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Use of Self-Interrogation Neutron Resonance Densitometry to Measure the Fissile Content in a BWR 9x9 Spent Fuel Assembly

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Abstract:

We have investigated the use of Self-Interrogation Neutron Resonance Densitometry (SINRD) to measure the fissile content in a BWR 9x9 spent fuel assembly in water via Monte Carlo N-Particle eXtended transport code simulations. In addition, the sensitivity and penetrability of SINRD to the removal of fuel pins from an assembly was also assessed. The sensitivity of this technique 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 fission chamber. These simulations utilize the ^{244}Cm spontaneous fission neutrons to self-interrogate the fuel pins. The amount of resonance absorption of these neutrons in the fuel can be measured using ^{235}U and ^{239}Pu fission chambers placed adjacent to the assembly. Ratios of different fission chambers were used to reduce the sensitivity of the measurements to extraneous material present in fuel. SINRD requires calibration with a reference assembly of similar geometry. However, since this densitometry method uses ratios of different detectors, most systematic errors related to calibration and positioning cancel in the ratios. The development of SINRD to measure the fissile content in LWR spent fuel is important to the improvement of nuclear safeguards and material accountability. Future work includes performing experimental measurements with a prototype SINRD detector pod on both fresh and spent LWR fuel in water.

Keywords: spent fuel; nuclear safeguards; non-destructive assay; plutonium

1. Introduction

The development of non-destructive assay (NDA) capabilities to directly measure the fissile content in spent fuel is needed to improve the timely detection of the diversion of significant quantities of fissile material. This NDA capability is crucial to the implementation of effective international safeguards by the International Atomic Energy Agency (IAEA) and would improve deterrence of possible diversions by increasing the risk of early detection [1]. Currently, the IAEA does not have effective NDA methods to verify the spent fuel and recover continuity of knowledge in event of a containment and surveillance systems failure [2]. Furthermore, this assay capability would also improve material accountability information at reprocessing plants prior to fuel dissolution and thus increase operational efficiency and reduce material unaccounted for (MUF) [3].

The primary objective of this research is to develop and assess the sensitivity of using Self-Interrogation Neutron Resonance Densitometry (SINRD) for nuclear safeguards measurements. Recent interest in this approach was stimulated by an IAEA request related to spent fuel verification. Prior measurements [4,5,6] and calculations [7] have demonstrated that the SINRD method gives quantitative results for the fissile concentration in metal plates, MOX fuel rods, and a PWR 17x17 fresh fuel assembly [8].The main application of SINRD is for use at a spent fuel storage facility for measurements in water, although SINRD could also be used for measurements in different mediums, such as air or sodium and at reprocessing facilities that have spent fuel pools. The focus of the work described in this paper was to investigate the viability of using SINRD to verify a BWR 9x9 spent LEU fuel assembly (FA) via Monte Carlo N-Particle eXtended transport code (MCNPX) [9] simulations. The following capabilities of SINRD were assessed: 1) ability to measure the ^{235}U and ^{239}Pu content in BWR spent LEU fuel and 2) sensitivity and penetrability to the removal of fuel pins from an assembly. 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 chamber because the effect of resonance absorption in the transmitted flux is amplified by the corresponding (*n,f*) reaction peaks in the fission chamber. For instance, a ^{235}U fission chamber has a high sensitivity to the neutron resonance absorption in ^{235}U present in the sample, and similarly for other fissile isotopes. SINRD uses spontaneous fission neutrons from ^{244}Cm to self-interrogate the spent fuel pins. Thus, the self-interrogation signature is a result of having the same fissile material in the fission chamber and the sample [4,8,10].

In Figure 1, the ^{239}Pu fission cross-section is compared to the resonance absorption lines in the neutron flux after transmission through a 0.11-mm Gd filter and ^{239}Pu metal samples 0.25-mm and 2.5-mm thick. It is important to note that as the sample thickness increases, the self-interrogation signature decreases due to self-shielding effects from saturation of the large ^{239}Pu fission resonance at 0.3-eV [4]. The results shown for the transmitted flux through ^{239}Pu metal samples of different thicknesses were obtained from MCNPX simulations and the ^{239}Pu fission cross-section was obtained from the JANIS ENDF-VII cross-section database [11].

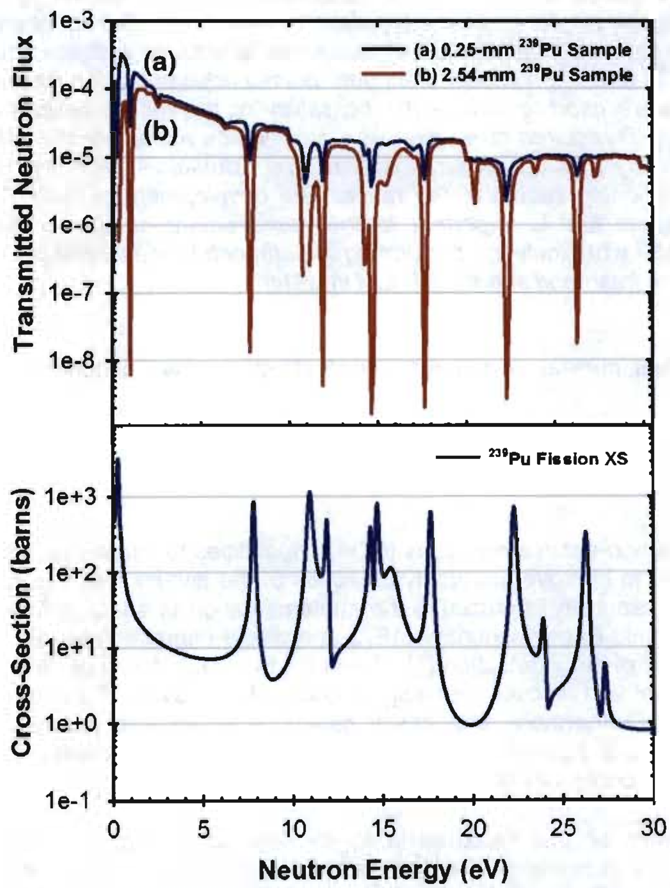


Figure 1. Comparison of absorption lines in neutron flux after transmission through Gd filter and (a) 0.25-mm and (b) 2.5-mm ^{239}Pu metal sample (upper plot) to ^{239}Pu fission cross-section (bottom plot).

2. Description of SINRD Measurement System

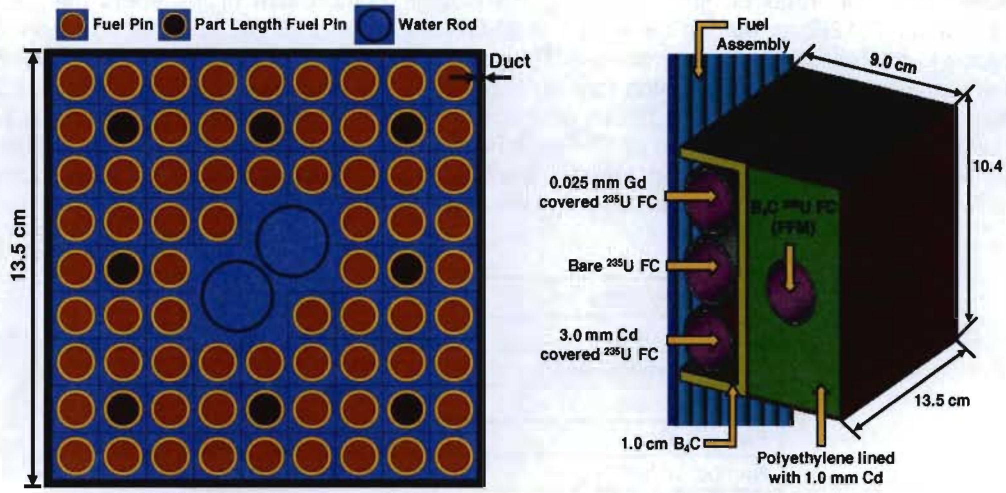
We have simulated the use of SINRD to quantify ^{235}U and ^{239}Pu in spent fuel and detect possible diversion scenarios for a BWR 9x9 spent LEU fuel assembly in water with 0%, 40%, and 70% void fractions (VF). This required first calculating the isotopic composition of the spent fuel assemblies using TransLAT [12] over burnup range of 0 to 50-GWd/MTU (in 10-GWd increments). Then, SINRD's response to each assembly was simulated. The concentration of ^{235}U and ^{239}Pu in the spent fuel pins was determined by tallying the fission rate in ^{235}U and ^{239}Pu fission chambers (FCs) located adjacent to the fuel assembly. Spontaneous fission neutrons from ^{244}Cm were used to self-interrogate the spent fuel pins in the MCNPX simulations of SINRD. It is important to note that the spent fuel isotopics were assumed to be homogeneously distributed in the fuel pins in the simulations. The specifications used to model a BWR 9x9 spent LEU fuel assembly are given in Table 1.

