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# Lead Slowing-Down Spectroscopy for Spent Fuel Assay: FY12 Status Report

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October 2012

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# Executive Summary

The Office of Nuclear Energy, Fuel Cycle R&D, Material Protection and Control Technology (MPACT) Campaign is supporting a multi-institutional collaboration to study the feasibility of using Lead Slowing Down Spectroscopy (LSDS) to conduct direct, independent and accurate assay of fissile isotopes in used fuel assemblies. The collaboration consists of Pacific Northwest National Laboratory (PNNL), Los Alamos National Laboratory (LANL), Rensselaer Polytechnic Institute (RPI), and Idaho State University (ISU). There are three main challenges to implementing LSDS to assay used fuel assemblies. These challenges are the development of an algorithm for interpreting the data with an acceptable accuracy for the fissile masses, the development of suitable detectors for the technique, and the experimental benchmarking of the approach. This report is a summary of the progress in these areas made by the collaboration during FY2012.

Significant progress was made on the project in FY2012. Extensive characterization of a "semi-empirical" algorithm was conducted. For example, we studied the impact on the accuracy of this algorithm by the minimization of the calibration set, uncertainties in the calibration masses, and by the choice of time window. Issues such as lead size, number of required neutrons, placement of the neutron source and the impact of cadmium around the detectors were also studied. In addition, new algorithms were developed that do not require the use of plutonium fission chambers. These algorithms were applied to data from measurements taken by RPI and shown to determine the $^{235}$U mass to within 4%. For detectors, a new concept for a fast neutron detector involving $^{4}$He recoil from neutron scattering was investigated. The detector has the potential to provide a couple of orders of magnitude more sensitivity than $^{238}$U fission chambers. Progress was also made on the more conventional approach of using $^{232}$Th fission chambers as fast neutron detectors. For benchmarking measurements, we continue to improve our understanding of the experimental setup by studying issues such as the effect of room return and impurities in the lead. RPI performed a series of experiments with a fresh fuel pin and various $^{235}$U and $^{239}$Pu sources. A comparison between simulations and measurements shows significant deviations after 200 μs for both $^{235}$U and $^{239}$Pu samples, as well as significant deviations at earlier times for the $^{239}$Pu sample.

The FY2013 effort will shift focus to planning for a Technical Readiness Level 5 demonstration. The primary deliverable for the year will be a plan on how to do this demonstration. The plan will include measurement design, sample acquisition, sample handling, cost estimate, schedule and assumptions. Research will continue on the $^{4}$He detector, algorithms development, thorium fission chambers and benchmarking measurements involving sub-assemblies of fresh fuel.

# Symbols, Acronyms and/or Initialisms

| | |
|---|---|
| GWd/MTU | Gigawatt days per metric ton of uranium |
| IAEA | International Atomic Energy Agency |
| ISU | Idaho State University |
| LANL | Los Alamos National Laboratory |
| LSDS | Lead Slowing-down Spectroscopy |
| NDA | Non-destructive assay |
| PNNL | Pacific Northwest National Laboratory |
| PWR | Pressurized water reactor |
| RMSE | Root Mean Square Error |

# Contents

# Figures

# 1.0 Introduction

Nondestructive assay (NDA) for quantifying the amount of the individual plutonium isotopes (e.g., $^{239}$Pu, $^{240}$Pu, $^{241}$Pu) in used-fuel assemblies is important for nuclear safeguards and used-fuel storage. Current NDA methods infer total plutonium mass using a combination of burnup codes for calculating isotopic inventories and passive measurements of easily-measured isotopes in used fuel (e.g., $^{137}$Cs and $^{244}$Cm). The International Atomic Energy Agency (IAEA) has determined that such methods typically carry a plutonium uncertainty of approximately 10% (Peter 2009). To address these issues, this work focuses on the application of lead slowing down spectroscopy (LSDS)—a well-established active interrogation technique having a long and extensive history for use in nuclear cross-section measurements (Danon et al. 2007; Romano et al. 2006)—to the measurement of isotopic masses in used fuel. The goal of this effort is to use LSDS to measure fissile isotopes (e.g. $^{235}$U, $^{239}$Pu, and $^{241}$Pu) directly in used-fuel assemblies with significantly better accuracy (at the ≤3% level), with minimal externally provided (operator-declared) information, and in a time-efficient manner. An example of an appropriate application of the LSDS instrument in a fuel cycle would be at the receiving end of a reprocessing facility.

Supported by funding from the Department of Energy Office of Nuclear Energy's Material Protection, Accounting, and Control Technology (MPACT) program, a collaboration was formed to study the technical challenges and advance the understanding of LSDS for fuel assay. The collaboration consists of Pacific Northwest National Laboratory (PNNL), Los Alamos National Laboratory (LANL), Rensselaer Polytechnic Institute (RPI) and Idaho State University (ISU).

This report provides a summary of the collaboration accomplishments in FY2012. A general introduction to the use of LSDS for used fuel assay can be found in *Lead Slowing-Down Spectrometry for Spent Fuel Assay: FY11 Status Report* (Warren et al. 2011). Detailed reports on the FY2012 accomplishments can be found in (Kulisek et al. 2012; Imel et al. 2012).

# 2.0 FY2012 Accomplishments

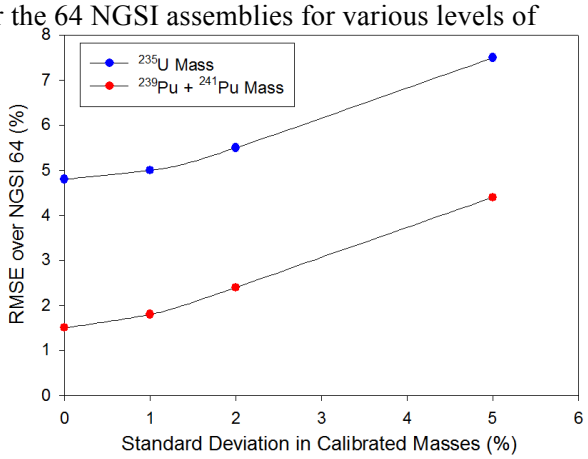
There are three main challenges for the implementation of LSDS techniques to the assay of used fuel. These challenges are the development of suitable algorithms, the empirical measurements, and development of fast neutron detectors. Accomplishments for each of these areas are listed below.

