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Determining Plutonium Mass in Spent Fuel with Non-destructive Assay Techniques – NGSI Research Overview and Update on NDA Techniques

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IAEA-CN-184/137

Determining Plutonium Mass in Spent Fuel with Non-destructive Assay Techniques – NGSI Research Overview and Update on NDA Techniques, Part II

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Abstract. The second paper on the non-destructive assay techniques investigated under the Next Generation Safeguards Initiative considers innovative instrument designs capable of producing isotope-specific responses for quantifying the fissile material content of spent nuclear fuel. The present research goal is to assess whether expected signatures can be confidently obtained in realistic spent fuel safeguards applications, contribute to the establishment of Pu inventories and determine fissile material diversions at fuel storage, handling and reprocessing facilities. In certain cases, theoretical concepts are supported by the experimental data obtained in the simplified setups. The overall effort will be concluded by nominating the most promising techniques and their combinations for the full-scale demonstration involving actual nuclear fuel assemblies. This paper provides an overview of the following 6 photon- and neutron-based techniques: Delayed Gamma, X-Ray Fluorescence, Nuclear Resonance Fluorescence, Passive Gamma, Lead Slowing-Down Spectrometry, and Neutron Resonance Transmission Analysis.

1. Introduction

The first paper accompanying this two-poster presentation introduces 6 out of 14 techniques developed for the non-destructive assay (NDA) of the spent nuclear fuel under the U.S. DOE Next Generation Safeguards Initiative (NGSI) [1, 2]. While extensive existing expertise and state-of-the-art modelling capabilities ensure an accurate design process, responses investigated in these neutron-based instruments are a convolution of multiple effects occurring in the isotopically and spatially complex nuclear fuel under interrogation conditions. Therefore, each of them will require an increasingly intricate analysis and often a combination of a few techniques in order to make fissile material inventory prediction.

The present article and the second poster consider 4 photon- and 2 neutron-based NDA techniques aimed at identifying and isolating the isotope-specific signatures available in the spent nuclear fuel assay. Although most of the techniques presented here are less mature than the ones in the preceding publication, these concepts potentially offer direct information on the fissile and fertile material inventory and parameters of the spent fuel such as residual fission rate, burnup estimation, etc. It is expected that some of them will be capable of achieving the NGSI goal of quantifying Pu content as stand-alone assay instruments or can be easily combined with any of the other NGSI project techniques. It is also probable that the innovative nature of these concepts will make them attractive for safeguards of fissile materials in a variety of forms other than spent nuclear fuel.

The overall design complexity of the instruments discussed here governed the somewhat specific approach and pace at which they were developed under the NGSI project. Often, a theoretical analysis of each technique required significant modifications and alterations of the existing modeling codes. In some cases, completely new algorithms and calculation schemes had to be introduced. Hence, a strong emphasis was placed on

verification and validation of these modeling capabilities before approaching the spent nuclear fuel assay applications. For certain techniques, the NGSI project has initiated specific experimental campaigns and data exchanges that supplied benchmarking information when literature sources were scarce.

The importance of benchmarking in simplified setups was elevated to ensure that the design models accurately account for all the complexities of the spent fuel assay setups rarely encountered in past experiments. In case of the neutron techniques, such factors as periodic structure of the fuel assembly, interrogation media and presence of the structural materials are all capable of affecting the expected response. The photon techniques require careful introduction of the high-resolution detectors to extremely intense exposure fields around the assembly. In cases with active interrogation, there is a direct correlation between type, strength and position of the source to the possibility of detecting the desired signatures. This type of analysis is generally computationally intense and any optimization has to be performed without compromising the accuracy.

At the current stage of the NGSI project, the primary design objective for the techniques discussed below is to confirm that the signatures nominated for the subsequent analysis can be securely retrieved under the optimal interrogation setup and parameters. It is important to demonstrate that an instrument provides sufficient accuracy, sensitivity and isotope specificity (if applicable) as is required in the NDA of spent nuclear fuel. The results of these findings will be used to determine the expected importance of each concept in a variety of safeguards scenarios and conclude whether the technique will be promoted to the next stage of the project entailing full-scale experimental verification.

2. Delayed Gamma (DG)

Capabilities of the spent nuclear fuel assay utilizing the delayed gamma signatures is currently being investigated at the Los Alamos National Laboratory, with contributions from the UC Berkeley Nuclear Engineering Department, Lawrence Berkeley National Laboratory, and Pacific Northwest National Laboratory [3, 4].

