LA-UR-08-03798

# USE OF SELF-INTERROGATION NEUTRON RESONANCE DENSITOMETRY (SINRD) TO MEASURE THE FISSILE CONTENT IN NUCLEAR FUEL

A.M. LaFleur[1], W.S. Charlton[1], H.O. Menlove[2], and M. Swinhoe[2]

[1]Nuclear Security Science and Policy Institute, 3133 TAMU, College Station, TX 77843
[2]Los Alamos National Laboratory, Los Alamos, NM 87545
amlafleur@tamu.edu; wcharlton@tamu.edu; hmenlove@lanl.gov; swinhoe@lanl.gov;

## ABSTRACT

We investigated a new method to verify the concentration of fissile material in a PWR fresh fuel assembly using Self-Interrogation Neutron Resonance Densitometry (SINRD). Monte Carlo N-Particle eXtended transport code (MCNPX) was used to simulate these measurements. The sensitivity of SINRD is based on using the same fissile materials in the fission chambers as are under investigation because the effect of resonance absorption lines in the transmitted flux are amplified by the corresponding (*n,f*) reaction peaks in fission chamber. This simulation utilizes the $^{238}$U 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 fission chambers placed adjacent to the assembly. We used ratios of different fission chambers to reduce the sensitivity of the measurements to extraneous material present in fuel. The development of SINRD to measure the fissile content in spent fuel is of great importance to nuclear safeguards and accountability. Future work includes the use of this technique to measure the concentration of fissile materials in spent fuel and pyroprocessing heavy metal product.

## INTRODUCTION

The development of nondestructive assay (NDA) capabilities to measure the fissile content in nuclear fuels is crucial to the implementation of effective international safeguards. The use of self-interrogation neutron resonance densitometry (SINRD) for the assay of fissile materials is a promising technique for nuclear safeguards and material accountability measurements. The neutron resonance cross-section structure is unique for each of the fissionable isotopes such as $^{235}$U, $^{233}$U, $^{239}$Pu, and $^{241}$Pu, and the resonance structure can provide a signature for the measurement of these materials of importance for safeguards and non-proliferation. The sensitivity of this technique is based on using the same fissile materials in the sample and fission chamber 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, a $^{235}$U fission chamber has high sensitivity to the neutron resonance absorption in $^{235}$U that is in the sample, and similarly for the other fissile isotopes. The self-interrogation signature is a result of having the same fissile material in the fission chamber as in the sample.[1]

The comparison of the $^{235}$U fission cross-section to the absorption lines in the neutron flux after transmission through a Gd filter and 0.25-mm [curve (a)] and 1.5-mm [curve (b)] $^{235}$U metal sample is shown in Fig. 1. It is important to note that as the sample thickness increases, the self-

interrogation signature decreases due to self-shielding effects occurring from saturation of the larger resonances.[2]

