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Integration of Self-Interrogation Neutron Resonance Densitometry and Differential Die-Away Self-Interrogation to Quantify Plutonium in a PWR 17x17 Spent Fuel Assembly

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ABSTRACT: The integration of Self-Interrogation Neutron Resonance Densitometry (SINRD) and Differential Die-away Self-Interrogation (DDSI) to more accurately quantify plutonium mass in a PWR 17x17 spent fuel assembly in water has been investigated via Monte Carlo N-Particle eXtended transport code simulations. Both of these instruments utilize the ^{244}Cm spontaneous fission neutrons to self-interrogate the fuel pins. The sensitivity of SINRD is based on using the same fissile materials in the fission chambers as are present in the fuel because the effect of resonance absorption lines in the transmitted flux is amplified by the corresponding (n,f) reaction peaks in the fission chamber. Thus, the ^{235}U and ^{239}Pu content in the spent fuel can be measured using ^{235}U and ^{239}Pu fission chambers placed adjacent to the assembly. DDSI uses time correlation neutron counting in a ^{3}He-based detector system, with triggering on the spontaneous fission events, to determine the spontaneous fission rate and the induced fission rate in the spent fuel. The time separation of counts from spontaneous fission (early gate) and induced fission (late gate) neutrons enables DDSI to independently measure the spontaneous fission mass and fissile mass in spent fuel. Based on the complementary characteristics of SINRD and DDSI, we believe that the integration of these instruments will provide a more robust verification method for spent fuel assemblies by improving the ability to measure plutonium. This research is part of the Next Generation Safeguards Initiative (NGSI) of the U.S. DOE and is being conducted in collaboration with several other national laboratories and universities throughout the U.S. Future work includes performing experimental measurements with both SINRD and DDSI on LWR fresh and spent fuel in water.

KEYWORDS: *spent fuel, plutonium, nuclear safeguards, non-destructive assay*

I. INTRODUCTION

Non-destructive assay (NDA) methods to directly measure the fissile content in spent fuel would improve the ability of the safeguards community to detect the diversion of significant quantities of fissile material. This NDA capability has the potential to improve safeguards for verification of commercial spent fuel assemblies by national authorities or the International Atomic Energy Agency (IAEA) and would improve deterrence of possible diversions by increasing the risk of early detection.[1] Effective NDA methods would also provide a capability to recover from the loss of continuity of knowledge in the event of a containment and surveillance (C/S) systems failure.[2] Furthermore, this assay capability would improve material accountability information at repository facilities prior to disposal or at reprocessing plants prior to fuel dissolution and thus increase operational efficiency, reduce material unaccounted for (MUF), and reduce shipper-receiver differences (SRD).[3]

The U.S. Department of Energy National Nuclear Security Administration established the Next Generation Safeguards Initiative (NGSI) to revitalize U.S. safeguards technology and human capital base.[4] In 2009, NGSI began funding a five-year research effort to develop and assess 14 potential NDA techniques for quantifying plutonium in commercial spent fuel. The first two years of this research effort was mainly focused on Monte Carlo modeling and the following three years will include the fabrication of hardware and measuring spent fuel assemblies.[5,6,7,8]

Two new NDA techniques called Differential Die-Away Self-Interrogation (DDSI) and Self-Interrogation Neutron Resonance Densitometry (SINRD) are currently being developed at Los Alamos National Laboratory (LANL) as part of this research effort. The main application of DDSI and SINRD is for use at a spent fuel storage facility for measurements in water, although both techniques could also be used for measurements at reprocessing facilities that have spent fuel pools. The focus of the work described in this paper was to investigate the viability of combining DDSI and SINRD to quantify the fissile content in a PWR 17x17 spent LEU fuel assembly via Monte Carlo N-Particle eXtended transport code (MCNPX)[9] simulations. For both instruments, the following characteristics were assessed:

(1) Dependence of measurement signatures on operator declared parameters such as initial enrichment (IE), burnup (BU), cooling time (CT)

(2) Expected uncertainty in results based on counting statistics for a count time of 30-minutes or less

(3) Sensitivity of measurement signatures to changes in the fissile mass in a spent LEU fuel assembly

