Sixth American Nuclear Society International Topical Meeting on Nuclear Plant Instrumentation, Control, and Human-Machine Interface Technologies
NPIC&HMIT 2009, Knoxville, Tennessee, April 5-9, 2009, on CD-ROM, American Nuclear Society, LaGrange Park, IL (2009)

FUEL ASSEMBLY SELF SHIELDING OF INTERROGATION NEUTRONS IN A LEAD SLOWING-DOWN SPECTROMETER

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ABSTRACT

Recent attention to nuclear safeguards has stepped up the need for additional non proliferation safety measures. One of these safeguards is the tracking of ^{239}Pu and other fissile materials in spent nuclear fuel. Noninvasive methods are being investigated, including neutron interrogation. RPI has modeled a spent fuel assembly assay method in its lead slowing-down spectrometer. The fuel assembly is interrogated with neutrons from a neutron source in the center of the lead. As the interrogation neutrons slow down in the lead, they create fissions in the fuel assembly. An array of ^{238}U detectors can then detect the fission neutrons from the ^{235}U and ^{239}Pu in the fuel as a function of slowing down time. The focus of this MCNP modeling is to determine the sensitivity and self shielding effects in a 16 x 16 pin fuel assembly. The results show significant shielding of interrogation neutrons from the fuel pins located further from the source up to 80%. The shielding is more significant for slower neutrons than fast. Also, secondary fissions in the assembly greatly affect the detector response and create a nonlinear response to quantities of fissile materials. The system easily identified missing fuel pins, but the response is not proportional to the quantity of fuel missing and depends on the location of the missing pins. The error in determining the quantity of ^{239}Pu was greater than 100% when using a linear fitting model. New fitting procedures and sources of data used to benchmark measurements must be further investigated.

Key words: Spent fuel, Assay, Lead spectrometer

1 INTRODUCTION

With the introduction of the GNEP plan and the need to reprocess spent nuclear fuel, there is an increased need for the security of fissile material. Therefore, it is necessary to find an efficient, non-invasive way to measure the mass of ^{235}U and ^{239}Pu in spent fuel assemblies in order to track fuel inventories and to prevent proliferation. It is possible to use interrogation neutrons to create fission in the fuel and detect the resulting fission neutrons. Each actinide has its own signature reaction rate as a function of incident neutron energy, or slowing-down time, in the lead. With proper fitting procedures, these signatures can be used to determine the quantity of Plutonium in the spent fuel. This process is being studied at Rensselaer Polytechnic Institute and at University of Nevada, Las Vegas [1].

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The MCNP calculations at RPI model measurements of spent fuel assemblies in the RPI Lead Slowing-Down Spectrometer (LSDS). Extensive analysis of the neutron slowing down in the lead has been done in the past which provides a sound foundation for these models [2,3]. Experimental fission data obtained in the LSDS is also available for comparison to calculation [4]. This is critical because calculations tend to assume ideal conditions and do not take into account the realities encountered in the experiment. In particular, this project focuses on the self shielding problems encountered when testing an entire assembly of 256 fuel pins.

2 THE RPI LEAD SLOWING-DOWN SPECTROMETER

The RPI Lead Slowing-Down Spectrometer is a 75 ton, 1.8 m cube of lead. The lead is covered with a thin layer of Cadmium to prevent neutrons that have escaped and thermalized from reentering the lead. The 60 MeV linac creates neutrons through a (γ,n) reaction when the electrons interact with an air cooled tantalum target in the center of the lead.

The neutrons slow down by scattering in the lead creating a large isotropic flux. The resulting neutron flux is about 4 orders of magnitude larger than an equivalent (5.6 m) time-of-flight experiment. The neutron energy, E as a function of slowing down time, t of the neutron flux in the LSDS is determined by the equation [3].

$$E(t) = \frac{k}{(t+0.3)^2} \tag{1}$$

where k = 165000 eV- μs^2, t is in units of μs and E is in units of eV. The energy dependent neutron flux is given as [3]

$$\phi(E) = E^{-0.776} e^{-\sqrt{0.214/E}}. \tag{2}$$

These equations can then be used to determine the expected reaction rate as a function of neutron slowing-down time. The neutron energy resolution as fitted to MCNP simulations in the LSDS is [3]

$$\left[\frac{dE}{E}\right]_{FWHM} = \left[0.0835 + \left(\frac{0.128}{E}\right) + 3.05\times10^{-5} E\right]^{1/2}. \tag{3}$$

The neutron energy resolution (FWHM) is approximately 35% between 1 eV and 10 keV which are the energies of interest in fuel assay measurements.

