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NONDESTRUCTIVE MEASUREMENTS OF FISSILE MATERIAL USING SELF-INDICATION NEUTRON RESONANCE ABSORPTION DENSITOMETRY (SINRAD)

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ABSTRACT

The use of self-indication neutron resonance absorption densitometry (SINRAD) to nondestructively measure the concentration of fissile material was investigated using Monte Carlo N-Particle eXtended transport code (MCNPX) and results were benchmarked against experimental data from 1968 and 1969. This technique utilizes the unique resonance structure in the fission cross-section of different fissile isotopes by passing a neutron beam through an absorber filter, fissile sample, and then to a set of fission chambers. The sensitivity of this technique is based on using the same fissile materials in the sample and fission chamber because the effect of the resonance absorption lines in the transmitted flux is amplified by the corresponding (*n*,*f*) reaction peaks in the fission chamber. The simulated 1968 experiment consisted of modeling fissile metal plates of ^{235}U and ^{239}Pu with thicknesses ranging from 0.254 to 3.05 mm. The simulated 1969 experiment consisted of modeling a MOX fuel rod containing pellets of different Pu enrichments ranging from 12 % Pu to 27% Pu. The agreement of MCNPX results with results from 1968 and 1969 experimental measurements confirms the accuracy of the MCNPX models used. Future work includes the use of the SINRAD to measure the ^{235}U and ^{239}Pu content in LWR spent fuel and pyroprocessing materials.

Key Words: neutron resonance absorption, fissile material, ^{235}U, ^{239}Pu

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-indication neutron resonance absorption densitometry (SINRAD) 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 resonance absorption lines in the transmitted flux are 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 the similarly for the other fissile isotopes. The self-indication term is a result of having the same fissile material in the fission chamber as in the sample.

The comparison of the ^{239}Pu fission cross-section to the absorption lines in the neutron flux after transmission through a Gd filter and 0.254-mm [curve (a)] and 2.54-mm [curve (b)] ^{239}Pu sample is shown in Figure 1. It is important to note that as the sample thickness increases, the self-indication effect decreases due to self-shielding effects occurring from saturation of the larger resonances.[1]

The primary objective of this research is to develop and assess the sensitivity of using SINRAD for nuclear safeguards measurements. Recent interest in this approach was stimulated by an IAEA request related to spent fuel verification. The first step in developing this technique was to simulate the use of SINRAD for the assay of fissile materials using Monte Carlo N-Particle eXtended transport code (MCNPX);[2] these results were benchmarked against 1968 and 1969 experimental measurements. Benchmarking simulated results against experimental data is of significant importance to validate the use of MCNPX as a computational tool and assess the accuracy of the MCNPX models used. Furthermore, an accurate computational model of SINRAD can be used as a learning tool because parameters in the model can be easily varied to obtain a better understanding of the underlying physics of the measurement technique.

The simulated 1968 experiment consisted of modeling fissile metal plates of ^{235}U and ^{239}Pu with thicknesses ranging from 0.254 to 3.05 mm. The simulated 1969 experiment consisted of modeling MOX fuel rods containing pellets of different Pu enrichments ranging from 12% Pu to 27% Pu. The agreement of MCNPX results with 1968 and 1969 experimental measurements confirms the accuracy of the MCNPX models used and enables these models of SINRAD to be applied to more complex geometries such as LWR fuel assemblies and pyroprocessing materials.