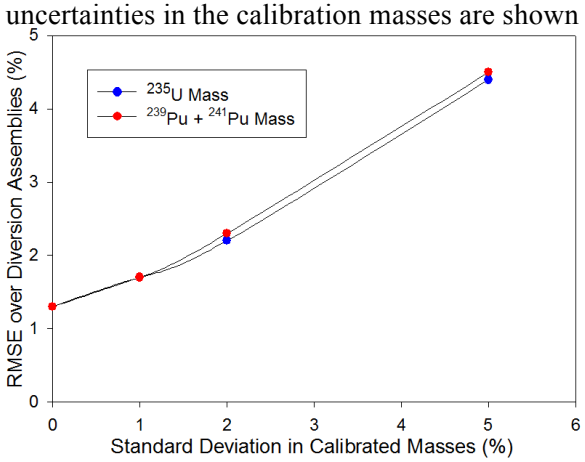
## 2.1 Algorithms

A few different algorithms have been pursued in FY 2012. PNNL has pursued two algorithm approaches. The semi-empirical approach uses singular value decomposition to determine the self-attenuation, and then extract the fissile masses, while the empirical approach uses linear algebra to relate the unknown masses to the measured time spectra (Kulisek et al. 2012). The PNNL models are typically applied to a widely diverse range of fuel assemblies. LANL has pursued a perturbation model, in which they examine sensitivity to small differences in the fissile mass (Gavron et al. 2011).

- PNNL applied their algorithms to data from measurements taken by RPI. The semi-empirical algorithm was able to determine the masses of $^{235}$U in the measurements within a root mean square error (RMSE) of 4.2%, while the second-order empirical algorithm had an RMSE of 2.3%. It is believed that better measurement statistics might improve on these results.
- LANL performed initial testing of a perturbation method to analyze the quantities of $^{235}$U, $^{239}$Pu and $^{241}$Pu based on time spectra of fuel assemblies with similar material inventories. Limiting the analysis to the time window that maximizes the differences between the response to $^{235}$U and $^{239}$Pu, we significantly improved the predictive capability of a perturbation approach over small changes in a particular fissile material (i.e., up to 10%).
- Both PNNL and LANL investigated the possibility of scaling isotopic fission chamber responses to avoid the use of exotic fission chambers based on $^{239}$Pu and $^{241}$Pu. PNNL found that the scaling based on the average response over the NGSI 64 significantly increased the RMSE of the calculated masses over the NGSI 64. LANL suggested techniques to tailor the scaling functions for given subsets of the NGSI 64, such as burnup-dependent scaling functions, that might improve upon the uncertainties found by PNNL.

- PNNL conducted an analysis to determine the sensitivity of the algorithms to uncertainties in the calibration set masses using the semi-empirical algorithm with all three isotopic fission chambers. The RMSE between the calculated and true masses over the 64 NGSI assemblies for various levels of uncertainties in the calibration masses are shown in




- **Figure 2-1**. This calculation was repeated for the 27 diversion assemblies. For a 2% standard deviation level of uncertainty in the calibration set masses, the RMSE for the sum of the $^{239}$Pu and $^{241}$Pu masses is still less than 3% for both of these assembly sets. Therefore, the accuracy of the calculated masses is still acceptable, allowing for realistic uncertainties of the calibration set masses.

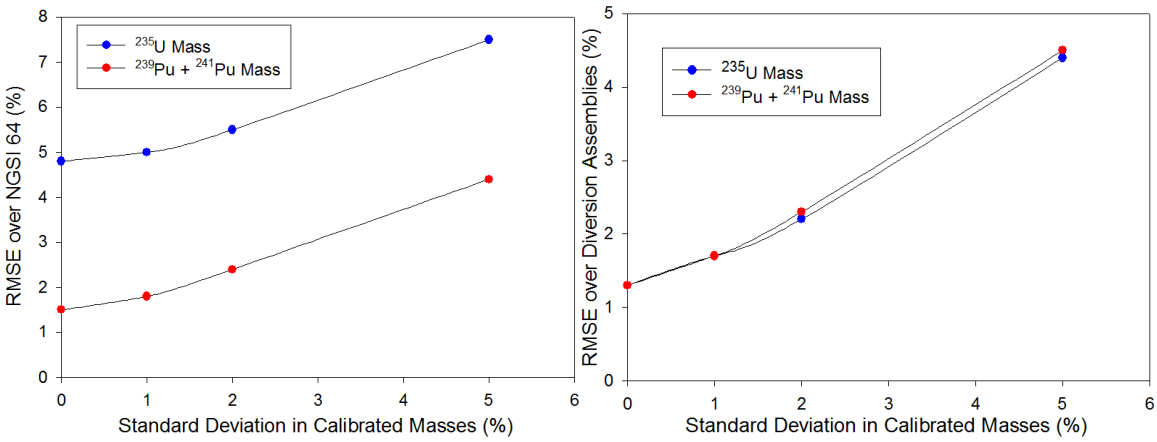



**Figure 2-1:** RMSE for $^{235}U$ and $^{239}Pu + ^{241}Pu$ mass estimates applying the semi-empirical algorithm on MCNPX-simulated assay. The left figure is for the NGSI 64 fuel assemblies, while the right figure is for the 27 diversion fuel assemblies.

- PNNL evaluated the effect of the number of calibration fuels used in the semi-empirical algorithm on the errors in the fissile isotopic mass estimates over the NGSI 64. The results of this analysis are shown in Figure 2-2. The effect of the calibration set size is not very pronounced for the $^{235}$U mass estimates; however, the $^{239}$Pu mass estimates improve monotonically with increasing calibration set size.