Assembly Data	
Lattice geometry	9 x 9 (square)
Assembly width (outer)	13.5 cm
Duct Thickness	0.25 cm
Fuel pin pitch	1.44 cm
Number of fuel pins	74 (8 Part-Length)
Inter-Assembly Gap	1.49 cm
Moderator	Light Water
Fuel Pin Data	
Fuel material	UO_2
Cladding material	Zircaloy-2
Initial ^{235}U Enrichment	3% and 4.5% ^{235}U
Fuel pellet density	10.01 g/cm^3
Fuel pellet diameter	0.975 cm
Outer pin diameter	1.118 cm
Cladding Thickness	0.071 cm
Active Fuel Height	371 cm
Partial Pin Fuel Height	244 cm

Table 1: Specifications for BWR 9x9 spent fuel assembly.

A top-down view of the BWR 9x9 fuel assembly (a) and the SINRD detector configuration (b) modeled in MCNPX are shown in Figure 2. SINRD consists of four FCs: Bare ^{235}U FC, boron carbide (B_4C) ^{235}U FC (located behind B_4C shield), 0.025-mm Gd covered ^{235}U FC, and 3.0-mm Cd covered ^{235}U FC. 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). The SINRD detector unit is approximately 10.4-cm high, 9.0-cm long, and 13.5-cm wide. In practice, SINRD would be located adjacent to the fuel assembly. To increase counting statistics, the FFM was embedded in polyethylene to thermalize the fast neutrons that penetrated the boron shielding. The polyethylene was covered with 1.0-mm of Cd to reduce the background from thermal neutrons reentering the SINRD unit. The neutron flux entering the detector pod was measured using two FCs. The Bare ^{235}U FC was used to monitor the entire neutron flux spectrum with thermal neutron domination, and the FFM was used to monitor the fast neutron flux above the B_4C absorption cutoff energy (3.8-keV). A fissile loading of 1.5-mg/cm^2 was modeled in the SINRD FCs using a 2-layer deposit thickness typical of standard commercial fission chambers. The ^{235}U FCs contained 93 wt% ^{235}U metal (19.1-g/cm^3) and the ^{239}Pu FCs contained 94 wt% ^{239}Pu metal (19.8-g/cm^3) [8].

Ratios of different fission chambers were used to reduce the sensitivity of the measurements to extraneous material present in fuel (e.g. fission products). This also reduces the number of unknowns we are trying measure because the neutron source strength and the detector-fuel assembly coupling cancels in the ratio. It is important to note that SINRD requires calibration with a reference assembly of similar geometry. However, since this densitometry method uses the ratios of different FCs, 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 in spent fuel [8,10].



(a) BWR 9x9 Fuel Assembly **(b) SINRD Detector Configuration**

Figure 2. (a) Top-down view of BWR 9x9 fuel assembly, (b) SINRD detector configuration modeled in MCNPX.

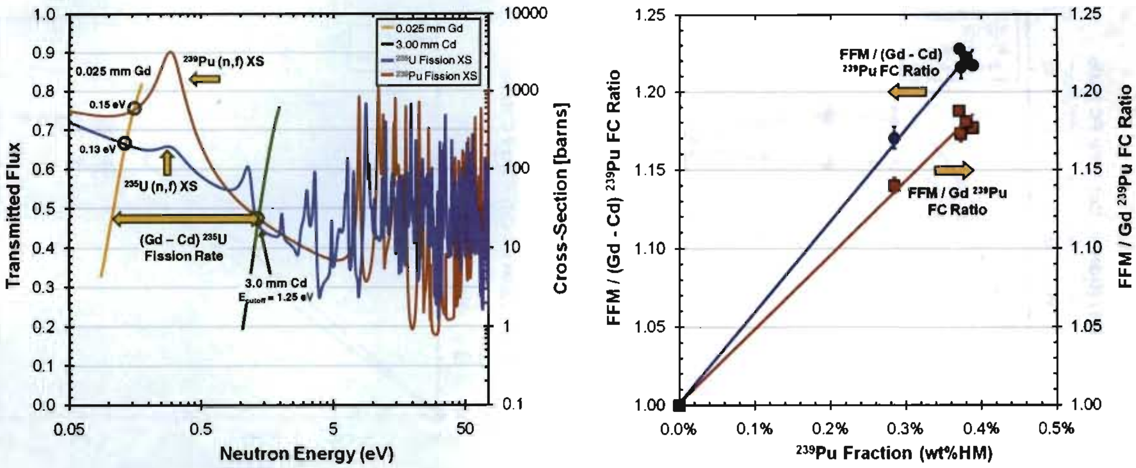
3. Analysis of SINRD for ^{239}Pu and ^{235}U Measurements

The SINRD detector configuration was optimized for quantifying ^{239}Pu and ^{235}U in a BWR 9x9 spent LEU fuel assembly with 0%, 40%, and 70% void fractions. To assess the sensitivity of SINRD to changes in the distribution of Pu isotopics in the spent fuel, we varied the initial enrichment (IE) from 3% to 4.5% ^{235}U. The cooling time was fixed at 5-yrs. The use of Gd and Cd ^{239}Pu FCs was investigated to determine how using ^{239}Pu FCs affects the sensitivity of the SINRD detector ratios to the ^{239}Pu content in spent fuel. This is important for LEU spent fuel because the ^{239}Pu and ^{235}U fractions are nearly equal at burnups greater than 30-GWd/MTU. However, it is important to note that ^{239}Pu FCs are not commercially available and would have to be specially manufactured. This could greatly increase the overall cost of SINRD. Therefore, we also investigated the use of all ^{235}U FCs in SINRD to quantify ^{239}Pu and ^{235}U in BWR spent LEU fuel.

3.1. SINRD Results for Quantifying ^{239}Pu

First, the use of different SINRD detector ratios were investigated for quantifying ^{239}Pu in BWR spent LEU fuel. Figure 3(a) shows how the large ^{239}Pu resonance at 0.3 eV can be windowed in energy by using the (Gd – Cd) ^{239}Pu fission rate based on the location of Gd and Cd absorption cut-off energies relative to the ^{239}Pu and ^{235}U fission cross-sections. The thick Cd filter (3-mm) absorbs the majority of neutrons in the low energy region of the ^{239}Pu resonance whereas the thin Gd filter (0.025-mm) transmits the majority of these lower energy neutrons.

