Non-Destructive Assay of Plutonium and Uranium with the RPI LSDS

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INTRODUCTION

Lead Slowing-Down Spectroscopy (LSDS) is a candidate for non-destructive assay of spent nuclear fuel (SNF) [4]. Research was collaborated via the Material Protection, Accounting, and Control Technology (MPACT) campaign under the Department of Energy. Researchers include: Pacific Northwest National Laboratory (PNNL), Los Alamos National Laboratory (LANL), University of Nevada Las Vegas (UNLV), Rensselaer Polytechnic Institute (RPI), and Idaho State University (ISU).

The development of an LSDS assay system would provide a characterization method for the direct assay of the fissile content of spent nuclear fuel (SNF) to the benefit of nuclear safeguards and nuclear material accountability. Additionally, the LSDS could be used to determine the burn up level for the purpose of reprocessing and recycling SNF. The three main tasks of this project involve development of an algorithm for interpreting fissile masses, development of suitable detectors, and the experimental benchmarking of the system.

THE RPI LSDS

The LSDS assay method requires the use of a pulsed neutron source to populate a lead mass with neutrons. At RPI, the source is driven by the electron linear accelerator (LINAC) tuned to 180 Hz and an electron energy of ~50 MeV. Subsequent (e,γ) and (γ,n) reactions on an air-cooled tantalum target result in neutron spallation with a maximum energy of 0.46 MeV [5]. Once these neutrons are produced in the lead, they scatter on the heavy lead nuclei, losing energy as they pass through the fission resonance region. Because scattering neutrons lose a fraction of their energy during each collision and faster neutrons undergo a faster collision rate, there is a correlation between neutron energy and time after neutron generation. The average neutron energy can be determined by the relation in equation 1:

$$E = \frac{k}{(t_0 + t)^2}. \qquad (1)$$

where $k \approx 165$ keVμs^2 and $t_0 \approx 0.3$ μs for the RPI LSDS. These interrogation neutrons are used to probe uranium and plutonium samples in the LSDS assay channel, resulting in the fission of ^{235}U, ^{239}Pu, and ^{241}Pu. The consequent fission neutrons can be detected and discriminated from the interrogation neutrons using threshold fission chamber consisting of ^{238}U and ^{232}Th. These detectors—along with fast-counting Time of Flight clocks—produce a response with distinguishable resonance structure corresponding to the quantity and isotopic makeup—i.e. burn up—of the fuel sample.

MEASUREMENTS AT RPI

Previously, the Rensselaer Polytechnic Institute (RPI) LSDS was used to assay fresh uranium fuel rods for ^{233}U and ^{235}U [2,3]. However, the presence of plutonium isotopes in SNF adds depth to the challenge of SNF assay.

Experiments were performed at the Gaerttner Laboratory LINAC at RPI in order to investigate the sensitivity of the assay method to incremental fissile material. Additionally, the measurements serve as benchmarks for Monte Carlo simulations used to develop the assay algorithms. The measurements were performed with incremental samples of plutonium and uranium contained in the RPI LSDS channel, illustrated in Figure 1.

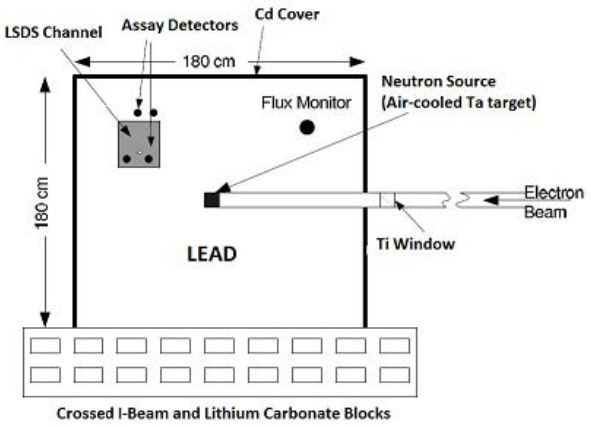

Figure 1: Diagram of RPI LSDS

Due to the current inability to handle SNF at the Gaerttner Laboratory, the uranium samples used included: an enriched uranium fuel pin from the RPI Walthousen Critical Facility and ten highly-enriched uranium (HEU) disks (between 200-300 mg each). The two plutonium samples were plutonium-beryllium (PuBe) sources, containing 96g and 47g of total Pu.