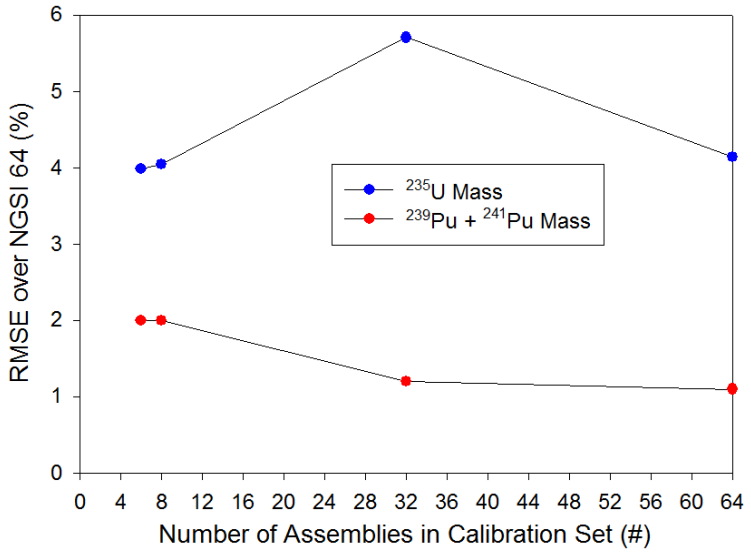



**Figure 2-2:** Results for $^{235}$U and $^{239}$Pu + $^{241}$Pu mass estimates for various calibration set sizes

- PNNL studied the sensitivity of the semi-empirical algorithm to the time window used in the analysis by varying the initial time of that window. These results are tabulated in **Table 2-1**. The data in the time window from 60 to 336 μs is clearly critical to an accurate extraction of the plutonium masses. The reason for the sharp drop in RMSE for $^{235}$U mass going from the 20 μs – 2000 μs to the 60 μs – 2000 μs time range is unknown.

**Table 2-1:** Average RMSE values obtained for $^{235}$U and $^{239}$Pu + $^{241}$Pu masses in the NGSI 64 using various slowing-down time ranges in the semi-empirical algorithm

| Time Range (μs) | RMSE of $^{235}$U Mass | RMSE of $^{239}$Pu + $^{241}$Pu Mass |
|---|---|---|
| 20 – 2000 | 11.2 % | 1.8% |
| 60 – 2000 | 4.4 % | 2.1 % |
| 336 – 2000 | 39.9 % | 11.7 % |
| 1125 – 2000 | 241% | 34.9% |

- PNNL studied the sensitivity of the LSDS technique over the geometry of the entire fuel assembly. Figure 2-3 shows the relative intensity of the interrogating flux and the relative assay signal per fissile atom as a function of the row, or depth into the assembly. The inner fuel rods observe about 90% of the neutron flux relative to the outer fuel rods. Also, these inner fuel rods contribute about 55% of the assay signal (per fissile atom) relative to the outer fuel rods. It should be noted that this 55% contribution from the inner row is likely an underestimate due to how the assay signal was recorded in MCNP. A similar investigation conducted by RPI indicated significantly more self-shielding (Romano et al. 2009), although the RPI investigation was performed on fresh fuel having a much higher enrichment of 9.8 wt% $^{235}$U.

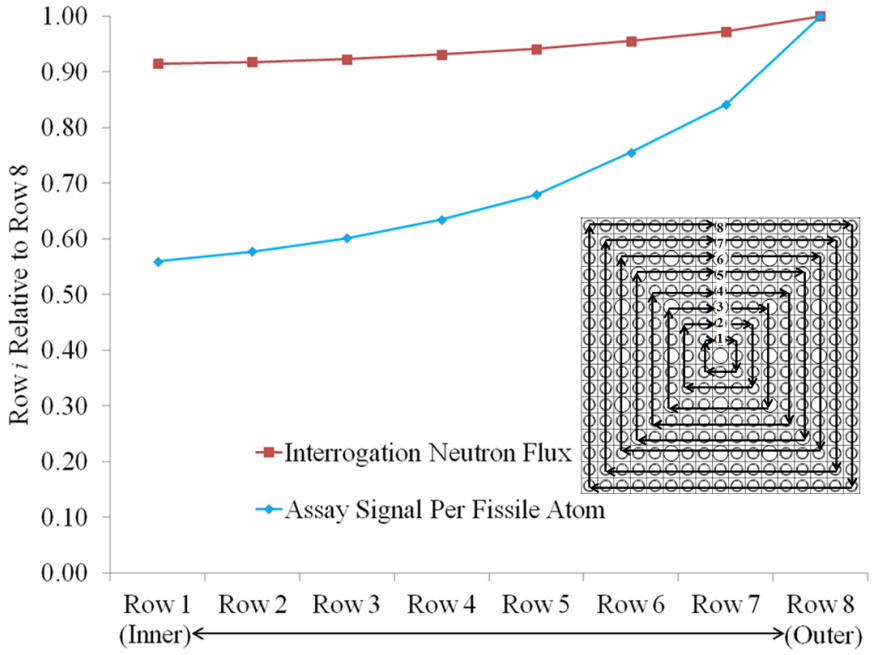



**Figure 2-3:** Assay signal per total number of $^{235}$U, $^{239}$Pu, and $^{241}$Pu atoms as a function of depth from the inner to the outer rows of fuel pins of the assembly. Also shown is the relative interrogation neutron flux as a function of depth in the assembly.

- PNNL simulated the assay signal from LSDS measurements on a single fresh fuel pin and nine-fuel-pin arrays for various levels of initial enrichment. The results for the single fuel pin are shown in Figure 2-4, while the results for the nine fuel pin assembly are shown in Figure 2-5. The assay signals are not proportional to the $^{235}U$ (fissile material) present in the fresh fuel pins, indicating the effect of self-shielding even for a single pin.