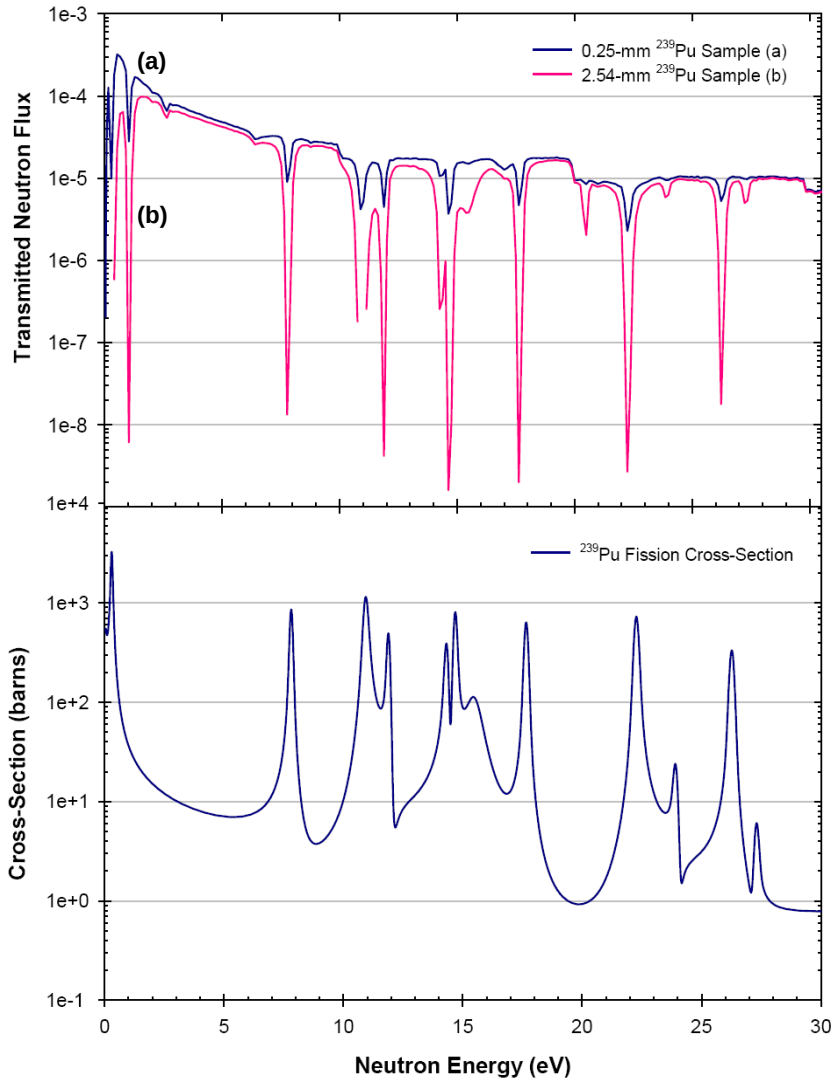

Figure 1: Comparison of the absorption lines in the neutron flux after transmission through a 0.114-mm Gd filter and 0.254-mm [curve (a)] and 2.54-mm [curve (b)] ^{239}Pu sample (upper plot) to the ^{239}Pu fission cross-section at neutron energies ≤ 30 eV (bottom plot).[1]

FISSILE METAL PLATES - 1968 EXPERIMENT

1968 Experimental Setup and Procedure

A diagram of the MCNPX simulated experimental setup used for 1968 measurements is shown in Figure 2. The neutron source used in the 1968 experiment was a collimated neutron beam from the LASL Water Boiler Reactor. The neutron beam first passed through a thin foil of Gd or Cd to remove thermal neutrons, then through the fissile sample (^{239}Pu or ^{235}U), and finally through three parallel-plate fission chambers containing deposits of ^{239}Pu, ^{235}U, and ^{10}B. The fissile samples consisted of 5.0 cm diameter metallic disks of 94.2 wt % ^{239}Pu and 93.0 wt % ^{235}U with thicknesses ranging from 0.24 to 3.05 mm. [1]

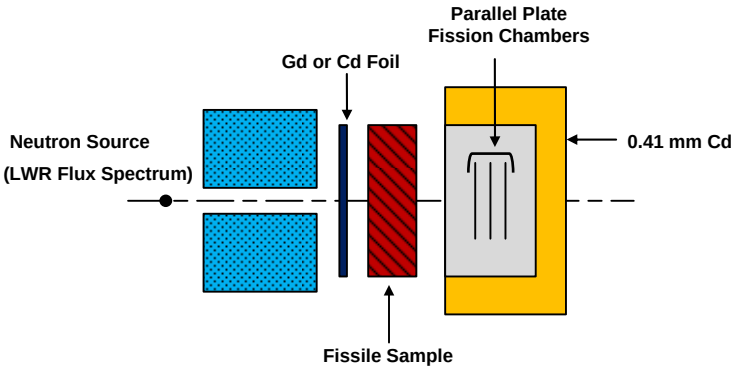

Figure 2: Simulated 1968 experimental setup.

In order to simulate the neutron source, MCNPX was used to calculate the energy-dependent flux spectrum of a typical Light Water Reactor (LWR) by modeling a fresh UO_2 fuel pin (3 w/o ^{235}U) surrounded by water. The resulting neutron flux spectrum as a function of energy (Figure 3) was then input into the MCNPX model of the 1968 experiment.