The isotope-specific delayed gamma assay is based on utilizing the difference in rates at which beta-delayed gamma-emitting isotopes are produced in fission events. The distribution of fission product yields can serve as a signature for each fissile element. The method requires preliminary data on the delayed gamma decay energies indicative of fissions in target isotopes present in the material under investigation. Gamma peaks corresponding to these energies are then isolated in the photon spectrum collected following the cool-down period after exposure by an active source.

In order to address the variety of aspects associated with the delayed gamma assay of the spent fuel, an innovative modeling technique was developed under the NGSI project. The calculation algorithm combines the analytical transmutation CINDER [5] code with the specifically developed DGSDEF [6] code producing the spatially- and time-dependent discrete gamma source. An arbitrary transport code, currently MCNPX [7], is utilized to calculate the interrogating neutron fluxes and predict acquired spectra accounting for the interrogation geometry and detector effects. This calculation scheme approach was subsequently extended to the design of other photon-based NGSI techniques (Passive Prompt Gamma and X-Ray Fluorescence).

The complexity of the delayed gamma modeling algorithm was verified in an extensive benchmarking effort involving the analysis of literature data and a specifically organized experimental campaign at the Idaho Accelerator Center of the Idaho State University. In this effort, the validity of the calculation technique and the quality of the associated physical data libraries was confirmed. Figure 1 demonstrates a good agreement between the measured and calculated spectra for the interrogation of the depleted uranium sample containing 0.2% U-235 with a LINAC-driven thermal neutron source. The developed modeling technique provides an accurate estimation of isotopic contributions to the final spectrum and produces the discrete gamma source term in the irradiated target.

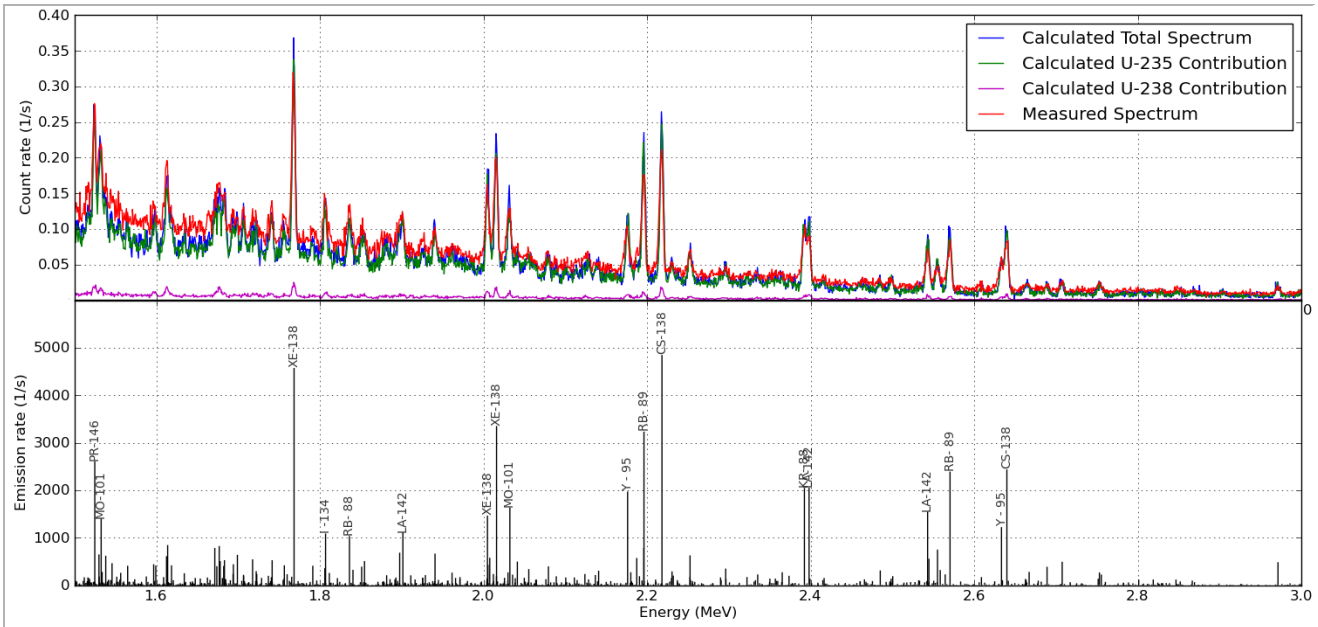

Fig. 1. A fragment of calculated and measured spectra for depleted uranium delayed gamma experiment (background subtracted) with calculated isotopic contributions (top). Calculated emission spectrum in the irradiated target (bottom).

