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

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Task 30

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BACKGROUND

Monitoring of higher actinides (HA; 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 Initiative. Recycling of used fuel by chemically separating it into uranium, fission products, and HA 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. The objective of this MPAC project is to develop technology to detect and accurately measure quantities of higher actinides in used fuel assemblies and processing systems without taking frequent samples. Process systems may include separations batches, pipelines, storage tanks, and fuel fabrication equipment. A variety of measurements may be combined to calculate flow rates of actinide elements with a to-be-determined precision.

RESEARCH OBJECTIVES AND METHODS

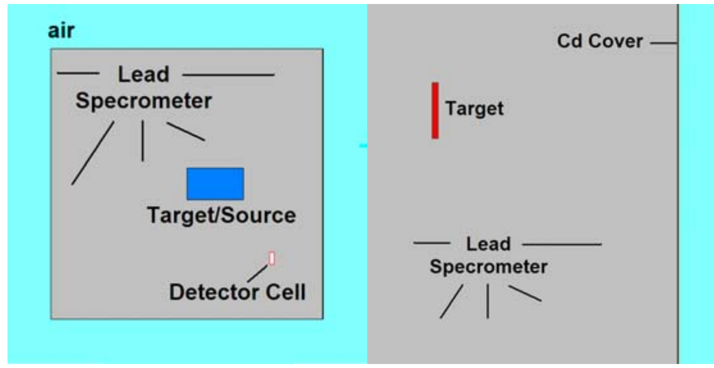
In the MPAC project, faculty and students are investigating the potential to use combined neutron and gamma-ray detector systems to measure quantities and isotopic constituents contained during separations and intermediate storage. This will require knowledge of the nuclear and decay characteristics 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 HA, or for stimulating emissions processes (e.g., X-ray fluorescence).

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. Planned experiments have not been conducted.

RESEARCH ACCOMPLISHMENTS

Lead and Carbon Slowing Down Spectrometers

Lead Slowing Down Spectrometer (SDS) and Carbon SDS (CSDS) were modeled. This modeling was expanded to include the UNLV ^{3}He Neutron Multiplicity Detector System (NMDS, which was developed as part of Task 6) with multiple neutron detectors in up to four layers surrounding used nuclear fuel rods or assemblies. Modeling also included simulation of decay gamma rays for coincidence counting investigations.



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.

In preparation for future experiments, MCNPX was used to model neutron transport characteristics in LSDS and CSDS. 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, SDS configurations must be studied before conducting experiments. A Los Alamos National Laboratory (LANL) LSDS was modeled to benchmark computational methods for determining energy-time correlation constants. The benchmark exercise was completed and published. Multiplicity measurements for spontaneous fission and spontaneous-fission induced fission for isotopes of U, Pu, and Cf were verified within Monte Carlo statistical uncertainty limits. Simulations were conducted for other isotopes (Am, Np, and others) for multiplicity measurement experiments using a subset of the NMDS detectors.

In future work, the MCNPX code will be used to design an experiment using the ISU CSDS and electron linac, followed by experiments conducted at ISU. During this period, plans will be developed to conduct experiments at ISU with the CSDS to develop technology for assaying fuel rods and/or assemblies.

Neutron Multiplicity Detector System

Collaboration with the V.G. Khlopin Radium Institute (KRI) was developed for an upgrade to and maintenance on the Neutron Multiplicity Detector System. KRI completed design studies for a coincidence/anti-coincidence using a multi-plate plastic scintillator system to reduce background and provide greater neutron-muon discrimination. KRI procured components for the upgrade, installed them and coupled outputs to their NMDS channels, and began measurements in St. Petersburg, Russia.

In preparation for the KRI visit, the NMDS was reconnected to the original Russian data acquisition system and several neutron multiplicity counts were conducted to confirm its performance. The data files were transmitted to KRI where they were evaluated. All detectors appear to be operating as they were when the system was initially delivered and set up at UNLV several years ago. Two specialists from KRI visited UNLV in November to service the NMDS; they cleaned and secured connections, replaced some parts, and adjusted bias voltages on some detectors.