II. DESCRIPTION OF SPENT FUEL SIMULATIONS

The use of DDSI and SINRD to quantify the fissile content in spent fuel was analyzed for a standard Westinghouse PWR 17x17 spent LEU fuel assembly in water (no boron). A spent fuel isotopic composition library of 64 different spent fuel assemblies was generated using MCNPX CINDER[10]. The main purpose for using this code was to calculate the spent fuel isotopic fractions as a function of fuel pin radius. This is needed in order to account for the large spatial gradient of actinide buildup across the fuel pellet. Four radial zones were used in each of the 264 fuel pins. In the NGSI spent fuel library[11], different combinations of the following IE, BU, CT, and surrounding medium parameters were used to characterize each assembly:

- **Initial Enrichment:** 2, 3, 4, and 5 wt% ^{235}U
- **Burnup:** 15, 30, 45 and 60 GWd/MTU
- **Cooling Time:** 1, 5, 20, and 80 yrs from discharge
- **Medium:** water, borated water (2200-ppm), and air

The response of DDSI and SINRD to each spent fuel assembly configuration was then simulated using MCNPX. Spontaneous fission neutrons from ^{244}Cm were used to self-interrogate the spent fuel pins in the MCNPX simulations of both instruments. It should be emphasized that these simulations were performed in water (no boron) with the CT fixed at 5-yrs. The specifications used to model the PWR spent fuel assembly are given in TABLE I.

TABLE I

Characteristics of PWR 17x17 spent LEU fuel assembly.

Assembly Data	
Lattice geometry	17 x 17 (square)
Assembly width	21.4 cm
Fuel pin pitch	1.26 cm
Number of fuel pins	264
Moderator	Light Water
Fuel Pin Data	
Fuel material	UO_2
Cladding material	Zircaloy-2
^{235}U initial enrichment	2% – 5% ^{235}U
Fuel pellet density	10.454 g/cm^3
Fuel pellet diameter	0.820 cm
Outer pin diameter	0.950 cm
Cladding Thickness	0.065 cm

II.A. Differential Die-Away Self-Interrogation (DDSI)

The DDSI technique is similar to traditional differential die-away analysis but it does not require an external pulsed neutron source. DDSI uses time-correlated neutron counting in a ^{3}He-based detector system, with triggering on the spontaneous fission events, to determine the fast and slow neutron distributions from the spontaneous and induced fission rate in the spent fuel.[12,13]

Therefore, the time separation of the spontaneous fission and fast fission neutrons with a fast die-away time from the thermal induced fission neutrons with a slow die-away time forms the basis of the DDSI technique. Fig. 1 shows the neutron capture distributions in the early and late gate windows used for DDSI. Region 1 is the exterior part of the instrument where neutrons are detected and Region 2 is the interior region where reflected neutrons interrogate the sample. The early gate window with fast die-away time is used to determine the spontaneous fission and fast induced fission rates. In the late gate window with slow die-away time, thermal induced fission neutrons are used to determine the fissile content in the sample.

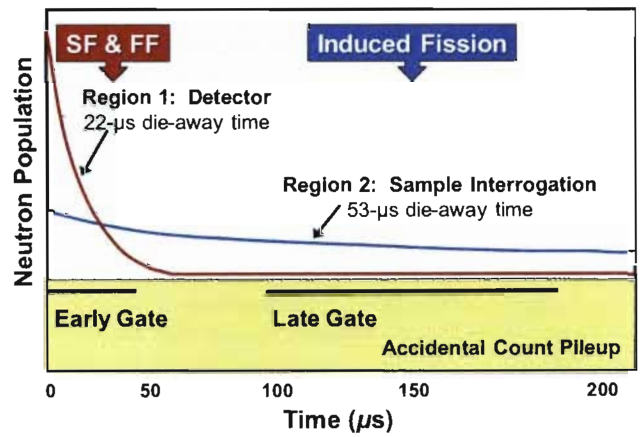

Fig. 1. Neutron capture distributions in the early and late time gate windows used for DDSI.

The design of the DDSI instrument was optimized using enhanced Particle Track Output (PTRAC) capture file capability in MCNPX.[14] Fig. 2 shows the optimized DDSI detector configuration modeled in MCNPX. **Region 1**, the detector region, consists of 58 ^{3}He tubes with 6-atm pressure and 40-cm active length. The ^{3}He tubes are arranged in two concentric rings and embedded in a 7-cm thick annulus of polyethylene (CH_2) surrounding the centrally located spent fuel assembly.