Satisfactory benchmark results provided sufficient grounds for the application of the calculation technique to simulations of the spent nuclear fuel delayed gamma assay. The conceptual modeling considered one of the 17x17 PWR assemblies from the NGSI spent nuclear fuel library [8]. In order to avoid interference from the passive continuum due to intrinsic spent nuclear fuel radioactivity, the region of interest for the delayed gamma peaks is extended to a wide high-energy gap between 2 and 6 MeV. The interrogation setup involves a Deuterium-Tritium neutron source compatible with most of the NGSI active interrogation techniques, and a high-efficiency HPGe detector for spectra acquisition. The current effort concentrates on optimization of the detector setup with the goal of reducing the count rate on the detector to a realistic level without degradation in the target delayed gamma peaks.

3. X-Ray Fluorescence (XRF)

The XRF assay technique is being developed by the Los Alamos National Laboratory in collaboration with the Oak Ridge National Laboratory and Texas A&M University [9].

This technique focuses on the feasibility of using the passively excited x-ray emission peaks from Pu and U to gather information about their relative quantities in the spent fuel. XRF is unique among the other techniques since it utilizes element-specific signatures rather than isotopic. It also presents multiple challenges when applied to spent nuclear fuel assay, mainly its penetration issues, since the low energy x-rays desired are effectively shielded by the few millimeters of a fuel material. Another concern is that these low energy x-ray signatures are located above a large background from intrinsically radioactive fuel and Compton scattering effects in the detector. This is particularly a concern with plutonium signal because in a PWR spent fuel its content is only within 1 to 2%. In case of low burnup with less Pu, the signal-to-noise ratio may be too low to get an accurate x-ray measurement.

For the XRF response prediction in the assay conditions the same modeling scheme as used for the Delayed Gamma instrument was adopted. By combining results of CINDER, DGSDEF, and MCNPX codes, the detailed source term and detected spectra were calculated for pre-defined material inventories. The modeling approach was benchmarked against the actual XRF measurements of North Anna spent fuel pin with some results shown in Figure 2. In this spectrum comparison the calculated peaks are shifted over from the measured result. This effect is due to a shift in U and Pu x-ray energies encoded in MCNPX that was used on the last stage of calculation. Overall, a good agreement in the peak-to-background ratio between measured and simulated spectra can be observed.

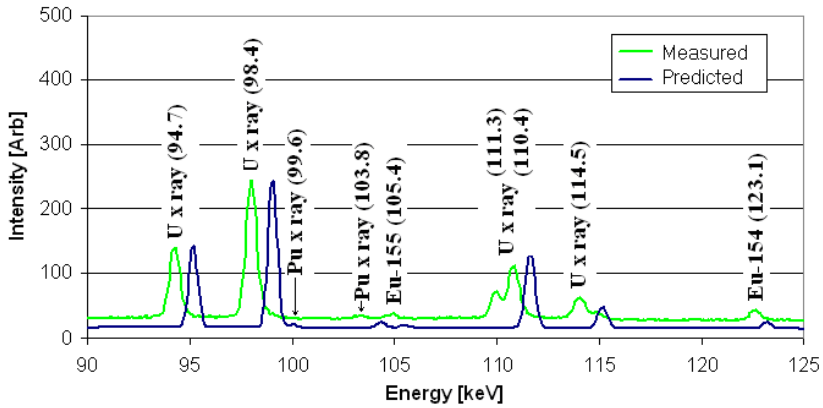

Fig. 2. Comparison of simulated and measured XRF spectra of North Anna spent fuel pin.

Both modeling and experimental results demonstrate that the plutonium x-rays can be collected for spent fuel; however there are several reservations about XRF capabilities for measurement of complete assemblies. First, the mean free path of low energy x rays in nuclear fuel is only a fraction of a mm. This is a serious drawback when considering assembly assay, since the measurement will likely be blind to diversions in all pins that do not have line-of-sight with the detector. Furthermore, the x-ray signal will increase proportional only to the first row of pins, the continuum is expected to increase proportional to the whole depth of the assembly, further degrading the already poor signal-to-noise ratio of the plutonium peaks.