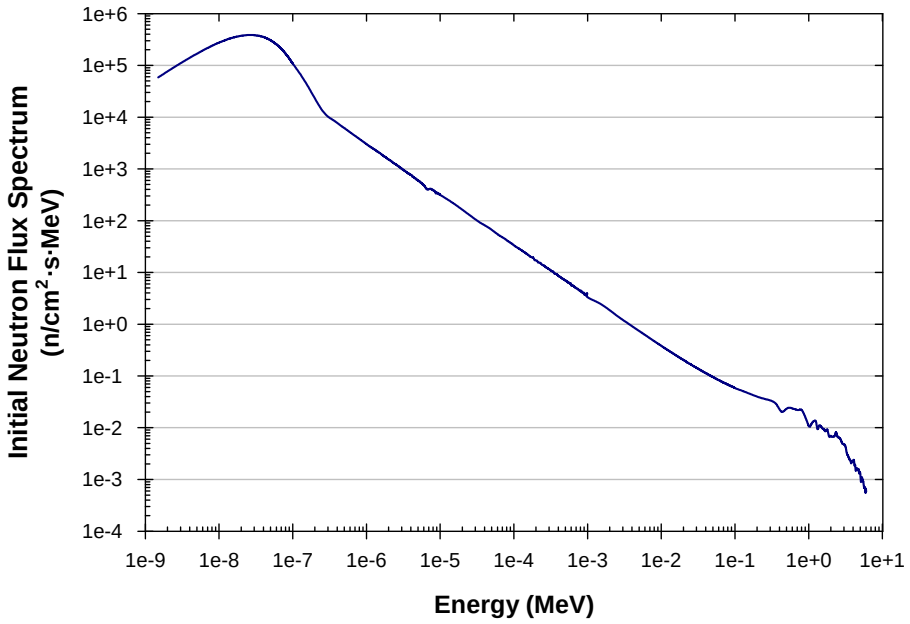

Figure 3: Initial energy-dependent neutron flux spectrum used in MCNPX model.

The three parallel-plate detectors used in the 1968 experiment are shown in Figure 4. These detectors were operated as gas flow proportional counters during sample irradiation using gas mixture of 90% Ar and 10% CH_4. In order to reduce the background from neutrons, the sides and back of the detector pod were covered with 0.41 mm of Cd. Since the attenuation of neutron flux between the parallel-plate fission foils was small, the geometry of these foils was not explicitly modeled in the MCNPX simulation. Instead, a void region was used to tally the ^{239}Pu and ^{235}U fission rates and the ^{10}B (n,α) rate. The count rate the three detectors was recorded both with and without the fissile sample present for normalization purposes.[1]

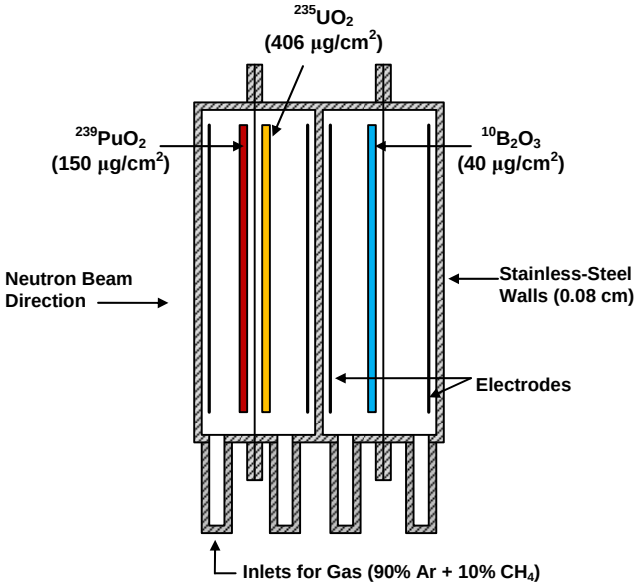

Figure 4: Parallel-plate ionization chambers used for self-indication measurements in 1968 experiment. [1]

Benchmark Results for Metal Plates

The fissile sample thicknesses shown in the following results were corrected to account for the 5.8 w/o ^{240}Pu in the ^{239}Pu samples and the 7.0 w/o ^{238}U in the ^{235}U samples. In the following plots, all results have been normalized to zero sample thickness. The response of ^{235}U and ^{239}Pu fission chambers as a function of fissile sample thickness is shown in Figure 5. The dashed lines correspond to the 1968 measured results. A 0.114-mm Gd filter was used to obtain these results. The self-indication effect is greater for ^{239}Pu because ^{239}Pu has a large resonance at 0.3-eV which readily absorbs neutrons near this energy region.

The ratios of fission chamber response rates to the ^{10}B detector response rate for various sample thicknesses of ^{239}Pu are shown in Figure 6. The dashed lines correspond to the 1968 measured results. A 0.076-mm Gd filter was used to obtain these results. Using the ratio of detector fission rates to the ^{10}B (n,α) rate significantly reduces the sensitivity of the measured response to extraneous material present in fissile sample; this occurs because the presence of extraneous material reduces both the ^{10}B detector and fission chamber response rates by approximately same amount. The shapes of the curves for the ^{239}Pu and ^{235}U detectors in Figure 6 show that ^{239}Pu is the fissile sample in the neutron beam. The curves obtained from the ^{239}Pu fission chamber decrease with increasing ^{239}Pu sample thickness clearly exhibiting the self-indication effect. It should also be noted that for ^{239}Pu sample thicknesses greater than 1.0-mm the ratio of the ^{239}Pu fission rate to ^{10}B (n,α) rate levels off due to self-shielding effects occurring from saturation of the large ^{239}Pu resonance at 0.3-eV. For thicker samples, a Cd filter can be used to eliminate neutron from the 0.3-eV region and to extend the sensitivity to the higher energy resonance region. The MCNPX results follow the same general trend as the measured results given the 1968 report.[1]