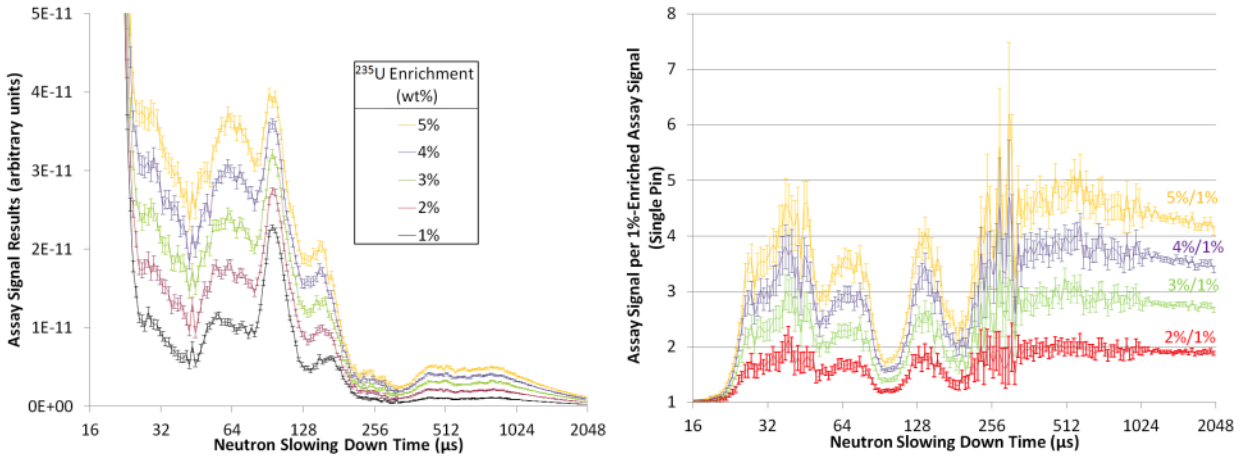



**Figure 2-4:** Assay signal for a single fresh fuel pin for various levels of initial enrichment. On the left, the $^{238}U$ fission chamber signal obtained from the simulated assay. On the right, the $^{238}U$ fission chamber signals are shown normalized to the signal for the 1 wt% enriched fuel pin.

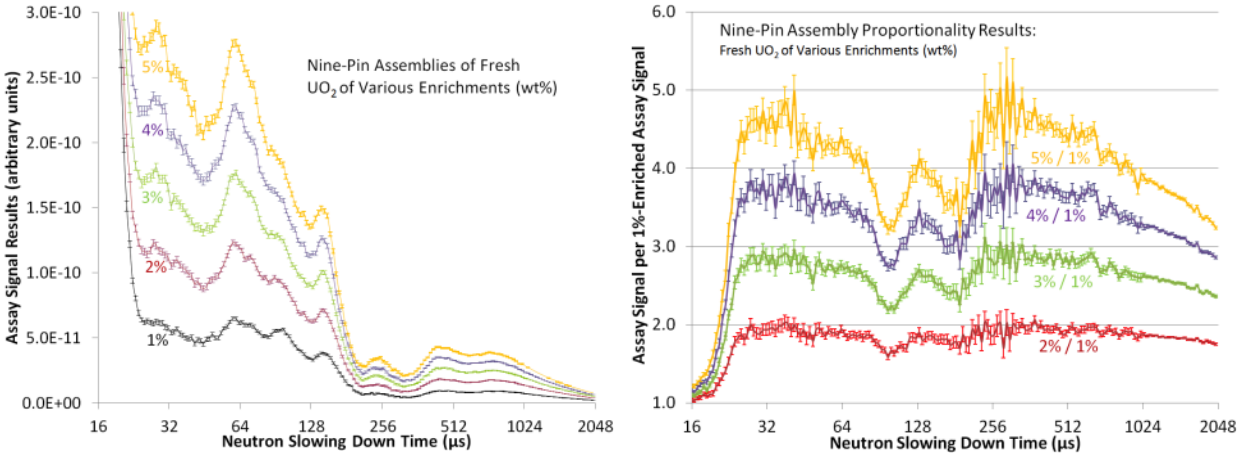



**Figure 2-5:** Assay signal for a nine-fresh-fuel-pin assembly for various levels of initial enrichment. On the left, the $^{238}U$ fission chamber signal obtained from the simulated assay. On the right, the $^{238}U$ fission chamber signals are shown normalized to the signal for the 1 wt% enriched fuel pin.

- PNNL studied the sensitivity to water in the central row (row 1 in Figure 2-3) of the fuel pins. The results of this analysis are shown in Figure 2-6. Water has a modest effect on the assay signal. No attempt has been made to quantify the impact of the water on the algorithms as calibrated against the NGSI-64 set of assemblies.

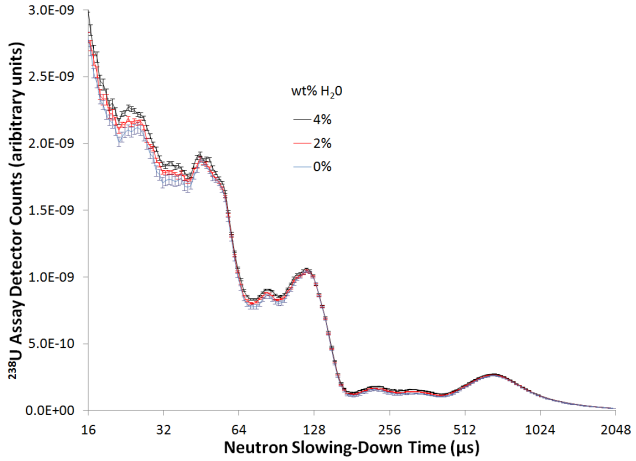



**Figure 2-6:** Simulated $^{238}U$ fission chamber signals for various wt% of water of each pin in the central row of fuel pins in the 30 GWd/MTU, 3 wt% enriched, 20 year cooling time assembly

- PNNL studied the contribution to uncertainties of the simulated assay signal. The number of source neutrons required in a notional LSDS instrument to achieve the same level of uncertainties in the results of the current MCNPX simulations is shown in Figure 2-7. The current statistical uncertainties of the simulations present a challenge for interpretation of some of the results. In the algorithm, it is assumed that the simulated signals are effectively noise free, so that a noise equivalent to $10^{16}$ neutrons is added to the signals. When the MCNPX simulations have greater statistical uncertainty than the Poisson noise characteristic of $10^{16}$ neutrons, the simulation uncertainties dominate the analysis, potentially creating artifacts in the analysis (Smith et al. 2011). This result indicates that significant effort will be required to reduce the simulation uncertainties for times less than 1 ms.

