5) Nuclear Chemistry, European Commission, JRC, Institute for Transuranium Elements, B.O.Box 2340, 76125 Karlsruhe, Germany, (6) Department of Radiation Physics, University of Gent

The radioimmunotherapy (RIT) with alpha-emitters is attractive because of the high LET and short path length allowing higher tumor cell kill and lower toxicity to healthy tissues. We assessed the relative biological efficacy (RBE) of alpha RIT (in vitro) compared with external gamma irradiation with respect to induction of apoptosis in B-CLL and induction of chromosomal damage in B and T lymphocytes. The dose range experiments demonstrated $^{213}\text{Bi}$-anti-CD20 was more effective than equivalent doses by external gamma irradiation to introduce apoptosis. The RBE for induction of apoptosis in B-CLL ranged from 2.2 at 2 Gy to 1 at 10 Gy. The micronucleus yield on lymphocytes was measured to assess the late toxicity by induction of chromosomal instability. While gamma-radiation induced a steady increase in micronucleus yields in B and T cells, the damage inflicted by $^{213}\text{Bi}$ was more dramatic with RBE ranging from 5 to 2 between 0.1 Gy and 2 Gy. In contrast to gamma irradiation, $^{213}\text{Bi}$ inhibited forced mitosis almost completely at 2 Gy.
Radioimmunotherapy with bismuth-213 as nonmyeloablative conditioning for marrow transplantation. Wolfgang A. Bethge, D. Scott Wilbur, Donald K. Hamlin, Erlinda B. Santos, Martin W. Brechbiel, Rainer Storb, and Brenda M. Sandmaier. (1) Transplantation Biology, Fred Hutchinson Cancer Research Center, 1100 Fairview Avenue North, P.O. Box 19024, D1-100, Seattle, WA 98102, Fax: 206-667-6124, wbethge@fhcrc.org, (2) Department of Radiation Oncology, University of Washington, (3) Radiation Oncology Branch, National Cancer Institute, NIH

To lower treatment-related toxicity and mortality of conventional marrow transplantation, a nonmyeloablative conditioning regimen using 2 Gy total body irradiation (TBI) and mycophenolate mofetil (MMF) combined with cyclosporine (CSP) for postgrafting immunosuppression was developed in a canine model. To specifically target cells responsible for graft rejection and to circumvent potential toxic effects of external beam irradiation, we investigated replacing TBI by radioimmunotherapy with the alpha-emitting radionuclide bismuth-213 (213Bi). 213Bi was coupled to monoclonal antibodies, either to the panhematopoietic antigen CD45 or the T-cell receptor , using the metal binding chelate CHX-A”-DTPA. The radioimmunoconjugates were administered in 6 injections on days -3 and -2 followed by transplant of DLA-identical marrow on day 0 and postgrafting immunosuppression with MMF and CSP. Targeting either CD45 or TCR with the radioimmunoconjugates in combination with postgrafting immunosuppression allowed stable marrow engraftment in a canine model, thus replacing TBI.

Radioimmunotherapy of infections using alpha- and beta-emitting radionuclides. Ekaterina Dadachova, Ruth Bryan, Antonio Nakouzi, Annie Frenkel, Joshua D. Nosanchuk, Andrew D. Schweitzer, and Arturo Casadevall. (1) Nuclear Medicine, Albert Einstein College of Medicine, 1695A Eastchester Rd., Bronx, NY 10461, Fax: 718-405-8457, edadacho@aecom.yu.edu, (2) Microbiology and Immunology, Albert Einstein College of Medicine

Fungal infections are notoriously difficult to treat in immunosuppressed patients and novel therapeutic modalities are urgently needed to eradicate infection in the setting of severe immune dysfunction. We have investigated the potential of radioimmunotherapy of C. neoformans and H. capsulatum fungal infections with organism-specific monoclonal antibodies (mAb) 18B7 and 9C7, respectively, radiolabeled with alpha-emitter 213-Bismuth and beta-emitter 188-Rhenium. Both C. neoformans and H. capsulatum proved to be extremely radioresistant to external gamma-radiation with significant killing of the cells observed only above 70,000 rad. On the contrary, two-log reduction in survival of fungal cells was achieved in vitro with 213-Bi- and 188-Re-labeled mAb’s 18B7 and 9C7. This study demonstrates that Antimelanoma antibody 9.2.27 was labeled with Bi-213 - an alpha emitter with 46-minute half-life. Labeling was done using two different chelators namely cyclic dihydride of DTPA (Cy-DTPA) and CHX-A” - DTPA. Stability of the labeled product was compared (by serum stability and DTPA challenge tests) for two chelators and was found to be similar. In vivo dose tolerance was tested up to 12 mCi / kg. Maximum tolerated dose (MTD) was found to be ~10mCi/kg. Tumours at various stages of growth (2, 4, 8, 16 and 26-days post inoculation) were given local intra-lesional Bi-213 - labeled-9.2.27 injections. At 2-days post-inoculation, complete tumour inhibition was observed whereas progressive response (inhibition or delay) was obtained with later time points. Systemic injections showed a significant delay in tumour growth compared to controls. These encouraging results are the basis of an ongoing phase-I dose escalation trial of intra-lesional targeted alpha therapy in patients with advanced melanoma.
RIT with organism-specific antibody can be an effective weapon against opportunistic fungal infection and suggests the potential usefulness of this modality against other infectious diseases.

4:30 —83. Cold and alpha-conjugated anti-CD20 induce different in vitro apoptosis responses in B-chronic lymphocytic leukemia and splenic lymphoma with villous lymphocytes. Katia Vandenbulcke, Fritz Offner, Guido Slegers, Filip De Vos, Virginie de Gelder, Annick Willems, Simona Martin, Roger Molinet, Christos Apostolidis, Tuomo K. Nikula, Willem Janssens, Ann Janssens, Hubert Thierens, Roger Molinet, Christos Apostolidis, Tuomo K. Nikula, Willem Janssens, Ann Janssens, Hubert Thierens, Rudi A. Dierckx, and Jan Philippe. (1) Department of Radiopharmacy, University of Gent, Gent, Belgium, katia.vandenbulcke@rug.ac.be, (2) Department of Hematology, University of Gent, (3) Department of Nuclear Medicine, Gent University Hospital, (4) Department of Radiation Physics, University of Gent, (5) Nuclear Chemistry, European Commission, Joint Research Centre, Institute for Transuranium Elements, B.O.Box 2340, 76125 Karlsruhe, Germany

Radiotherapy, fludarabine, anti-CD20 antibody and glucocorticoids can kill malignant lymphocytes in B-CLL and SLVL. To evaluate the relation of the number of CD20 molecules per cell and effect of radioconjugated and cold anti-CD20 antibodies we compared CD20 expression in B-CLL and SLVL and related this to apoptosis in response to 6 different cytotoxic treatments. Peripheral blood lymphocytes were isolated. Cells were treated in vitro with (1) 213Bi-anti-CD20, (2) external 60Co gamma irradiated, (3) fludarabine, (4) methylprednisolone, (5) cold anti-CD20 and human serum as a complement source or (6) cold anti-CD20 + gamma-radiation were intrindividually compared with non treated cells. Apoptosis was evaluated by flow cytometry using Annexin V and 7-AAD. In conclusion no differences were observed between CLL and SLVL in apoptosis induced by fludarabine, gamma radiation and prednisolone. Both cold and radioconjugated antiCD20 induced more apoptosis in SLVL than in B-CLL, due to the much higher expression of CD20 molecules on SLVL cells. Cold anti-CD20 plus gamma radiation induced less apoptosis than 213Bi conjugated anti-CD20 in CLL (p<0.01) but not in SLVL.

4:50 —84. Molecular mechanisms of alpha-particle-induced cytotoxicity in human leukemic cells as revealed by gene-expression profiling. Simona Martin, Ralf Kronenwett, Katia Vandenbulcke, Fritz Offner, Slawomir Kliszewski, Christos Apostolidis, Ramon Carlos, Willem Janssens, Maurice Leroy, Tuomo Nikula, Jan Philippe, and Rainer Haas. (1) Nuclear Chemistry, European Commission, Joint Research Centre, Institute for Transuranium Elements, PO Box 2340, Karlsruhe 76125, Germany, Fax: 0049-7247-951593, simona.martin@itu.fzk.de, (2) Department of Hematology, Oncology and Clinical Immunology, University of Duesseldorf, (3) Department of Radiopharmacy, University of Gent, (4) Department of Hematology, University of Gent

Alpha-particle emitters such as 213Bi with high LET and short path length kill cells by few nuclear hits. Little is known about the molecular pathways of alpha-induced cytotoxicity in human cancer cells. Our study was aimed to identify the transcriptional responses induced in peripheral blood lymphocytes from 12 patients with B-chronic lymphocytic leukemia (CLL) after in vitro treatment with 213Bi-CHX-A''-DTPA-anti-CD20. Using cDNA arrays comprising 1,185 genes we intraindividually compared expression profiles of treated and non-treated cells and identified 66 genes significantly altered after treatment. These include genes coding for DNA-replication proteins, basic transcription factors, cell cycle regulators, adhesion and death receptors, growth factors, chemokines. As B-CLL cells differ according to the immunoglobulin (Ig) mutational status, we found significantly different responses between cells from patients with mutated and unmutated rearranged Ig genes. Our results give insights into the molecular effects of alpha-irradiation and provide information on cell-type-specific molecular responses.

5:10 —85. Long-term survival and toxicity in Balb/c mice after systemic administration of alpha emitters. Horst Wesch, Steffen Heeger, Frauke Spiecker, Folker Amelung, Martin W. Brechbiel, Gerhard Moldenhauer, and Tuomo Nikula. (1) Department of Radiology, German Cancer Research Center, Im Neuenheimer Feld 280, 69120 Heidelberg, Germany, Fax: +49 6221 42 2572, H.Wesch@dkfz.de, (2) Department of Molecular Immunology, German Cancer Research
In recent years radioimmunotherapy (RIT) with beta- or alpha-particles emitting radionuclides has been used in experimental and clinical settings for cancer therapy. To assess possible late effects of the novel therapeutic modality together with the comparison of acute versus chronic alpha radiation a long-term animal experiment was performed. Two groups were injected intravenously with a short-lived Bi-213-labeled antibody (4.7 MBq/animal and 1 MBq/animal), one with a long-lived alpha-emitter (Th-232 dioxide) and one untreated group. Dose dependent reduction in survival was observed in all groups receiving the treatment. Significant morphological differences compared to the controls were seen in the liver, spleen, lungs, heart and the adrenal glands in the 4.7 MBq group. Furthermore, in this group the formation of thrombi in the aorta was observed causing embolic damages. All these effects occurred more than one year after treatment. Future experiments should clarify whether these observations are radiation or mouse strain specific.

5:30 — Discussion.

Section B
Convention Center -- Room 391

Analytical Chemistry in Nuclear Technology
Cosponsored with Division of Analytical Chemistry

Developments in Techniques for Organic Compounds and Organic-Bearing Waste Streams

J. A. Campbell, Presiding
C. J. Coleman, D. T. Hobbs, J. C. Griffin, and D. E. Hobart, Organizers

1:30 — Introductory Remarks.

1:35 — 86. Role of mass spectrometry in the analysis of Hanford tank wastes. James A. Campbell, Eric W. Hoppe, Gary M. Mong, and Richard B. Lucke, Box 999, MS P8-08, Pacific Northwest National Laboratory*, Battelle Blvd., Richland, WA 99352, Fax: 509-376-2329

The Hanford Site in southeastern Washington State has 177 underground storage tanks containing approximately 55 million gallons of mixed hazardous waste. Mass spectrometry has played an important role in the identification and quantification of organics in the Hanford tank wastes. Chelators such as ethylenediaminetetraacetic acid (EDTA) and N-(2hydroxyethyl) ethylenediaminetriacetic acid (HEDTA) were used in large quantities during different process campaigns. In addition, phosphate-related components including tributyl phosphate, dibutylbutyl phosphate, and bis-(2ethylhexyl) phosphoric acid were used as extractants. These compounds and degradation products form an important part of the organic load found in the radioactive chemical wastes.