To balance the need for short die-away time for time separation purposes and a high efficiency for adequate neutron coincident counting statistics, 1-mm thick Cd fins were used. The Cd fins protrude from the 1-mm thick Cd liner surrounding the polyethylene annulus. This provides a single exponential neutron die-away time in the detector region of 22-μs. In addition, since the fins do not completely surround the ^{3}He tubes, a neutron can potentially be detected in one of several tubes resulting in a higher overall detector efficiency compared to surrounding each tube with Cd. For a ^{252}Cf point source located in the center of the DDSI instrument in air, the detector efficiency was estimated to be 13%. The detector region of DDSI is surrounded by iron (5-cm thick) in order to reflect fast neutrons back into the detector and shield gammas from adjacent assemblies in the spent fuel pool.

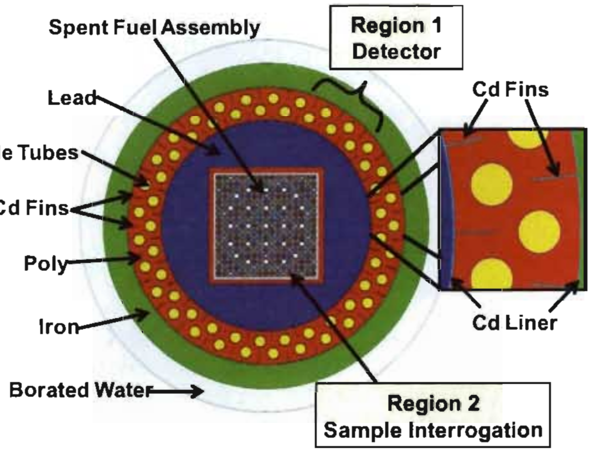

Fig. 2. Horizontal cross-section of the DDSI detector configuration for MCNPX simulations is shown.

Region 2, the sample interrogation region, consists of the spent fuel assembly surrounded by 1-cm thick polyethylene to increase the induced thermal fission rate within the assembly from reflected neutrons (e.g. thermal albedo). The thickness of this CH_2 layer was optimized to maximize the thermal neutron albedo without significantly reducing the detection efficiency of neutrons from the assembly. There is also a 1-mm thick removable Cd liner that can be inserted directly around the assembly for Passive Neutron Albedo Reactivity (PNAR) measurements, an alternate approach for quantifying fissile content that uses the same hardware as DDSI.[15] The Cd liner was not used in the DDSI simulations because no Cd liner increased the die-away time of the sample region to 53-µs. A longer die-away time is needed to maximize the time separation between spontaneous and induced fission count rates. In order to minimize gamma-ray pileup in the ^{3}He tubes, the spent fuel assembly is surrounded by 41-mm thick lead shield.

II.B. Self-Interrogation Neutron Resonance Densitometry (SINRD)

SINRD is another promising technique for the assay of fissile materials. The neutron resonance cross-section structure is unique for different fissile isotopes such as ^{235}U, ^{239}Pu, and ^{241}Pu. This resonance structure can provide a signature for the measurement of materials of importance to safeguards and non-proliferation. The sensitivity of SINRD is based on using the same fissile materials in the sample and fission chambers because the effect of resonance absorption in the transmitted flux is amplified by the corresponding *(n,f)* reaction peaks in the fission chamber (FC). Thus, the self-interrogation signature is inversely proportional to the amount of resonance absorption in the sample. For instance, a ^{235}U FC has a high sensitivity to neutron resonance absorption in ^{235}U present in the sample, and similarly for other fissile isotopes.[16,17,18]

The SINRD detector system was designed for measuring LWR fuel assemblies. Since the fuel assemblies considered in this research have square lattices, we designed the SINRD detector unit to be rectangular where the width of the unit is equal to the width of the assembly. SINRD consists of four FCs: Bare ^{235}U FC, boron carbide (B_4C) ^{235}U FC (located behind B_4C shield), Gd covered ^{235}U FC, and Cd covered ^{235}U FC. The SINRD detector unit (Fig. 3) is located adjacent to the assembly and is approximately 10.4-cm high, 9.0-cm long, and 21.4-cm wide. It should be noted that throughout the rest of this paper, we refer to the B_4C ^{235}U FC as FFM (or Fast Flux Monitor).[18]