3 THE MCNP MODEL OF THE LSDS

The geometry of the LSDS is configured in the MCNP files so that the electron beam tube enters the front of the 180 cm cube of lead along the positive x axis into the neutron target located at the origin. The assay channel is 30 cm behind and 30 cm above the target. It is 23.4 x 23.4 cm and extends completely through the lead from left to right along the y axis and perpendicular to the beam axis (x axis).

The source neutrons created in the LSDS are typically assumed to have an evaporation spectrum described by the equation [5]

$$p(E) = Ee^{-E/a} \tag{4}$$

where *a* is 0.46 MeV. This is the source model used in the MCNP calculations.

The value of k in equation (1) is derived from the average lethargy gain and the mean free path of the neutrons in the lead. Therefore, this value is heavily dependent on the hydrogen content in the lead or any materials introduced due to the measurements, particularly those of low atomic number. As a result of this, the resolution of the neutron energy can be substantially degraded, and careful consideration must be made when evaluating the MCNP calculations as to the materials that will be present when conducting actual experiments. Past experiments have shown a neutron energy resolution of 100% when 0.134 w% of water was introduced into a powdered U_2O_8 fuel [6]. With a resolution of 50%, a structure in the data can still be seen, which is sufficient to determine plutonium levels [6]. Fig. 1 compares the average neutron energy as a function of slowing-down time in the assay channel. The two sets of data compare the average neutron energy in the channel if the channel is filled with lead or voided. The neutron energy cut off was set at 10^{-8} MeV so no data is shown at lower energies.

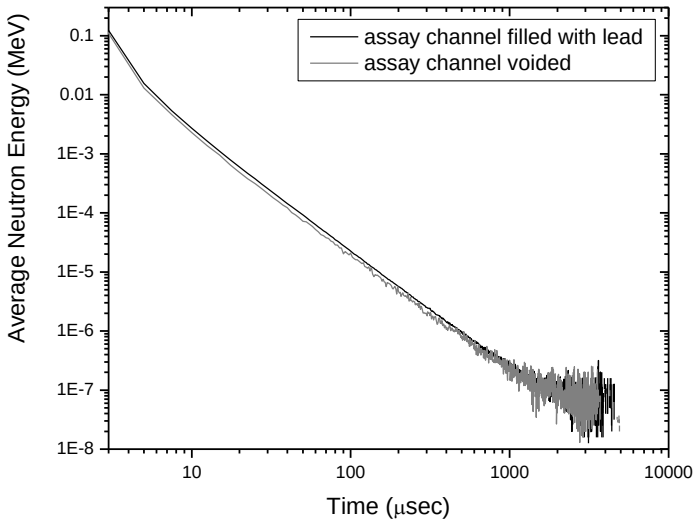


Figure 1. Average neutron energy as a function of slowing down time in the assay channel.

3.1 Compare Calculation to Experiments

A tally 5 detector was placed in the assay channel of the lead and the tally was multiplied by the ^{235}U fission cross section. This data was compared to experimental fission data for ^{235}U collected in the LSDS with 24.9 μg of ^{235}U this last year [4] and is shown in Fig. 2. The differences seen between the calculated data and the experimental data are due to the broadening of the neutron energy resolution in the experiment when compared to the ideal conditions of the calculation. This broadening is caused by equipment placed in the lead opening as well as the small quantities of impurities in the lead bricks. Also, the differences in the dip at about 300 μs, is theorized to be room return of neutrons when detectors are placed in large openings in the lead that cannot be completely shielded from neutrons that have left the lead, slow down in the room and return to the lead.