*This work was supported by the Department of Energy under contract DE-AC06-76RLO 1830.
Pacific Northwest National Laboratory is operated by Battelle Memorial Institute.

1:55 — 87. Study of cyclization of chelating compounds using electrospray mass spectrometry. James A. Campbell, Box 999, MS P8-08, Pacific Northwest National Laboratory*, Battelle Blvd., Richland, WA 99352, Fax: 509-376-2329, and Ying Shi, WP78-106, Merck & Co

Organic chelating compounds such as N-(2hydroxyethyl)ethylenediaminetriacetic acid (HEDTA) and ethylenediaminetriacetic acid (EDTA) have been identified as components in high level waste at the Hanford Site in southeastern Washington State. In the derivatization reaction with boron trifluoride/methanol to form methyl esters, they were found to cyclize and form six-membered rings (intramolecular lactone and lactam, respectively).

Positive and negative ion mode electrospray mass spectrometry methods were developed to identify an open form of the chelators (free acids) as well as their corresponding cyclized forms (lactams or lactones). Experimental studies were undertaken to understand the chelator cyclization process and kinetics.

*This work was supported by the Department of Energy under contract DE-AC06-76RLO 1830.
Pacific Northwest National Laboratory is operated by Battelle Memorial Institute.

Several organic-phosphorus compounds and chelating agents were used in producing various materials at the Hanford site located in southeastern Washington. Large quantities of these materials are stored in mixed hazardous waste tanks at the site. Analytical methods have been developed for the analysis of phosphates, organic acids, chelators and their degradation products. However, these methods are time-consuming, labor intensive, and costly.

MALDI/TOFMS has distinct advantages over the previously developed methods including minimal sample preparation, rapid analysis, and minimal waste production. MALDI/TOFMS has been used to analyze chelators, degradation products, organic acids, and phosphates. The chelators and phosphates produce positive ion mass spectra, while the organic acids produce negative ion spectra.

This work was supported by the Department of Energy under contract DE-AC06-76RLO 1830. Pacific Northwest National Laboratory is operated by Battelle Memorial Institute.

Analysis of chelators and degradation products using derivatization gas chromatography/mass spectrometry. Gary M. Mong, Richard B. Lucke, Blandina R. Valenzuela, and James A. Campbell, Box 999, MS P8-08, Pacific Northwest National Laboratory*, Battelle Blvd., Richland, WA 99352, Fax: 509-376-2329

Chelators such as ethylenediaminetetraacetic acid (EDTA), N-(2-hydroxyethylenediaminetriacetic acid (HEDTA), and nitrioltriacetic acid (NTA) are not part of the EPA’s list of targeted compounds; however, they are receiving renewed environmental interest. Large quantities of chelators were used in the processes at the Hanford site and are now stored in underground storage tanks.

Analytical techniques have been developed for the analysis of chelators and their degradation products.

The Hanford Site in southeastern Washington has 177 underground storage tanks which contain large quantities of highly radioactive mixed wastes. Several of these tanks contain significant amounts of organic materials such as ethylenediaminetetraacetic acid (EDTA), nitrioltriacetic acid (NTA) and N-(2hydroxyethyl)ethylenediaminetriacetic (HEDTA). Hundreds of tons of these chelating compounds were used in processing activities at the Hanford Site.

An ion exchange method was developed for reducing the radioactivity level of Hanford tank matrices; the wastes can then be analyzed for carboxylates and chelator components. Studies have shown that eluting the water-soluble fraction of tank waste through an ion exchange column (Na form) does not reduce or increase the organic carbon.

This work was supported by the Department of Energy under contract DE-AC06-76RLO 1830. Pacific Northwest National Laboratory is operated by Battelle Memorial Institute.

Analysis of organic acids in simulated and actual tank waste using ion chromatography. Gary M. Mong, James A. Campbell, and Amit K. Sharma, Box 999, MS P8-08, Pacific Northwest National Laboratory*, Battelle Blvd., Richland, WA 99352, Fax: 509-376-2329

Analytical techniques have been developed for the analysis of chelators and their degradation products.
The U.S. Department of Energy has 177 underground storage tanks at the Hanford Site in southeastern Washington that contain mixed hazardous wastes. Experiments with simulated waste have indicated that low-molecular weight organic acids are formed when ethylenediaminetetraacetic acid (EDTA) and N-(2-hydroxyethyl)ethylenediaminetriacetic acid (HEDTA) are exposed to radiolysis.

The IC method developed was used to detect and quantify organic acids in simulated and actual tank waste samples. The components identified and quantified in actual Hanford tank waste include formic, oxalic, and citric acids. The results demonstrate that ion chromatography can be effectively used to identify and quantify organic acids in the presence of the high nitrate and nitrite concentrations found in Hanford tank waste samples.

*This work was supported by the Department of Energy under contract DE-AC06-76RLO 1830. Pacific Northwest National Laboratory is operated by Battelle Memorial Institute.

4:10 —93. Analysis of phosphate-related components in Hanford tank wastes. Eric W. Hoppe, Gary M. Mong, James A. Campbell, Mary F. Hultgrenn, Mathew Thornton, and Ester Mintzer, Box 999, MS P8-08, Pacific Northwest National Laboratory*, Battelle Blvd., Richland, WA 99352, Fax: 509-376-2329

Several organophosphorus compounds were used as extractants in various processes at the Hanford Site located in southeastern Washington. Tributyl phosphate, dibutylbutyl phosphate, and bis-(2ethylhexyl) phosphate were used and now form an important part of the organic load found in the radioactive chemical wastes stored in underground storage tanks at the site.

This paper describes an interpretation of the mass spectral properties of organophosphorus compounds. The mass spectral fragmentation patterns have been extended to predict the identity of phosphorus-related components in tank waste. The presence of phosphorus-related components in tank waste was verified by comparing the mass spectrum obtained from standards when available.

*This work was supported by the Department of Energy under contract DE-AC06-76RLO 1830. Pacific Northwest National Laboratory is operated by Battelle Memorial Institute.

4:30 —94. Analysis of phosphates, sulfonates, antifoam degradation products, and anionic detergents in tank wastes. Eric W. Hoppe¹, James A. Campbell¹, Richard B. Lucke¹, Gary M. Mong¹, Blandina R. Valenzuela¹, Douglas Walker², and Major Thompson². (1) Box 999, MS P8-08, Pacific Northwest National Laboratory*, Battelle Blvd., Richland, WA 99352, Fax: 509-376-2329, (2) Savannah River Site

High-level waste was generated during processing of nuclear materials using solvent extraction with tributyl phosphate and by ion exchange with both anion and cation exchange resins. Residual portions of these organics as well as gelatin and Alconox were transferred to the high-level tanks along with aqueous solutions. Subsequent degradation of these organics has produced degradation products such as dibutyl phosphoric acid, trimethylamine, and other organics due to the intense radioactivity levels of the waste.

*This work was supported by the Department of Energy under contract DE-AC06-76RLO 1830. Pacific Northwest National Laboratory is operated by Battelle Memorial Institute.
Methods were developed for the analysis of volatile organic components such as trimethylamine, butanol, and toluene using headspace gas chromatography/mass spectrometry. Other organics were analyzed using derivatization with diazomethane to form methyl esters with subsequent analysis using gas chromatography/mass spectrometry.

**This work was supported by the Department of Energy under contract DE-AC06-76RLO 1830. Pacific Northwest National Laboratory is operated by Battelle Memorial Institute.

### WEDNESDAY MORNING

#### Section A

Convention Center -- Room 390

**Development of High-LET Therapeutic Radiopharmaceuticals**  
**Cosponsored with Division of Inorganic Chemistry, and Division of Medicinal Chemistry**

**Auger Emitters and At-211**

M. J. Mattes and H. Lundqvist, *Presiding*  
M. W. Brechbiel and D. S. Wilbur, *Organizers*

**8:00 — Introductory Remarks.**

**8:05 —95. Preclinical research on Auger therapy.** *Hans Lundqvist*, E. Bohl, J. Carlsson, L. Gedda, V. Tolmachev, A. Orlova, A. Bruskin, and S. Sjoberg, Organic Chemistry Department, Uppsala University, Uppsala, Sweden, hans.lundqvist@bms.uu.se

To improve conditions for Auger therapy we are doing pre-clinical work in the following areas: (a) Radionuclide production, (b) Synthesis of intercalators for halogen and metal radiolabelling, (c) In vitro studies to quantify double strand breaks caused by the Auger cascade, and (d) In vivo delivery systems, two step targeting.

Beside fairly pure Auger-emitters like I-125, we have focused our interest on a cocktail approach with in vivo generator systems like In-114m/In-114 and Ce-134/La-134 that give Auger electrons as well as high-energy beta radiation. One aproach is to label internalising peptides and antibodies and to study cell killing ability in cell culture and animal experiments. Another is to use DNA intercalators coupled to chelators. Here we have synthesised and tested the DNA-binding ability of several such systems. Promising results have been obtained in cell culture to demonstrate the delivery of Auger-emitters to the cell nucleus by the use of targeting liposomes loaded with intercalators.

**8:35 —96. Comparison of antibodies conjugated to I-125, In-111, Ga-67, or I-131 for single-cell kill.** *M. Jules Mattes⁴*, Martin W. Brechbiel², and Rosana B. Michel¹. (¹) Center for Molecular Medicine and Immunology, 520 Belleville Avenue, Belleville, NJ 07109, Fax: 973-844-7020, mjmatte@gscancer.org, (²) National Institutes of Health

We previously found that 67-Ga was more potent and more specific in single cell kill than other Auger electron emitters and ß-particle emitters, using an anti-CD74 Ab (MHC class II invariant chain). Similar experiments have now been performed with other, more typical Abs. Target cells were Raji B-lymphoma cells, and the Abs tested were anti-mature MHC class II antigen, and anti-CD20. Labeling was with 125-I, 131-I, 111-In, or 67-Ga. Abs were incubated with the cells for 2 days, uptake of radioactivity was assayed at various times, and toxicity was assayed by a clonogenic assay. The Fraction Surviving was plotted vs. cumulative dpm/cell to determine relative potency. The ranking of the radionuclides for potency was 131-I > 67-Ga > 125-I > 111-In. 67-Ga was approximately 2- to 3-fold more potent than 111-In. This was very similar to previous results with anti-CD74. Dosimetry calculations were consistent with the level of toxicity observed.