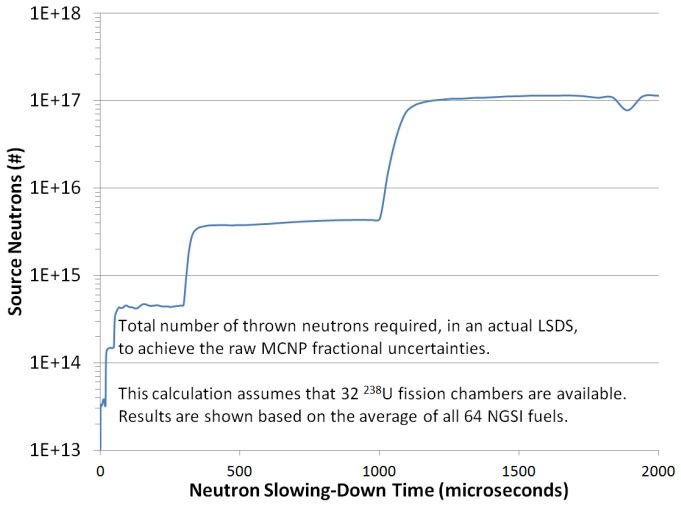



**Figure 2-7:** Number of source neutrons required in an LSDS instrument to achieve the same level of fractional uncertainties in the MCNPX-simulated $^{238}$U fission chamber signal

- PNNL studied the appropriate outer dimensions of lead required for the LSDS instrument. The simulated assay signals for the same fuel but for various outer dimensions of the lead are shown in Figure 2-8. The 2 m by 1 m size (the nominal lead dimensions) performs very similarly to the 1.75 m by 1.75 m. However, increasing the lead to 2 m by 2 m significantly enhances the assay signal. The appropriate way to understand the impact of the lead dimensions is to run the process for all geometries for all assemblies of the NGSI 64 set and determine the mass uncertainties. This effort is computationally intensive and has not been completed.

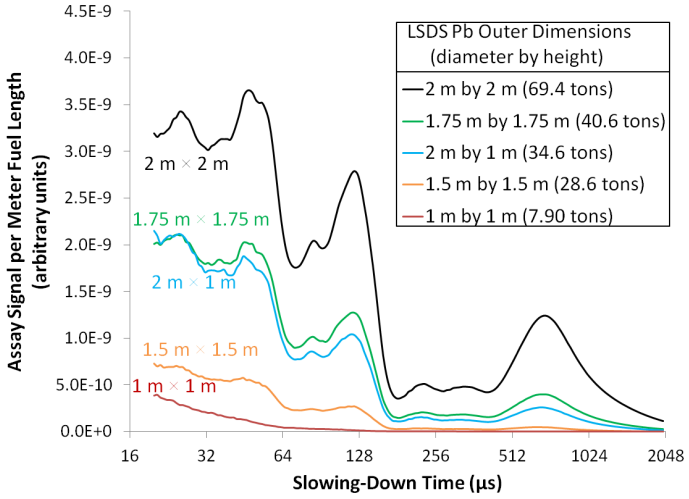



**Figure 2-8:** Simulated $^{238}U$ fission chamber signal for cylindrical LSDS instruments of various size (lead outer dimensions). The source location was kept constant at ~60 cm from the fuel.

- PNNL also studied the impact of the neutron source location with respect to the fuel. Results are shown in Figure 2-9 for the LSDS instruments with different source-to-fuel distances. The neutron source is assumed to be isotropic in this study. These results show a large increase in signal strength and time-spectral resolution when the source is moved deeper in to the LSDS instrument and closer to the fuel.

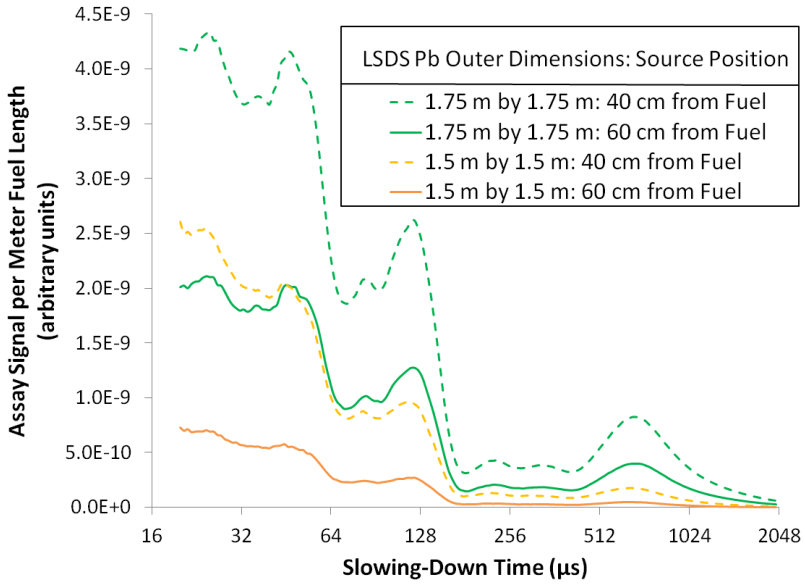



**Figure 2-9:** Simulated $^{238}$U fission chamber signal for cylindrical LSDS instruments of various size (lead outer dimensions). The source distance from the fuel was varied between 40 and 60 cm.

- PNNL explored the concept of using cadium around the assay signal detectors as a means to reduce the influence of $^{235}U$ contamination in the $^{238}U$ fission chambers. It was found that cadmium is not very beneficial. It does not significantly reduce the sensitivity to the level of $^{235}U$ impurities in a $^{238}U$ fission chamber, and it significantly alters the shape of the response, possibly destroying some of the signal. The alteration of the signal can be seen in Figure 2-10.