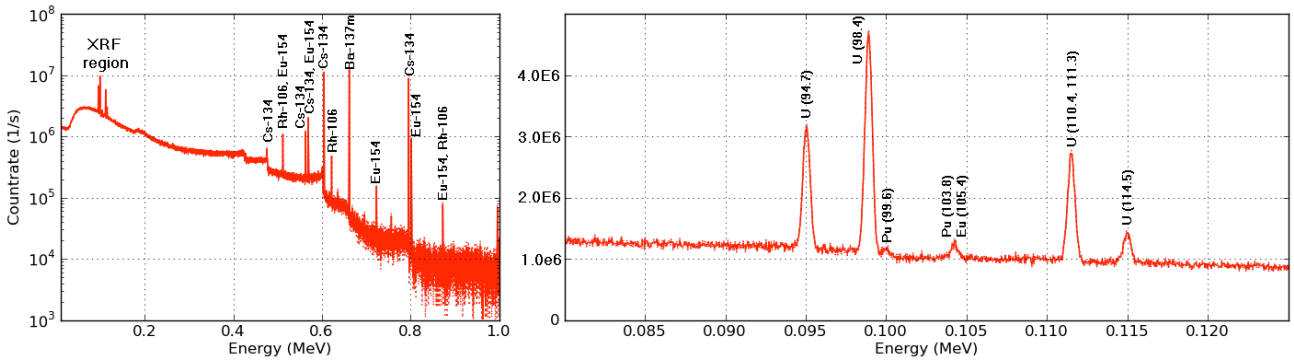

Fig. 3. Simulated XRF response for a spent fuel assembly assay with a planar detector: full spectrum (left) and XRF region (right).

Results of preliminary modelling of the XRF response from the spent nuclear fuel assembly using a planar HPGe detector are shown in Figure 3. Despite a number of assumptions adopted in this modelling, it is conceivable that the detector setup can be optimized to overcome the count rate issues and isolate Pu x-ray peaks. However, since XRF is only able to measure the outer layer of the assembly, the measured signature must be extrapolated to represent the plutonium content in the whole assembly. This presents a challenge since Pu content varies with burnup across individual pins and the whole assembly. Even if the signal-to-noise obstacle of assaying an assembly can be overcome, the plutonium distribution must be known to utilize the XRF assay technique.

4. Nuclear Resonance Fluorescence (NRF)

The Nuclear Resonance Fluorescence application to the spent nuclear fuel assay is investigated at the Lawrence Berkeley National Laboratory in collaboration with the UC Berkeley Nuclear Engineering Department and Los Alamos National Laboratory [10].

In NRF measurements specific isotopes are identified by actively induced characteristic lines observed in recorded high-resolution gamma spectra. The concentration of an isotope in a material can be determined from measured NRF signal intensities if NRF cross sections and assay geometries are defined. The resulting gamma spectra measured exhibit nuclide-specific, narrow lines that allow the identification and quantification of isotopic content. Nuclear resonances for U-235, U-238, and Pu-239 have been found in the 1.5 MeV to 2.5 MeV range with cross sections that are typically several to tens of eV·barn. Since photons with energies

around 2 MeV can penetrate a fuel assembly, NRF potentially enables the non-destructive, direct measurement of isotopic mass (fissile and actinides) without the need for operator-declared information.

In case of the spent nuclear fuel assembly interrogation, the direct measurement of NRF signatures proved to have several disadvantages. Spent fuel assay requires the measurement of a small isotopic fraction resulting in a signal to (scattered) background ratio that is quite small. In addition, the background produced by the radioactive decay of the fission products in spent fuel is difficult to overcome. Furthermore, it may be difficult to correct for the depth dependence of the isotopic measurement. As a more promising approach for the spent fuel assembly assay, a transmission measurement method was proposed. In this technique, schematically shown in Figure 4, an interrogating photon beam is transmitted through the assembly, in which photons with resonant energies are preferentially attenuated. This attenuation can be measured with the aid of a so-called witness foil that consists of the isotope of interest and intercepts the transmitted beam. The NRF spectra are measured at backwards angles where the detectors are shielded from the background produced by the assembly. The decrease in the observed NRF peaks when compared to a reference standard indicates the concentration of the isotope of interest in the assembly.