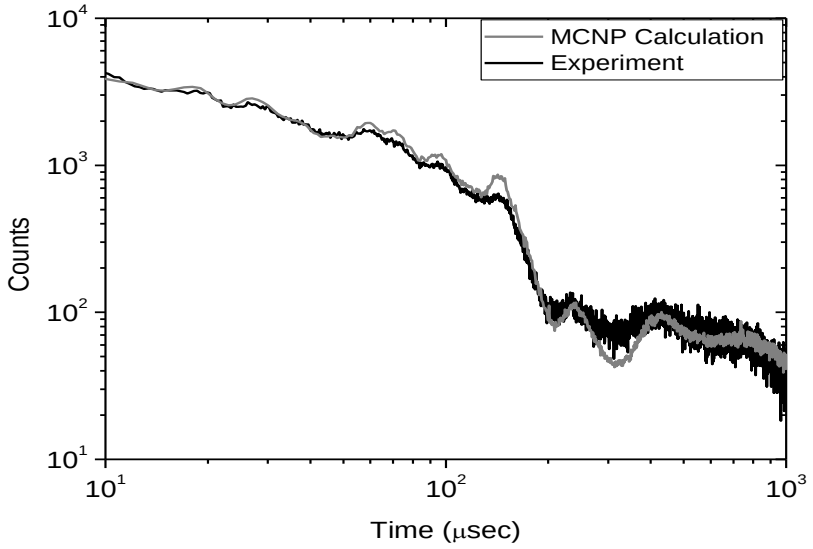


Figure 2. MCNP fission result compared to actual experimental results of ^{235}U fission in the LSDS.

4 ^{238}U DETECTORS

The detectors for the MCNP calculations are five tally 5 detectors with a radius of exclusion of 1 cm located above the fuel assay channel. The tally 5 was multiplied by the ENDF/B-VII.0 cross section for ^{238}U as well as the number of ^{238}U atoms in a 200 mg sample to simulate a ^{238}U threshold detector. The detectors used for spent fuel assay experiments at RPI contained 200 mg of ^{238}U with less than 4ppm ^{235}U, but the impurities were not included in the calculations [5].

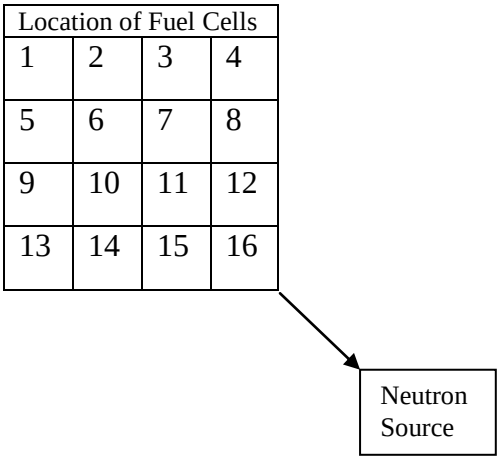
Ideally, the threshold detectors should be located as close to the fuel as possible in order to increase statistics and reduce the number of collisions of the fission neutrons prior to reaching the detector. The ^{238}U fission chambers used at RPI are small gas filled detectors and are stable in a gamma dose rate of up to 10^5 R/hour [5]. MCNP calculations were completed with the tally 5 detectors located next to the fuel, 5 cm into the lead, and 10 cm into the lead. There were no significant visible changes seen in the shape of the detector response; however, the statistical accuracy was decreased due to geometry and attenuation in the lead. Calculations used in this study were completed with the detectors 1.5 cm into the lead. Experiments at RPI placed the detectors 2.5 cm into the lead above the fuel assembly [6,7].