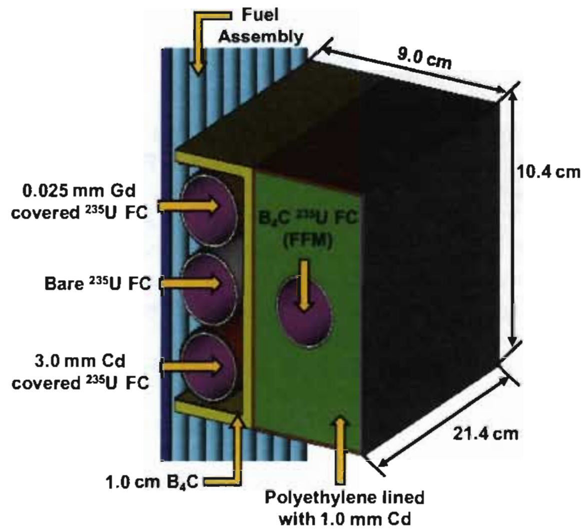

Fig. 3. SINRD detector configuration modeled in MCNPX.

The neutron flux entering the detector pod was measured using two FCs. The Bare ^{235}U FC is used to measure the gross neutron leakage from the fuel assembly dominated by thermal neutrons. The FFM measures the fast neutron flux from the assembly at energies above the B_4C absorption cutoff energy (3.8-keV). The Gd and Cd covered FCs are used to measure the resonance absorption from ^{235}U and ^{239}Pu in the spent fuel pins. The transmitted flux through Gd, Cd, and B_4C relative to ^{235}U and ^{239}Pu fission cross-sections is shown in Fig. 4.[18]

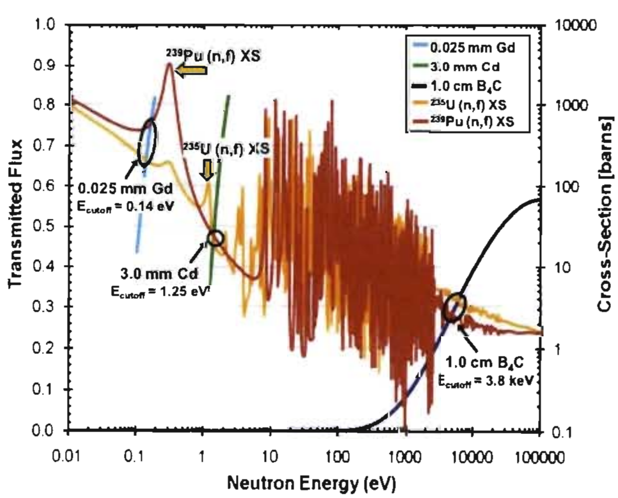

Fig. 4. Cut-off energies of absorber filters relative to ^{235}U and ^{239}Pu fission cross-sections.

Both Gd and Cd have large absorption cross-sections in the resonance energy region. The thickness of these absorber filters was chosen based on the desired absorption cutoff energy relative to the ^{235}U and ^{239}Pu fission cross-sections. We chose to use 0.025-mm thick Gd with cutoff energy of 0.13-eV and 3-mm thick Cd with cutoff energy of 1.25-eV. Based on the location of the Gd and Cd absorption cut-off energies shown in Fig. 4, we see that the thick Cd filter (3.0 mm) absorbs the majority of neutrons in the low energy region of the ^{235}U and ^{239}Pu fission resonances whereas the thin Gd filter (0.025 mm) transmits the majority of these lower energy neutrons.