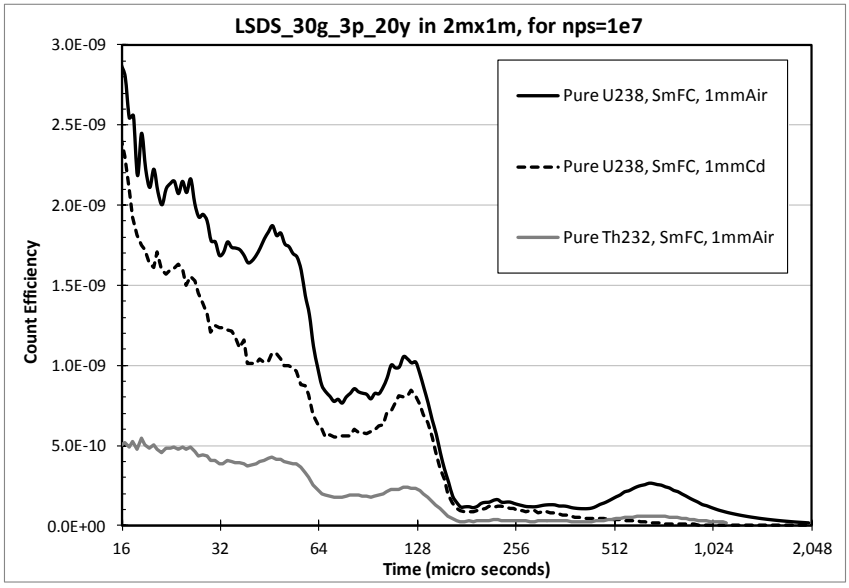



**Figure 2-10:** Comparison of assay signal response for a pure $^{238}U$ chamber and a pure $^{232}Th$ chamber with different material immediately outside of the chambers. The solid lines are for geometries in which there is 1 mm of air outside of the chamber, while the dashed line is for 1 mm of cadmium around the outside of a $^{238}U$ chamber. The "SmFC" indicates a specific fission chamber geometry that is not relevant for this discussion.

- PNNL studied the sensitivity of a $^{238}$U-based assay detector to $^{235}$U impurities. A first qualitative attempt to answer that question has been completed and is shown in the top of Figure 2-11. The assay signals of a $^{238}$U fission chamber with various levels of impurities of $^{235}$U are illustrated. No impurities and 4 ppm $^{235}$U produce essentially the same signal. While the higher levels of impurities do change the response, they do not dramatically alter the shape of the response. As a result, it may be possible that significantly higher levels of $^{235}$U impurities than 4 ppm would be tolerable and provide reasonable uncertainties once applied across the NGSI 64 library.

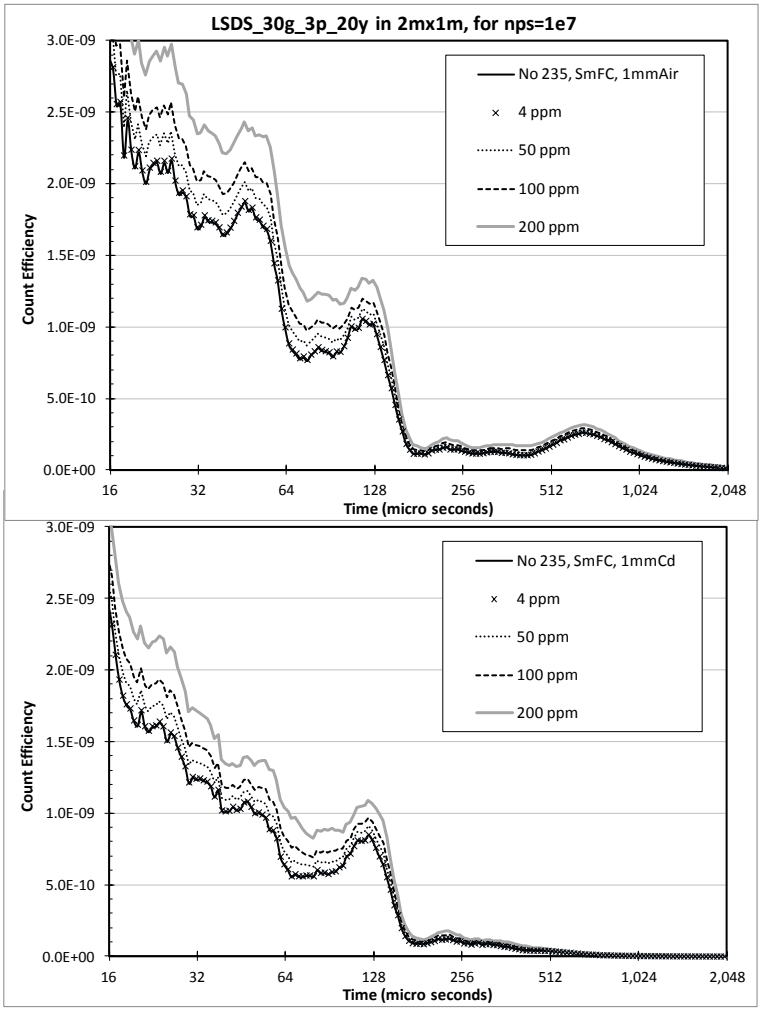



**Figure 2-11:** Sensitivity to $^{235}$U impurities in a $^{238}$U fission chamber. The top figure is the count efficiency of the nominal $^{238}$U fission chamber with given levels of impurities. The bottom figure is similar, except that the fission chamber has been surrounded by 1 mm of cadmium. The "SmFC" indicates a specific fission chamber geometry that is not relevant for this discussion.

## 2.2 Empirical Measurements

- LANL completed benchmarking of a $^{235}U$ and a weapons-grade plutonium (WGPu) source at the LANSCE LSDS instrument using a compensated thorium-plated ion chamber. As shown in Figure 2-12, some discrepancies between the experimental data and simulation calculations were observed. For both sources, a discrepancy occurs for slowing-down times greater than 120 microseconds. This discrepancy is also evident in background runs (no fissile source) where the natural uranium in the thorium detector responds to the slow neutron flux. In addition, there is a discrepancy between the WGPu data and calculations between 30 and 80 microseconds. We have not yet resolved the origin of this discrepancy. Similar benchmarking has been performed at RPI. The RPI LSDS instrument is considerably larger than the LANSCE LSDS instrument (1.8 meters long compared to 1.2 meters at LANSCE). RPI does not find a discrepancy at 30-80 microseconds.