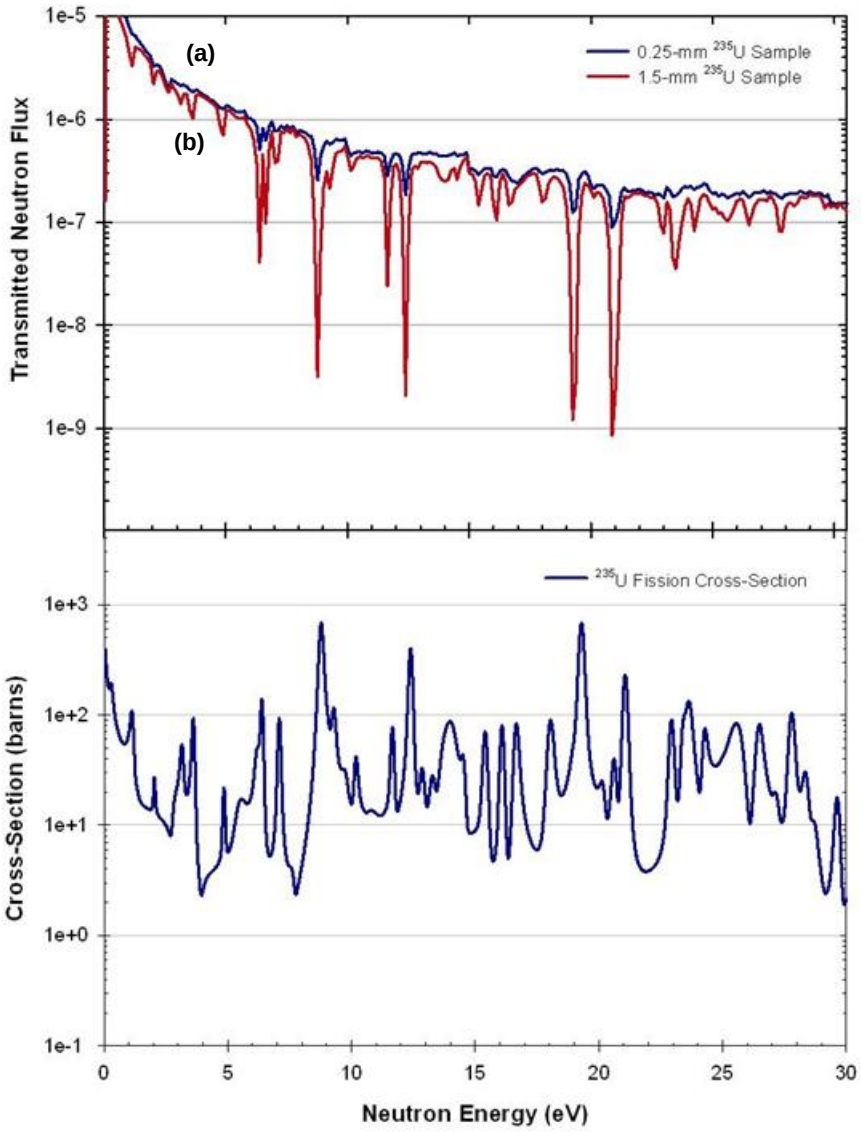


**Figure 1.** Comparison of the absorption lines in the neutron flux after transmission through a 0.114-mm Gd filter and 0.25-mm [curve (a)] and 1.5-mm [curve (b)] $^{235}$U metal sample (upper plot) to the $^{235}$U fission cross-section at neutron energies ≤ 30 eV (bottom plot).[2]

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[3,4] and calculations[1] have demonstrated that the SINRD method gives quantitative results for the fissile concentration in metal plates and in MOX fuel rods. The work described in this paper is focused on investigating the use of SINRD to measure the total $^{235}$U content in a PWR fresh fuel assembly via Monte Carlo N-Particle eXtended transport code (MCNPX)[5] simulations. The results from these simulations were used to optimize the detector configuration and obtain a better understanding of the underlying physics of this measurement

technique. Additionally, an accurate computational model of SINRD for a fresh fuel assembly can be applied to simulations of this technique to measure the fissile content in LWR spent fuel assemblies.

The enrichment of the $UO_2$ fuel in a PWR assembly was varied from 0.25 to 5.0 a/o $^{235}U$ to observe how the measured response changes as a function of the $^{235}U$ content in the fuel. The sensitivity of this measurement technique was assessed by modeling partial defects (i.e. missing pins) in the PWR fuel assembly. The number of pins removed from the assembly was varied from 6% to 50% to determine the limiting case that SINRD is capable of detecting.

## DESCRIPTION OF MCNPX SIMULATIONS

The PWR fresh fuel assembly was modeled in water (with and without 2200-ppm of boron) and in air (surrounded by water) to determine how the scattering of neutrons in water affects the measured response. Spontaneous fission neutrons from $^{238}U$ were used to self-interrogate the fresh fuel pins in the MCNPX simulations of SINRD. The concentration of $^{235}U$ in the fuel pins was determined by measuring the distinctive resonance absorption lines from $^{235}U$ using a set of $^{235}U$ fission chambers (FC) placed adjacent to the side of the fuel assembly. Ratios of different fission chambers were used to reduce the sensitivity of the measurements to extraneous material present in fuel and the neutron source strength cancels in the ratio. The specifications used to model the fuel assembly are given in Table I.