A commercial control program was installed to automate the collection and storage of data on the 60-channel NMDS. It will automatically take data and has operated reliably since this upgrade.

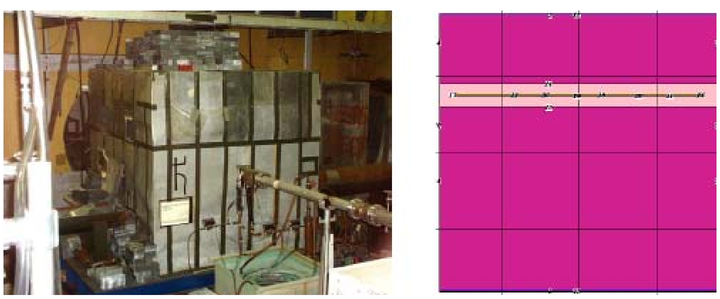
<u>Rensselaer Polytechnic Institute collaboration</u>

Rensselaer Polytechnic Institute (RPI) collaborators investigated neutron slowing down spectroscopy for quantitative analysis of ^{239}Pu, ^{235}U and possibly ^{241}Pu in used fuel assemblies or rods. Several fuels, configurations, and parametric studies were modeled.

RPI completed various parametric studies to compare computed response to measurements in the LSDS. First order calculations were conducted with one fuel pin based on data from an AP1000 reactor. A detector was placed in the assay channel of the lead and the tally was convoluted with the ^{235}U fission cross section. This data was compared to experimental fission data for ^{235}U collected in the LSDS in 2007. Differences between calculated and experimental data are due to the broadening of the neutron energy resolution at lower neutron energies. Also, the reaction rate decreases as the slowing-down time increases, so that background becomes a larger factor as the neutron energy decreases.

Other RPI results:

- Simulation of ^{235}U fission chamber response was compared to measured data indicating that hydrogen content and impurities in the lead must be included to accurately simulate the measured response.
- Simulation of a single SPERT fuel pin and a ^{238}U assay detector were performed.
- Analysis of optimization and biasing methods for LSDS calculations demonstrated that, due to the nature of the LSDS, all areas of the lead as well as all neutron energies are critical to the problem and that even biasing the source will affect the resulting spectrum in the ^{238}U detectors.
- A model of an AP1000 fuel assembly was completed to study effects of self shielding within the assembly. These studies demonstrated that MCNP can produce results that agree with measurements of time-dependent fission rates in the LSDS; however, great effort must continue to be made to optimize the calculations. Particular effort must be made to increase efficiency to allow for smaller statistical error with a minimum of calculation time.



RPI's LSDS (left), and a cross section at x = -37.5cm showing the assay channel and fuel pin (the x axis is positive along the beam line).

ACADEMIC YEAR HIGHLIGHTS

- A.A. Rimsky-Korsakov and N.A. Kudryashev, "General Upgrade Design, Instrumentation Upgrade Of Neutron Multiplicity Detector System," Progress Report 1, V.G. Khlopin Radium Institute, St. Petersburg, Russia, September 2007.
- A.A. Rimsky-Korsakov and N.A. Kudryashev, "Detailed Equipment Specifications and List Of Materials and Components to be Purchased, Instrumentation Upgrade Of Neutron Multiplicity Detector System," Progress Report 2, V.G. Khlopin Radium Institute, St. Petersburg, Russia, September 2007.
- Timothy Beller graduated with an M.S. in Materials and Nuclear Engineering, December 2007.
- Ryan LeCounte graduated with an M.S. in Materials and Nuclear Engineering, December 2007.
- Lawrence Lakeotes graduated with an M.S in Materials and Nuclear Engineering, May 2008.
- Sandra De La Cruz graduated with a B.S.M.E. (nuclear option), May 2008.
- Luis Durani graduated with a B.S.M.E. (nuclear option), May 2008.

FUTURE WORK

UNLV is discussing follow-on research with Pacific Northwest National Laboratory, Idaho National Laboratory, LANL, ISU, and RPI. Current work includes continuation of studies at RPI as well as an undergraduate research internship at LANL during the summer 2008 to model fuel assemblies.

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