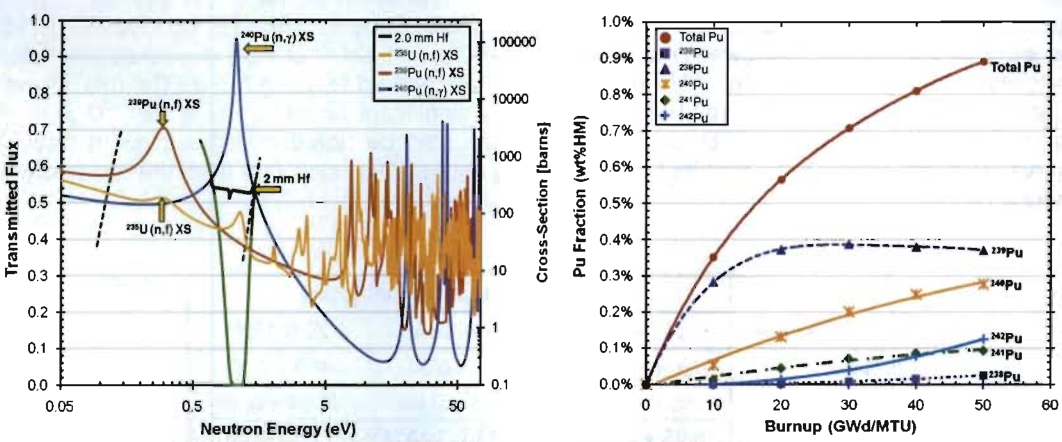
In Figure 3(b), the FFM / Gd ^{239}Pu FC ratio is compared to the FFM / (Gd – Cd) ^{239}Pu FC ratio versus ^{239}Pu fraction in 3% ^{235}U spent LEU fuel. These results were normalized to the fresh fuel case (3% IE). Using the (Gd – Cd) ^{239}Pu fission rate in the detector ratio, increased the slope of the SINRD signature by 18%. It is also important to note that the linearity of the curves shown in Figure 3(b) indicates that the SINRD ratio is accurately tracking the ^{239}Pu concentration in BWR spent LEU fuel. It should be noted that these results have been normalized to the fresh fuel case because in practice SINRD could be calibrated using a fresh fuel assembly.



(a) (Gd – Cd) energy window for ^{235}U & ^{239}Pu **(b) Gd vs (Gd – Cd) ^{239}Pu FC Ratio**

Figure 3. (a) ^{235}U and ^{239}Pu *(n,f)* cross-sections within (Gd – Cd) absorption cut-off energy window, (b) comparison of FFM / (Gd – Cd) to FFM / Gd ^{239}Pu FC ratios versus wt% ^{239}Pu.

To determine how the absorption of low energy neutrons by ^{240}Pu affects the SINRD FC ratio, a 2-mm Hf filter was added inside the Gd filter. The transmitted flux through a 2-mm Hf filter relative to the ^{240}Pu (n,γ) cross-section and buildup of Pu isotopics in BWR spent LEU fuel are shown in Figure 4(a) and (b), respectively.



(a) Transmitted flux through 2.0 mm Hf **(b) Plutonium Isotopics versus Burnup**

Figure 4. (a) transmitted flux through 2-mm Hf relative to ^{240}Pu *(n,γ)* cross-section, (b) buildup of Pu isotopics in BWR spent LEU fuel (No Void, 3% ^{235}U IE, 5-yrs cooled).

In Figure 5, the effect of using 2-mm Hf on FFM / (Gd – Cd) ^{239}Pu FC ratio is shown as a function of (a) burnup and (b) ^{239}Pu fraction. These results have been normalized to the fresh fuel case (3% IE). Adding 2-mm Hf to the Gd ^{239}Pu FC increased the slope of the SINRD signature by 6%. This is because the Hf filter absorbs the majority of neutrons in the same energy region as the ^{240}Pu (n,γ) resonance reducing the ^{240}Pu effect on the SINRD ratio.

Referring to Figure 5(a), it is important to note that the results for the SINRD ratio with 2-mm Hf closely follow the curve for the ^{239}Pu fraction in LEU spent fuel over the burnup range of 0 – 50-GWd. However, when no Hf is used the SINRD ratio continues to increase with burnup even though the ^{239}Pu fraction decreases for burnups >30-GWd. The purpose for plotting the (Gd+Hf – Cd) ^{239}Pu FC ratio results versus burnup in Figure 5(a) and ^{239}Pu fraction in (b) was to illustrate that similarity of the curves in (a) translates to linear curves in (b) when the SINRD ratio was plotted versus ^{239}Pu fraction.

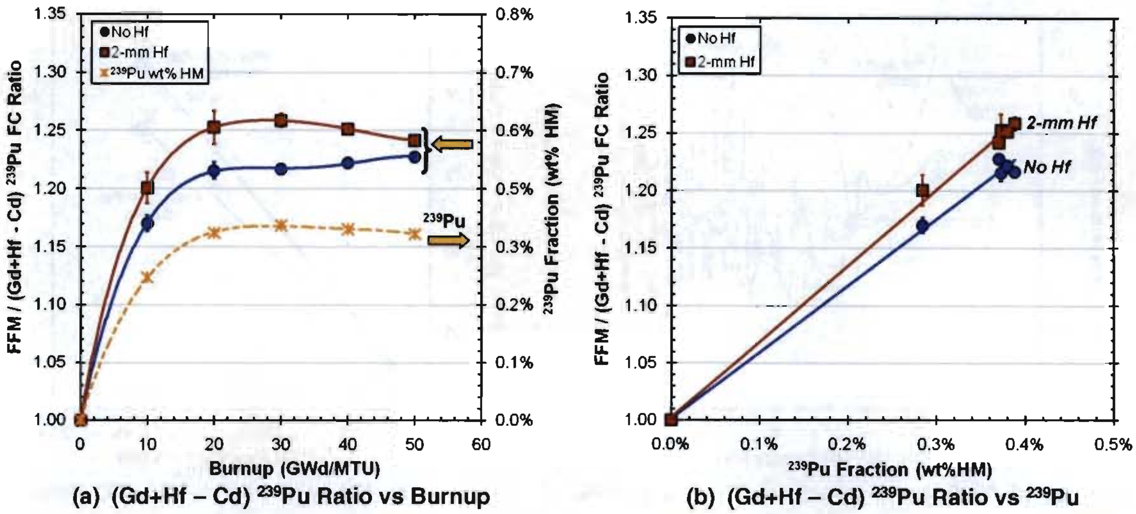


Figure 5. Optimized SINRD ratio for ^{239}Pu: FFM / (Gd+Hf – Cd) ^{239}Pu FC ratio versus (a) burnup and (b) ^{239}Pu wt%HM with no Hf and 2-mm Hf.

It is also important to note that the error bars shown on all results represent the calculated uncertainties in the SINRD ratios obtained via error propagations of expected counting statistics [7,9]. The expected count rates in the SINRD FCs are given in Table 2 for a BWR spent LEU fuel assembly with 40-GWd burnup (3% ^{235}U IE, 5-yrs cooled). The neutron source terms were 9.3E+07 n/s for 0% VF, 1.4E+08 n/s for 40% VF, and 1.9E+08 n/s for 70% VF. The use of Hf in the Gd ^{239}Pu and ^{235}U FCs reduced the count rate by 44% and 24%, respectively. The effect of using Gd and Cd ^{235}U FCs compared to ^{239}Pu FCs decreased the count rates in the Gd FC by 59% (no Hf) and Cd FC by 10%. Using error propagations, the lower count rates in the Gd and Cd ^{235}U FCs increased the relative uncertainty in the FFM / (Gd – Cd) ^{235}U FC ratio by 67% compared to using ^{239}Pu FCs. It is important to note that this increase in the relative uncertainty is significant because using all ^{235}U FCs also decreased the slope of the SINRD signature. It should also be noted that these count rates are conservative because the (α,n) contribution to the total neutron emission rate from the assembly was not accounted for.

