Task 30

Combined Radiation Detection Methods for Assay of Higher Actinides in Separation Processes

D. Beller and W. Kernan

BACKGROUND

Monitoring of transuranic actinides (TRU–includes neptunium, plutonium, americium, and curium) during the separation of used nuclear fuel has been identified as a critical research area in the U.S. Advanced Fuel Cycle Research and Development program (AFC R&D). Recycling of used fuel by chemically separating it into uranium, fission products, and TRU would be the first step in this new fuel cycle. Material Protection, Accounting, and Control (MPAC) is necessary for materials accounting, criticality monitoring, and assurance of proliferation resistance.

In the MPAC project, faculty and students are investigating the potential to use combined neutron and gamma-ray detector systems to measure quantities and isotopic constituents contained during separations and intermediate storage. This will require knowledge of the nuclear and decay characteristics of materials during processing, the development of conceptual designs of monitoring systems, radiation transport studies to develop an understanding of operational regimes, and experiments to confirm performance. In addition, both passive and active concepts will be investigated, including collaborations with the Idaho Accelerator Center at Idaho State University (ISU) to use electron linear accelerators for producing photoneutrons in situ, for photon activation of TRU, or for stimulating emissions processes (e.g. x-ray fluorescence).

RESEARCH OBJECTIVES AND METHODS

The ultimate objective of this project is to develop technology to detect and accurately measure quantities of higher actinides in processing systems without taking frequent samples. These systems include used fuel receipt, separations batches, and pipelines. A variety of measurements may be combined to calculate flow rates of actinide elements with a to-be-determined precision. Nuclear and decay characteristics of materials during processing will be acquired, conceptual designs of monitoring systems will be developed, radiation transport studies will be conducted to develop an understanding of operational regimes, and experiments will be performed to confirm performance. Radiation transport and scoping studies will be conducted to investigate combined gamma-ray, neutron, and active and passive detection techniques to measure quantities and isotopic constituents contained during separations and intermediate storage. Scoping and design studies will first be performed using validated data sets (decay properties and reaction cross sections) and the radiation transport code MCNPX. Basic measurements will then be performed and compared to predictions. Experiments to be conducted in subsequent work are to be determined, but may include small quantities of radioactive actinides at UNLV in addition to accelerator-coupled experiments at ISU.

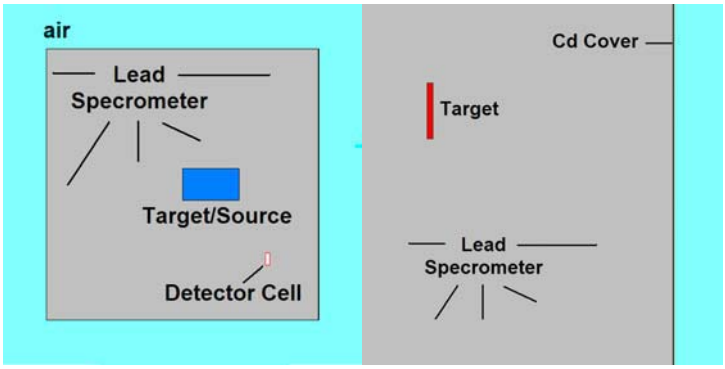
RESEARCH ACCOMPLISHMENTS

Students continued to develop concepts and models for use of the ^{3}He Neutron Multiplicity Detector System (NMDS, which was developed as part of Task 6) for MPAC. Technology for assaying fuel rods and/or complete assemblies, and the use of the NMDS and other techniques to assay dilute quantities of TRU in waste and other process streams were investigated.

Lead and Carbon-based Slowing Down Spectrometers

Neutron slowing down spectrometers (SDSs) were modeled and plans were developed to conduct experiments at ISU with their carbon-based SDS. This work will be used to investigate technology for assaying fuel rods and/or complete assemblies. In preparation for these experiments, MCNPX was used to model neutron transport characteristics in lead and carbon-based SDSs. The energy of neutrons measured in a neutron detector with an SDS can be characterized by the equation $E = K/(t+t_0)^2$, where E is the energy of the neutron when it was created at the source (not its energy when detected), t is the time of the radiation detection (count) after a source event, and K and t_0 are characteristics of the particular SDS. Thus, if an accelerator pulse is used to generate neutrons which then initiate fission, the time history of the spectrum can be de-convolved to yield the energy spectrum. In addition, the neutron multiplicity can simultaneously be measured by the NMDS.