**9:05 —97. Photonuclear production of In-111.** *Herbert A. Moore¹*, Richard F. Testa¹, Robert C. Block², Nathaniel Bobeck², and Yaron Danon². (¹) AlphaMed Inc, 4410 Lindholm Road, Metchosin, BC V9C 3Y1, Canada, hamoore@direct.ca, (²) Mechanical, Aerospace and Nuclear Engineering Department, Rensselaer Polytechnic Institute

Indium-111 has been used as a label for diagnostic radiopharmaceuticals for some time, most recently...
as the imaging surrogate for a newly approved Y-90 labeled therapeutic. In addition, Indium-111 is being examined for the potential that its very short range high LET Auger electrons may have in radiotherapeutic treatments of micro-tumors. The authors have been exploring the practicality of producing a number of medically important isotopes, such as In-111, via nuclear reactions promoted by photons with energies in the region of the Giant Dipole Resonance of requisite target isotopes. In the instance of In-111, we have demonstrated the feasibility of producing large quantities of radionuclidically pure, no carrier added, radionuclide by taking advantage of the two-stage sequence: Sn-112(photon,n)Sn-111 --> In-111

9:25 — 98. Evaluation of astatine-211-labeled targeted radiotherapeutics. Michael R. Zalutsky, Oscar Pozzi, Xiao-Guang Zhao, and Kevin L. Alston, Department of Radiology, Duke University, Box 3808, Durham, NC 27710, Fax: 919-684-7121, zalut001@mc.duke.edu

Although the therapeutic potential of alpha particle emitters such as astatine-211 has long been recognized, there are many hurdles which must be overcome in order to initiate clinical investigation of targeted therapeutics labeled with these radionuclides. Recently, we began a clinical trial of an At-211-labeled monoclonal antibody labeled using the N-succinimidyl 3-[At-211]astatobenzoate reagent in brain tumor patients. The chemical and biological studies that were needed in order to initiate this trial will be discussed as an example of the issues which must be addressed to evaluate a promising alpha-emitting compound in patients. These include the development of high-level radionuclide production methods and radiosynthetic approaches that are reliable at high activity levels, with radiolytic effects being a critical factor. Evaluation of the in vivo stability, normal organ toxicity, and therapeutic efficacy of the labeled molecule in animal models also will be addressed.

9:55 — Intermission.

10:15 — 99. Small astatinated molecules in tumor targeting. Per Hoff, Department of Chemistry, section 4, Nuclear Chemistry group, University of Oslo, P.O.Box 1033 Blindern, N-0315 Oslo, Norway, Fax: (+47) 22 85 54 77, Per.Hoff@kjemi.uio.no

The number of small astatinated molecules with potential for direct therapeutic tumor targeting increases slowly but steadily. Various methods have been tried out, but procedures involving (tributyl)-stannyl or (trimethyl)-silyl analogs as precursors seem to be superior. Hence, the synthesis of such compounds is of importance for the progress of this strategy. Bisphosphonates, with affinity for regions with pathologically increased bone turnover (targeting skeletal metastases and osseous osteosarcoma) and different analogs of melanin building blocks (targeting melanotic melanoma) are examples of compounds which may be astatinated through this type of methods. The main advantage of using small molecules for targeting is rapid penetration, uptake and clearance of the compound. The main disadvantages are enrichment in "problem organs", in vivo instability, and tumorous regions with low uptake. It is imperative to choose molecules which avoid the known mechanisms of enzymatic de-halogenation.

10:45 — 100. Optimizing methods for labeling biomolecules with astatine-211. D. Scott Wilbur¹, Donald K. Hamlin¹, Ming-Kuan Chyan¹, Catherine Foulon², Michael R. Zalutsky², Tim Wedge³, and M. Frederick Hawthorne³. (1) Department of Radiation Oncology, University of Washington, 2121 N. 35th Street, Seattle, WA 98103, Fax: 206-685-9630, dswilbur@u.washington.edu, (2) Radiology, Duke University Medical Center, (3) Department of Chemistry and Biochemistry, Univ. of California, Los Angeles

At present, astatine-211 is one of only a few alpha-emitting radionuclides that appear to be suitable for Targeted Radionuclide Therapy of cancer. Unfortunately, methods for coupling At-211 with cancer targeting carrier molecules are very limited, and in many examples result in the release of astatide in vivo. To develop new labeling methods, we have been conducting an investigation of the labeling and in vivo stability of a series of benzoyl and boron cage molecules. At-211 labeling of all compounds studied was fairly efficient (>50%). Varying degrees of stability were observed in the in vivo evaluations, but appeared to be related to the rate of blood clearance (metabolism) of the compound. A comparison of the preparation, physical properties, and in vivo stabilities of the astatinated compounds will be made.
Based on previous work utilizing the guanidine-containing acylation agent, N-succinimidyl 4-guanidinomethyl-3-[^131I]iodobenzoate ([^131I]SGMIB), we have now investigated the potential utility of its astato analogue for labeling the anti-epidermal growth factor receptor variant III (EGFRvIII) monoclonal antibody (mAb) L8A4. N-succinimidyl 3-[^211At]astato-4-guanidinomethylbenzoate ([^211At]SAGMB) in its Boc-protected form was prepared in 61.7 ± 13.1% radiochemical yield, in situ deprotected to[^211At]SAGMB, which was coupled to L8A4 in 36.1 ± 1.9% yield. Paired-label internalization assays demonstrated that tumor cell retention of radioactivity for L8A4 labeled using[^211At]SAGMB was almost identical to L8A4 labeled using [^131I]SGMIB, and 3-4-fold higher than for mAb radioiodinated using Iodogen. Paired-label biodistribution of L8A4 labeled using[^211At]SAGMB and [^131I]SGMIB in athymic mice hosting U87MG EGFR xenografts resulted in identical uptake of both[^211At] and [^131I] in tumor tissues over 24 h. These results suggest that[^211At]SAGMB may be a useful acylation agent for labeling internalizing mAbs with[^211At].

We have investigated the effect of treatment with a cytostatic agent combined with[^211At]-EGF targeting in vitro using two EGFR-rich cell lines; A431, which responds to the cytotoxic agent, and U343MGaCl2:1, found not to respond. In both cell lines, the combined treatment markedly increased the uptake of[^211At]-EGF. The survival was investigated using both a clonogenic survival assay and a cell growth assay. With the combined treatment, the survival of U343 cells was at least 3.5 times lower than with[^211At]-EGF treatment alone. For the A431 cells, however, the combined treatment increased the survival.

Engineered anti-tumor antibody fragments exhibit rapid and highly specific tumor targeting for the delivery of therapeutic radioisotopes. Successful RAIT strategies depend upon selecting radioisotopes with physical properties complementary to the biological properties of the antibody fragment. The C6.5 diabody has a T1/2 alpha (equilibration phase) of 0.7 hrs, a T1/2 beta (elimination phase) of 6 hrs and a T1/2 in tumor of ~30 hrs which favors the use of short-lived radioisotopes. In particular,[^211At] (T1/2=7.2 hrs) was hypothesized to be very effective in diabody-directed RAIT. This hypothesis was tested in immunodeficient mice bearing established HER2/neu positive MDA-MB-361 DYT2 tumors treated with[^211At]-SAPS –C6.5 diabody at below the maximum tolerated dose. Results of experiments treating at single i.v. doses of 20 mCi, 30 mCi and 45 mCi will be presented. Supported by U.S. Dept. of Energy Grant DE-FG02-01ER63190.
Section B
Convention Center -- Room 391

Analytical Chemistry in Nuclear Technology
Cosponsored with Division of Analytical Chemistry

Developments in Techniques for Analyzing Solid Samples

T. Barber and T. P. O’Holleran, *Presiding*

8:30 — Introductory Remarks.

8:40 — 104. Analysis of the glass phase of a ceramic high-level waste form by analytical electron microscopy. T. P. O’Holleran, and Jan-Fong Jue, Engineering Technology Division, Argonne National Laboratory - West, P. O. Box 2528, Idaho Falls, ID 83403-2528, Fax: 208-533-7863, tom.oholleran@anl.gov

A ceramic waste form has been developed to immobilize the salt waste stream from electrometallurgical treatment of spent nuclear fuel. The waste form consists of sodalite grains (the host phase for the waste salt) bonded together with a borosilicate glass. The waste form is produced by first occluding waste salt in dehydrated LTA zeolite powder. The salt loaded zeolite powder is mixed with a borosilicate glass frit and fired at temperatures up to 1220 K. The firing converts the zeolite to sodalite and consolidates the mixture, with the glass acting as a binder. At elevated temperatures, complex reactions between the glass and aluminosilicate phase significantly alter the composition of the glass. Characterization results are reported, and mechanisms are proposed to explain the experimental observations.

9:10 — 105. Microscopy studies of cation incorporation into uranium(VI) solid phases. Matthew Douglas¹, Sue B. Clark¹, Satoshi Utsunomiya², and Rodney C. Ewing². (1) Department of Chemistry, Washington State University, P.O. Box 644630, Pullman, WA 99164-4630, matt_douglas@wsu.edu, (2) Department of Nuclear Engineering and Radiological Sciences, University of Michigan

Uranium(VI) silicates such as uranophane, Ca[(UO2)(SiO3OH)]2•5H2O, are expected to be one of the long-term solubility limiting phases in the Yucca Mountain high-level waste repository, and may control dissolved concentrations of fission products and other actinides that become incorporated into them. To evaluate its capacity for incorporation, uranophane was synthesized in the presence of a variety of metal ions anticipated to be present in the repository. The resulting crystalline phases were characterized by X-ray powder diffraction (XRD), scanning electron microscopy (SEM), transmission electron microscopy (TEM), and analytical electron microscopy (AEM). In some cases, cations such as Cs, Sr, and Eu show some association with uranophane. In addition, polycrystalline phases of differing structure and elemental composition have been observed, and in most cases contain the majority of the foreign cations. Results and their implications for understanding the solid phase chemistry of U(VI) will be discussed.

9:40 — 106. Identification of UO2+3 species formed by aqueous corrosion of uranium metal. Jacqueline E. Fonnesbeck, and Jeffrey F. Berg, Nuclear Technology Division, Argonne National Laboratory-West, PO Box 2528, Idaho Falls, ID 83403, Fax: 208-533-7755, jacqueline.fonnesbeck@anlw.anl.gov

Aqueous corrosion of uranium metal can produce hydrogen, uranium hydride, and uranium oxide. Specifically, this study focused on the composition of the uranium oxide corrosion product formed from this process. Previous research has shown that this corrosion product is composed primarily of uranium dioxide (UO2). However, recent studies at Argonne National Laboratory-West have shown an indication of the presence of higher valence uranium oxides in this corrosion material, i.e. U3O8, UO3, etc. Analytical measurements used for this study included X-Ray powder Diffraction analysis for general product identification, Thermal Ionization Mass Spectrometry for uranium concentration, LECO™ O2 analysis for oxygen concentration as well as for temperature dependent oxide decomposition profiles to confirm the presence of higher level uranium oxides.
10:10 — Intermission.

10:30 — 107. Identification of \( \text{UO}_2^{+x} \) species formed by aqueous corrosion of uranium metal. Jacqueline E. Fonnesbeck, and Jeffrey F. Berg, Nuclear Technology Division, Argonne National Laboratory-West, PO Box 2528, Idaho Falls, ID 83403, Fax: 208-533-7755, jacqueline.fonnesbeck@anlw.anl.gov

Aqueous corrosion of uranium metal can produce hydrogen, uranium hydride, and uranium oxide. Specifically, this study focused on the composition of the uranium oxide corrosion product formed from this process. Previous research has shown that this corrosion product is composed primarily of uranium dioxide (\( \text{UO}_2 \)). However, recent studies at Argonne National Laboratory-West have shown an indication of the presence of higher valence uranium oxides in this corrosion material, i.e. \( \text{U}_2\text{O}_5 \), \( \text{UO}_3 \), etc. Analytical measurements used for this study included X-Ray powder Diffraction analysis for general product identification, Thermal Ionization Mass Spectrometry for uranium concentration, LECO \( \text{O}_2 \) analysis for oxygen concentration as well as for temperature dependent oxide decomposition profiles to confirm the presence of higher level uranium oxides.