SINRD Detectors	BWR Spent LEU Fuel [cps]		
	No Void	*40% Void*	*70% Void*
Bare ^{235}U	308 ± 0.29	415 ± 0.34	489 ± 0.37
FFM ^{235}U	896 ± 0.50	1451 ± 0.63	2280 ± 0.80
Gd ^{235}U	95 ± 0.16	145 ± 0.20	207 ± 0.24
Gd+Hf ^{235}U	72 ± 0.14	110 ± 0.17	158 ± 0.21
Cd ^{235}U	54 ± 0.12	86 ± 0.15	134 ± 0.19
Gd ^{239}Pu	241 ± 0.26	358 ± 0.32	479 ± 0.36
Gd+Hf ^{239}Pu	134 ± 0.19	199 ± 0.24	271 ± 0.27
Cd ^{239}Pu	60 ± 0.13	96 ± 0.16	148 ± 0.20

Table 2: Expected count rates in SINRD FCs for 40-GWd BWR spent LEU fuel.

In BWR spent LEU fuel, the relative concentrations of ^{235}U, ^{239}Pu, and ^{240}Pu change significantly for different void fractions. As a result, we have examined the effect of void fractions on our optimized SINRD ratio for measuring ^{239}Pu using ^{239}Pu FCs with 2-mm Hf and all ^{235}U FCs with no Hf. The results are shown in Figure 6 for (a) 0%, (b) 40% and (c) 70% void fractions in BWR spent LEU fuel with 3% ^{235}U IE. For no void fraction, the use of all ^{235}U FCs not only decreased the SINRD signature but the results no longer linearly track the ^{239}Pu fraction. This negative effect on our SINRD signature may be attributed to the fact that the concentration of ^{235}U relative to ^{239}Pu is large at low fuel burnups

(≤ 30-GWd) and nearly equal at high burnups. As a result, the competing effects from the burnup of ^{235}U and buildup ^{239}Pu are wiping out our signature. Thus, all ^{235}U FCs cannot be used to determine the ^{239}Pu content in BWR spent LEU fuel with no void fraction. Referring to Figure 6(b) and (c), the results for 40% and 70% void fractions clearly show that all ^{235}U FCs can be used to quantify ^{239}Pu in spent LEU fuel. The ability to use all ^{235}U FCs to measure ^{239}Pu in spent fuel with 40% and 70% void fractions may be attributed to the much larger amount of ^{239}Pu relative ^{235}U and that the ^{239}Pu content continues to increase over burnup range of 0 to 50-GWd.

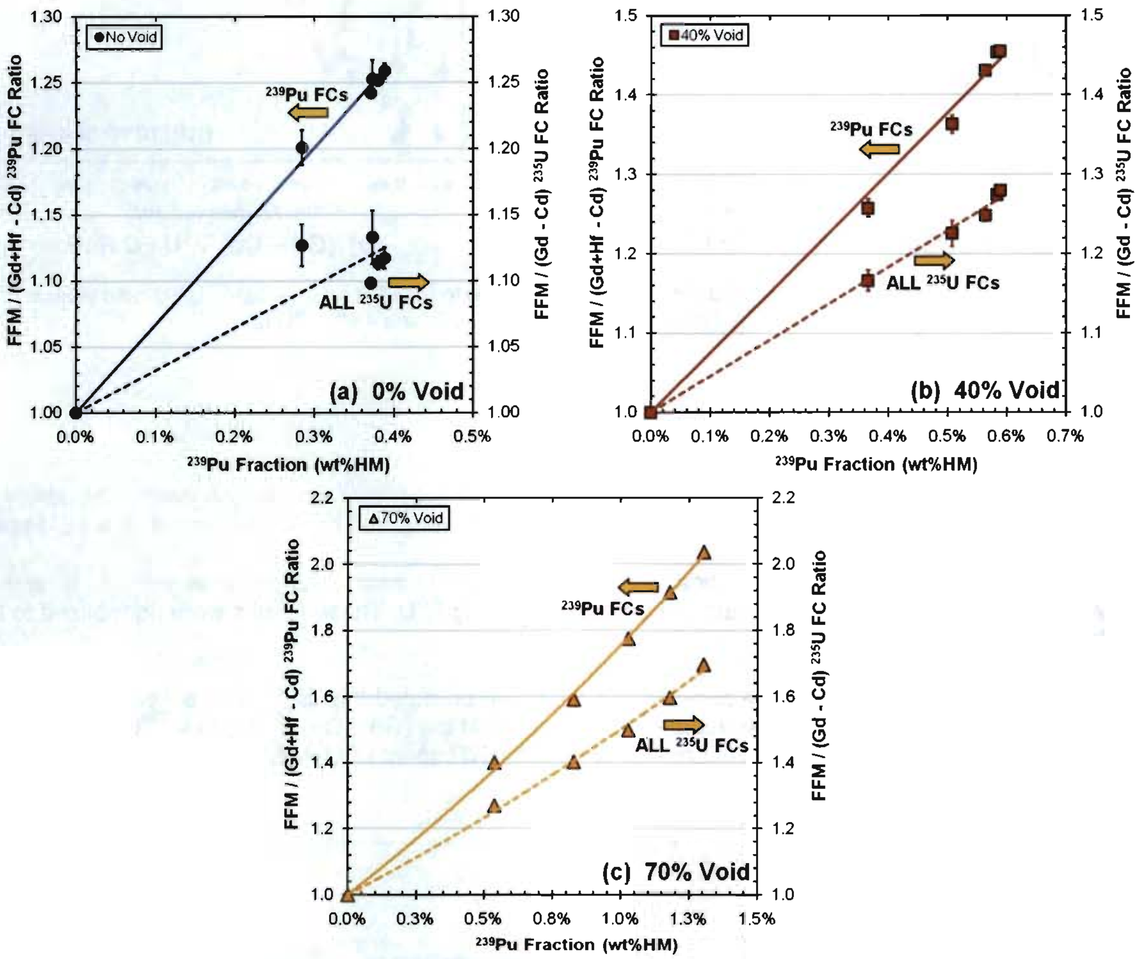


Figure 6. Effect of using all ^{235}U FCs on the FFM / (Gd+Hf – Cd) FC ratio versus ^{239}Pu fraction in BWR spent LEU fuel (3% IE) with (a) 0%, (b) 40% and (c) 70% void fractions.

The sensitivity of our SINRD detector ratio signature to initial enrichment was investigated using ^{239}Pu FCs and all ^{235}U FCs. The FFM / (Gd+Hf – Cd) ^{239}Pu FC ratio is compared to the FFM / (Gd – Cd) ^{235}U FC ratio versus ^{239}Pu fraction in Figure 7(a) and (b), respectively, for 3% and 4.5% ^{235}U IE. These results were not normalized to fresh fuel case. The maximum change in the SINRD ratio from varying the IE was 7.5% for the case with no void and all ^{235}U FCs; however, the sensitivity to IE decreases as the void fraction increases. Referring to Figure 7(b) for all ^{235}U FCs, the large scatter in the results for 0% and 40% void fractions confirms our conclusion that ^{239}Pu FCs are needed to accurately measure the ^{239}Pu content in BWR spent LEU fuel at low void fractions.