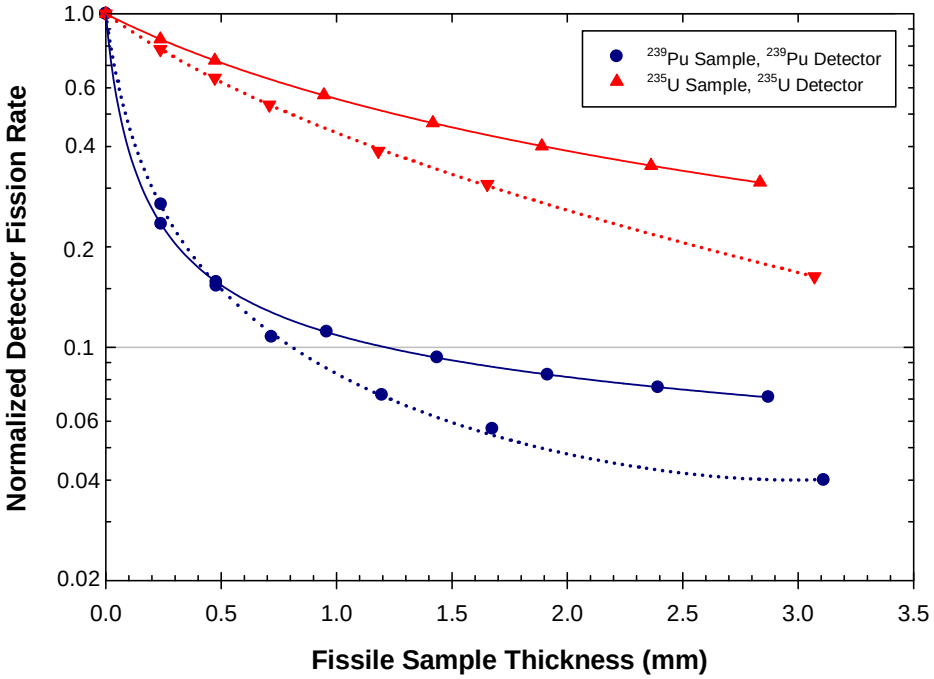

Figure 5: Normalized detector fission rates as a function of corresponding fissile sample thickness using a 4.5 mil Gd filter. The dashed lines correspond to 1968 measured results.

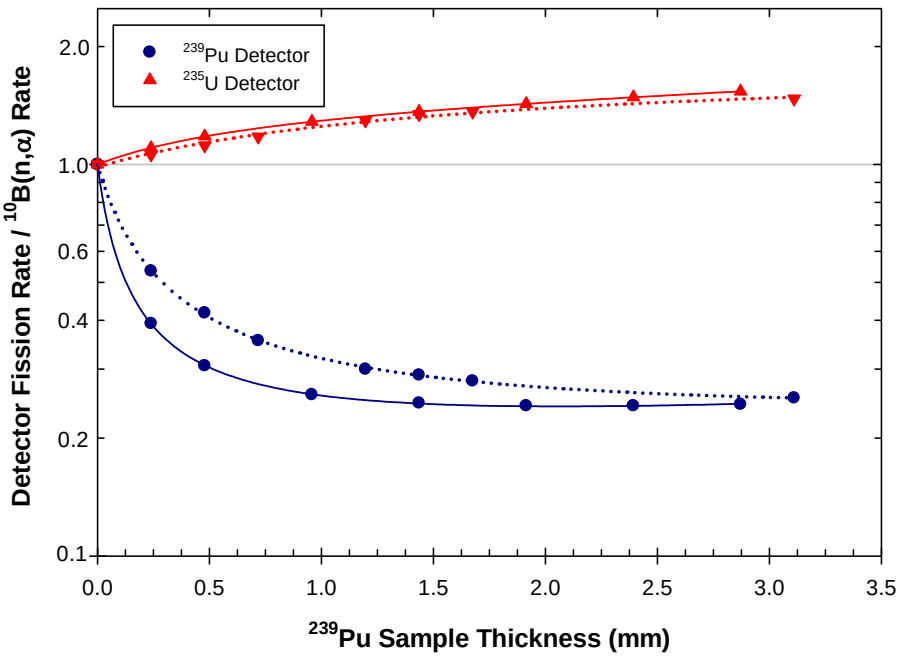

Figure 6: Ratio of the ^{239}Pu and ^{235}U detector fission rates to the ^{10}B detector (n,α) rate versus ^{239}Pu sample thickness using 3.0 mil Gd filter. The dashed lines correspond to 1968 measured results.