TABLE I. Characteristics of PWR fresh fuel assembly

| | |
|---|---|
| Assembly width (square) | 212 mm |
| Lattice dimensions | 17 x 17 |
| Number of pins per assembly | 264 |
| Fuel material | $UO_2$ |
| $^{235}U$ enrichment | 0.25 - 5 at% |
| Cladding material | Zircaloy 2 |
| Outer fuel diameter | 9.00 mm |
| Outer clad diameter | 10.0 mm |
| Fuel element pitch | 12.5 mm |
| Moderator | Light Water |

The detector configuration located adjacent to the assembly is shown in Fig. 2. The detector unit is approximately 20-cm long and 9.2-cm high. In order to reduce the background from neutrons, the sides and back of the detector pod were covered with either 4.0-mm of boron carbide ($B_4C$) or 1.0-mm of Cd. The outer $^{235}U$ fission chamber was embedded in polyethylene to thermalize the fast neutrons and increase counting statistics. The neutron flux at the detector pod was monitored using two fission chambers. The bare $^{235}U$ fission chamber was used to measure the entire neutron spectrum with thermal-neutron domination, and the covered (outer) fission chamber located behind the $B_4C$ shield was used to monitor the fast neutron flux above the neutron resonance region energy.

Figure 3 shows a top view of the PWR 17x17 assembly and locations of 20.0% and 6.25% partial defects. Since the detector pod can be placed on any of the four sides of the assembly, the defects were made with four sided symmetry.