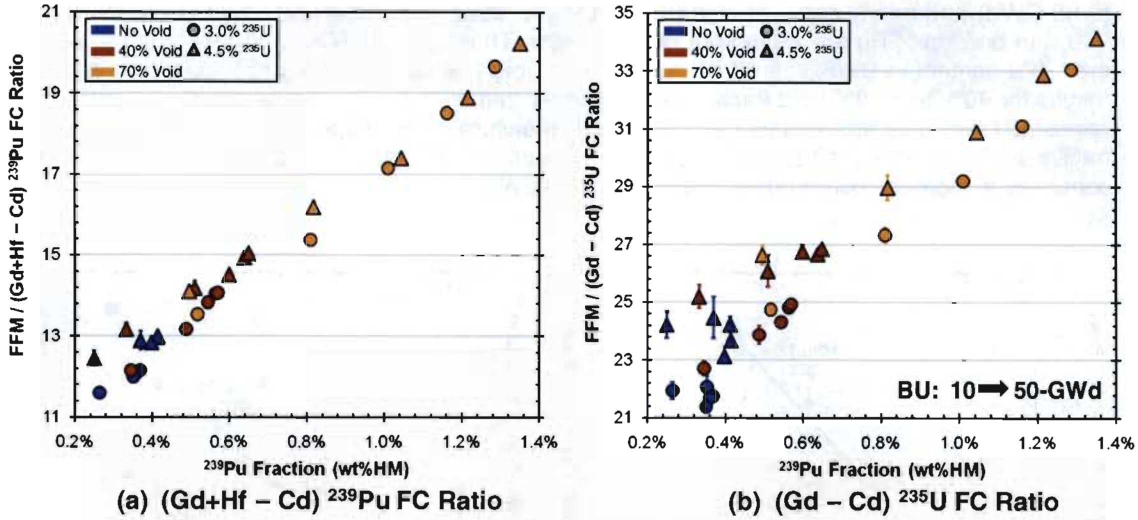


Figure 7. Comparison of (a) FFM / (Gd+Hf – Cd) ^{239}Pu FC ratio to (b) FFM / (Gd – Cd) ^{235}U FC ratio versus ^{239}Pu fraction for BWR spent LEU fuel with 3% and 4.5% ^{235}U IE.

3.2. SINRD Results for Quantifying ^{235}U

The use of SINRD to quantify ^{235}U in BWR spent LEU fuel was also investigated. The ability to measure ^{235}U using SINRD is important to verifying the burnup and initial enrichment of a LEU spent fuel assembly. In
Figure **8**, seven different SINRD ratios are shown versus ^{235}U fraction for the case with 3% IE and no void fraction to determine which ratio is best for quantifying ^{235}U. These results were normalized to the fresh fuel case. The ratios shown in
Figure **8**(a) have the FFM in the denominator and in
Figure **8**(b) the Bare FC is in the denominator. It should be noted that all ^{235}U FCs were used in all of the SINRD ratios. Based on these results, we can see that the (Gd – Cd) ^{235}U / Bare ^{235}U FC ratio [
Figure **8**(b)] is the only ratio that linearly tracks ^{235}U in BWR spent LEU fuel.

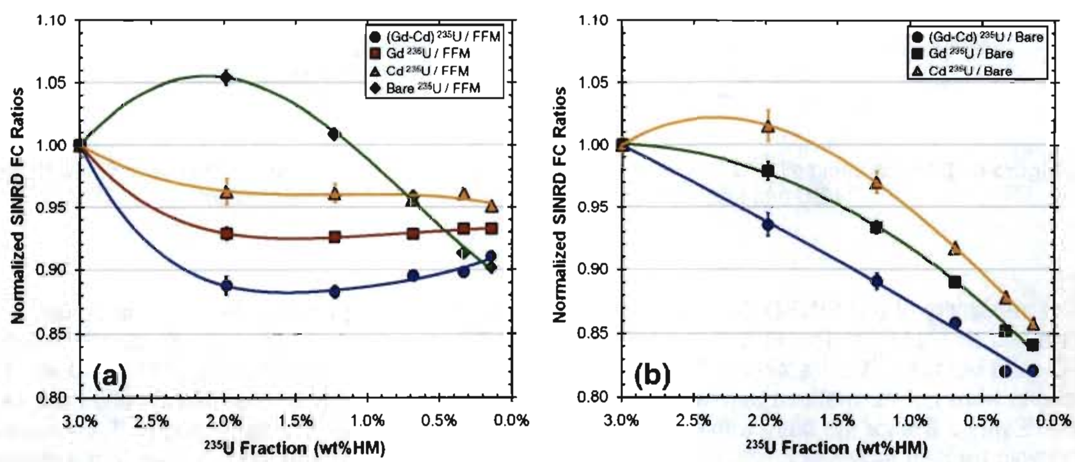


Figure 8. Comparison of different SINRD ratios versus ^{235}U fraction in BWR spent LEU fuel with 3% IE and no void fraction.

In order to determine if resonance absorption by ^{239}Pu within the (Gd – Cd) energy window is contributing to our SINRD signature, the (Gd – Cd) ^{235}U / Bare ^{235}U FC ratio is shown in Figure 9 versus (a) ^{235}U fraction and (b) ^{235}U + ^{239}Pu fraction. These results are shown with 0%, 40%, and 70%

void fractions to assess the sensitivity of this ratio different void fractions. It is important to note the change in the slope of the (Gd – Cd) ^{235}U / Bare ^{235}U FC ratio for different void fractions when plotted versus only ^{235}U compared to ^{235}U + ^{239}Pu in LEU spent fuel. These results show that the effect of ^{239}Pu on our SINRD ratio increases as the void fraction increases. This was expected because the concentration of ^{239}Pu in BWR spent LEU fuel increases by a factor of 3 from 0% to 70% void fraction. Thus, the ability to quantify ^{235}U decreases as the void fraction increases due to the competing effects of the burnup of ^{235}U and buildup of ^{239}Pu.

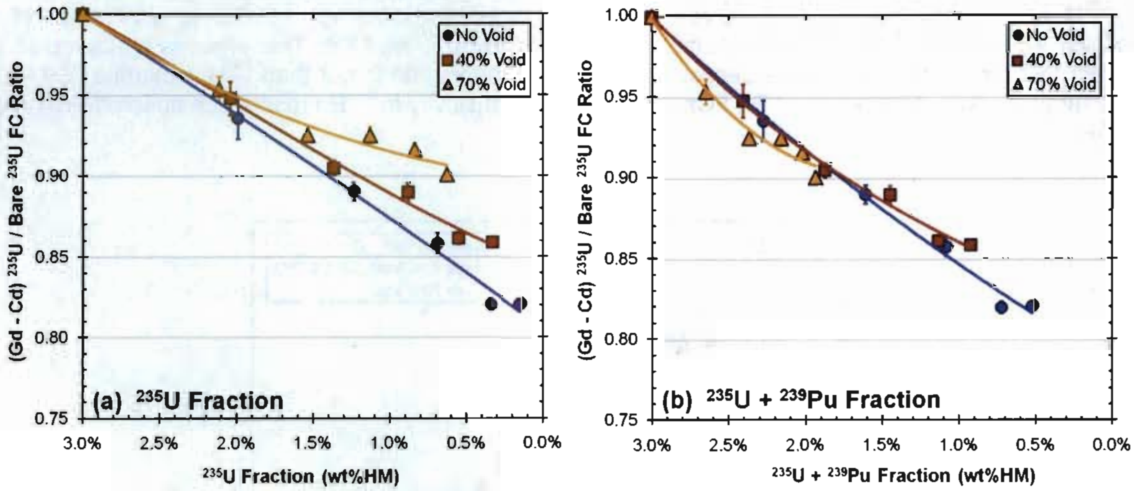


Figure 9. (Gd – Cd) ^{235}U / Bare ^{235}U FC ratio versus (a) ^{235}U fraction and (b) ^{235}U + ^{239}Pu fraction in BWR spent LEU fuel with 3% IE for different void fractions.