5 FUEL PIN AND ASSEMBLY MODELS

The pins and assembly are based on data from an AP1000 reactor but are shortened to 180 cm for the MCNP calculations because this matches the width of the LSDS [8]. Each fuel pin contains 953 g of UO_2 fuel. The composition of the fresh fuel is 11.8 w% O_2, 79.6 w% (759 g) ^{238}U, and 8.6 w% (82 g) ^{235}U for an enrichment of 9.8%. For calculations of spent fuel, some of the ^{235}U is replaced with ^{239}Pu and the combined quantity of ^{235}U plus ^{239}Pu is referred to as the fissile material. The fissile material is either 100 a% ^{235}U, a mixed fuel where 10 a% or 30 a% of the ^{235}U is replaced by ^{239}Pu, or 100 a% ^{239}Pu. The pin gap is filled with He and the cladding is

Zircaloy-2 and the assay channel contains air. For the preliminary calculations comparing an assembly to a single fuel pin a 17 x 17 pin assembly with a pitch of 1.26 cm is modeled. The assembly for the sensitivity and shielding calculations was reduced to 16 x 16 pins for ease of computation. The assembly is then grouped into 16 cells of 4 x 4 pins each as shown in table I.

Table I. Fuel assembly grouped into 16 cells of 16 fuel pins each. The neutron source is about 43 cm from cell 16.



6 SELF SHIELDING AND FUEL SENSITIVITY

For the shielding and sensitivity analysis, the detector response to an assembly is compared to that of a single fuel pin. Next, the neutron slowing-down time regions of interest are determined and, finally, the fissions in each fuel cell and the detector responses are examined over the total neutron slowing-down time spectrum from 10 – 2000 μs.

6.1 Single Pin Compared to Assembly Response

Calculations using a single pin serve as a baseline for the assembly calculations and can be used to determine self shielding issues within the assembly. Fig. 3 below shows the difference in the detector response for a single pin multiplied by 289 (the number of pins in an assembly) compared to the assembly itself. If there were no self shielding, the spectra would have the same shape. However, the spectra are strikingly different; not only are the magnitude of spectra different, but the peaks are shifted to earlier slowing-down times. This is due to the increased energy loss with each scatter in the assembly. The high energy neutrons are not affected, but as the fission cross section of the fuel increases, the shielding increases, particularly in the low energy regions. The important point to notice is that the reaction rate (fission neutrons counted in the detectors) is highly suppressed by about an order of magnitude in the 1000 μs region (about 0.3 eV) where ^{239}Pu has a strong resonance. The effect is also seen in ^{235}U, but not as strong. As will be shown later in this report, the resonances greatly affect the sensitivity of the system to missing fuel, and the strength of the effect is dependent on the ratio of ^{235}U to ^{239}Pu.

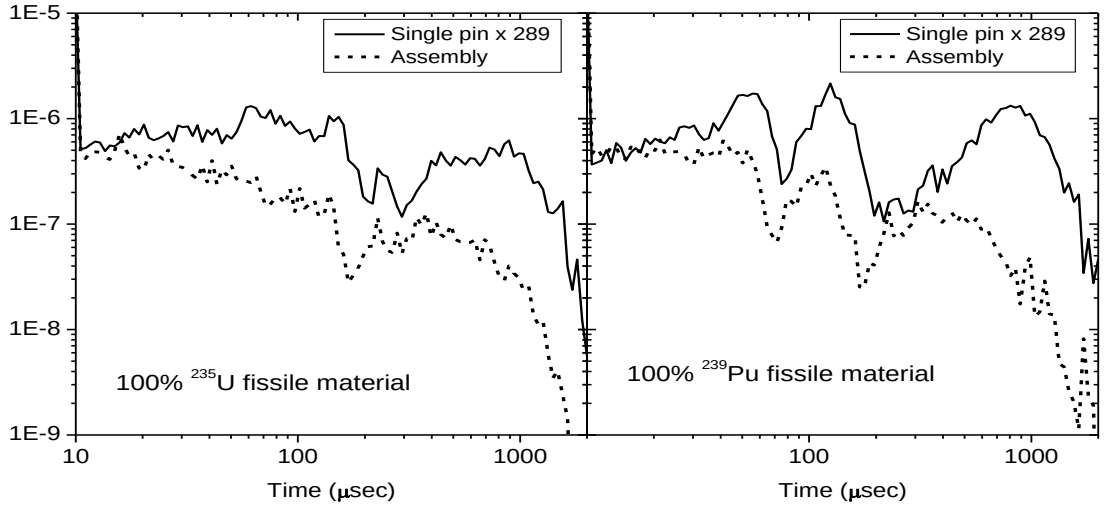


Figure 3. Comparison of the detector response to a single fuel pin and a fuel assembly showing the effect of self shielding. The left plot includes fuel pins with 100% ^{235}U as the fissile material, and the right plot includes fuel pins with 100% ^{239}Pu as the fissile material.