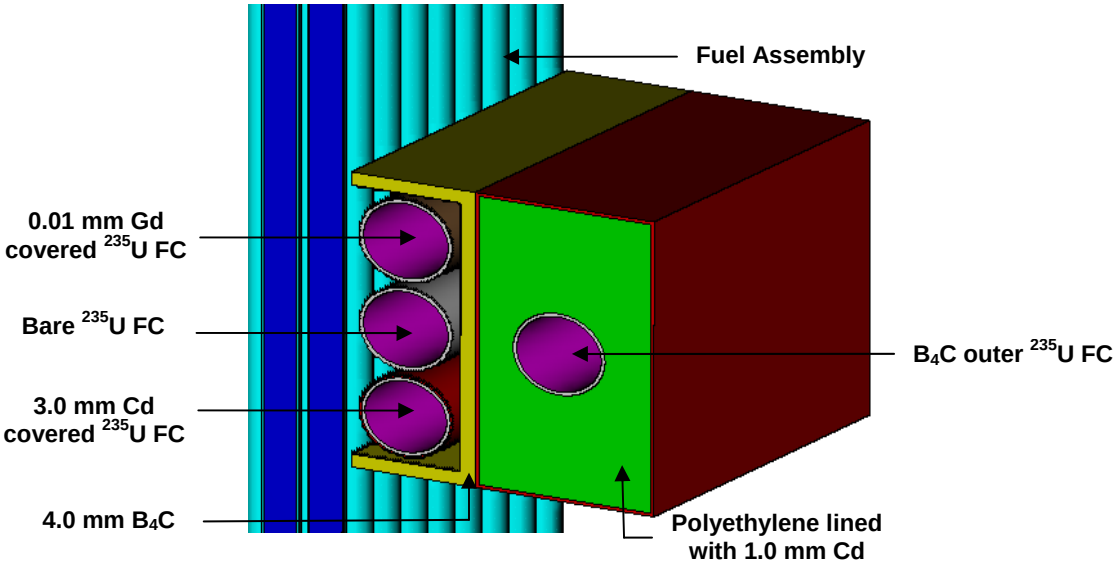


**Figure 2.** Detector configuration located adjacent to assembly.

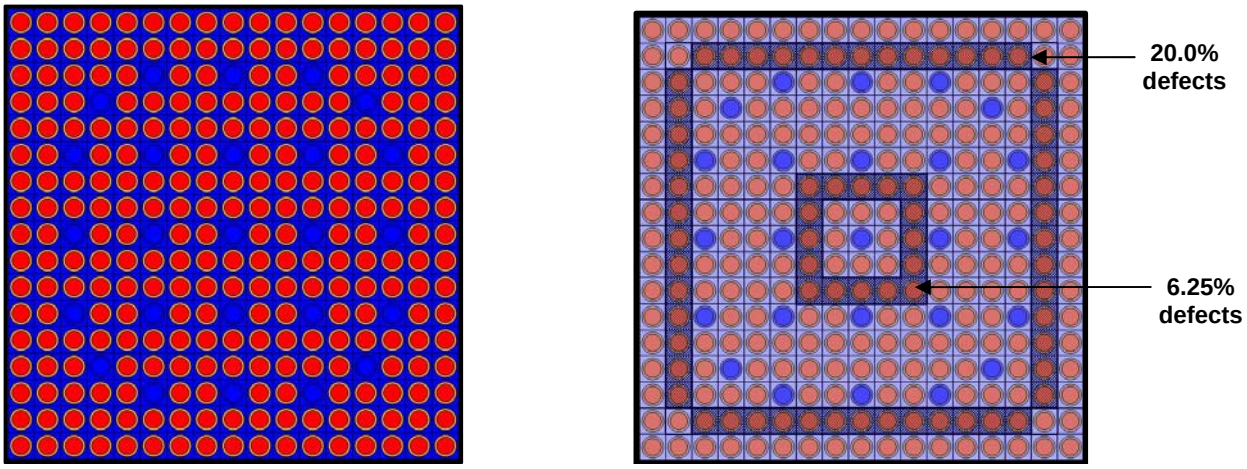


**Figure 3.** Top view of PWR 17x17 assembly (left) and partial defects locations (right).

## FRESH FUEL RESULTS

In order to assess how the addition of boron in the water affects the detector ratio signature, MCNPX was used to simulate the PWR assembly with no boron, 1000-ppm, and 2200-ppm boron in water. The ratio of the $B_4C$ outer $^{235}$U FC to bare $^{235}$U FC for different $^{235}$U enrichments as a function of the boron concentration in water is shown in Fig. 4. These results show the increase in thermal absorption from increasing the $^{235}$U content in the fuel. The detector ratio varies with $^{235}$U enrichment because the detector pod is located directly next to the assembly. The $B_4C$ outer $^{235}$U FC to the bare $^{235}$U FC measurement could be used to determine the concentration of boron in a spent fuel pool.

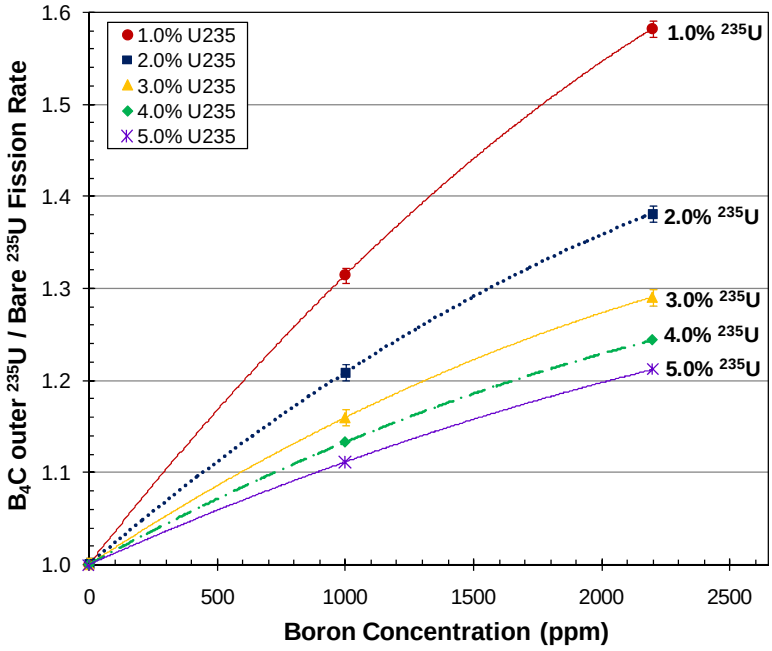


**Figure 4.** Comparison of $B_4C$ outer $^{235}U$ FC to bare $^{235}U$ FC ratio for different $^{235}U$ enrichments versus boron concentration in water.

### *Pin Removal Results*

The sensitivity of the SINRD ratios to partial defects was investigated using the MCNPX simulations where the PWR assembly was fixed at different enrichments ranging from 0.25% to 5.0%. In each case, fuel pins were removed and replaced by stainless steel (SUS 304) pins to give a defect varying from 6.25% to 50%. The fission chamber ratios that can be used for SINRD consist of two filters (Gd and Cd) covering $^{235}U$ FCs and two neutron flux monitors (bare $^{235}U$ FC and $B_4C$ outer $^{235}U$ FC). This study investigated the sensitivity of the SINRD method to different combinations of filters and monitors. The Gd filter removes thermal neutrons at a lower cutoff energy than the Cd filter, and the study showed that the Gd filter provided better results than the Cd filter for the present configuration.