11:00 — 108. Multivariate calibration for the analysis of alloys by laser-induced breakdown spectroscopy. Scott R. Goode, Richard Hoskins, and Stephen L. Morgan, Department of Chemistry and Biochemistry, University of South Carolina, Columbia, SC 29208, goode@sc.edu

Laser Induced Breakdown Spectroscopy (LIBS) has been utilized as an analytical tool for many years. It is a rapid method for analysis of many types of samples including alloys, geologic specimens, precious art works and requires almost no sample preparation. Although LIBS has enjoyed acceptance as an analytical tool in many areas it does have several interferences effecting the technique. With multivariate calibration, multiple matrix effects can be reduced making it possible to produce calibration curves for alloys. In this paper, we will present results that indicate multivariate calibration can obviate the need for matrix-matched standards for analyzing alloys and other materials.

WEDNESDAY AFTERNOON

Section A
Convention Center -- Room 390

Development of High-LET Therapeutic Radiopharmaceuticals
Cosponsored with Division of Inorganic Chemistry, and Division of Medicinal Chemistry

Other Alpha Emitters and Applications

T. J. Ruth and T. M. Behr, Presiding
M. W. Brechbiel and D. S. Wilbur, Organizers

1:30 — 110. Production of high-LET radioisotopes at TRIUMF-ISAC. Thomas J. Ruth, TRIUMF, 4004 Wesbrook Mall, Vancouver,
The future growth area in nuclear medicine will be in use of radiotoxic isotopes for therapy (RIT). The idea of a radioisotope used in therapy is based on the desire to link a radionuclide which has a high linear energy transfer associated with its decay products (Auger electrons, -particles or -particles) to a biologically active molecule that can be directed to a tumor site. Most of the radionuclides of interest are produced in reactors. The difficulty with reactor produced radioisotopes is that they have, in general, low specific activity, limiting the ability to deliver sufficient radiotoxic affects. Thus alternative methods of production need to be found. The TRIUMF online isotope separator facility (ISAC) has the capability of producing extremely high purity isotopes, which makes it possible to explore the advantages of ultra high specific activity in the context of selecting the appropriate radioisotope and its production for RIT.

2:00 —111. Biomedical application of alpha-particle-emitting radium-223. Roy H. Larsen, Anticancer Therapeutic Inventions AS, Kjelsåsveien 172 A, 0884 Oslo, Norway, Fax: +47 23007991, roy.larsen@ati-as.no

Radium-223 (t1/2=11.4 days) decays via a chain of short-lived daughter nuclides to stable lead, producing four alpha particles. Significant efforts have been made to link 223Ra to tumor seeking carrier molecules. An alternative way to utilize 223Ra has recently been explored. This is the use of cationic 223Ra against skeletal metastases of prostate- and breast cancer. Studies in a nude rat skeletal metastases model of human breast cancer, resistant to high dose cisplatin and doxorubicin as well as pamidronate and 131I-labeled bisphosphonate treatment, indicated a significant antitumor effect from 223Ra. A phase I dose escalating study showed that therapeutic relevant activity levels could be given to patients without dose limiting toxicity. Responses as pain relief and/or reduction in the alkaline phosphatase level, a biomarker often elevated in patients with skeletal metastases, were observed in the majority of the patients.

2:30 —112. Targeted alpha-particle radioimmunotherapy for prostate cancer. Dangshe Ma, Christine Hopf, Jason P. Gardner, Gerald P. Donovan, Norbert Schülke, Danielle Fisch, and William C. Olson, PSMA Development Company, 777 Old Saw Mill River Road, Tarrytown, NY 10591, Fax: 914-789-2807, dma@progenics.com

Prostate cancer causes the deaths of over 30,000 Americans annually, and there is no effective therapy for recurrent disease. Monoclonal antibodies (mAbs) labeled with beta-particle emitting isotopes have been shown to target metastatic disease in man; however, meaningful clinical responses remain to be seen. Compared to beta-emitters, alpha particle-emitting radioisotopes may offer improved potency and specificity due to the short path length and high linear energy transfer of alpha particles. A few alpha particles can kill a single cancer cell. We have generated fully human mAbs with sub-nanomolar affinity for prostate-specific membrane antigen (PSMA), a tumor-associated antigen whose expression increases with disease progression. The mAbs possess high immunoreactivity, rapidly internalize, and selectively kill PSMA-expressing target cells when labeled with Ac-225, an alpha-emitter with radiologic properties that are attractive for therapy. Alpha-particle radioimmunotherapy holds great promise for treatment of metastatic prostate cancer.

2:50 —113. Controlling the fate of Ac-225 and its daughters. Michael R. McDevitt, Molecular Pharmacology and Chemistry Program, Memorial Sloan-Kettering Cancer Center, 1275 York Avenue, New York, NY 10021, Fax: 212-717-3622, m-mcdevitt@ski.mskcc.org

The alpha particle emitting Ac-225 nuclide is extremely cytotoxic compared to other therapeutic radionuclides. Our laboratory has developed methods to target molecular-sized generators of alpha-emitting isotope cascades to the inside of cancer cells using Ac-225 coupled to internalizing monoclonal antibodies. Treatment of mice bearing xenografts of solid prostate carcinoma, disseminated lymphoma, and ovarian carcinoma with single doses of the constructs at nanocurie levels induced tumor regression and prolonged survival in a substantial fraction of animals without toxicity. While
there has been no toxicity observed in these studies, the fate of the radionuclidic daughters emitted through the Ac-225 decay process presents a concern that is being addressed. The ongoing development of these constructs involves the examination of methods to abrogate the potential toxicity of the Ac-225 radionuclidic daughters. Strategies to accomplish this by controlling the clearance of parent and daughters will be presented.

3:10 — Intermission.

3:30 — 114. Radiopeptide therapy with gastrin-receptor ligands: Toxicity and therapeutic efficacy of alpha, Auger electron, and beta emitters. Thomas M Behr, and Martin P. Behe, Dept. of Nuclear Medicine, University of Marburg, Baldingerstr, D-35043 Marburg, Germany, Fax: +49-6421-28-67025, tmbehr@mail.uni-marburg.de

In our previous work, we have developed CCK-B receptor ligands for therapy of CCK-B receptor expressing tumors (e.g., medullary thyroid cancer [MTC] or small cell lung cancer [SCLC]). The aim of this study was to assess their toxicity and anti-tumor efficacy, labeled with high-LET emitters (e.g., Auger electron or alpha) emitters as compared to low-LET beta-emitters. A minigastrin derivative, which rapidly internalizes after receptor binding, was derivatized with a novel DTPA-based chelator with improved stability for various radiometals, such as beta (Y-90, Sm-153), Auger electron (In-111, Ga-67, Nd-140) or alpha-emitters (Bi-213/Po-213, Ac-225). Biodistribution studies were performed in nude mice bearing MTC xenografts. The maximum tolerated doses (MTDs) of each agent were determined. Groups of mice were injected with the various radiopeptides, whereas controls were left untreated. Myelo-, 2nd organ toxicity and tumor growth were monitored. In accordance with uptake values of > 30 %ID/g, the kidney was the first dose-limiting organ with all radiolabels. With all beta- and alpha-emitters, renal doses of > 90 Gy led to acute, > 60 Gy to chronic radiation nephropathy. With the Auger electron emitters, renal doses approximately twice these thresholds were tolerated. Whereas no gastrointestinal (GI) toxicity was observed with beta-emitters despite their pronounced CCK-B receptor mediated uptake in the gastric mucosa, at their respective MTDs all high-LET (Auger electron and alpha) emitters led to hemorrhagic gastritis. At equitoxic dosing, all high-LET (i.e., Auger electron and alpha) emitters were significantly (~2-3-fold; p<0.01) more therapeutically effective than the beta-labeled conjugates. Permanent cures were exclusively observed with Auger electron and alpha, not with beta-emitters. These data suggest that high-LET emitters may be therapeutically more effective than beta-emitters. Surprisingly, MTDs were very similar between alpha- and beta-, but higher with the Auger-electron emitters, which may be due to their short path length, selectively irradiating the tubulus cells. The more local energy deposition of high-LET radiation in the gastric mucosa may explain why GI toxicity exclusively occurs with high-, but not with low-LET emitters.

4:00 — 115. Applications for short-lived alpha emitters. Stephen J. Kennel, Sandra M. Davern, and Saed Mirzadeh, Life Sciences Division, Oak Ridge National Laboratory, Knoxville, TN 37831-8077, Fax: 865-574-1274, kennelsj@ornl.gov

For cancer endo-radiotherapy, targeting to solid tumors with conventional means usually takes a day or more. Cancer radiotherapy with short half life alpha emitters Bi-213, Bi-212 and At-211 has focused on treating tumor cells found chiefly in the blood stream. Rapid targeting to solid tumors has utilized small targeting agents including diabodies and scFv, two step targeting including streptavidin-biotin systems and bispecific antibodies and targeting to tumor blood vessels. We have shown that targeting of Bi-213 and At-211 to tumor blood vessels is therapeutic for small tumors in a model system. The challenge now is to find targeting agents that bind selectively to tumor vasculature. We are using in vivo selection of phage display libraries to identify potential targeting agents. For the application of treating autoimmune disease, we are trying to use alpha emitters to delete circulating antigen specific T lymphocytes from immune animals. Tetrameric complexes of major histocompatabilitiy protein-antigen constructs have been prepared and covalently conjugated to CHX-A"-DTPA ligand. Specific binding of these complexes to immune spleen cells has been demonstrated. Immune mice treated with Bi-213 tetrarmers have been tested for antigen positive, CD8 positive T cells using flow cytometry. The results of
these experiments will be discussed.

4:20 —116. Peptide-targeted Pb-212/Bi-212 alpha radiation for melanoma therapy. Thomas P. Quinn1, JianQing Chen1, Weiwei Feng1, Mark Hylarides2, and Alan Fritzberg2. (1) Department of Biochemistry, University of Missouri-Columbia, 117 Schweitzer Hall, Columbia, MO 65211, Fax: 573-884-4812, quinnt@missouri.edu, (2) NeoRx Corporation

The incidence of malignant melanoma is on the increase, while the median survival for patients with metastatic disease remains low. In response to the need for new melanoma therapeutics, our laboratory has developed a novel family of metal-cyclized alpha-melanocyte stimulating hormone (MSH) peptide analogs that target alpha-MSH receptors present on melanoma tumor cells. In this study, the alpha-MSH peptide analog DOTA-ReCCMSH was examined for its ability to target Pb-212 and its alpha-particle emitting daughter Bi-212 to melanoma tumors. DOTA-ReCCMSH was radiolabeled with Pb-203, Bi-207 and Pb212/Bi-212. The RP-HPLC purified radiolabeled complexes were shown to be radiochemically stable in vitro over a 24 h period except for the Pb/Bi-212 complex that exhibited some instability associated with its decay. Initial biodistribution studies of the Bi-207, Pb-203, Pb212/Bi-212 labeled DOTA-ReCCMSH complexes performed in melanoma-bearing mice showed significant tumor uptake and high retention coupled with rapid clearance kinetics, highlighting the therapeutic potential of Pb-212/Bi-212[DOTA]-ReCCMSH.

4:40 —117. Radioimmunotherapy of intraperitoneal disseminated disease: Preliminary evaluation of Herceptin as a radioimmunoconjugate. Martin W. Brechbiel1, Diane E. Milenic2, Kayhan Garmestani3, Alia Abdulla3, Terrish Overstreet2, and Joe Flynn4. (1) National Cancer Institute, NIH, Building 10, Room B3B69, 10 Center Drive, Bethesda, MD 20892, Fax: 301-402-1923, (2) National Cancer Institute, (3) Radioimmune and Inorganic Chemistry Section, National Institutes of Health, (4) Walter Reed Medical Center

The effectiveness of Herceptin, a monoclonal antibody that recognizes the HER2 antigen, and approved by the FDA for the treatment of breast cancer could be expanded if used as a vector for therapeutic radionuclides. Herceptin has been conjugated with the acyclic and the macrocyclic bifunctional chelates, CHX-A° DTPA and TCMC, respectively, and the tumor targeting (In-111) and efficacy (Bi-213, Bi-212, Pb-212, and Y-90) evaluated in the treatment of disseminated, metastatic disease was employing two intraperitoneal tumor animal models: 1) the LS-174T cell line, a human colon carcinoma, and 2) the SHAW cell line, a pancreatic carcinoma. Results pertaining to targeting, MTD, and therapeutic efficacy with alpha-emitting radionuclides both as a single dose and in a fractionated regimen as well as in conjunction with a radiosensitizer will be presented.