The design of the SINRD instrument was optimized for quantifying the ^{235}U and ^{239}Pu content in a PWR spent LEU fuel assembly. We assumed a ^{235}U loading of 1.5-mg/cm^2 (93% ^{235}U metal) in the SINRD FCs using a 2-layer deposit thickness typical of standard commercial fission chambers.[19] It is important to note that only ^{235}U FCs were considered in this analysis. Previous work[20,21] has investigated the use of Gd and Cd ^{239}Pu FCs to quantify ^{235}U and ^{239}Pu in spent fuel. Using Gd and Cd ^{239}Pu FCs is advantageous because ^{239}Pu FCs have a much higher sensitivity to the mass of ^{239}Pu in spent fuel compared to ^{235}U FCs and the count rates are much higher which improves counting statistics. However, ^{239}Pu FCs are not commercially available and would have to be specially manufactured which could greatly increase the overall cost of SINRD. Thus, the IAEA may prefer to use all ^{235}U FCs for the actual implementation of SINRD. This densitometry method requires calibration with a reference assembly of similar geometry. However, SINRD uses the ratios of different FCs so most of the systematic errors related to calibration and positioning cancel in the ratios. In addition, SINRD can be calibrated with a fresh fuel assembly because it is not sensitive to neutron absorbing fission products.[18]

III. ANALYSIS OF RESULTS

The sensitivity of using DDSI and SINRD to quantify the fissile content in spent fuel was assessed via MCNPX simulations. The total neutron emission rate from a PWR 17x17 spent LEU fuel assembly changes by several orders of magnitude (10^4 to 10^9 n/s) depending on the burnup, initial enrichment, and cooling time of the assembly. This source term is further amplified by a factor of 2 – 3 by neutron multiplication in the assembly when in water. Thus, the use of ratios to quantify fissile content is very useful for spent fuel applications because it reduces the number of unknowns we are trying to measure since the neutron source strength and the detector-fuel assembly coupling cancels in the ratio. Using ratios also reduces the sensitivity of the measurements to extraneous material present in fuel (e.g. fission products). Since the signature ratios from SINRD and DDSI are complementary to one another, the results were integrated together to provide a more robust verification signature for spent fuel assemblies.

It is important to note that all of the results have been normalized to the response ratio for the 5% ^{235}U fresh LEU fuel assembly because in practice both DDSI and SINRD would be calibrated using a fresh fuel assembly. It should also be noted that the error bars shown on all results represent the calculated uncertainties in the ratios obtained via error propagations of expected counting statistics.

For DDSI, two viable ratios were established to quantify fissile content in the spent fuel assemblies:

(1) Late-to-early-gate doubles $(L/E)_D$ ratio

(2) Late gate doubles-to-singles D_L / S ratio

The enhanced PTRAC capability[14] in MCNPX was used to optimize the DDSI gate widths for the early gate (0 – 4μs) and late gate (72 – 164μs) doubles. For the purpose of integrating DDSI and SINRD, only the D_L / S results are discussed in this paper. In Fig. 5, the D_L / S ratio (a) is compared to the net multiplication and (b) versus total fissile fraction (^{235}U + ^{239}Pu + ^{241}Pu) in the PWR fuel assemblies with fixed CT of 5-yrs.