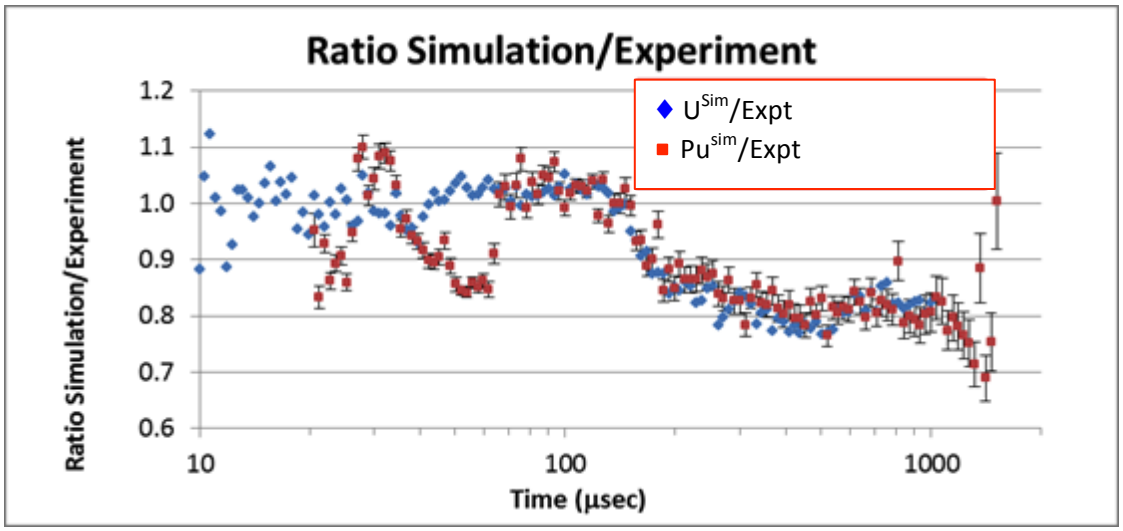



**Figure 2-12:** Ratio of simulated LSDS spectra to experimental spectra as a function of time. The diamonds are the $^{235}U$ data, and the squares—the $^{239}Pu$ data.

- RPI observed that the response of the $^{239}Pu$ fission chamber placed in the LSDS instrument is sensitive to room return in a region where the fission cross section is low. This observation enabled the adjustment of the simulation to include the proper room return.

- RPI obtained a new analysis of the impurities in the RPI lead. The analysis included new information on the content of strong absorbers such as boron (0 ppm), cadmium (0.01 ppm) and gadolinium (0.01 ppm). New calculations with these impurities were compared to the measured response of $^{239}$Pu and $^{235}$U fission chambers. The results show the visible but small effect of the impurities. The agreement between the experiments and calculations is good, as seen in Figure 2-13.

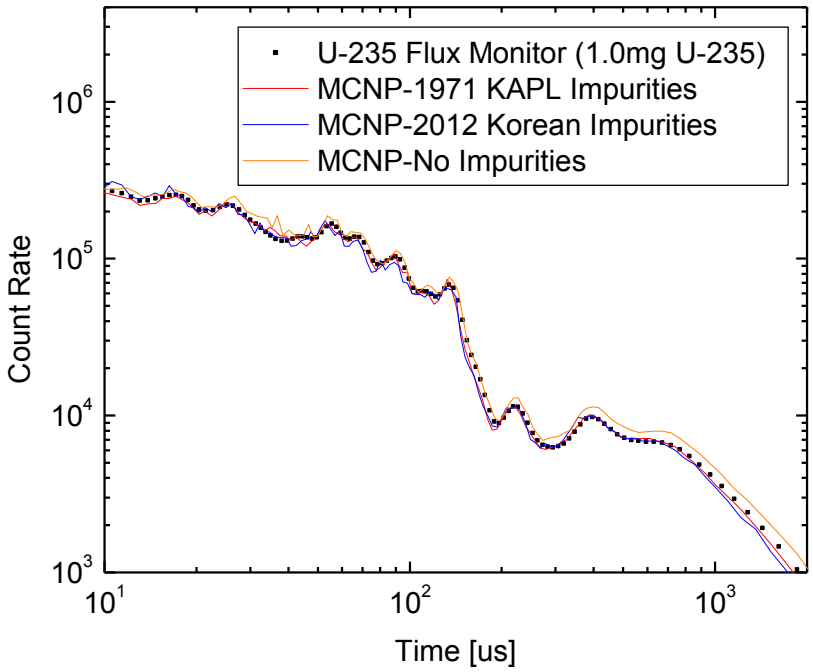



**Figure 2-13:** Comparison measurements of $^{235}$U fission chamber response to simulations with two sets of impurities in the RPI lead. Hydrogen content in the simulations is 1 ppm as measured in the new chemical analysis.

- RPI investigated the effect of room return on the response of a $^{239}Pu$ fission chamber placed in the LSDS instrument. They concluded that some room return affects the measurement and depends on how well the measurement location is covered with lead and neutron absorbers. Figure 2-14 shows measurements at different positions in the lead; the effect of room return is seen in the region of 200 µs to 400 µs. Covering the detector channel with $Li_2CO_3$ neutron absorber shows that the room return can be reduced.

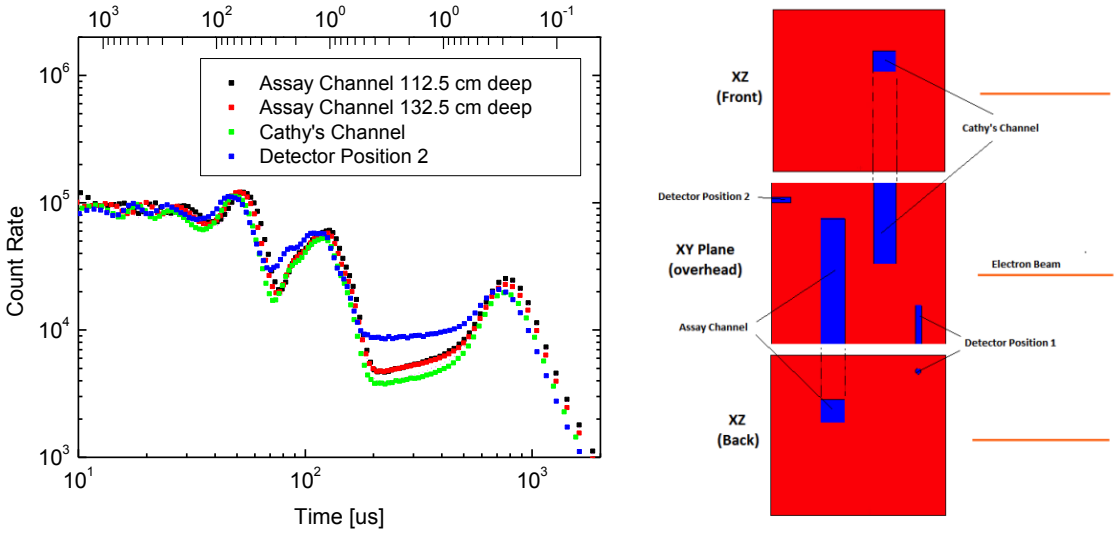



**Figure 2-14:** The slowing down spectrum at different locations in the RPI LSDS instrument. The data shows the effect of room return in the slowing down time of 200 µs to 400 µs.

