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

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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 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.

In this MPAC project, faculty and students will investigate 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 (IAC) at Idaho State University (ISU) to use electron linear accelerators for producing photon neutrons in situ, for photon activation of HA, 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 years 2 and 3 are to be determined, but may include small quantities of radioactive actinides at UNLV in addition to accelerator-coupled experiments at ISU.

RESEARCH ACCOMPLISHMENTS

To initiate the AFCI MPAC Project, a kick-off meeting was held that included representatives and technical staff from the DOE, LANL, INL, ANL, PNNL, UNLV, ISU, and others. A collaboration with Los Alamos, which supplied safeguards monitoring equipment for the Rokkasho reprocessing plant in Japan, was initiated. UNLV faculty met with group leaders and technical staff at LANL in conjunction with the MPAC PI and others from Idaho State University to develop collaborations for monitoring systems development. They also met with N-1 Safeguards Science & Technology Group and N-2 Advanced Nuclear Technology Group. In addition, technical staff from N-1, N-2, and N-4 Safeguards Systems Group visited UNLV to discuss ongoing and potential MPAC projects, to tour labs, and to meet with students.

One technique that has been used in a variety of MPAC applications is neutron multiplicity measurement. This is the coincidence measurement of multiple neutrons that are emitted in individual fission events. A Neutron Multiplicity Detector System (NMDS) was previously built in collaboration with the V. G. Khlopin Radium Institute (KRI) in St. Petersburg, Russia (see Task 6 on...
The system has since been extensively tested at UNLV and ISU. The NMDS is a modular system consisting of 64 \(^3\)He detectors (tubes), electronics, and lead and polyethylene bricks. The modular nature of the system allows it to be used in a variety of configurations and for a variety of purposes. Examples are measurement of neutron multiplicity from high-power, high-energy spallation targets, detection of fissile materials in cargo systems or luggage, and assay of actinides in fuel or storage containers. Count rate and data acquisition capabilities of the NMDS were expanded in TRP Task 6, “Neutron Multiplicity Measurements for the AFCI Program.” As part of Task 30, a new connector board was acquired to interface the remaining 32 detectors to the field programmable gate array (FPGA) board, such that the entire NMDS can now be operated with either the Russian DAQ or the new high-rate UNLV DAQ. Comparison testing is ongoing.

Data required for studying detection concepts includes elemental and isotopic constituents of discharged reactor fuel rods or assemblies, separations processes, storage systems, and fuel and waste-form fabrication systems. The RADDB code system, which is a user-friendly SCALE/ORIGEN-based code, was initially used to generate isotopic fractions for the higher actinides for initial scoping studies, in which the MCNPX radiation transport code system was used with the latest data libraries to model the NMDS. Elemental constituents of process flows and flow rates were later obtained from a detailed UREX+ flow sheet from ANL for use in designing conceptual detector systems. This information was then used in MCNP modeling. In one of these studies, the system was configured to surround a separations processing pipe to measure mixed transuranics. In addition, the effects of isotopic concentration on neutron multiplicity counting was studied. The NMDS model in MCNPX was reconfigured to surround a 2-inch process pipe containing actinides in solution (see the figure on the opposite page. Physical models were also assembled to ensure that there would be sufficient materials (e.g. polyethylene).

In addition to the NMDS, modeling of a germanium detector with MCNPX was initiated based on a report of similar work from Sandia National Laboratories. In addition, concepts for combined neutron-gamma and passive-active interrogation systems using the NMDS were developed.

**FUTURE WORK**

In the following year modeling will be completed, the NMDS will be tested further, and it will be employed in two separate projects. One is to develop a lead neutron slowing down spectrometer (SDS) to assay fuel assemblies upon receipt for separations, and the other is to assay a process pipe containing separated higher actinides. Both projects will involve testing using the NMDS. For fuel assembly assays, the use of a lead SDS (LSDS) will be investigated, and testing will involve a carbon (graphite) SDS (CSDS). The CSDS has been constructed at the Idaho Accelerator Center. Actinides for the pipe assay experiments are available at UNLV.