To obtain a better understanding of the physics of this SINRD ratio, Figure 10 shows the neutron flux multiplied by neutron energy, E·ϕ(E), at burnups of 10, 30, and 50-GWd relative to Gd and Cd cut-off energies for 3% IE BWR spent LEU fuel with (a) 0% VF and (b) 70% VF. Comparing the results shown in Figure 10, we see that the depression in the neutron flux within the (Gd – Cd) energy window (indicated by black arrow) increases as the burnup increases and is noticeably larger for the case with 70% VF. This depression in the flux is from ^{235}U and ^{239}Pu resonance absorption which increases with burnup due to the buildup of ^{239}Pu. The depression is larger for spent LEU fuel with 70% VF [Figure 10(b)] compared to (a) with 0% VF because the ^{239}Pu content is a factor of 3 greater. These results show that ^{239}Pu resonance absorption within the (Gd – Cd) energy window is contributing to our SINRD signature, especially at high void fractions, and thus should be accounted for.

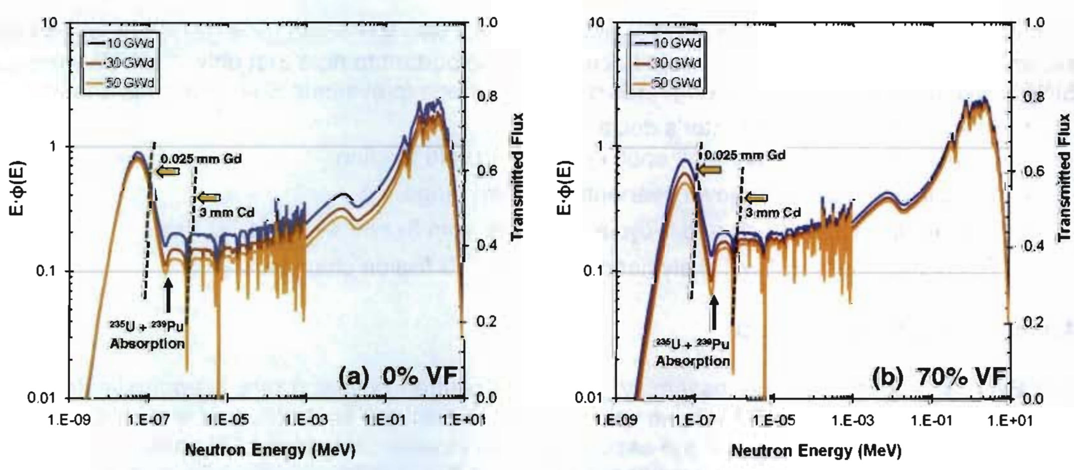


Figure 10. Comparison of E·ϕ(E) at burnups of 10, 30, and 50-GWd versus neutron energy relative to Gd and Cd cut-off energies for BWR spent LEU fuel with (a) 0% VF and (b) 70% VF.

The effect of varying the initial ^{235}U IE from 3% to 4.5% on the (Gd – Cd) ^{235}U / Bare ^{235}U FC ratio was also analyzed. The results are shown in Figure 11 versus ^{235}U + ^{239}Pu fraction in BWR spent LEU fuel for different void fractions. In contrast to previous results, these results were not normalized to the fresh fuel case. Varying the initial ^{235}U IE, changed the SINRD ratio by less than 5% over burnup range of 0 to 50-GWd/MTU and thus, is not sensitive to this parameter. For both 3% and 4.5% ^{235}U IE, the SINRD ratio linearly tracks the ^{235}U + ^{239}Pu content in BWR spent LEU fuel with 0%, 40%, and 70% void fractions. It should be noted that the slope of the SINRD FC ratio signature for determining ^{235}U + ^{239}Pu using all ^{235}U FCs decreased by a factor of ~9 for 0% VF, ~13 for 40% VF and ~16 for 70% VF compared to the slope for measuring ^{239}Pu using ^{239}Pu FCs. This effect is attributed to the fact that the ^{239}Pu fission cross-section is an order of magnitude larger than ^{235}U within the (Gd – Cd) energy window. As a result, ^{239}Pu FCs have a higher sensitivity to ^{239}Pu resonance absorption in spent fuel.

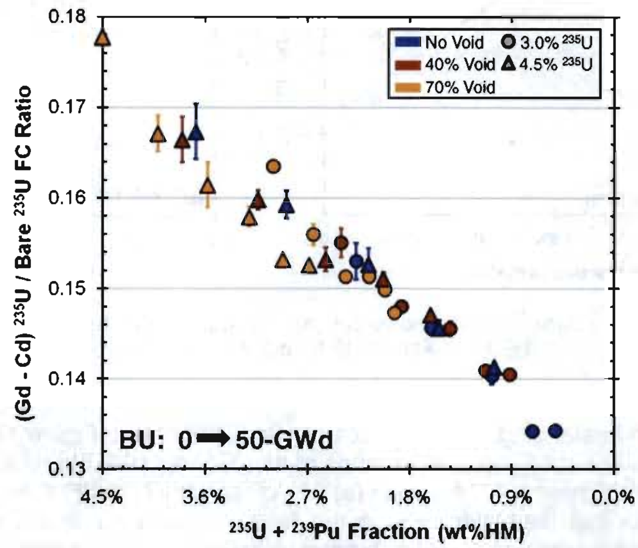


Figure 11. Comparison of (Gd – Cd) ^{235}U / Bare ^{235}U FC ratio versus ^{235}U + ^{239}Pu fraction in BWR spent LEU fuel with 3% and 4.5% ^{235}U IE.

4. Analysis of SINRD for Possible Diversion Scenarios

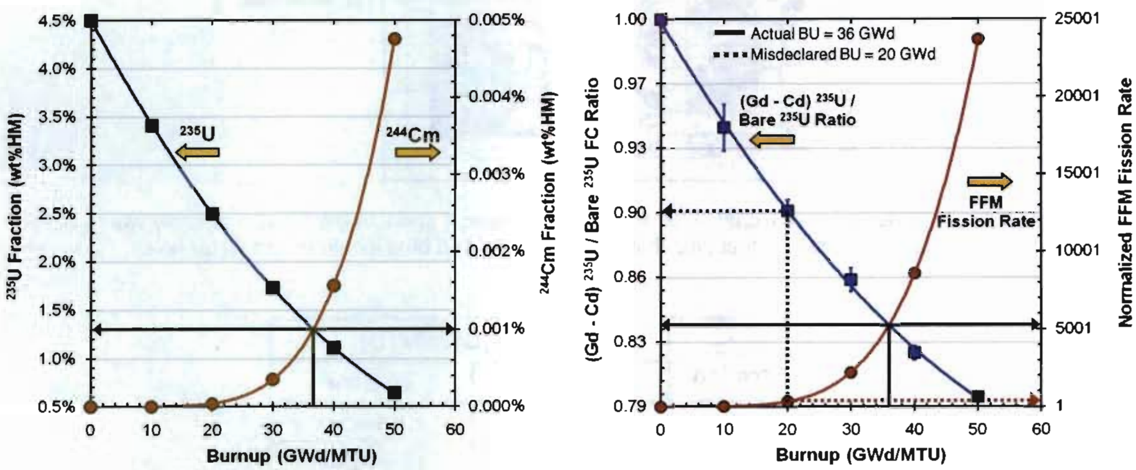
The sensitivity of SINRD to possible diversion scenarios was assessed for a BWR 9x9 spent LEU fuel assembly with 0%, 40%, and 70% void fractions. It is important to note that only ^{235}U FCs were used in SINRD. We used the following safeguards detection criteria to evaluate SINRD for this analysis:

- Independent of the Operator's declaration of:
  - burnup, initial enrichment, cooling time, and void fraction
- Sensitive to fuel pin removal over entire burnup range.
- Able to distinguish fresh and 1-cycle MOX fuel from 3- and 4-cycle LEU fuel.
- Recognize that IAEA will likely need to use all ^{235}U fission chambers.