Several different neutron filters with varying thicknesses of Gd and Cd were used in the 1968 measurements to determine the effect of the low-energy neutron cutoff energy on the fission chamber responses. The $^{235}U/^{239}Pu$ detector fission rate ratio as a function of ^{239}Pu sample thickness for different filters of Gd or Cd is shown in Figure 7. The dashed lines correspond to the 1968 measured results. The differences exhibited by the $^{235}U/^{239}Pu$ response curves shown in Figure 7 are predominantly caused by the large 0.3-eV resonance in ^{239}Pu. The neutron flux in the energy region of this ^{239}Pu resonance is attenuated by the 0.114-mm Gd filter, whereas an appreciable number of lower energy neutrons are transmitted by the 0.025-mm Gd filter. As a result, the $^{235}U/^{239}Pu$ response ratio decreases faster with ^{239}Pu sample thickness for the 0.025-mm Gd filter than for the 0.114-mm Gd filter. The 0.762-mm Gd filter absorbs nearly all of the neutrons in the energy region of the large ^{239}Pu resonance; on the other hand, the 0.508-mm Cd filter allows a large fraction of neutrons to be transmitted. This causes the $^{235}U/^{239}Pu$ response ratio to increase faster with ^{239}Pu sample thickness for the 0.508-mm Cd filter than for the 0.762-mm Cd filter. It may be noted that the MCNPX calculations reproduce the calculations that were done in the 1968 paper more closely than they reproduce the experimental measurements. This may indicate that some of the experimental parameters may be somewhat different from their assumed values.

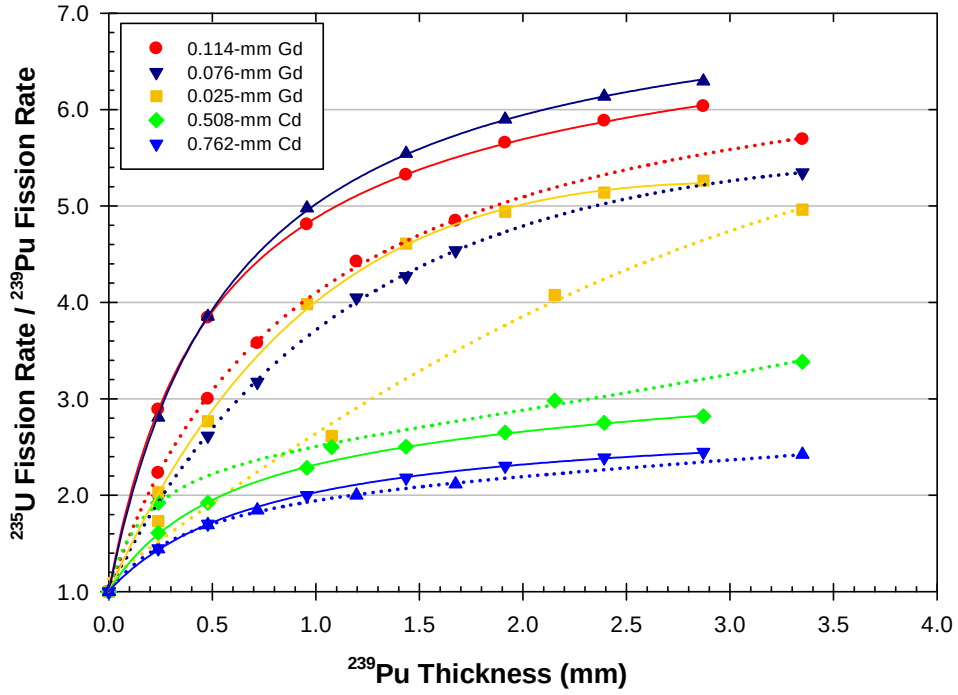

Figure 7: $^{235}U/^{239}Pu$ detector fission rate ratio as a function of ^{239}Pu sample thickness for Gd and Cd filters of varying thicknesses. The dashed lines correspond to 1968 measured results.

MOX FUEL RODS - 1969 EXPERIMENT

1969 Experimental Setup and Procedure

In 1969, General Electric and a consortium of research institutes, built the South East Fast Oxide Reactor (SEFOR) in Arkansas. This was a fast breeder reactor with liquid sodium coolant.

During the initial fuel fabrication, some of the fuel pellets contained an incorrect plutonium concentration in the MOX rods and these rods were transferred to LASL for NDA of the plutonium loading in the pellets. The rods had received a low level of irradiation in the SEFOR reactor prior to the NDA. Reactivity measurements of SEFOR MOX fuel rods indicated that the plutonium content in some of the rods was 5% - 40% less than the amount specified by the manufacturer. To help resolve this problem, the 1969 experiment was conducted. In this experiment, SINRAD was used to measure the pellet-to-pellet Pu distribution in MOX fuel rods.