This study involved the interrogation of more than 20 fissile sample configurations and provided detector responses for each 30-minute experiment. Each trial was

conducted with an electron beam at 180 Hz, an electron current of 14-15 μA, and an electron energy of about 50 MeV. This corresponds to a pulse intensity of about 10^{10} n/pulse and 10^{15} source neutrons per trial. The responses of each measurement provided input data for the assay algorithm.

EXPERIMENTAL RESULTS AND ANALYSIS

The data collected by the ^{238}U assay detector were normalized and plotted to show the time-dependent response corresponding to incremental fissile uranium and plutonium sample content. Figure 2 displays the ^{238}U detector response corresponding to assay measurements with the fuel pin and plutonium samples. The lowest response corresponds to the fuel pin and small PuBe source, and it has been previously demonstrated that Monte Carlo simulations are in good agreement with the experimental results [1]. Additionally, the three elevated responses are overlapping and correspond to the fuel pin, both PuBe sources, and 0, 5, and 10 HEU disks, respectively. Figure 2 demonstrates that incrementally adding small HEU disks in the presence of plutonium results in unobservable changes to the assay detector response, indicating the challenge of such an assay.

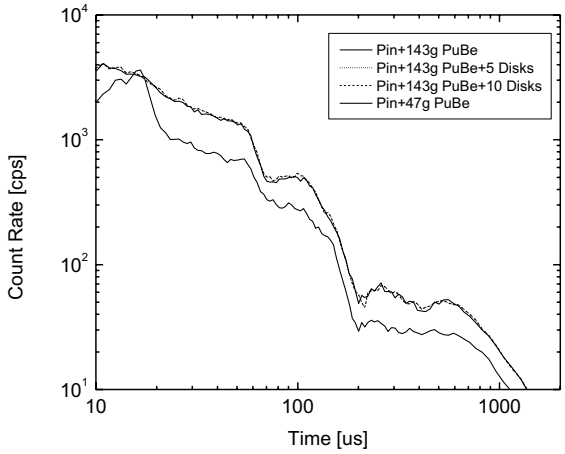

Figure 2: LSDS assay of (1) pin and small PuBe source and (2) pin, both PuBe sources, and 0, 5, and 10 HEU disks

The addition of plutonium into the LSDS channel was used to exhibit its effect on the assay of ^{235}U. Specifically, the plutonium structure is most significant near the thermal fission peak (t>200 μs). The fission resonance structure of ^{235}U can be seen in the LSDS between 20-100 μs, so the detector responses were integrated over this region. Although Figure 2 demonstrates that small changes in uranium are difficult to observe in the detector responses, Figure 3 reveals that the integral count rate increases proportionally to small increments of ^{235}U. Furthermore, this proportionality holds true in the presence of large quantities of plutonium, which is the requirement for measuring burn up of SNF.

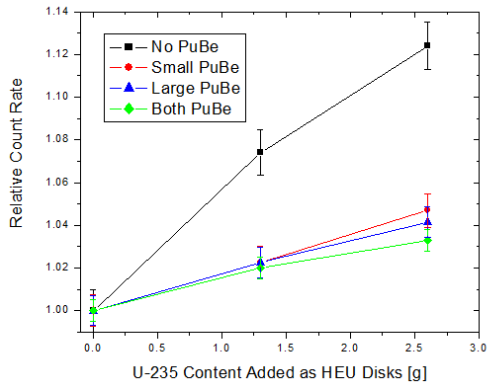

Figure 3: Changes in integral counts when adding HEU.

CONCLUSIONS

Experimental measurements were performed at RPI with various samples—a fresh fuel pin, two PuBe sources, and several HEU disks—to mimic the assay of SNF with specific burn up. By using these samples in various combinations, the precision of uranium and plutonium assay using the LSDS was investigated. It was demonstrated that the RPI LSDS is sensitive to small changes in ^{235}U content even in the presence of large quantities of plutonium.

Future work will involve further analyzing the data with the developed assay algorithm [4]. Additionally, it would be worthwhile to measure multiple fuel pins with variable plutonium content in order to simulate a full fuel assembly.

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