5:00 — Discussion.

5:15 — Concluding Remarks.

Section B
Convention Center -- Room 391

Analytical Chemistry in Nuclear Technology
Cosponsored with Division of Analytical Chemistry

General Developments

D. T. Hobbs and C. J. Coleman, Organizers, Presiding
J. C. Griffin and D. E. Hobart, Organizers

1:30 — Introductory Remarks.

1:35 —118. ESI-MS investigation of uranyl(VI) speciation in the presence of citrate. Sofie P Pasilis, Arpad Somogyi, and Jeanne E. Pemberton, Department of Chemistry, University of Arizona, 1306 E. University Blvd., Tucson, AZ 85721, Fax: 520-621-8248, pasilis@email.arizona.edu

The ability to generate gas–phase ions from solution species makes ESI–MS a promising technique for the investigation of actinide complexes with organic ligands. Here, ESI–MS was used with Raman spectroscopy to investigate complexes formed between uranyl(VI) and citrate in solution. Raman
spectroscopy was used to examine the pH dependence of complex formation. Complex stoichiometries were confirmed with ESI–MS by altering uranyl(VI) and citrate (Cit) ratios, pH, solvent, and solution counter anion. At pH < 5, (UO₂)₂Cit₂ is the predominant species, but at higher pH, (UO₂)₃Cit₂ and (UO₂)₃Cit₃ are prevalent. The UO₂–citrate species form singly–charged cluster ions with alkali metal cations, solvent molecules and counter anions in the gas phase. Preliminary results further suggest gas–phase electron capture by uranyl(VI) to form uranyl(V) in these systems.

2:05 — 119. Vanadium isotopic analysis by thermal ionization mass spectrometry. Yong-Hong Zhang, Department of Nuclear Engineering, Tokyo Institute of Technology, 2-12-1 O-okayama, Meguro-ku, Tokyo 152-8550, Japan, Fax: 81-3-5734-2958, yhzhang@nr.titech.ac.jp, Masao Nomura, Research Laboratory for Nuclear Reactors, Tokyo Institute of Technology, and Yasuhiko Fuji, Research Laboratory for Nuclear Reactors, Tokyo Institute of Technology

The geochemical and biological behaviors of vanadium have recently received considerable attention, because this element is classified as a trace bio-element. V consists of a mixture of two naturally occurring isotopes, V-50 and V-51. Significant isotopic fractionations in the ratio of V-50/V-51 were observed in some petroleum and certain ascidians, which have been well known to highly concentrate vanadium. For detecting vanadium isotopic fractionations in nature and studying vanadium isotope effects, it is necessary to establish a reliable vanadium isotope mass-spectrometric determination. To achieve these, the effects of the isobars, the quantities, the chemical forms of vanadium compounds, and the variation with time in mass spectrometry were investigated. Employing the obtained techniques and the optimum conditions, we determined the vanadium isotopic ratio in seawater after isolation of vanadium form many other kinds of metals especially uranium recovered with the Amidoxime adsorbents from seawater in the Shimokita Peninsula of Japan by JAERI. It is found that within the experimental uncertainty, there is no variation in the ratio of V-50/V-51 between the selected reagents and seawater.


Reprocessing of irradiated fuel and target rods at the Savannah River Site (SRS) discards small quantities of plutonium to the high-level waste (HLW) system. Due to the strongly alkaline conditions, most of the plutonium reports to the precipitated solids fraction of the waste, which is referred to as sludge. However, the HLW solutions do contain small concentrations (~1.0E-05 M) of plutonium, which require removal before disposal. SRS produces two types of plutonium materials, weapons grade plutonium, which is comprised chiefly of ²³⁹Pu, and heat source plutonium, which is comprised chiefly of ²³⁸Pu. The specific activities of these plutonium isotopes vary by a factor of 279 and, consequently, the alpha activity in HLW solutions due to plutonium can vary by several orders of magnitude depending on the concentration and isotopic distribution of the plutonium. This paper will discuss analytical methods used to determine the concentration and isotopic distribution of plutonium in alkaline solutions as well as findings from measurements with actual and simulated tank wastes.

3:05 — Intermission.

3:25 — 121. Quantitative gas analysis by a quadrupole mass spectrometer. Lily L. Wang, Los Alamos National Laboratory, Los Alamos, NM 87545, Fax: 505-667-5268, wangl@lanl.gov, and Quirinus G. Grindstaff, BWXT Y12 Development

A method to quantify gas components in small, low pressure samples has been developed using a quadrupole mass spectrometer. The method employed calibration of direct introduction of a small volume (1.47cc) of each pure gas of interested at pressure ranging from 0.05 to 1.0 torr to obtain its sensitivity factor. Relatively short analysis time for each determination, in the order of a few seconds, is required. The gas analysis system consists of a custom-designed manifold and a quadrupole mass
spectrometer. The analysis of samples can be performed using the same small volume or some larger know volumes. The stability of the sensitivity factors was evaluated with pure H2, CH4, and Ar gases for over a period of about two months. The reproducibility and accuracy of the method were demonstrated subsequently with a known gas mixture containing H2, CH4, and Ar over a 19-day period using sensitivity factors obtained previously. The method is applicable to quantitative analysis of radiological gas samples where the quantity of sample gas is to be minimized; moreover the method can be applied to quantifying gas species produced from ongoing processes with time.

3:55 —122. From conceptualization to "open for business": Analysis of uranyl nitrate solutions of varying enrichments and concentrations at the Savannah River Site. Michael J. Brisson1, Sheldon T. Nichols2, Joey J. Smiley2, and Jessica Urbanik-Coughlin2. (1) Analytical Laboratories Dept, Westinghouse Savannah River Co, Savannah River Site MS 704-17F, Aiken, SC 29808, Fax: 803-952-2147, mike.brisson@srs.gov, (2) Analytical Laboratories Department, Westinghouse Savannah River Co

The Savannah River Site (SRS) is located in Aiken, SC, and is operated by Westinghouse Savannah River Co. under contract to the U. S. Department of Energy. In 1999, conceptual design began for a program to blend down existing stocks of 61% enriched U-235 (in a uranyl nitrate matrix) to a nominal 5% enrichment for use as fuel in Tennessee Valley Authority (TVA) reactors. Analyses required include uranium content and isotopes, radiochemistry, and a wide variety of impurities. This presentation describes the process of selection of analytical methods and equipment, laboratory renovations, and method testing performed to prepare the SRS Central Laboratory to receive and analyze samples for this program. Method uncertainties, based on available testing performed to date, will be discussed.


Because WSRC site facilities use tributyl phosphate (TBP) and normal paraffin hydrocarbons (NPH) for separations processes, the need for simple and fast analyses of these materials on plant has always been great. Various methods have been developed for preparing and analyzing TBP in the semivolatile organics analysis (SVOA) lab to accommodate analysis of TBP solutions in a variety of sample matrices produced by various customer research studies and plant processes. A method has been developed for quantitation of TBP and NPH which uses isotopic dilution with TBP-d27 (C/D/N Isotopes, Inc., Pointe-Claire, Quebec, Canada H9R 1H1) and dodecane-d26 (Aldrich Chemical, Milwaukee), and analysis by Gas Chromatography / Mass Spectrometry (GC/MS) with selected ion monitoring. This technique is a modification of an earlier method developed for the analysis of only TBP. The standards are added to the sample aliquot prior to a single extraction, followed by GC/MS analysis. This method provides a fast and efficient way of accurately determining TBP and NPH in a variety of sample matrix types.

THURSDAY MORNING

Section A
Convention Center -- Room 390

Radiochemistry at RIA
Cosponsored with Division of Analytical Chemistry

P. F. Mantica and M. A. Stoyer, Organizers, Presiding

8:30 — Introductory Remarks.

8:35 —124. RIA facility overview. Richard C. York, National Superconducting Cyclotron Laboratory, Michigan State University, East Lansing, MI 48824-1321, Fax: 517-353-5967, york@nscl.msu.edu
RIA Facility Overview – The Rare Isotope Accelerator (RIA) will provide research opportunities with rare isotopes that are presently not available. RIA will utilize a superconducting driver linac capable of accelerating protons through uranium to energies >400 MeV/nucleon at power levels >100 kW. The primary stable beams will be used to produce at substantial intensities rare isotopes by any of several means providing isotopes that now exist only in the cosmos. RIA will enable major inroads into uncharted areas of nuclear physics improving our understanding of nuclear matter and supporting groundbreaking interpretations of astronomical observations. Because of RIA’s large discovery potential, the Nuclear Science Advisory Committee (NSAC) to the National Science Foundation and the Department of Energy has stated that RIA is the nuclear science community’s highest priority for major new construction. RIA will also provide basic information necessary for reliable nuclear weapons stockpile stewardship. An overview of the RIA concept and scope will be presented.

9:05 —125. Physics opportunities with RIA. Robert V. F. Janssens, Physics Division, Argonne National Laboratory, 9700 S. Cass Ave., Argonne, IL 60439, Fax: 630-252-6210, janssens@anl.gov

The field of nuclear physics with exotic nuclei is both rich and diverse. By providing access to the wider, uncharted landscape of unstable nuclei, RIA will provide the essential tools to address critical issues in three broad areas: the nature of nucleonic matter, the origin of the elements and of energy generation in stars and tests of the standard model and of fundamental conservation laws. Examples of important issues in these three areas will be discussed.

*This work was supported in part by the U. S. Department of Energy, Nuclear Physics Division, under Contract No. W31-109-ENG-38.

9:35 —126. Harvesting isotopes for neutron cross-section measurements at RIA. Larry E. Ahle, and L. A. Bernstein, Physis and Advanced Technologies, Lawrence Livermore National Laboratory, L-414, PO Box 808, Livermore, CA 94551, Fax: 925-422-5940, ahle1@llnl.gov

Neutron cross-section measurements on radioactive nuclei are of interest to the Stockpile Stewardship program. Performing these measurements directly requires radioactive isotopes to be produced, harvested, and processed into targets. The requirements for isotope harvesting are explored by looking at an experiment to measure the 89Zr(n,2n) and 89Zr(n,np) cross-sections as a function of beam energy to be performed at LBNL over the next couple of years. One of the most stringent requirements turns out to be purity, often implying a contamination of less than 10^-9 for certain isotopes. These requirements are then compared to the isotope production capabilities at RIA. Specifically, a production station at the first stripper location of the driver, the ISOL target facility, and the fragmentation line are all examined. Limitations of each are explored as well as possible enhancements that could be added to allow these production facilities to meet the requirements for neutron cross-section direct measurements. This work was performed under the auspices of the U.S. Department of Energy by the University of California, Lawrence Livermore National Laboratory under contract No. W-7405-Eng-48.