The MOX fuel composition was 12-27 w/o PuO_2-UO_2 (depleted) mixture that was formed into cylindrical pellets approximately 2.3-cm in diameter and 1.6-cm high. Figure 8 shows a schematic of the SEFOR fuel element and the corresponding profile of neutron transmission data. In the 1969 experiment, the curve shown in Figure 8 was measured using the neutron beam from the LASL Water Boiler Reactor filtered through a Gd foil, and measured with a collimated BF_3 neutron detector. This measurement was simulated in MCNPX by modeling each segment of the fuel element individually and tallying the transmitted neutron flux. The transmitted neutron flux is lowest at both ends of the fuel rod due to the dense nickel reflectors and is highest near the center of the rod due to the air gap that separates the fuel segments. The ^{238}U insulator pellets are clearly distinguishable from the MOX fuel segments, that may be used to eliminate the possibility of a rod containing only ^{238}U pellets.[3]

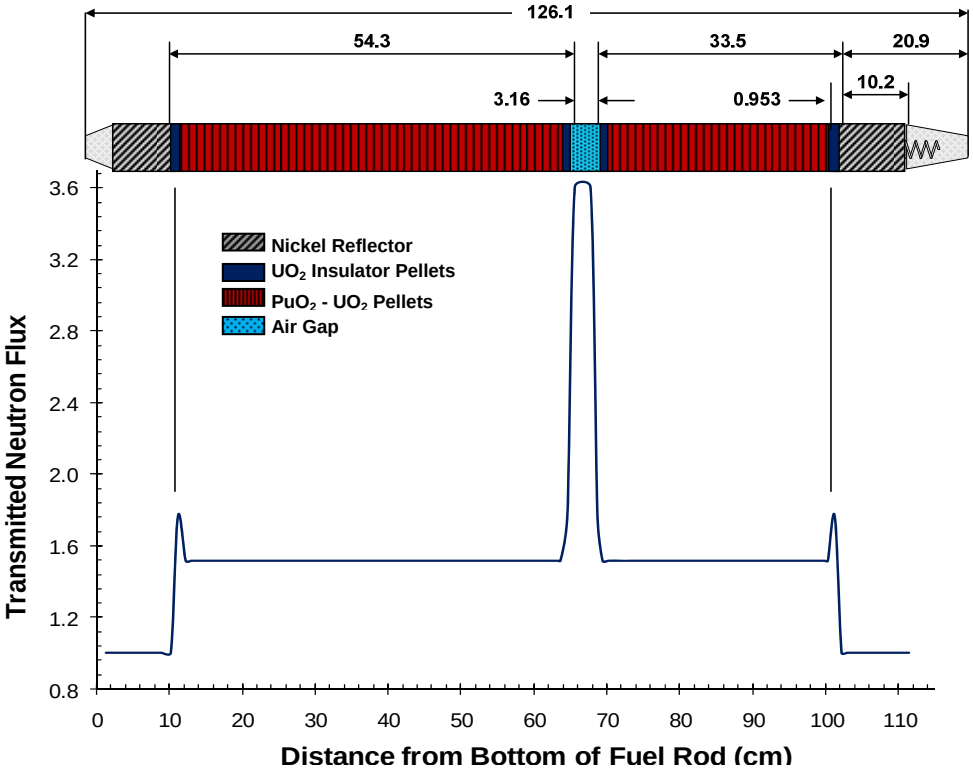

Figure 8: Schematic of SEFOR fuel element and profile of neutron transmission data.

The nondestructive assay results for six different SEFOR MOX fuel rods are given in Table I. Rods A13 and 473 were given as standards with known Pu enrichments of 27.3% and 20.4%, respectively. The delayed neutron yield technique was to measure the amount of fissile material (^{239}Pu + ^{241}Pu), as well as, the amount of fertile material (^{238}U) in the other four rods that had

previously been irradiated in the reactivity measurements. It should be noted that the amount of Pu in the four irradiated rods is significantly less than the amount of Pu in the two standard rods. The results given for the six different MOX fuel rods were used to calculate the initial composition of the MOX pellets used in the MCNPX model of the 1969 experiment. [3]

Rod Number	U + Pu (g)	^{239}Pu + ^{241}Pu (g)	Pu / (U + Pu)
A13[a]	2949	738	27.3%
473[a]	2930	549	20.5%
878[b]	2926 ± 29	484 ± 12	18.1%
919[b]	2862 ± 29	458 ± 11	17.5%
920[b]	2900 ± 29	445 ± 11	16.8%
873[b]	2956 ± 30	339 ± 9	12.5%

[a] *Specifications for the standard rods (A13 & 473)*
[b] *Rods previously irradiated in reactor reactivity measurments*

Table I: Nondestructive assay results for SEFOR fuel rods.