In the following figures, all results have been normalized to the ratio with all depleted uranium (DU) fuel rods in assembly (0.25 a/o $^{235}U$). The ratio of the Gd covered $^{235}U$ FC to the $B_4C$ outer $^{235}U$ fast flux monitor in pure water as a function of the $^{235}U$ enrichment is shown in Fig. 5. We see that the 6.25% partial defects case cannot be distinguished from the no defects case within the statistics of the Monte Carlo results but the 20% and 50% defects can be detected. For each case, error propagations were used to calculate the resulting uncertainties in the ratio of the Gd covered $^{235}U$ FC to the $B_4C$ outer $^{235}U$ FC. The uncertainties in this ratio were between 0.5% – 0.8% for approximately 2.0E+07 particle histories.

Next, we simulated the same detector configuration, but added 2200-ppm boron in the water that is typical for PWR spent fuel storage pools. The results are shown in Fig. 6 where the 6.25% partial defects case is barely distinguishable from the no defects case. The addition of boron in the water decreased the SINRD signature ratio by 18%. It should be emphasized that the data has been normalized to the DU case. Thus, this type measurement could show the departure from a reference

fuel assembly with no defects. In all fuel assembly measurements, a reference assembly for calibration is assumed.

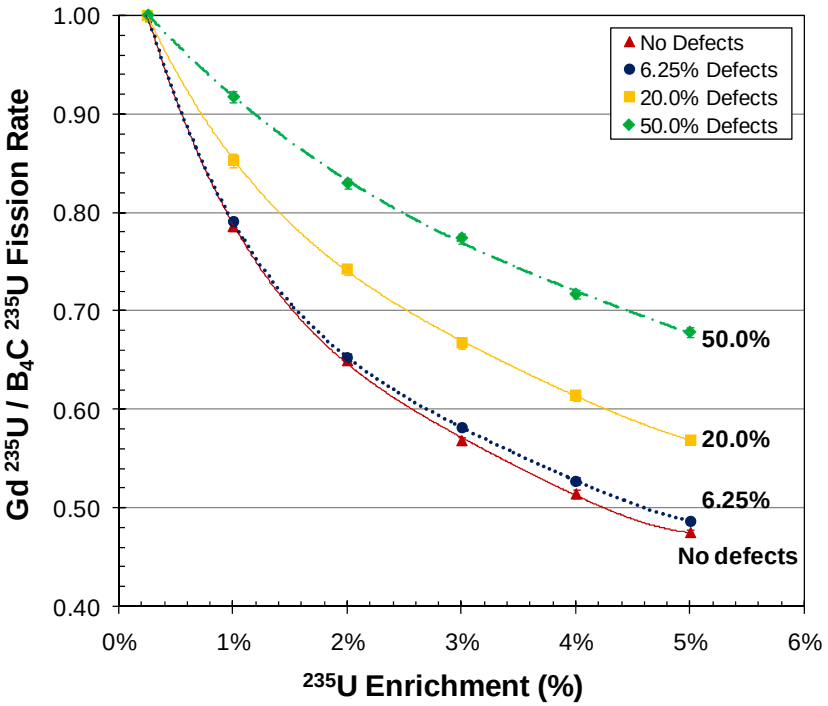


**Figure 5.** Effect of partial defects on ratio of Gd covered $^{235}$U FC to $B_4C$ outer $^{235}$U FC (epithermal to fast flux ratio) in *water* versus $^{235}$U enrichment.