10:05 —131. Who will do the nuclear science at RIA? Training the next generation of nuclear scientists for DOE. Sue B. Clark, Department of Chemistry, Washington State University, P.O. Box 644630, Pullman, WA 99164-4630, s_clark@wsu.edu

Recent projections of human resource needs at most national laboratories indicate a significant need for technical personnel, especially in the area of nuclear science. At the same time, recent surveys of colleges and universities demonstrate decreasing opportunities for academic training in these areas. Similar problems are reported in other countries. Although the national laboratories are accustomed to the necessity of providing on-the-job training in these areas, many of the brightest young students are lured away from considering these areas because nuclear science is so poorly represented in most academic technical disciplines. These situations combined leads to the question, "Who will do the work of research and discovery at new DOE facilities such as RIA?" This question and some of the current educational programs in the
nuclear sciences will be discussed. In addition, ideas for new programs will be suggested.

10:25 — Intermission.

10:45 —127. Requirements for a radiochemical harvesting facility. Kenton J. Moody, Analytical and Nuclear Chemistry Division, Lawrence Livermore National Laboratory, P.O. Box 808, L-231, Livermore, CA 94551

It is planned that RIA will provide for the harvesting of radionuclides, mainly for target preparation. This activity will require the use of an on-site radiochemistry facility, in which the chemical manipulation of the product radionuclides can be accomplished. Most of the chemical separation operations will have to be performed through the use of remote manipulators to reduce personnel exposure. The design of a facility in which the required separation and fabrication steps may be performed must take into account issues of equipment reliability, cross contamination and decontamination, efficiency of space utilization, transportation of short-lived species from one chemical step to another, methods of target fabrication, and the “clean” extraction of the final purified target. Examples of facilities in which remote chemical operations were performed will be given.

11:15 —128. Fundamental symmetry measurements with trapped radioactive atoms at RIA. David J. Vieira, Marc Hausmann, Jinwei Wu, and Xinxin Zhao, Isotope and Nuclear Chemistry, Los Alamos National Laboratory, Mail Stop J514, Los Alamos, NM 87545, Fax: 505-665-4955, vieira@lanl.gov

RIA will make possible a wide variety of fundamental symmetry / weak interaction studies. In particular, the very intensity mass- and recoil-separated beams that will be available at RIA will enable high-precision measurements of beta-neutrino correlations, atomic parity nonconservation, nuclear anapole moment, and the search for a nuclear electric dipole moment. These will provide important new constraints on the Standard Model.
A powerful, new tool with which to undertake these measurements is the ability to trap radioactive atoms in a magneto-optical trap (MOT) and pure optical dipole or magnetic traps. To illustrate the potential for such techniques, I will highlight the recent work that we have done in trapping radioactive atoms with high efficiency using a mass separator coupled MOT system. Once trapped in a MOT they are then polarized and loaded into a pure magnetic trap called a time-averaged orbiting potential (TOP) trap where they confined with lifetimes of 100+ seconds. We are presently pursuing two high-precision measurements using the TOP trap technology. The first is the high-precision parity-violation measurement involving the beta-asymmetric decay of polarized $^{82}$Rb ($t_{1/2}=76$ s). The second experiment is an atomic physics experiment involving the sympathetic cooling of radioactive $^{84}$Rb ($t_{1/2}=33$ d, fermionic) by ultracold $^{87}$Rb (stable, bosonic) atoms. The latest developments on these experiments and future experiments that are envisioned for RIA will be discussed.
Work performed under the auspices of the U.S. Department of Energy under contract W-7405-ENG-36.

11:35 —129. Production of radioisotopes for medical applications at RIB. Thomas J. Ruth, TRIUMF, 4004 Wesbrook Mall, Vancouver, BC V6T 2A3, Canada, Fax: (604) 222-1074, truth@triumf.ca

The production of radioisotopes for use in medicine is achieved through nuclear reactions in reactors or from charged particle bombardment in accelerators. The most important aspects in the field of nuclear medicine today are: to increase the specificity of functional imaging, the quantification of medical imaging techniques based on PET and SPECT and the development of new approaches in systemic radioisotope therapy (RIT). There are three main aspects which make radio-isotopes available at Radioactive Ion Beam facilities valuable for nuclear medicine: 1) availability of exotic or uncommon radio-isotopes, 2) the unprecedented high purity (isotopically separated and carrier-free) and 3) the ion beam quality of the radionuclides. The universal unspecific spallation reaction makes a complete range of isotopes having as complete a diversity of types and energies of radiation, of half-life, and of chemical properties available as one would wish.

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11:55 — 130.  New concept for exploiting RIA for radioisotope production. Jose R. Alonso, Lawrence Berkeley National Laboratory, MS71R259, 1 Cyclotron Road, Berkeley, CA 94720, Fax: 510-486-5788

The very high currents of diverse ion species available in the RIA Driver (eg 100 pA for Z 10) could provide an exciting new production source for isotopes of interest for radiopharmaceutical and other radioisotope applications. One simple concept would call for providing the capability of peeling off beams of energies less than 10 MeV/amu into a dedicated radioisotope production facility, and making use of the quite-sharp excitation functions for (HI,xn) reactions to enhance the yields of desired isotopes with a minimum of post-processing. Heavy-ion (or more appropriately – light-ion) accelerators have never been considered practical for commercial isotope production because of the low current usually available from such machines. The RIA Driver for the first time offers a system capable of overcoming this limitation, and so should be seriously viewed as a viable source of commercially-interesting isotopes. With energy widths of a few MeV/amu, the (xn) cross sections allow for targets of a few mg/cm² to cover cleanly the range of efficient production of a given isotope. Flexibility of choosing different ion species could provide significant reduction in feedstock cost by allowing more freedom to select odd-A (generally monoisotopic) targets, thus reducing need for isotopically enriched target materials. The flexibility offered by the RIA Driver should be carefully studied by the radioisotope community.

Section B
Convention Center -- Room 391

General Topics in Nuclear Chemistry
S. C. Srivastava, Organizer, Presiding

8:30 — Introductory Remarks.

8:35 — 132. First chemical investigation of hassium (Hs, Z = 108). Christoph E. Düllmann¹, Willy Brüchle², Rugard Dressler³, Klaus Eberhardt⁴, Bernd Eichler³, Robert Eichler², Heinz Gäggele³, Thomas N. Ginter⁵, Fredy Glaus³, Kenneth E. Gregorich⁶, Darlene C. Hoffman⁸, Egon Jäger², Dieter Jost¹, Uwe W. Kirbach⁶, Diana M. Lee⁶, Heino Nitsche⁶, Joshua B. Patin³, Valeria Pershina², Dave Piguett, Zhi Qin⁹, Matthias Schädel², Brigitta Schauten², Erwin Schimpf³, Hans-Joachim Schött², Sandra Soverna⁸, Ralf Sudowé⁸, Petra Thörle¹, Sergei N. Timokhin¹⁰, Norbert Trautmann⁴, Andreas Türlæ³, Annett Vahle¹¹, Gerhard Wirth², Alexander B. Yakushev¹⁰, and Peter M. Zieinski³.

¹ Present address: Lawrence Berkeley National Laboratory, 1 Cyclotron Road MS 88R0192, Berkeley, CA 94720, Laboratory for Radio- and Environmental Chemistry, Universität Bern and Paul Scherrer Institut, Freistrasse 3, CH-3012 Bern, Switzerland. Fax: 510-486-6707, CEDuellmann@lbl.gov, (2) Gesellschaft für Schwerionenforschung mbH, (3) Laboratory for Radio- and Environmental Chemistry, Paul Scherrer Institut, (4) Institut für Kernchemie, Universität Mainz, (5) Laboratory for Radio- and Environmental Chemistry, Paul Scherrer Institut and Bern University, (6) Nuclear Science Division, Lawrence Berkeley National Laboratory, (7) Laboratory for Micro- and Nanotechnology, Paul Scherrer Institut, (8) Nuclear Science Division, Lawrence Berkeley National Laboratory, and Department of Chemistry, University of California, Berkeley, (9) Institute of Modern Physics, Chinese Academy of Science, (10) Flerov Laboratory of Nuclear Reactions, Joint Institute for Nuclear Research, (11) Research Center Rossendorf e.V

We present the first successful chemical investigation of hassium (Hs, Z=108). Relatively long-lived (T½~10s) Hs nuclides were produced in the nuclear reaction 248Cm(26Mg;5,4n)269,270Hs. Fusion products were converted to volatile HsO₄, which was rapidly transported to a thermochromatography detection system. The deposition temperature of HsO₄ on the column surface material was measured using a column consisting of PIN diodes suitable for registering α-decaying and spontaneously-fissioning nuclides. A longitudinal negative temperature gradient from -20 to -170°C was established along the column. A dose of 1.0·1E18 Mg-26-projectiles was accumulated. Seven correlated decay chains were registered. Three were attributed to the decay of Hs-269, and two chains were tentatively assigned to the decay of the new isotope Hs-270. The two last decay chains were incomplete. From the deposition temperature,
the adsorption enthalpy was deduced as \((-46\pm2)\) kJ/mol for HsO₄ compared to \((-39\pm1)\) kJ/mol for OsO₄, indicating a comparable or slightly lower volatility for HsO₄. The formation of a highly volatile oxide justifies the classification of Hs into group 8 of the periodic table.

8:55 —133. Age determination of uranium materials in the context of illicit trafficking of nuclear materials. Alfred F. Morgenstern, Christos Apostolidis, Klaus Mayer, and Maria Wallenius, Nuclear Chemistry, European Commission, Joint Research Centre, Institute for Transuranium Elements, P.O. Box 2340, Karlsruhe 76125, Germany, Fax: +49(0)7247-95199618, morgenstern@itu.fzk.de

Over the last decade a total of about 500 cases of illicit trafficking involving nuclear material, radioactive sources and radioactively contaminated material have been confirmed world wide and have raised growing public concern about criminal acts involving nuclear materials. Consequently, research efforts in nuclear forensic science have been intensified in order to develop and improve methods for the identification of nature, origin and intended use of seized materials. Various sample characteristics are of relevance for the identification of nuclear materials, including isotopic composition, the content of chemical impurities, material properties and the date of production. In this work we demonstrate analytical methods for the age determination of uranium samples of different uranium enrichment using the parent/daughter relations U-234/Th-230 and U-235/Pa-231. Due to the relatively long half-lives of U-234 (2.46e5 years) and U-235 (7.04e8 years) only minute amounts of daughter nuclides are growing in during the time periods of interest for such samples (typically 5-50 years), therefore both separation of Th and Pa from uranium must be of high chemical recovery and must afford large decontamination factors. Thorium is separated from bulk uranium using extraction chromatography and subsequently measured by a-spectrometry and thermal ionization mass spectrometry (TIMS). Protactinium is separated by highly selective sorption of protactinium to silica gel followed by a-spectrometric measurement. The direct determination of U-234/Th-230 ratios using inductively coupled mass spectrometry (ICP-MS) without prior separation has also been investigated. The methods were tested and validated using uranium reference materials of known ages. The experimental results obtained with both parent/daughter relations were found to agree with the assumed ages of the reference materials within the combined uncertainty of the measurement calculated according to ISO/BIPM. The analysis exploiting the parent/daughter pair U-235/Pa-231 exhibits a slightly larger combined uncertainty and bias than the thorium method but was found highly valuable in validating the experimental results by means of a second, independent analysis.