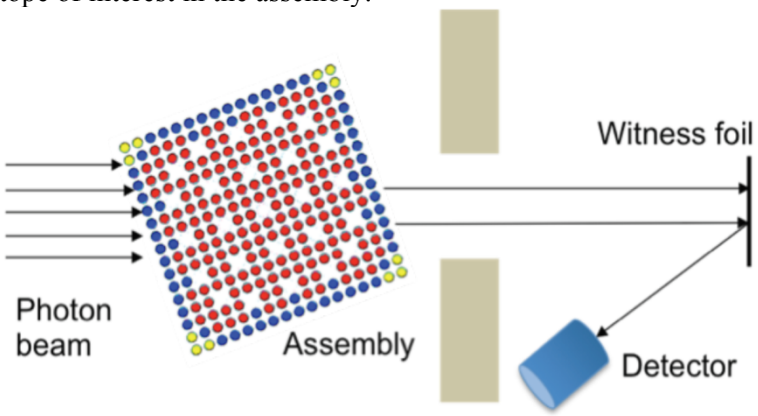

Fig. 4. Schematic setup for a transmission NRF measurement. The assembly is rotated with respect to the incoming beam to prevent the streaming of photons between rows of pins.

Accurate calculation of the NRF assay responses required considerable modification and update of the existing modeling techniques. The NRF physics was introduced to the MCNPX transport code and the Rayleigh scattering routine was corrected to ensure the correct prediction of the elastically scattered photon background important for signal-to-background ratios. The code improvements were verified in the proof-of-principle experiment performed at Massachusetts Institute of Technology High-Voltage Research Lab. Preliminary modeling of Pu-239 measurement in a spent fuel assembly with a bremsstrahlung source and HPGe detectors showed that source and count rate limitations lead to unsatisfactory long measurement times for precisely measuring the small concentrations of fissile isotopes. However, other gamma detectors with much higher count rate capabilities could be a better choice. Photon sources with very narrow energy spreads, such as based on laser Compton scattering, would greatly reduce the flux of low energy photons scattered into the detectors and, with sufficient beam intensities, allow much higher NRF count rates.

5. Passive Photon Assay

Photon techniques described in the previous sections involve advanced detector systems and sophisticated acquisition setups. To support the design process specific codes and modelling techniques were developed. The new capabilities providing accurate prediction of detector responses and associated source terms suggest a revision of the established spent fuel assay techniques based on passive signatures. Present research effort considers calculation and analysis of the high-resolution passive gamma spectra as shown in Figure 5.