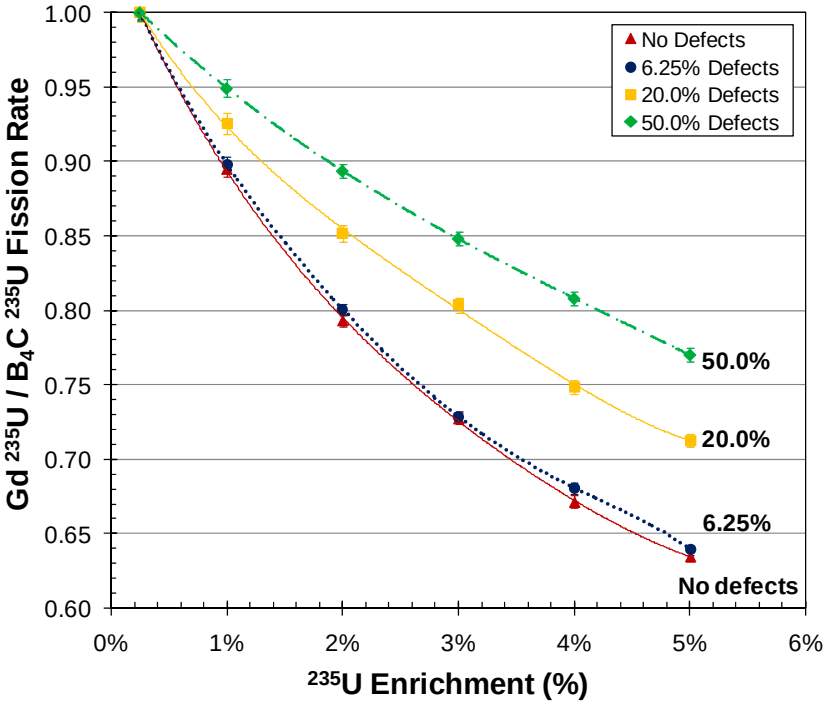


**Figure 6.** Effect of partial defects on ratio of Gd covered $^{235}$U FC to $B_4C$ outer $^{235}$U FC (epithermal to fast flux ratio) with *2200-ppm boron* in *water* versus $^{235}$U enrichment.

Previous measurements using the SINRD method have shown good sensitivity for metallic samples and MOX rods when applied in air.[3,4] For spent fuel assemblies, the storage is normally in water, and for PWR, there is typically ~ 2200-ppm boron in the water for criticality control. The water was expected to have a derogatory effect on the SINRD measurement because of neutron scattering by the hydrogen that will remove neutrons from the resonance absorption lines shown in Fig. 1. For present and advanced fuel cycle processing, the fuel will be removed from the water and the assembly or pins could be measured in air. Thus, we have simulated the fuel assembly in air to evaluate the SINRD method. For the air case, it was necessary to moderate the $^{238}$U spontaneous fission source term with external moderation because the water has been removed from the internal fuel pin area. The external water surrounding the assembly was used for this purpose.

Figure 7 shows the ratio of the Gd covered $^{235}$U fission rate to $B_4C$ $^{235}$U fission rate in air versus $^{235}$U enrichment. The change in the detector ratio signature versus enrichment increased by 27% in air than for the water case (no boron), however, the 6.25% defects case is still too small for inspection verification purposes.

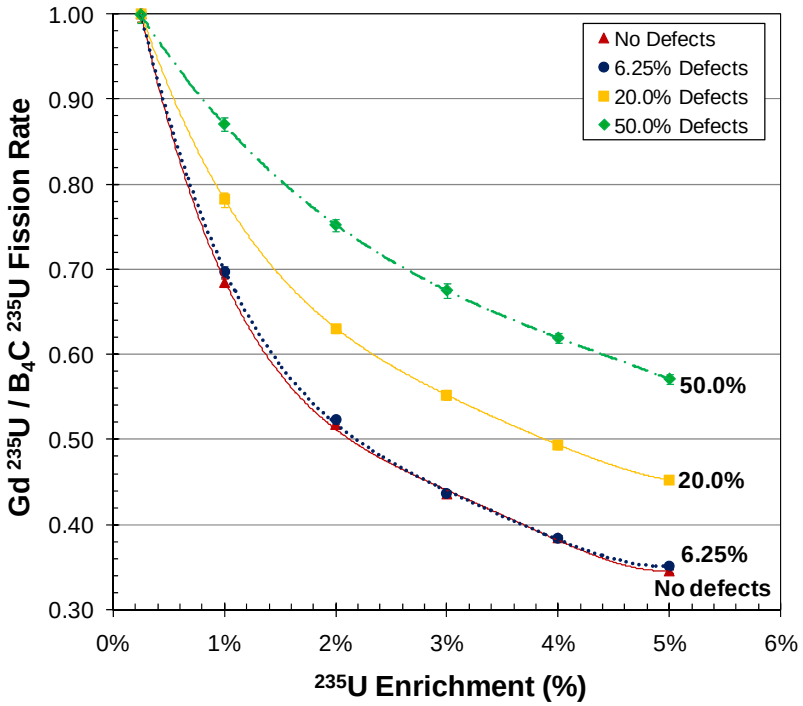


**Figure 7.** Effect of partial defects on ratio of Gd covered $^{235}$U FC to $B_4C$ outer $^{235}$U FC (epithermal to fast flux ratio) in *air* versus $^{235}$U enrichment.