6.2 Regions of Interest

Fig. 4, compares the detector response to the fuel with fissile material comprised of either 100% ^{239}Pu or 100% ^{235}U. Three regions of interest can be seen where the ^{239}Pu response

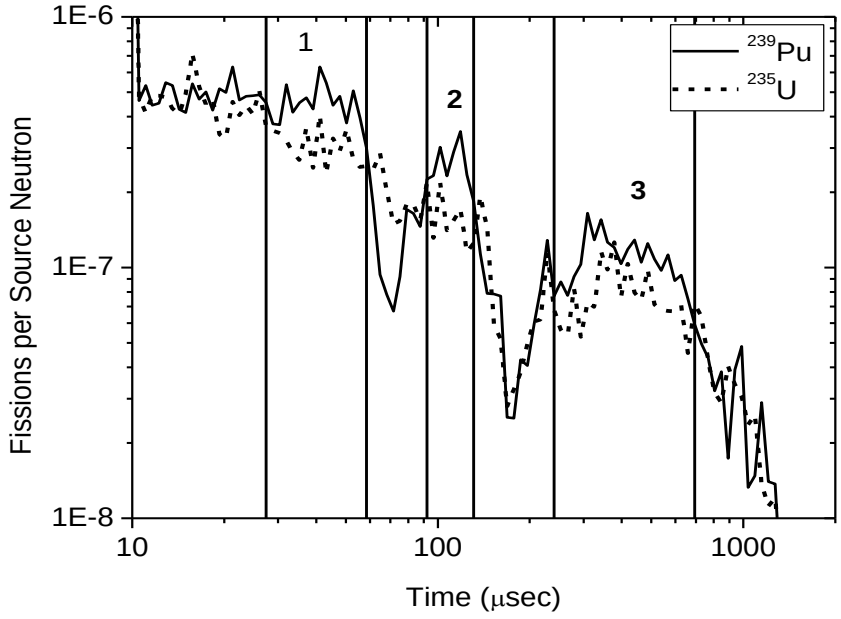


Figure 4. Comparison of the detector response for the fuel assembly with the fissile material content either 100% ^{235}U or 100% ^{239}Pu. The slowing-down time limits for resonance regions 1, 2, and 3 are shown.

is higher than the ^{235}U response due to resonances in the ^{239}Pu reaction rate. The first resonance group occurs at about 50 μs and corresponds to about 40 eV, the second resonance group occurs at 100 μs and corresponds to about 10 eV and the third resonance around 500 μs corresponds to

the large ^{239}Pu resonance at 0.3 eV. Because the 0.3 eV ^{239}Pu resonance is so large and distinct, it would seem that this would be the ideal energy region to compare ^{235}U and ^{239}Pu reaction rates. This is true for a single fuel pin as shown in Fig. 3, but in the assembly, the flux in this region is suppressed by absorption of the interrogation neutrons in the outer cells. The region which is the strongest indicator of ^{239}Pu quantities for a fuel assembly is explored later in this report.

6.3 Fissions in the Fuel

To quantify the self shielding in the assembly, the fissions due to interrogation flux only in each cell was calculated using fuel with 10% ^{239}Pu and 90% ^{235}U as the fissile material. The total fissions per source neutron were integrated over a slowing down time of 10 to 2000 μs and in resonance regions 1, 2 and 3 and is summarized in Table II. The fissions in each cell are also shown as a percentage of cell 16.

Table II. Fissions per source particle in each fuel cell due to interrogation flux only. Numbers displayed are absolute values and percent of the total in cell 16. The fissions are integrated over the entire collection time as well as resonance regions 1, 2 and 3. The fissile material consists of 90% ^{235}U and 10% ^{239}U.