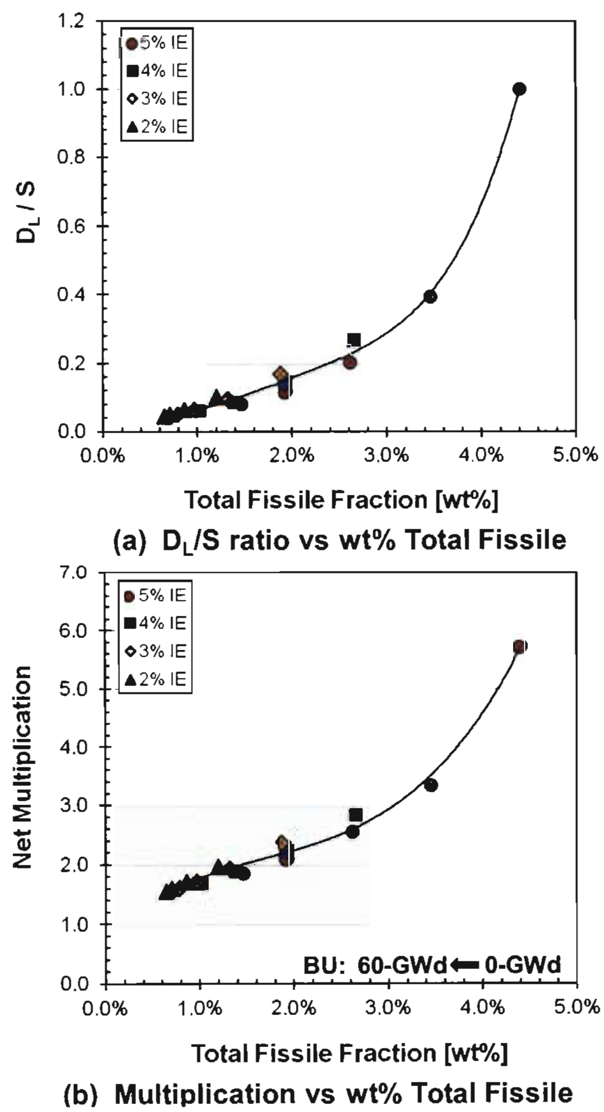

Fig. 5. Comparison of the (a) D_L / S ratio and (b) net multiplication versus total fissile fraction with no boron in water.

In the D_L / S ratio, the source strength cancels; however, the detector efficiency does not. It should also be noted that the D_L / S ratio has a second order dependence on the neutron leakage multiplication (net multiplication) from the spent fuel assembly.[22] Neutron multiplication, M, is defined as the ratio of the total number of neutrons produced from induced fission and spontaneous fission to the total number of neutrons produced from spontaneous fission. This term is a function of the system geometry and the composition of the fuel pins (e.g. ^{235}U, ^{239}Pu, and ^{149}Sm).[23]

Referring to Fig. 5(b), we see that the change in the net multiplication as a function of the total fissile fraction in the spent fuel has nearly the same structure shown in Fig. 5(a) for D_L / S. Thus, we can conclude that this ratio is strongly dependent on neutron multiplication in the assembly. This may be attributed to the fact that in over-moderated and highly multiplying mediums like water and borated water, the leakage multiplication is directly proportional to the total fissile content in the assembly. It follows that since the D_L / S ratio is strongly dependent on the net multiplication in the assembly, it is also proportional to the total fissile content in the assembly.

It is also important to note that the normalized D_L / S ratio [Fig. 5(a)] changed by 95% (1.0 to 0.045) over the range of fissile mass in the assemblies. Thus, we can conclude that this ratio has a high sensitivity to a broad range of total fissile mass. In addition, the D_L / S ratio is not a function of the initial fuel enrichment; specifically, a particular value for the D_L / S ratio does not correspond to more than one value for total fissile mass. This is significant because it means that our estimate of the total fissile mass in the spent fuel assembly does not require knowing the IE.

Similar to the DDSI results, we also established two viable ratios for SINRD to quantify ^{235}U and ^{239}Pu in the spent fuel assemblies:

(1) ^{235}U: (Gd – Cd) ^{235}U / Bare ^{235}U FC ratio

(2) ^{239}Pu: FFM / (Gd – Cd) ^{235}U FC ratio

In PWR spent LEU fuel, the main isotopes of significance to SINRD are ^{235}U, ^{239}Pu, ^{240}Pu, and ^{241}Pu. All of these isotopes have a large absorption resonance within the (Gd – Cd) cut-off energy window. The fission rate in Gd and Cd FCs is a function of the resonance absorption from all of these isotopes in spent fuel. Thus, the sensitivity of SINRD to a particular isotope is based on the magnitude of its cross-section within the (Gd – Cd) energy window and its concentration in spent fuel relative to the other isotopes.

The DDSI results described above showed that the D_L / S ratio can be used to quantify the total fissile mass in the spent fuel assemblies. However, additional information is required to determine the ^{235}U and total Pu mass separately and is thus the primary motivation behind the integration of SINRD and DDSI results. For SINRD, it should be emphasized that only ^{235}U FCs were considered in this analysis. Since ^{235}U FCs have a higher sensitivity to ^{235}U resonance absorption in the spent fuel pins compared to ^{239}Pu, only the results for the SINRD ratio used to quantify ^{235}U are discussed in this paper.