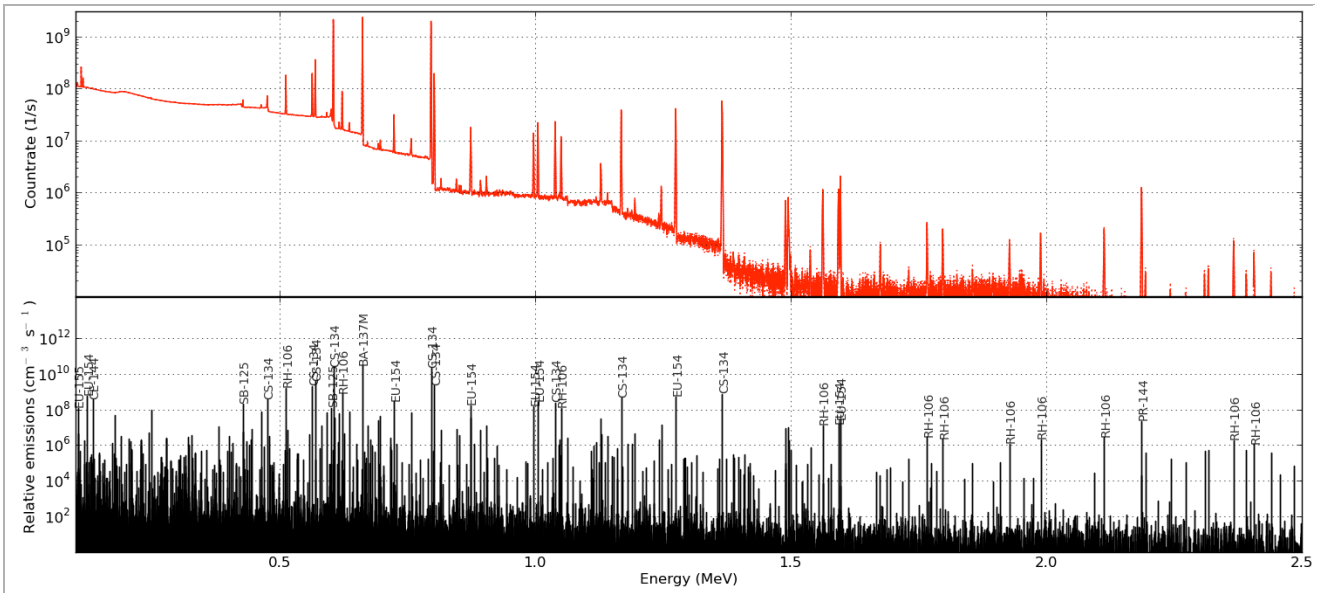

Fig. 5. Calculated high-resolution passive gamma spectrum of a spent nuclear fuel assembly obtained for the HPGe detector with unrestricted count rate (top). Associated discrete photon source term (bottom).

Detailed studies of high-resolution spectra can potentially improve existing and proposed spent fuel assay techniques. The concentration of individual fission products can be obtained by measuring specific gamma lines; these individual activities and activity ratios can be related to fuel burnup, initial enrichment and other parameters. This data can be used for additional verification of operator-declared values and identify assemblies with unusual irradiation history or possible pin diversion.

6. Lead Slowing-Down Spectroscopy (LSDS)

The assay instrument for Lead Slowing-Down Spectroscopy was developed at the Pacific Northwest National Laboratory with contribution from the Los Alamos National Laboratory [11].

LSDS is an active interrogation technique that has been used extensively in cross-section measurements, but has seen relatively limited application to the non-destructive assay of nuclear materials. The assay principle is based on the time-dependent production of fission neutrons in the fuel as a pulse of interrogation neutrons is moderated in the region surrounding the interrogation chamber as shown in Figure 6. Prior work in the application of LSDS to spent fuel demonstrated its potential for direct measurement of fissile isotopes such as U-235, Pu-239 and Pu-241 in full assemblies, but also noted the severity of self-shielding and the resulting system nonlinearities. More recently, an analytical model for self-shielding was developed, which draws on tabulated nuclear data but leaves the mass of fissile and attenuating isotopes as free variables to fit. This new time-spectrum analysis technique also allowed direct assay of non-fissile Pu-240, an important isotope for quantification of total Pu in international safeguards.

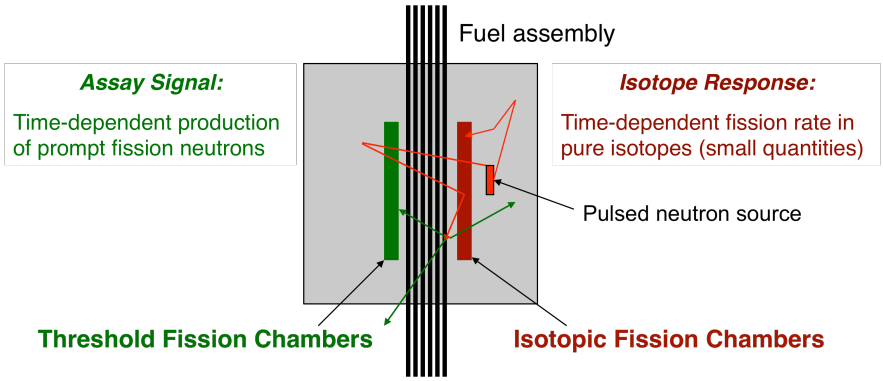

Fig. 6. Schematic of a nominal lead-slowing-down spectrometer for spent nuclear fuel assay.

According to preliminary simulations, LSDS instrument interrogates the entire depth of the assembly and as a result, mass assay is largely insensitive to the spatial distribution of target isotopes within the assembly. Along

with a capability to accurately detect Pu and U isotopic mass distributions, it makes LSDS assay extremely sensitive to pin diversions from the fuel assembly. However, these unique characteristics are dependent on a large volume of high-purity lead (likely greater than 25 tons), an intense pulsed neutron source, and a low concentration of low-mass isotopes such as hydrogen in the media. Therefore, LSDS potential applications are limited and can include the head-end of a reprocessing plant, mixed-oxide fuel fabrication plants (particularly for those fuels where gamma-ray and neutron signatures from recycled actinides overwhelm Pu emissions), and interim or long-term dry storage facilities.