However, the detector system must be able to resolve the time dependence of the neutron signal. Because ^{3}He detectors, such as those contained in the NMDS, have a slow response, the SDS configurations must be studied before conducting experiments. Those studies are currently ongoing, beginning with benchmarking computational methods. A Los Alamos lead SDS was mod-



Cross sections of the UNLV benchmark of the LANL lead SDS taken from MXNPX models. (Left) The overall geometry, (Right) A close-up of a cross section of the thin target. The response to neutrons of various energies (time constants) is influenced by the position and isotopic composition of the detector.

eled to benchmark computational methods for determining energy-time correlation constants, and energy-time correlation constants are now close to those obtained in LANL experiments. The next step will be to use the MCNPX code to design an experiment using the ISU carbon-based SDS and electron linac, followed by experiments conducted at ISU in the next year. During this period, plans were developed to conduct experiments at ISU with the carbon-based SDS to develop technology for assaying fuel rods and/or assemblies.

MPAC for UREX+ Processes

Another application of NMDS to MPAC might utilize the measurement of both passive and active neutron multiplicity in very dilute concentrations of higher actinides, such as in a non-actinide waste stream. This is being investigated since it may be more feasible to monitor where actinides are not supposed to be than where they are. The NMDS will be used to assay actinides in pipes, tanks, etc. in a UREX+ fuel separations plant to measure quantities and isotopic constituents in such a stream. Sources and configurations were investigated to simulate a waste pipe that contains residual higher actinides from UREX-plus for use in upcoming experiments.

Neutron Multiplicity Detector System

Several programming and maintenance issues with the NMDS that were revealed during testing in December were repaired or resolved. New detector wiring that was installed on one detector group of NMDS significantly reduced background count rates (noise). As a result, all of the of standard cables were replaced with coaxial cables. In addition, as a result of differences in detector response between the original Russian data acquisition system and the new LabView-based system, detector response is being re-mapped with the data collection and analysis software. These improvements are expected to significantly improve the performance of the NMDS.

Isotope	Resonance energy (eV)	K (keV*μsec²) LANL	K (keV*μsec²) UNLV
In-115	1.5	164	
Ta-181	4.3	161	148
Au-197	4.9	158	194
Ag-109	5.2	161.5	191
Ta-181	10.4	162	153
Ag-107	16.3	162.5	166
Cd-111	27.5	163	167
Au-197	59	159	174

Comparison of UNLV MCNPX computed time constants versus LANL's values for the LANL LSDS benchmark.

ACADEMIC YEAR HIGHLIGHTS

- D. Beller, W. Kernan, M. Schanfein, T. Ward, A. Rimsky-Korsakov, F. Harmon, Q. Newell, L. Lakeotes, P. Attur, T. Beller, and R. LeCounte, "Combined Radiation Detection Methods for AFCI MPAC Project," *Proceedings*, 47th Annual Meeting of the Institute of Nuclear Materials Management, Nashville, TN, July 16-20, 2006.
- A collaboration with the V. G. Khlopin Radium Institute was developed for an upgrade to and maintenance on the Neutron Multiplicity Detector System.

Neutron Multiplicity Detector System configured in a cubic geometry for background and source counting.

FUTURE WORK

In the final year of this project modeling will be completed, the NMDS will be tested further, and it will be employed in two separate experiments. One is with a carbon-based neutron slowing down spectrometer to develop lead-based SDS technology to assay fuel assemblies upon receipt for separations, and the other is to assay a process pipe containing dilute concentrations of separated higher actinides. The carbon-based SDS has been constructed at the Idaho Accelerator Center. Actinides for the pipe assay experiments are available at UNLV, how they will be used in these studies is yet to be determined.

Research Staff

Denis Beller, Principal Investigator, Research Professor, Mechanical Engineering Department
Warnick Kernan, UNLV Instructor, Physics Department; Principal Scientist, Remote Sensing Laboratory

Students

Quinten Newell, Lawrence Lakeotes, and Timothy Beller, Graduate Students, Mechanical Engineering Department

Collaborators

Mark Schanfein, Safeguards and Security Group, Los Alamos National Laboratory
James Laidler, National Technical Director for Separations and Waste, Argonne National Laboratory
Thomas Ward, UNLV Science Adviser for Russian Collaborations, TechSource, Inc.