Fig. 6 shows the (Gd – Cd) ^{235}U / Bare ^{235}U FC ratio versus the ^{235}U fraction in PWR spent LEU fuel for varying IE and BU with no boron in the water. These results were normalized to the response ratio for the 5% ^{235}U fresh LEU fuel assembly. Compared to the results shown in Fig. 5(a) for the D_L / S ratio, the (Gd – Cd) / Bare ^{235}U FC ratio only changed by 27% (1.0 to 0.72) over the range of ^{235}U mass in the assemblies. However, it is important to note that the results for this SINRD ratio linearly track the ^{235}U content in LEU spent fuel independent of IE with a maximum relative uncertainty of 2%. Thus, we conclude that the (Gd – Cd) / Bare ^{235}U FC ratio can be used to quantify the ^{235}U content in a broad range of fuel assemblies. This is a very important result because it means that the integration of SINRD and DDSI measurements can be used to independently determine the mass of ^{235}U and fissile Pu (^{239}Pu + ^{241}Pu) in PWR spent LEU fuel assemblies. Furthermore, the ability to measure ^{235}U can also be used to verify the operator's declaration of BU and IE.

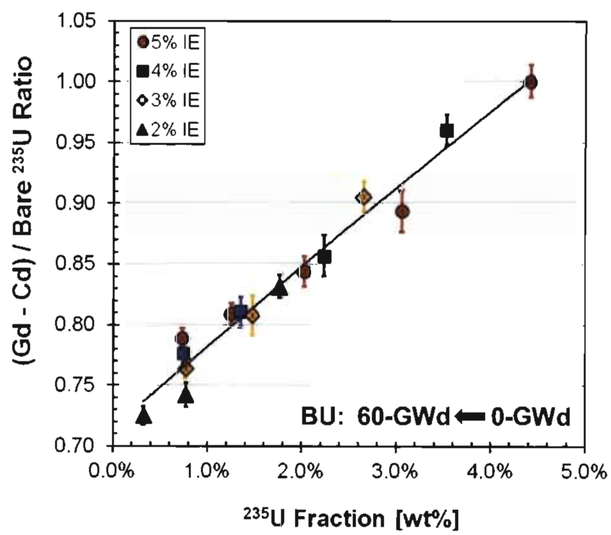

Fig. 6. (Gd – Cd) ^{235}U / Bare ^{235}U FC ratio versus the ^{235}U fraction in PWR spent LEU fuel.

IV. CONCLUSIONS

We have simulated the change in the response of both DDSI and SINRD instruments for different PWR 17x17 spent LEU fuel assemblies in water from the NGSI spent fuel library using MCNPX. The results from these MCNPX simulations were used to assess the viability of integrating SINRD and DDSI measurements to independently quantify the mass of ^{235}U and fissile Pu (^{239}Pu + ^{241}Pu) in the assemblies. It is important to note that both DDSI and SINRD require calibration with a reference assembly of similar geometry.

A summary of the final detector design and the optimized detector ratio used for the integration of DDSI and SINRD instruments is given below:

DDSI: Total Fissile ($^{235}U + ^{239}Pu + ^{241}Pu$) Measurements

- Final detector design
  - Early gate = (0 – 4μs), Late gate = (72 – 164μs)
  - Used 1-mm Cd fins in detector region to minimize die-away time (22μs) and maximize efficiency
- Optimized DDSI ratio
  - Late gate doubles-to-singles, D_L / S, ratio
  - 95% change in DDSI ratio (1.0 to 0.045)

SINRD: ^{235}U Measurements

- Final detector design
  - All ^{235}U FCs
  - 0.025-mm Gd, 3.0-mm Cd
- Optimized SINRD ratio
  - (Gd – Cd) / Bare ^{235}U FC ratio
  - 27% change in SINRD ratio (1.0 to 0.72)

The expected measurement uncertainties in the DDSI and SINRD ratios were calculated via error propagations of counting statistics. The relative uncertainties ranged from 0.31% to 1.5% for the D_L / S ratio and from 0.96% to 2.0% for the (Gd – Cd) / Bare ^{235}U FC ratio. It is also important to note that the integration of SINRD and DDSI measurements can also be used to verify the operator's declaration of BU and IE, a capability with safeguards relevance in its own right. In practice to independently quantify the mass of ^{235}U and total Pu in PWR spent fuel, it is first necessary to verify the burnup and initial enrichment of the assembly using a known reference assembly for calibration. Then, the optimized DDSI and SINRD ratios can be used in conjunction with burnup codes and MCNPX simulations to estimate the ^{235}U and total Pu content. Future work includes performing experimental measurements with both SINRD and DDSI on LWR spent fuel in water.

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