- RPI performed a series of experiments with a fuel pin (~4.8% enriched in $^{235}$U), several $^{235}$U disks and two PuBe sources. The uranium pin contains approximately 34.8 g $^{235}$U and 689 g $^{238}$U. The 10 HEU disks contain a total of 2.6 g $^{235}$U. The large PuBe source contains approximately 90 g of plutonium, while the smaller PuBe has about 47 g of plutonium. Both of these sources have a considerable amount of fissile material, complicating the assay of uranium within the fuel pin and HEU disks. These experiments were designed to provide a limited set of data to benchmark the unfolding algorithms. The measured response of a $^{238}$U assay detector is shown in Figure 2-15. The measurements containing the PuBe source have a significantly higher count rate.

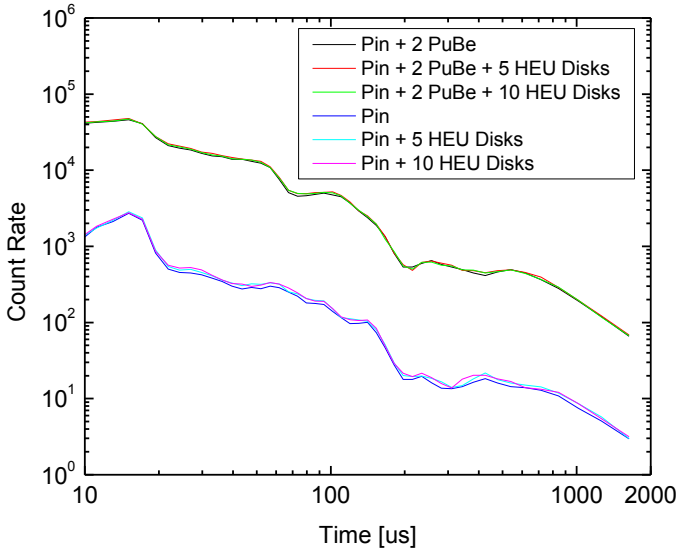



**Figure 2-15:** The $^{238}$U assay detector response corresponding to a fuel pin and variable HEU and fuel pin with two PuBe sources and variable HEU disks

## 2.3 Fast Neutron Detector

- LANL tested an alternative detector for fast neutrons. The detector was a $^{4}$He-filled proportional counter, which could provide a three orders-of-magnitude higher efficiency than the efficiency of the thorium detector. However, it saturated with the initial pulse from the proton storage ring and became inoperable. We are now examining the possibility of using a $^{4}$He scintillator, which involves some complex steps in construction that will make it initially expensive. We hope that the budget in the next fiscal year will support its construction. We are also looking in to the possibility of collaborating with Duke University, where they have some experience constructing such a detector (Tornow et al. 2011). This system would require a gated power supply for the phototube that drops the HV during the first few microseconds after the pulse to prevent saturation.

- ISU built back-to-back fission chambers in early 2012, as shown in Figure 2-16. These detectors will enable the calibration of thorium-plated foils. The simple geometry enables calibration of an unknown foil against a known foil. It is also possible to use these chambers to subtract the gamma response of the detectors by using one plated foil and one unplated foil.

**Figure 2-16:** Photos of back-to-back fission chambers. Left image shows the detector when the chambers are closed, while the right shows the detector when the chambers are open.

- ISU is currently investigating available organizations that could deposit the thorium on the metal foils. The university was working with Idaho National Laboratory (INL) on this in 2011 and early 2012, but recent personnel changes at INL have created a delay. There are other possibilities, including PNNL and Lawrence Livermore National Laboratory (LLNL).

# 3.0 Summary and Future Work

Significant progress was made on the project in FY2012. Extensive characterization of a "semi-empirical" algorithm was conducted. For example, we studied the impact on the accuracy of this algorithm by the minimization of the calibration set, uncertainties in the calibration masses, and by the choice of time window. Issues such as lead size, number of required neutrons, placement of the neutron source and the impact of cadmium around the detectors were also studied. In addition, new algorithms were developed that do not require the use of plutonium fission chambers. These algorithms were applied to data from measurements taken by RPI and shown to determine the $^{235}$U mass to within 4%. For detectors, a new concept for a fast neutron detector involving $^{4}$He recoil from neutron scattering was investigated. The detector has the potential to provide a couple of orders of magnitude more sensitivity than $^{238}$U fission chambers. Progress was also made on the more conventional approach of using $^{232}$Th fission chambers as fast neutron detectors. For benchmarking measurements, we continue to improve our understanding of the experimental setup by studying issues such as the effect of room return and impurities in the lead. RPI performed a series of experiments with a fresh fuel pin and various $^{235}$U and $^{239}$Pu sources. A comparison between simulations and measurements shows significant deviations after 200 μs for both $^{235}$U and $^{239}$Pu samples, as well as significant deviations at earlier times for the $^{239}$Pu sample.

The FY2013 effort will shift focus to planning for a Technical Readiness Level 5 demonstration. The primary deliverable for the year will be a plan on how to do this demonstration. The plan will include measurement design, sample acquisition, sample handling, cost estimate, schedule and assumptions. Research will continue on the $^{4}$He detector, algorithms development, thorium fission chambers and benchmarking measurements involving sub-assemblies of fresh fuel.

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