Fissions in fuel - Total time					Fissions in fuel – Resonance 1			
1 0.0055 46.4%	2 0.0058 48.7%	3 0.0066 55.3%	4 0.0083 69.2%		1 0.00047 37.0%	2 0.00047 36.8%	3 0.00058 45.0%	4 0.00084 65.5%
5 0.0058 48.7%	6 0.006 50.1%	7 0.0068 56.9%	8 0.0087 72.6%		5 0.00048 37.2%	6 0.00045 35.1%	7 0.00054 42.2%	8 0.00085 66.1%
9 0.0066 55.4%	10 0.0068 56.8%	11 0.0077 64.5%	12 0.0098 81.8%		9 0.00057 44.6%	10 0.00054 42.5%	11 0.00065 50.6%	12 0.00096 75.3%
13 0.0083 69.3%	14 0.0086 72.4%	15 0.0097 81.6%	16 0.0119 100%		13 0.00085 66.0%	14 0.00084 65.5%	15 0.00096 75.2%	16 0.00128 100%
Total time – 0.123					Total Res 1 – 0.011			
Fissions in fuel – Resonance 2					Fissions in Fuel –Resonance 3			
1 0.00014 25.6%	2 0.00014 25.3%	3 0.00018 32.6%	4 0.00031 57.7%		1 0.00019 30.8%	2 0.00019 31.1%	3 0.00023 38.5%	4 0.00036 60.0%
5 0.00013 23.9%	6 0.00012 22.1%	7 0.00016 29.8%	8 0.0003 55.8%		5 0.00018 30.5%	6 0.00018 29.6%	7 0.00023 37.3%	8 0.00037 60.6%
9 0.00018 33.5%	10 0.00015 28.6%	11 0.0002 36.7%	12 0.00036 66.8%		9 0.00023 38.4%	10 0.00022 37.0%	11 0.00027 45.2%	12 0.00043 71.1%
13 0.00031 57.5%	14 0.00029 53.9%	15 0.00036 66.5%	16 0.00054 100%		13 0.00037 61.3%	14 0.00037 60.9%	15 0.00043 71.5%	16 0.0006 100%
Total Res 1 – 0.0039					Total Res 2 – 0.0049			

The flux is reduced as the neutrons travel through the assembly due to absorption in the fuel. The largest absorption is in cell 16 in the low energy region. The flux is reduced a full 35% from cell 16 to 11 and another 25% from cell 11 and 6 as the flux travels diagonally through the

assembly. Cell 1 has only a slight reduction in flux compared to cell 6 due to slow neutrons entering the cell from the surrounding lead. In resonance region 1, the self shielding is not as great as the lower energy resonances because of the lower cross section value. Resonance region 2 shows the greatest self shielding of nearly 80% from cell 16 to cell 6. The flux through the assembly is reduced significantly after passing through the first cell (16) and the absorption is the greatest at the ^{239}Pu resonances and at low energies. The reduction in the total flux is about 60% from cell 16 to 11.

Due to the large absorption in cell 16, a calculation was made with the fuel contents in cell 16 voided. In this case, the fuel content is decreased by 6.3%, but the fissions in the fuel is decreased by only 4.4% when integrating over the entire collection time, and only 1.8% in resonance 1 and 2 regions. This is due to the fact that the entire assembly sees a higher interrogation flux when cell 16 is voided, particularly in the resonances. It is important to note, that this phenomenon is a function of the source location and cannot be avoided by placing detectors around the entire perimeter of the fuel assembly. It is recommended that this calculation be rerun with detectors surrounding the assembly. Due to the increased fissions in the center cells and cells 15 and 12, any missing pins in cell 16 will not be easily detected.