A diagram of the MCNPX simulated experimental setup used for 1969 measurements is shown in Figure 9. Similar to the 1968 experiment, the neutron beam from the LASL Water Boiler Reactor was used for 1969 measurements. This beam was collimated through a 1.6-cm hole and filtered by 0.025-mm-thick Gd foil before passing through the MOX fuel rod and to the ^{239}Pu fission chamber. A mechanical scanner was used to advance the MOX fuel rod across the neutron beam path in 0.76-cm increments. [4]

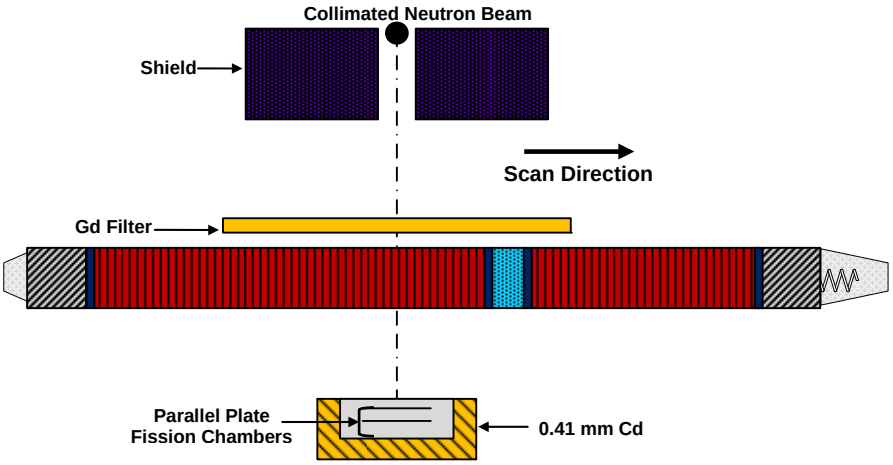

Figure 9: Simulated experimental setup of 1969 experiment.

Benchmark Results for MOX Fuel Rods

Figure 10 shows the axial Pu distribution in SEFOR fuel rod 878 obtained from the self-indication neutron scan. This measurement was simulated in MCNPX for selected data points and the corresponding results are also shown in Figure 10. The excellent agreement between the 1969 measurements and the MCNPX results for selected data points confirms the accuracy of MCNPX model used.

The integral along the length of the rod of the inverse ^{239}Pu fission rate as a function of total fissile plutonium mass in five different MOX fuel rods is shown in Figure 11. In the 1969 experiment, the total ^{239}Pu content was obtained by integrating the area under the axial Pu scan. These values were normalized to MCNPX results for inverse ^{239}Pu fission rate to obtain the calibration curves shown in Figure 11.

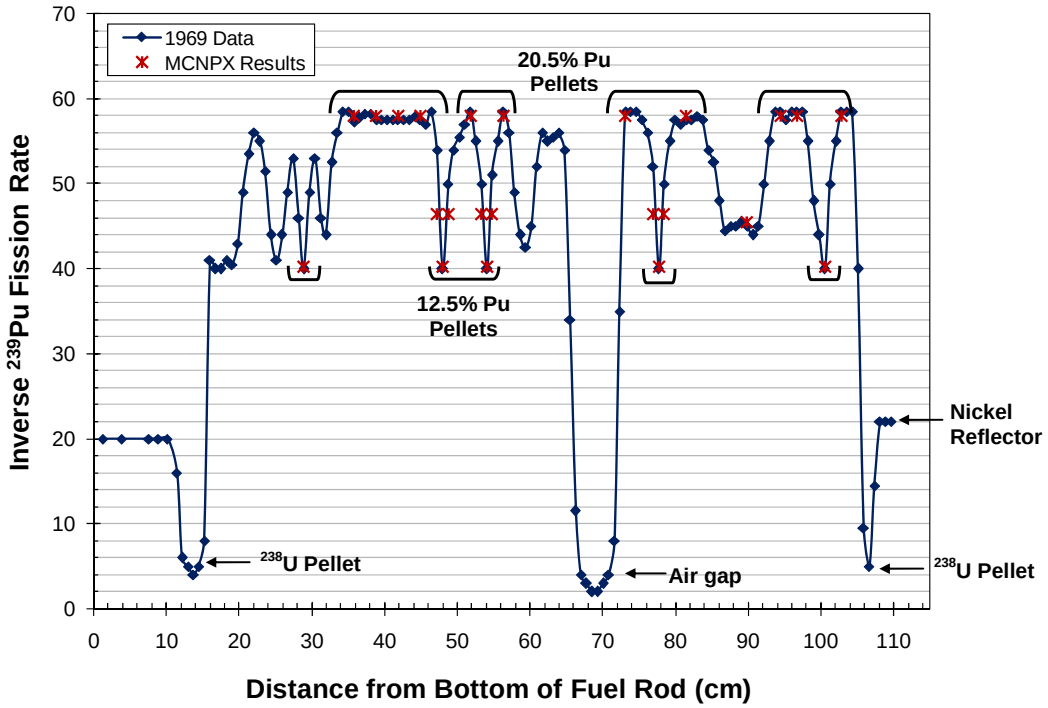

Figure 10: Axial Pu distribution in SEFOR fuel rod 878 obtained from "self-interrogation" neutron scan.