9:15 —134. Determination of input parameters for estimation of annual residential radiation doses from routine nuclear reactor releases. Daechul Cho¹, In Hyoung Rhee¹, and Gab-Bock Lee². (1) Environmental Engineering, Soonchunhyang University, 646 Eupnae-Ri, Shinchang-Myon, Asan, South Korea, Fax: 82-41-530-1659, daechul@sch.ac.kr, (2) Radiation safety & Chemistry Group, Korea Electric Power Research Institute

This research is to develop a standard methodology for determining the input parameters that impose a substantial impact on radiation doses of residential individuals in the vicinity of four nuclear power plants in Korea. We have selected critical nuclei, pathways and organs related to the human exposure via simulated estimation with K-DOSE 60 based on the updated ICRP-60 and sensitivity analyses. From the results, we found that 1) the critical nuclides were found to be H-3, Xe-133, Co-60 for Kori plants and C-14, Ar-41 for Wolsung plants. The most critical pathway was 'vegetable intake' for adults and 'milk intake' for infants. However, there was no preference in the effective organs, and 2) sensitivity analyses showed that the chemical composition in a nuclide much more influenced upon the radiation dose than any other input parameters such as food intake, radiation discharge, and transfer/concentration coefficients by more than 10² factor. The effect of transfer/concentration coefficients on the radiation dose was negligible.

Pyrochemical operations at Los Alamos Plutonium Facility use high temperature melts of calcium chloride for the reduction of plutonium oxide to plutonium metal and high temperature melts of sodium chloride and potassium chloride mixtures for the electrorefining purification of plutonium metal. The remaining plutonium and americium are recovered from these salts by dissolution in hydrochloric acid followed by either solvent extraction or ion exchange recovery. Aqueous chloride operations generate copious quantities of secondary wastes and are difficult to manage from a materials corrosion perspective. To minimize these problems, a precipitation of the dissolved salts followed by removal of the interstitial chloride would allow for subsequent nitrate-based processing. This would overcome many of the difficulties of the current aqueous chloride operations. Results of the precipitation and chloride washing tests will be discussed from the perspective of plutonium recovery, chloride removal efficiency and filterability.

9:55 — Intermission.

10:15 — 136. Development of radioactive dendrimer nanocomposites to treat tumor microvasculature. Lajos Balogh¹, Shraddha S. Nigavekar², Andrew T. Cook³, Leah Minc³, and Mohamed K. Khan⁴. (1) Center for Biologic Nanotechnology, University of Michigan, Ann Arbor, 200 Zina Pitcher Pl, 4010 Kresge-II, Ann Arbor, MI 48109-0533, Fax: (734) 615-0621, baloghli@umich.edu, (2) Department of Radiation Oncology, University of Michigan, (3) Michigan Memorial Phoenix Project, University of Michigan

We report on nanocomposite devices (NCDs), which exploit differences between normal and tumor vasculature and deliver radioisotopes to tumors. These NCDs are synthesized as monodisperse and stable hybrid nanoparticles composed of gold and poly(amidoamine) (PAMAM) dendrimers. The \{Au(0)\} nanodevices are spherical with a well-defined in a range of 5-75 nm, with a specific surface to permit both size- and/or surface recognition targeting and may also be used to map certain cell compartments making both imaging and therapy possible. Biodistribution of \{Au(0)\} nanocomposites was determined in a mouse tumor model system using isotope activation analysis. \{Au(0)\} was activated into \{Au-198\} by irradiation of the NCDs in a neutron beam. This technique enables us to first synthesize the nanomaterials, then fabricate and characterize the complex device, and finally activate the NCDs. This approach will allow the use of other short half-life radionuclides thereby shortening the development of new active NCD systems.

10:35 — 137. PET imaging of N-(3-[¹⁸F]fluoropropyl)-2b-carbomethoxy-3b-(4-bromophenyl)nortropane ([¹⁸F]FPCBT, BRL-306) in baboon. Ronald M. Baldwin¹, Thomas Chaly², Gilles Tamagnan¹, J.K. Staley¹, M.S. Al-Tikriti¹, Y-K. Hou¹, Xiao-Hui Gu³, R. Zong³, Ross J. Baldessarini⁴, John L. Neumeyer³, and P. Garg¹. (1) Department of Psychiatry, VA Medical Center 116A2, Yale University, 950 Campbell Ave, West Haven, CT 06516, ronald.baldwin@yale.edu, (2) PET Research, North Shore University Hospital, (3) Medicinal Chemistry Laboratory, Alcohol and Drug Abuse Research Center, McLean Hospital, Harvard Medical School, (4) Department of Psychiatry, Harvard Medical School and McLean-Mailman Research Center

The cocaine analog N-(3-fluoropropyl)-2b-carbomethoxy-3b-(4-bromophenyl)nortropane (FPCBT, BRL-306) is a promising candidate for imaging the dopamine transporter (DAT) because of its in vitro potency (Ki 3.3 nM for DAT) and 10:1 selectivity for DAT over serotonin transporters. [¹⁸F]FPCBT was synthesized in two steps by nucleophilic fluorination of propanediol ditosylate followed by reaction with norCBT to give 4-5% overall yield. Radioactivity accumulated rapidly in the brain, with highest uptake in the DAT-rich corpus striatum. In plasma, a metabolite fraction (less lipophilic than parent) reached 50% of the total plasma activity within 30 min. The in vivo properties of [¹⁸F]FPCBT in nonhuman primate demonstrated preferential uptake in DAT-rich areas of the brain as predicted by its in vitro binding properties. (Supported in part by NS40587.)
Production and concentration of high-purity rhenium-188 using a multicolumn selectivity inversion generator technology.

Daniel R. McAlister, Andrew H. Bond, and E. Philip Horwitz, PG Research Foundation, 8205 S. Cass Ave., Ste. 111, Darien, IL 60561, Fax: 630-963-6432, dmcalister@pgrf.com

Rhenium-188 is a useful therapeutic radionuclide for applications including radioimmunotherapy, bone pain palliation and radiation synovectomy. Most current technologies for the production of $^{188}$Re are based on methods developed for $^{99m}$Tc, where macroquantities of isotopically enriched $^{188}$WO$_4^{2-}$ are adsorbed onto alumina and $^{188}$ReO$_4^-$ is eluted with saline solution. Such methods suffer from decreased yields of $^{188}$Re over time and often produce comparatively low specific activity $^{188}$Re per unit volume. Multicolumn selectivity inversion generator (MSIG) technologies employing combinations of ion-exchange, extraction chromatography and related separations methods have been developed to produce a $^{188}$Re generator (or concentration system) that produces high purity $^{188}$Re in 2-5 mL of solution amenable for conjugation. The $^{188}$Re generator and concentrator chemistries have been examined using the Automated Radionuclide Separator, which is a low pressure chromatography system that decreases separation time and provides a consistently pure product.

THURSDAY AFTERNOON

Section A
Convention Center -- Room 390

Radiochemistry at RIA
Cosponsored with Division of Analytical Chemistry

M. Kreisler, Presiding
M. A. Stoyer and P. F. Mantica, Organizers


Neutron reaction cross-sections of radionuclides off-stability are needed for the interpretation of both archival data from nuclear weapons testing and isotope abundances from s-process nucleosynthesis. The neutron induced reactions of interest are (n, gamma), (n,xn), (n,fission), and (n,p). The Device for Advanced Neutron Capture Experiments, DANCE, has been constructed at the Lujan Center of the Los Alamos Neutron Science Center, LANSCE, for the measurement of neutron capture. This spectrometer is capable of measuring neutron capture cross sections using targets of less than one milligram at neutron energies ranging from thermal to 100 keV. A lead slowing down spectrometer is being planned for installation at LANSCE. This spectrometer will allow the measurement of fission cross sections using targets less than 10 nanograms. RIA will provide new opportunities for measurements that could be of benefit to the Stockpile Stewardship Program and the study of nucleosynthesis. Recent results from DANCE will be presented along with future plans including possibilities at RIA.

1:50 —140. Techniques for obtaining neutron-induced reaction cross sections at RIA. L. A. Bernstein, and Larry E. Ahle, Physics and Advanced Technologies, Lawrence Livermore National Laboratory, 7000 East Avenue, L-414, Livermore, CA 94551, Fax: (925)-422-0883, labernstein@llnl.gov

Neutron-induced reaction cross sections on unstable nuclei are of great interest to both the Stockpile Stewardship and the Nuclear Astrophysics communities. Direct and indirect measurements of
these cross sections at RIA will fall into two general categories: prompt and decay counting. Decay counting experiments place the most stringent requirements on the isotopic purity of the targets harvested (see the previous talk by L. Ahle). In contrast, prompt experiments require only that the target contain a majority of only one particular isotope. However, the background from radioactive decay of the beam places special considerations on the spectrometers used for these measurements. This talk will cover several prompt measurement techniques and describe how they might be performed at RIA using a tunable mono-energetic neutron source. It will also discuss an indirect method for obtaining "surrogate" neutron-induced reaction cross sections using rare isotope beams on charged particle targets.

This work was performed under the auspices of the U.S. Department of Energy by the University of California, Lawrence Livermore National Laboratory under contract No. W-7405-Eng-48.

2:10 —141. Collecting mass- and recoil-separated reaction products at RIA. David J. Vieira, Roland A. Bibeau, Marc Hausmann, and Wayne A. Taylor, Isotope and Nuclear Chemistry, Los Alamos National Laboratory, Mail Stop J514, Los Alamos, NM 87545, Fax: 505-665-4955, vieira@lanl.gov

The Rare Isotope Accelerator (RIA) will produce copious amounts of radioactivity. In particular, for long-lived ($t_{1/2} > 2$ days), mass- or recoil-separated beams of $\sim 10^{11}/s$ in intensity, one will accumulate $\sim 10^{16}$ atoms/day or $\sim 3$ microgm/day at A=200. Consequently there is high interest in collecting such separated samples for the preparation of radioactive targets and/or the production of high-specific activities for biomedical or other applications (see Ref. 1). In many cases it will be more feasible to do a stable beam + radioactive target experiment as opposed to the other way around and this should not overlook as an option. In this talk, I will explore the parasitic collection of long-lived activities at RIA. In particular, we have investigated the collection of off-mass species after the first stage of mass separation at RIA (i.e., before the isobar separator) or at the first intermediate focus of the fragment separator where magnetic rigidity and range selection can be used to effect an A/Q separation. After a suitable collection period, the samples will be dissolved and radiochemically processed to prepare pure samples - radioactive targets. Facilities needed to do this work will be outlined. To further illustrate these points, I will also highlight a new mass separator at Los Alamos, called the radioactive species isotope separator (RSIS), that we are developing for the production of mass-separated radioactive targets to be used in neutron-capture experiments with the DANCE detector array at LANSCE. Work performed under the auspices of the U.S. Department of Energy under contract W-7405-ENG-36.


2:30 —142. A measurement of $^{89}$Zr(n,2n)$^{88}$Zr and $^{89}$Zr(n,np)$^{88}$Y cross sections. M.A. McMahan¹, Larry E. Ahle², J. A. Becker², L. A. Bernstein², Joseph Cerny¹, D. M. Moltz³, K. J. Moody⁴, Edward Morse⁵, James P. O'Neil⁶, J. Powell¹, Kevin Roberts², and Mark A. Stoyer⁴. (1) Nuclear Science Division, Lawrence Berkeley National Laboratory, MS 88, 1 Cyclotron Rd, Berkeley, CA 94720, Fax: 510-486-7983, p_mcmahan@lbl.gov, (2) Physics and Advanced Technologies, Lawrence Livermore National Laboratory, (3) Department of Chemistry, University of California at Berkeley, (4) Nuclear and Analytical Chemistry Division, Lawrence Livermore National Laboratory, (5) Department of Nuclear Engineering, University of California @ Berkeley, (6) Biomedical Isotope Facility, Center for Functional Imaging, Lawrence Berkeley National Laboratory.