6.4 Detector Response

Once the fissions are created in the fuel, the neutrons must travel through the fuel collision free to the ^{235}U detectors where the fission threshold is about 1 MeV. The detector response data is normalized to 5 ^{238}U detectors with 200 mg of ^{238}U each. Table III shows the total detector response to each fuel cell integrated over the entire slowing down time and for resonance regions 1, 2 and 3. The detector response is also shown as a percent of the response of cell 4 from which the most neutrons arrive at the detectors. In every case, the detectors see only 3 % of the neutrons from cell 13 compared to cell 4. This is mostly due to geometric attenuation of the fission neutrons which would cause the cell 13 response to be 5.3% of cell 4. The remaining 1.7% of the reduced response is due to fission neutron collisions in the fuel. This is a good result in that the fuel self shielding of fission neutrons is minimal can be considered negligible if the assembly is surrounded on all sides by detectors. This leads to the conclusion that detectors must be located around the entire perimeter of the fuel assembly. If this is the case, then cell 6 would be vulnerable cell to missing pins due to the lack of interrogation flux (50% of cell 16) combined with its distance from the detectors.

The self shielding causes a non-linearity in the detector response. The slowing down regions that show the greatest differences in reaction rate between ^{239}Pu and ^{235}U also see the least interrogation flux which reduces the fissions in the fuel. At the same time, the fissions that occur also create secondary fission in the fuel due to the tightly packed assembly. So the center pins fission more due to secondary neutrons, particularly in the resonance region. These two phenomena greatly complicate the problem of interpreting the detector response.

Table III. Detector response per source particle to each fuel cell and as a percent of the response of cell 4. The response is integrated over the entire collection time as well as resonance regions 1, 2, and 3. The fissile material consists of 90% ^{235}U and 10% ^{239}U. Results are x10^{-6}.

Detector Response x10^{-6} -Total time					Detector Response x10^{-6} - Region 1				
Det1	Det2	Det3	Det4	Det5	Det1	Det2	Det3	Det4	Det5
1	2	3	4		1	2	3	4	
0.079	0.258	0.463	0.496		0.0069	0.0228	0.0407	0.045	
16.0%	52.0%	93.3%	100%		15.3%	50.7%	90.5%	100%	
5	6	7	8		5	6	7	8	
0.051	0.106	0.168	0.183		0.0044	0.0088	0.0135	0.0167	
10.4%	21.4%	33.9%	37.0%		9.8%	19.6%	30.0%	37.2%	
9	10	11	12		9	10	11	12	
0.029	0.051	0.07	0.083		0.0025	0.0041	0.0059	0.0078	
5.9%	10.3%	14.1%	16.8%		5.6%	9.1%	13.2%	17.2%	
13	14	15	16		13	14	15	16	
0.016	0.025	0.033	0.041		0.0015	0.0024	0.003	0.004	
3.1%	5.1%	6.6%	8.2%		3.4%	5.3%	6.6%	9.0%	
Total time response – 4.307x10^{-6}					Total Region 1 response -0.380x10^{-6}				
Detector Response x10^{-6} – Region 2					Detector Response x10^{-6} – Region 3				
Det1	Det2	Det3	Det4	Det5	Det1	Det2	Det3	Det4	Det5
1	2	3	4		1	2	3	4	
0.0019	0.006	0.0131	0.0171		0.0028	0.0081	0.016	0.0211	
11.3%	35.1%	76.8%	100%		13.4%	38.6%	75.7%	100%	
5	6	7	8		5	6	7	8	
0.0014	0.0025	0.0045	0.0061		0.0016	0.0034	0.0057	0.0073	
8.1%	14.8%	26.3%	35.9%		7.7%	16.3%	27.1%	34.9%	
9	10	11	12		9	10	11	12	
0.0008	0.0014	0.0021	0.0029		0.001	0.0018	0.0025	0.0036	
4.8%	8.0%	12.1%	17.0%		5.0%	8.5%	12.1%	16.8%	
13	14	15	16		13	14	15	16	
0.0005	0.0008	0.0011	0.0016		0.0007	0.001	0.0013	0.0018	
3.1%	4.8%	6.4%	9.3%		3.1%	4.8%	6.4%	8.6%	
Total Region 2 response -0.128x10^{-6}					Total Region 3 response -0.160x10^{-6}				

7 MISSING FUEL PINS

To further investigate the sensitivity of the system, calculations were completed with fuel pins missing from various locations. Although these calculations include an array of detectors only across the top of the fuel assembly, the response to fuel missing in the center cells should be similar for all sides. Table IV shows the change in the detector response compared to the response with all pins present. When 2 pins are removed, 0.7% of the fuel is missing; therefore, it is important that the detector responses show at least a 0.7% decrease in fissions. The exception is the case of Cell 16 where all the pins are missing which is the equivalent to 5.5% of the fuel.