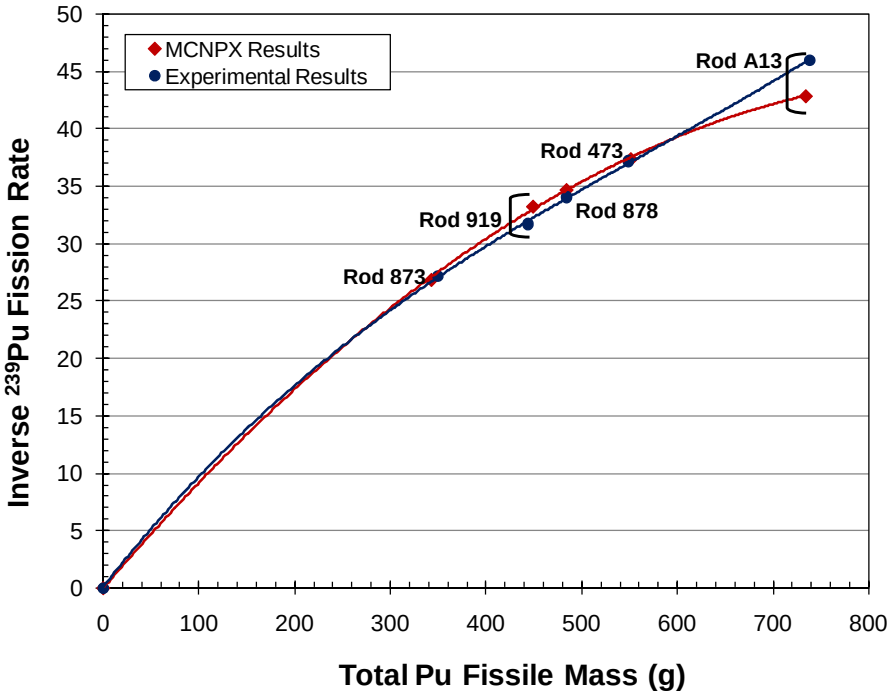

Figure 11: The integral of the inverse ^{239}Pu fission rate as function of total fissile plutonium mass. The 1969 experimental measurements were normalized to MCNPX results.

CONCLUSIONS

The MCNPX simulation of the 1968 results shows the same behavior as the experimental data. Interestingly, there is reasonable agreement with the 1968 calculated results. It should be emphasized that using ratios of different fission chambers has a clear advantage over a single-detector measurement by significantly reducing the sensitivity of the measured response to extraneous material present in the fissile sample mixture. The SINRAD method assumes that the detector ratios can be calibrated with a sample of the same geometry.

For the SEFOR reactor MOX fuel rods, the agreement of MCNPX results with 1969 experimental measurements confirms the accuracy of the MCNPX models used. A primary advantage of using SINRAD in this type of geometry is that, unlike gamma scanning which only sees the outer surface of the rod, the self-indication neutron scan is able to see through the entire pellet. Furthermore, SINRAD can be used to measure both fresh and spent fuel elements since this technique is insensitive to the high gamma radiation levels in spent fuel.

The accuracy of the SINRAD measurements for the SEFOR fuel was good enough to identify the plutonium enrichment of the misplaced pellets, and to show that the production pellets of 12% plutonium had been accidentally loaded into the rod of 20% pellets.

The SEFOR results have special importance in that the plutonium is mixed with much larger numbers of ^{238}U and oxygen atoms typical of spent FBR fuel. The IAEA is interested in developing verification methods for spent fuel from FBR and LWR reactors. There is currently work in progress at LANL to evaluate the same basic physics signature as SINRAD for spent

fuel. However, for the spent fuel case, there is adequate neutron emission from the curium in the spent fuel to be self-interrogating and no reactor beam is necessary. Thus, the IAEA introduced the modified acronym of self-interrogation neutron resonance densitometry (SINRD) for the spent fuel case using the curium self-contained neutron source.

Future work at LANL and TAMU includes the use of SINRD to measure the ^{235}U and ^{239}Pu content in LWR spent fuel assemblies, pins, and pyroprocessing materials.

ACKNOWLEDGEMENTS

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