7. Neutron resonance transmission analysis (NRTA)

Neutron Resonance Transmission analysis for spent fuel assay is investigated at the Idaho National Laboratory [12].

NRTA uses a pulsed accelerator and time-of-flight technique to characterize spent fuel. The accelerator produces short, intense pulses of fast neutrons. These fast neutrons are moderated with a low-Z material and are thermalized to the 0-40 eV resonance energy range. These thermal neutrons impact the spent fuel pins and are scattered, absorbed, or transmitted. Neutron transmission through the assembly is monitored as a function of neutron energy (time after the pulse). Neutron detection is performed using a high-rate sensor, such as a lithiated-glass scintillation detector. Results are read from the detector as count rate versus time, with faster neutrons arriving first and slower neutron arriving later.

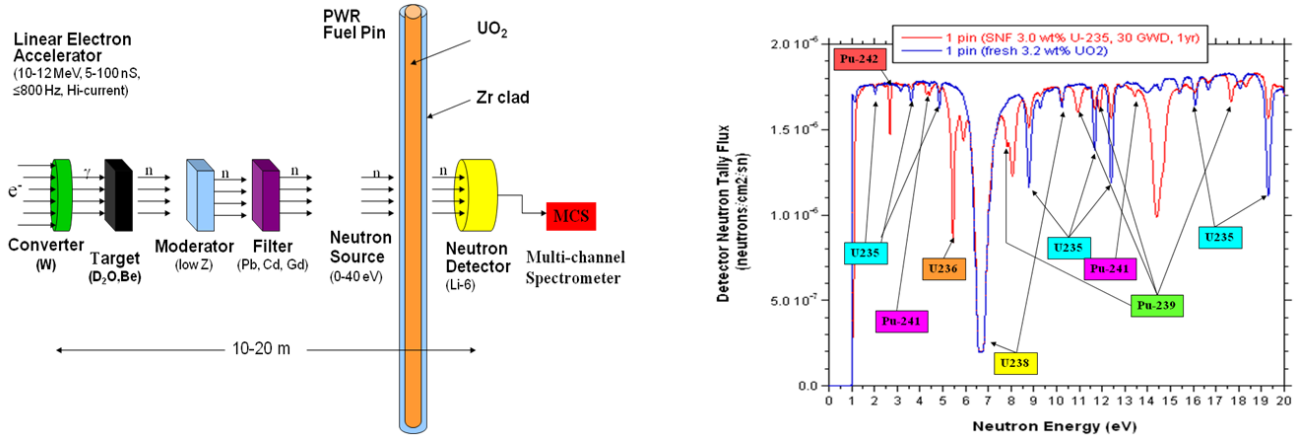

Fig. 7. Basic components of the NRTA technique (left). Comparison of calculated resonance transmission spectra for a single PWR fuel pin with fresh and spent UO_2 fuel (right).

The low-energy elastic scattering and absorption resonances of the major uranium and plutonium isotopes and some fission products modulate the transmitted neutron spectrum by creating resonance depressions in the transmitted signal. The resonance depressions can be analyzed to determine the uranium and plutonium content in the fuel. Actinide resonances in the 0-40 eV range are well-spaced and in sufficient number to allow actinide identification, concentration, and fuel burnup estimation. Figure 7 demonstrates two MCNP-calculated transmission spectra through a single PWR fuel pin; these spectra are very similar to an actual measured spent fuel spectrum.

8. Summary

Six NGSI non-destructive assay techniques considered in this paper offer an attractive capability to directly measure isotopic signatures of fissile materials in spent nuclear fuel. Instruments based on these principles can considerably enhance efforts of safeguarding Pu included in irradiated fuel assemblies. However, some of them are sensitive to the interrogation setup parameters and may require complex implementations to obtain the optimal response. In the complementary paper to this one, the capabilities of several neutron-based concepts were discussed. Several of the techniques rely on preliminary information of the fissile material mass ratios that can be obtained using methods discussed here. It is appropriate to expect that a combination of instruments can be effectively utilized for the spent fuel assay. The current research effort concentrates on advancement of the most promising techniques and integrated setups to the next stage: the full-scale proof-of-principle experiments.

9. Acknowledgments

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