4.1. Verification of Burnup

In a BWR 9x9 spent LEU fuel assembly, the ^{244}Cm neutron emission rate is approximately 9.3E+07 n/s for burnup of 40-GWd/MTU with no void fraction. This source term is further amplified by a factor of 2 – 3 by neutron multiplication in the assembly when in water. For spent LEU fuel, this high neutron source term provides adequate counting statistics in the fission chambers to give better than 1% precision in a few minutes for the SINRD ratios.

The use of SINRD to verify the burnup of a BWR spent LEU fuel assembly was investigated. In Figure 12, the ^{235}U and ^{244}Cm fractions are compared to the (Gd – Cd) ^{235}U / Bare ^{235}U FC ratio and FFM fission rate versus burnup for the diversion scenario where the burnup is misdeclared low. These results were normalized to the fresh fuel case with 4.5% IE. Since the ^{239}Pu content increases with burnup in LEU spent fuel, a proliferator is more likely to misdeclare the burnup low. Comparison of the results in Figure 12 (a) to (b), clearly shows that the FFM fission rate is directly proportional to ^{244}Cm and that the (Gd – Cd) ^{235}U / Bare ^{235}U FC ratio is proportional to ^{235}U in LEU spent fuel over the burnup range of 0 – 50-GWd/MTU.



(a) ^{235}U & ^{244}Cm Fraction **(b) Misdeclare Burnup LOW**

Figure 12. Comparison of (a) ^{235}U and ^{244}Cm fraction to (b) the (Gd – Cd) ^{235}U / Bare ^{235}U FC ratio and FFM fission rate versus burnup for diversion scenario where burnup is misdeclared low.

The fact that ^{235}U fraction decreases as a function of burnup, whereas the ^{244}Cm fraction increases enables us to verify the burnup of the BWR spent LEU assembly because the proliferator can only get one of these curves right. Referring to Figure 12(b), the solid black line indicates the actual burnup of the assembly (36-GWd) and the solid black arrows point to the expected measured values at this burnup. The misdeclared burnup (20-GWd) is shown by the black dotted line. The dotted red and blue lines correspond to the expected measured values for the misdeclared burnup. When the burnup is misdeclared, the expected measured values move in opposite directions. Thus, comparing a set of measurements where the burnup is misdeclared to a reference measurement with known burnup would clearly indicate an anomaly in the declaration.

4.2. Sensitivity of SINRD to Partial Defects

To assess the sensitivity and penetrability of SINRD, partial defects were modeled in a BWR 9x9 spent LEU fuel assembly with fuel burnups of 10 and 40-GWd/MTU. We uniformly removed 4 and 18 fuel pins (5% and 24% of the total pins, respectively) from two different radial regions of the assembly and replaced them with DU pins. The initial fuel enrichment was fixed at 3% ^{235}U for this analysis. The fuel pin removal locations of partial defects for Regions 1 and 2 are shown in Figure 13. Region 1 consists of the second row from the outer surface of assembly and Region 2 consists of rows in the center of the assembly. The average depth from the outer surface is 2.16-cm for Region 1 and 5.75-cm for Region 2.

To assess the penetrability of SINRD to partial defects, the percent change in the SINRD ratios was calculated for each region to determine if the diverted pins can be detected within 3σ uncertainty. The count times used for the diversion cases are given in Table 3. These count times are conservative because they do not account for the contribution to the neutron emission rate from (α,n) neutrons.

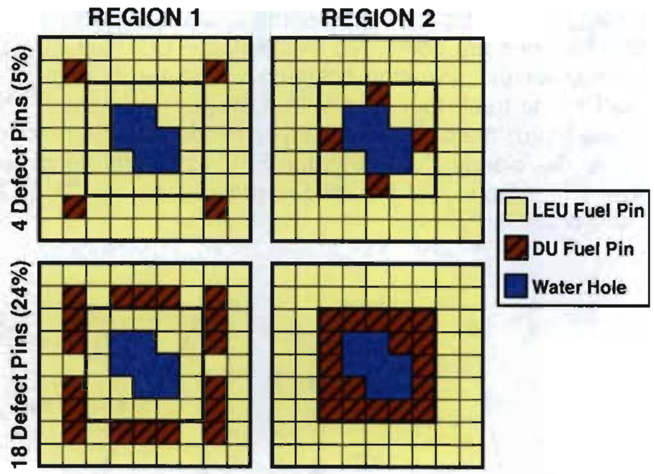


Figure 13. Fuel pin removal locations of defects for Regions 1 and 2 in BWR 9x9 assembly where red pin locations represent fuel pins that were removed and blue locations are water holes.

Void Fraction	**Burnup [GWd/MTU]**	
	10-GWd	***40-GWd***
No Void	5 hours	20 minutes
40% Void	4.5 hours	10 minutes
70% Void	4.5 hours	10 minutes

Table 3: Count times used to detect pin diversions within 3σ uncertainty for BWR spent fuel.

The sensitivity of different SINRD ratios with 5% and 24% of the total number of pins removed from Regions 1 and 2 are given in Table 4 for BWR spent LEU and MOX fuel, respectively. The highlighted values correspond to the maximum positive and negative percent change in ratios that are within 3σ uncertainty for 5% and 24% pins removed from each region. The cells that are shaded gray correspond to the percent in change detector ratios that are not within 3σ uncertainty of a spent fuel assembly with no diverted pins. It should be emphasized that all ^{235}U FCs were used to obtain these results where no Hf was used for spent LEU fuel and 1-mm Hf was used for spent MOX fuel. Error propagations (Appendix A) were used to calculate the uncertainties in the percent change in the SINRD ratios for all diversion cases. These uncertainties were between 0.2% – 1% for the FFM / Bare ^{235}U FC ratio using the count times given in Table 3. Thus, this type of measurement could show the departure from a reference fuel assembly with no defects.

It is important to note that for a BWR spent LEU fuel assembly with burnup of 10-GWd none of the SINRD ratios can detect 5% pin diversions within 3σ in Region 2. If the count time was increased to 40-hrs for 0% void, 12-hrs for 40% void, and 25-hrs for 70%, then only the FFM / Bare ^{235}U FC ratio could detect 5% pin diversions within 3σ in Region 2. A summary of the results shown in Table 4 is given below:

- All SINRD ratios have the highest sensitivity to pin removal in Region 1.
- For BWR spent LEU fuel, the FFM / Bare ^{235}U FC ratio is the most sensitive SINRD ratio for detecting fuel pin diversions within 3σ from Regions 1 and 2.
  - This ratio is sensitive to reactivity changes in the fuel assembly due to changes in the concentration of thermal absorbers.
  - The percent change in this ratio is positive for pin removal from Regions 1 and 2.