## CONCLUSIONS

We have simulated the change in the ratio of $^{235}$U FC rate to the $B_4C$ $^{235}$U fast fission monitor using MCNPX. This ratio is sensitive to the $^{235}$U content in the fuel assembly. The SINRD signature for $^{235}$U concentration has not saturated for the $^{235}$U loadings up to 5%. For a factor of two change in the $^{235}$U concentration, the signature ratio changes by 21% in water and 32% in air. Therefore, the sensitivity of the method to partial defects is limited to significant (> 10%) changes in the $^{235}$U linear loading. This densitometry method requires a calibration with a reference assembly of similar geometry.

The purpose of this paper was to study the SINRD method for the PWR fresh fuel assembly configuration. The fission detector package illustrated in Fig. 2 can be applied to any of the four sides of the assembly, so four quadrant symmetry was assumed in the fuel loading and fuel removal. There are generally two different models for fissile material diversion from a fuel assembly. The first is to mis-declare the $^{235}U$ enrichment for the assembly, and the second is to remove fuel pins and to replace them with depleted uranium or iron pins. In the first model, the fissile material distribution is the same as for the calibration standard; however, for the second diversion model, the position of the pin diversion will affect the measurement. This paper assumed the pin removal positions shown in Fig. 3.

For PWR fuel assemblies, the fuel pins present a "plum pudding" target with approximately 264 pins. For the fresh fuel assemblies simulated in this paper, the pins in the assembly present an approximate uniform sample to the transmitted neutrons because the self-shielding is small for individual pins. For spent fuel assemblies, the initial $^{235}U$ enrichment is tailored for the pin positions so that the in-growth of Pu (and burnup) is similar for the different pin positions. For the normal application of neutron (or gamma-ray) densitometry techniques, the sample is assumed to be homogeneous so that the transmitted beam provides the average concentration of the isotope of interest. This homogeneity is the normal condition for solutions and bulk powder, but not for fuel assemblies. Future work will look at different geometric configurations, pin removal distributions, and the use different filters to cover the fission chambers to maximize the SINRD signature. In addition, the use of the SINRD method to measure the fissile content in LWR spent fuel assemblies and pyroprocessing heavy metal product will also be investigated.

## ACKNOWLEDGEMENTS

We would like to acknowledge the Program of Technical Assistance to the IAEA (POTAS) and the Department of Energy National Nuclear Security Administration's Office of Global Security Engagement and Cooperation (NA-242) for their support in the development of the SINRD method. The IAEA has provided useful guidance and support for the activity.

## REFERENCES

1. A. M. LAFLEUR, W. S. CHARLTON, H. O. MENLOVE, M. T. SWINHOE, "Nondestructive Measurements of Fissile Material Using Self-Indication Neutron Resonance Absorption Densitometry (SINRAD)," *Proceedings of 8th International Conference on Facility Operations – Safeguards Interface,* Portland, OR, March 30 – April 4, 2008.
2. H. O. MENLOVE, C. D. TESCHE, M. M. THORPE, R. B. WALTON, "A Resonance Self-Indication Technique for Isotopic Assay of Fissile Materials," *Nucl. Appl.*, **6**, 401 (1969).
3. "Nondestructive Assay of SEFOR Fuel Rods," *Los Alamos National Laboratory Program Status Report April-June 1969*, LA-4227-MS (1969).
4. "Neutron Self-Indication Assay of SEFOR Fuel Rods," *Los Alamos National Laboratory Program Status Report July-September1969*, LA-4315-MS (1969).
5. D. B. PELOWITZ ed., "MCNPX User's Manual, Version 2.5.0," *Los Alamos National Laboratory Report*, LA-CP-05-0369 (2005).