A collaboration between LBNL (Nuclear Science and Life Science Divisions), U.C. Berkeley (Departments of Chemistry and Nuclear Engineering) and LLNL (Physics and Nuclear Chemistry Divisions) are tackling the challenging problem of measuring the (n,2n) and (n,np) cross sections on $^{89}$Zr. The experiment is a multi-step process: 1) The $^{89}$Zr with a half-life of three days will be made at the Biomedical Isotope Facility at LBNL using the reaction $^{89}$Y(p,n)$^{89}$Zr at a proton energy of 10 MeV. 2) The Zr will be chemically separated from the Y target to a high degree of purity. 3) The material will be transported to
campus and irradiated with 14.8 MeV neutrons at the Rotating Target Neutron Source. 4) 75 days after bombardment, gamma rays from $^{89}$Zr will be counted and the half-life measured. 4) 100 days after bombardment, gamma rays from $^{88}$Zr and $^{88}$Y will be counted.

* This work supported in part by the U.S. DOE Stewardship Science Academic Alliance program.

2:50 — Intermission.

3:10 — 143. Radioactive targets for nuclear astrophysics experiments. Jeff C. Blackmon, Physics Division, Oak Ridge National Laboratory, P.O. Box 2008, MS 6354, Bldg. 6010, Oak Ridge, TN 37831-6354, Fax: 865-576-8746, blackmon@ornl.gov

Reactions involving radioactive nuclei play an important role in many astrophysical environments. Neutron captures on neutron-rich isotopes are important for the synthesis of heavy elements in AGB stars by the s process, and reactions on neutron-deficient nuclei are important for production of the p nuclides, whose astrophysical origins are still unclear. The opportunities at an advanced radioactive ion beam facility for producing long-lived isotopes of interest for nuclear astrophysics are discussed. Production yields are compared to capabilities at current stable beam facilities and high-flux reactors. It is shown that reactor-based production is favorable for long-lived, neutron-rich isotopes, but a few exceptions are described. Neutron-deficient nuclei for p-isotope synthesis studies are produced quite competitively at radioactive ion beam facilities. Parasitic collection of isotopes is desired, but online production with a radioactive beam is preferable for isotopes with shorter lifetimes.

3:30 — 144. Nuclear astrophysics at the ISAC Radioactive Beams Facility: Prelude for RIA. John D’Auria, Department of Chemistry, Simon Fraser University, Burnaby, BC V5A 1S6, Canada, Fax: 604 291 3765, dauria@sfu.ca

RIA is planned to be the next generation, very high intensity RB facility and will eventually be useful for nuclear astrophysics. It is important to gain experience at operating facilities in order to plan properly for RIA. At present, ISAC is the new, high intensity radioactive beams facility, operational in Vancouver. One of its primary functions is for studying nuclear astrophysics especially related to explosive scenario. Studies of radiative proton and alpha capture reactions involving radioactive reactants are now being performed using the DRAGON recoil mass separator facility. In this presentation a review of the studies presently completed, in progress or planned at ISAC and ISAC II in nuclear astrophysics will be presented along a review of some of the other experimental facilities operational or planned.

3:50 — 145. Nuclear structure of transactinide nuclei with RIA. Irshad Ahmad, Physics Division, Argonne National Laboratory, 9700 S. Cass Ave., Argonne, IL 60439, Fax: 630-252-6210, ahmad@phy.anl.gov

Single-particle spectra are needed both to predict the limit of the periodic table and also to test theoretical models for a system with the very large Coulomb energy. Nuclear structures of nuclei up to Z=100 are well studied because these nuclides are available in sufficient quantities. Thus the locations of single-particle states occupied by the 101st proton and 162nd neutron are experimentally known. In the last decade, sensitive techniques like recoil mass separators, segmented focal plane Si detectors for alpha particles and electron detectors have been developed which allow spectroscopic measurements of few-atom sources. The advantage of RIA will be in the production of neutron-rich isotopes which is not possible with stable beams. Thus using neutron-rich beams, it would be possible to produce and study Rf and Sg isotopes with neutron numbers beyond the N=162 gap. Possible experiments will be discussed. This work was supported by the U.S. Department of Energy, Division of Nuclear Physics under contract No. W-31-109-ENG-38.

4:10 — Panel Discussion.

5:00 — Concluding Remarks.
Section B
Convention Center -- Room 391

General Topics in Nuclear Chemistry
S. C. Srivastava, Organizer, Presiding

1:30 — Introductory Remarks.

1:35 — 146. Development of a U-235m generator for measurement of neutron cross sections. Dawn A. Shaughnessy, Yves X. M. Dardenne, Kenton J. Moody, Mark A. Stoyer, and John F. Wild, Analytical and Nuclear Chemistry Division, Lawrence Livermore National Laboratory, P.O. Box 808, L-231, Livermore, CA 94551, Fax: 925-422-5565, shaughnessy2@llnl.gov

There is interest by the defense and nuclear technology field in measuring cross sections from 235mU(n,f) reactions. We are currently developing an online 235mU generator in order to produce quantities sufficient for targets in neutron bombardments. The alpha decay of 239Pu populates the 235mU isomer and members of the 235mU-based rotational band almost exclusively (approximately 99.9%). By using an online production method, a continuous supply of 235mU can be generated with a favorable ratio of 235mU isomer to 235U ground state. The generator consists of a thin layer of 239Pu electroplated on the inside of a tube. The 239Pu layer is thin enough such that 235mU recoil is efficiently emitted into the center of the tube. The recoils are collected in a stream of gas passed through the tube and deposited where needed. Initial tests and optimization of the generator parameters will be discussed.

This work was performed under the auspices of the U.S. Department of Energy by the University of California, Lawrence Livermore National Laboratory under Contract No. W-7405-Eng-48.

1:55 — 147. Use of actinide oxides for the partial oxidation of methane. Jonathan W. Plaue, and Kenneth Czerwinski, Department of Nuclear Engineering, Massachusetts Institute of Technology, 138 Albany St., NW13-220, Cambridge, MA 02139, Fax: 617-253-7300, jplau@mit.edu

The heterogeneous reaction of methane gas and solid actinide oxides will be explored. In the first step of the proposed cycle, the actinide compound is reduced, thereby oxidizing methane to synthesis gas, a mixture of carbon monoxide and hydrogen. Synthesis gas has a variety of uses as precursor material for synthesis in the chemical industry. The second step involves the re-oxidation of the actinide materials using water vapor, which is reduced to form hydrogen gas. Previous work by others has demonstrated this cycle using cerium oxide compounds. Beneficial similarities in the redox properties and hydrolysis constants for the actinide elements suggest them as better reactant materials. Actinide oxide materials will be synthesized, characterized, and evaluated for reactivity with methane. Reactivity with methane will be evaluated using a heated tube reactor coupled with an inline gas chromatograph.

2:15 — 148. Determining the exact mass of neutrinos through logical mass analysis. Andrew C. Angus, Absolute Foundation, 1435 Olympic Drive, Milpitas, CA 95035, andrewangus@yahoo.com

The primary purpose of this paper is to demonstrate that a neutrino has a definite mass. Secondly, the purpose of this paper is to demonstrate through reaction equation the genesis of neutrino from nuclear reactions induced by electricity.

The author used a method called logical mass analysis to determine theoretically the exact mass of a neutrino. The author determined the exact mass of three types of neutrinos: a. neutrino-p has a rest mass almost 1/2 of an electron mass or 0.2395 MeV, b. neutrino-d has a rest mass about 1/10 of an electron mass or 0.059 MeV, c. neutrino-t has a rest mass about 9/10 of an electron mass or 0.444 MeV. The author formally defines neutrino as a leptonic particle having a mass ranging from 1/10 of an electron mass to 9/10 of an electron mass.

The importance of this paper lies in the fact that the exact mass of neutrino has eluded scientists for 60 years since 1930 when the neutrino was postulated as a massless particle. Secondly, this paper is important because this paper serves as a basis for experimental scientists to identify different types of neutrinos and their respective definite masses during experimentations.
2:35 — Intermission.

2:55 — 149. Investigation of single-bubble sonoluminescence produced by acoustic cavitation of D$_2$O. Kenneth C McGill Sr.¹, Ralph H. France III¹, Curtis Harris¹, James Black Jr.¹, Suzanna Williams¹, and Caroline A. Loglisci². (1) Chemistry & Physics, Georgia College & State University, CBX 82, GC&SU, Milledgeville, GA 31061, Fax: 478-445-1092, kmcgill@gcsu.edu, (2) Physics, University of Connecticut

The acoustic cavitation of D$_2$O was measured using an ocean optics ultra violet spectrometer. Walls of a container were constructed for the D$_2$O using 2mm thick by 6cm long quartz cylindrical cavity. The upper and lower transducers were gold plated piezoelectric quarts crystals. Compressing a Teflon seal between the piezoelectric crystal and quartz tube created a watertight seal. Argon was bubbled through a solution of D$_2$O to replace any other existing dissolved gasses, as single bubble sonoluminescence is known to work best with dissolved noble gasses. The container was immersed in the D$_2$O and sealed using a clamp. A standing wave in the cavity causes cavitations of the fluid to create bubbles. Once the bubble collapses the emitted light spectrum was measured using Ocean Optic ultraviolet spectrometer. By measuring the emitted light spectrum a close approximation of the actual temperature can be obtained, however, as water is opaque to ultraviolet light, this may provide only a lower limit. If the temperature is high enough (~ few MK) fusion neutrons may be emitted, which will be measured at a future experiment panned to take place in collaboration with Yale University.

3:15 — 150. A new physical constant and its applications to nuclear energy production. Andrew C. Angus, Absolute Foundation, 1435 Olympic Drive, Milpitas, CA 95035, andrewangus@yahoo.com

The purpose of this paper is to introduce a new physical constant called the Angus' constant which is represented by the Greek letter lambda in upper case ($\Lambda$). The Angus' constant or $\Lambda$ is equal to 1.98 x 10^-25 J-m. The author explains how he derived the new physical constant. Then, the author explains the importance of this new physical constant to nuclear science. The new physical constant helps to affirm the dual nature of the photon. Furthermore, this paper shows that there are two ways to represent mathematically the photon, namely in the form of the "Einstein's photon equation" and the "Angus' photon equation". This paper reveals that the new physical constant can be used to give an alternative explanation to Einstein's Law of photoelectric effect. Finally, the author reveals the applications of this new physical constant to nuclear energy production.


Mathematical development of Lin's theory of flux is presented. Based on the theory, when a chemical reaction system is subjected to a high time rate of temperature change, it changes from equilibrium to non-equilibrium conditions. It is proved mathematically that, when a gas system is subjected to a high time rate of temperature increase, the activities of particles (molecules, atoms or nuclei, and electrons) are increased. The acceleration of particles in the dynamic condition can lead to nuclear reactions. In the pilot plant studies conducted at Research Triangle, NC, USA, for SO$_2$ conversion to SO$_3$ by rapid heating, a 10-ft high vertically fired combustor (VFC) was used. It is observed that, as the air passing through the VFC during the idle period of sixteen hours (no external heat is added to the system), the temperature of the flowing air consistently rises up rapidly from ambient temperature (90 °F) at inlet of the VFC to an average temperature as high as 582 °F (in the range of 840 °F to 455 °F) at one section of the VFC, an increase of about 500 °F. The airflow temperature increase of such large magnitude and long duration indicates that nuclear reactions are present in VFC. It is also found that the water vapor in the air stream has completely disappeared in the VFC, for no sulfuric acid formation resulting from the reaction of water and SO$_3$ is detected there. Evidently, the water vapor in the air is converted to hydrogen and oxygen by rapid heating. It is proposed that in the dynamic condition, electrons are driven off from their orbits, and protons are produced from the hydrogen ions. The mutual bombardments and
direct impacts between the elements in the plasma fluid produce various nuclear reactions including nuclear fusion. The possible nuclear reactions are shown in the body of the paper. The large amount of heat released from nuclear reactions in the air offers an inexhaustible source of energy supply. Unlike conventional fuels, it does not contribute air, water and solids pollutions.