Region Defects	Burnup	SINRD Ratios *BWR Spent LEU*	REGION 1 0% Void	REGION 1 40% Void	REGION 1 70% Void	REGION 2 0% Void	REGION 2 40% Void	REGION 2 70% Void
5% Pin Defects (4 pins)	***10 GWd***	FFM / (Gd - Cd) ^{235}U	*2.13%*	*2.35%*	*3.21%*	*-0.59%*	*0.70%*	*-0.16%*
		FFM / Bare ^{235}U	4.49%	4.85%	4.67%	*1.07%*	1.53%	0.75%
		Bare ^{235}U / Gd ^{235}U	*-3.51%*	*-3.56%*	-3.21%	*-1.81%*	*-1.86%*	*-1.14%*
		Bare ^{235}U / Cd ^{235}U	*-4.32%*	*-4.25%*	-4.38%	*-1.91%*	*-2.62%*	*-1.30%*
	40 GWd	FFM / (Gd - Cd) ^{235}U	2.87%	5.20%	4.13%	*0.92%*	*1.71%*	*1.10%*
		FFM / Bare ^{235}U	3.76%	4.60%	4.84%	0.86%	1.24%	0.97%
		Bare ^{235}U / Gd ^{235}U	-2.38%	-2.50%	-3.01%	-1.13%	-1.35%	-1.35%
		Bare ^{235}U / Cd ^{235}U	-3.54%	-4.73%	-4.29%	-2.07%	-2.62%	-2.17%
24% Pin Defects (18 pins)	***10 GWd***	FFM / (Gd - Cd) ^{235}U	*9.03%*	*9.10%*	11.3%	*-1.60%*	*-1.28%*	*0.72%*
		FFM / Bare ^{235}U	15.6%	14.3%	14.7%	4.58%	5.01%	5.33%
		Bare ^{235}U / Gd ^{235}U	-13.8%	-11.8%	-11.5%	-8.84%	-9.09%	-8.77%
		Bare ^{235}U / Cd ^{235}U	-18.8%	-16.4%	-17.1%	-10.7%	-11.0%	-11.6%
	40 GWd	FFM / (Gd - Cd) ^{235}U	9.03%	11.5%	12.7%	*-0.30%*	*1.68%*	*0.83%*
		FFM / Bare ^{235}U	12.9%	14.8%	16.3%	4.21%	5.67%	6.73%
		Bare ^{235}U / Gd ^{235}U	-10.1%	-11.5%	-12.9%	-7.66%	-8.52%	-10.0%
		Bare ^{235}U / Cd ^{235}U	-14.9%	-17.4%	-18.2%	-10.1%	-11.6%	-12.1%

Table 4: Percent change in SINRD ratios with 5% and 24% fuel pins removed from Regions 1 and 2 for BWR spent LEU fuel (No Hf).

In Figure 14, the fuel pin removal results for FFM / Bare ^{235}U FC ratio as a function diversion case are shown for BWR spent LEU fuel with no void fraction and burnup of (a) 10-GWd and (b) 40-GWd. The solid line represents the signal from the case with no diversions; the dashed lines represent ±1% change in the SINRD ratio to account for systematic errors. We chose to use the FFM / Bare ^{235}U FC ratio in this analysis because it was the most sensitive ratio for detecting fuel pin diversions within 3σ from Regions 1 and 2. These results show that the SINRD ratio has the highest sensitivity to fuel pin diversions from Region 1. The diversion of 4 pins (5% of total number of pins) from Region 2 for both 10 and 40-GWd/MTU are the only cases that are not clearly within ±1% of the no diversion signal.

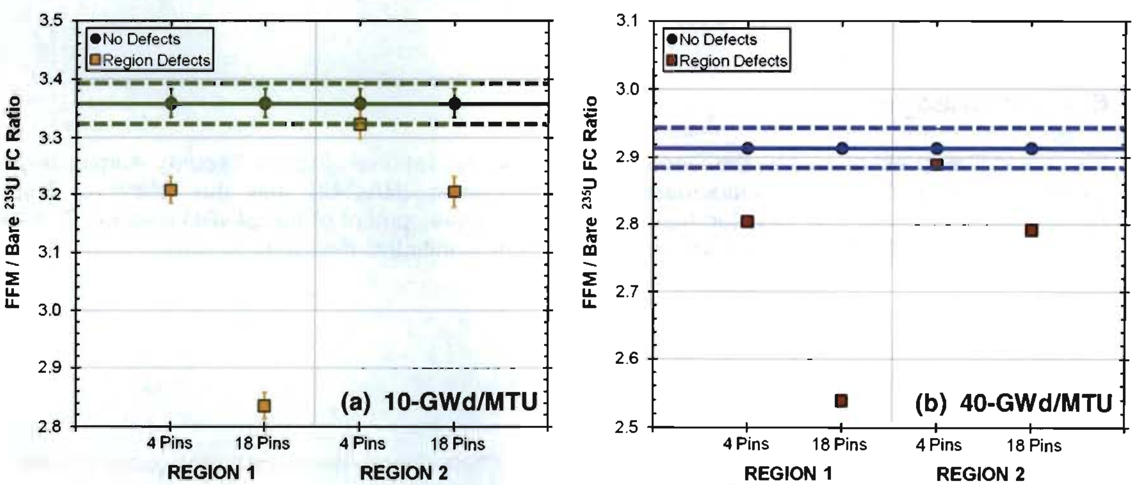


Figure 14. Pin removal results for FFM / Bare ^{235}U FC ratio as a function diversion case for BWR spent LEU fuel with burnup of (a) 10-GWd and (b) 40-GWd (no void fraction).

5. Conclusions

We have simulated the change in different SINRD detector ratios over a burnup range of 0 – 50 GWd using MCNPX. For a BWR spent LEU fuel assembly with void fractions of 0%, 40%, and 70%, the FFM / (Gd+Hf – Cd) ^{239}Pu FC ratio was optimized for determining ^{239}Pu using 2-mm Hf. This SINRD ratio is proportional to the ^{239}Pu mass in the assembly over the burnup range of 0 to 50-GWd. Due to the fact that the IAEA will likely need all ^{235}U FCs, the use of the FFM / (Gd – Cd) ^{235}U FC ratio to determine ^{239}Pu was also investigated. All ^{235}U FCs cannot be used to quantify the ^{239}Pu content in BWR spent LEU fuel with 0% void fraction but can be used for 40% and 70% void fractions. The ability to use all ^{235}U FCs to quantify ^{239}Pu in spent fuel with 40% and 70% void fractions may be attributed to the much larger amount of ^{239}Pu relative ^{235}U and that the ^{239}Pu content continues to increase over burnup range of 0 to 50-GWd.

The sensitivity and penetrability of SINRD was assessed by modeling partial defects in a BWR 9x9 spent LEU fuel assembly. It is important to note that all ^{235}U FCs were used in this analysis. The percent change in the SINRD ratios was calculated for Regions 1 and 2 to determine if the diverted pins can be detected within 3σ. It should be noted that for a BWR spent LEU fuel assembly with burnup of 10-GWd none of the SINRD ratios can detect 5% pin diversions within 3σ in Region 2. Based on the results from these calculations, the FFM / Bare ^{235}U FC ratio is the best ratio for detecting pin diversions from a BWR spent LEU fuel assembly. This is because the FFM / Bare ^{235}U FC ratio has the lowest uncertainty of all the SINRD ratios which is important for spent LEU fuel because neutron source term is very low at low burnups (<20-GWd). These uncertainties were between 0.2% – 1% for the FFM / Bare ^{235}U FC ratio. Thus, this type of measurement could show the departure from a reference fuel assembly with no defects.

The purpose of the BWR spent fuel simulations was to assess the ability of SINRD to measure the fissile content in spent fuel and the sensitivity and penetrability of SINRD to partial defects in an assembly. Based on the results from these simulations, we have concluded that SINRD provides a number of improvements over current IAEA verification methods. These improvements include:

1) SINRD provides absolute measurements of burnup independent of the operator's declaration.
2) SINRD is sensitive to pin removal over the entire burnup range and can verify the diversion of 5% of fuel pins within 3σ from BWR spent LEU and MOX fuel.
3) The calibration of SINRD at one reactor facility carries over to reactor sites in different countries because it uses the ratio of FCs that are not facility dependent.
4) SINRD can distinguish fresh and 1-cycle spent MOX fuel from 3- and 4-cycles spent LEU fuel without using reactor burnup codes.

6. Acknowledgements

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