Table IV. Percent change in detector response due to missing fuel pins.

Location of Missing Pins	% fuel missing	Change in Detector Counts			
		Total time	Region 1	Region 2	Region 3
Center pins	0.7%	-1.6%	+0.003%	-26.9%	-12.2%
Cell 6 Top left corner	0.7%	-2.8%	-0.035%	-21.1%	-11.3%
Cell 13 Bottom left corner	0.7%	-2.3%	+0.03%	-23.7%	-5.0%
Cell 4 Top right corner	0.7%	-1.4%	-0.007%	-10.4%	-8.1%
All of Cell 16	5.5%	-0.9%	+0.125%	-21.1%	-11.3%

The results show that in all cases except resonance region 1, the decrease in detector response exceeds the decrease in fuel. This is due to the reduction in secondary fissions created by chain reactions in the assembly itself. Evidently, the pin worth in the assembly is greater than its individual worth. This fortunate increase in pin worth makes the missing pins much easier to detect. The exception is resonance region 1 where the fission cross section is lower than other resonance regions and secondary fissions are not as important. It is recommended that resonance region 2 counts be used for missing fuel detection because of the high pin worth in this resonance region. It is interesting to note that the response to the absence of 16 pins in cell 16 (which saw the greatest interrogation flux) creates the same response as 2 missing pins in cell 6 (which saw about 50% less interrogation flux than cell 16) . This is due to the lack of absorption of interrogation neutrons in cell 16 which creates a higher interrogation flux in the rest of the fuel.

This result means that the location of the missing pins also changes the detector response. Therefore, the location of the missing pin must be determined in order to next determine the missing quantity. Actual experiments have shown that the location of perturbations in fissile fuel quantities can be detected [6], but the density and size of the assembly was much smaller than the present case.

8 PLUTONIUM QUANTITY FITTING

To determine the quantity of ^{239}Pu in the fuel, additional data was taken with the fissile material in the fuel as 30% ^{239}Pu and 70% ^{235}U. The change in fissions in the fuel and detector response were observed and compared to the 10% ^{239}Pu fuel. Table V shows the fissions in the fuel from interrogation neutrons. The self shielding is slightly greater when compared to Table III showing the 10% ^{239}Pu fuel. The total fissions in the 30% ^{239}Pu fuel due to interrogation neutrons is 8.3% less than that of the 10% ^{239}Pu fuel. In resonance region 1, the fissions are only 2.2% greater, in the resonance region 2, the fissions are 6.7% greater and in resonance region 3, the fissions are unchanged. This surprising result is due to the increase self shielding in the fuel as the ^{239}Pu levels increase.

Table V. Fissions in the fuel per source particle due to interrogation flux only for fuel with 30% ^{239}Pu and 70% ^{235}U as the fissile material. Data is total integrated over entire collection time as well as resonance regions 1, 2 and 3.

Fissions in fuel - Total time					Fissions in fuel – Region 1			
1 0.0054 45.9%	2 0.0056 48.2%	3 0.0064 54.8%	4 0.008 69%		1 0.00049 36.8%	2 0.00048 36.4%	3 0.00059 44.5%	4 0.00087 65.7%
5 0.0056 48.1%	6 0.0058 49.4%	7 0.0065 56.2%	8 0.0084 72%		5 0.00048 36.2%	6 0.00045 33.7%	7 0.00055 41.2%	8 0.00086 64.9%
9 0.0064 55%	10 0.0066 56.3%	11 0.0074 63.8%	12 0.0095 81.1%		9 0.00059 44.5%	10 0.00055 41.6%	11 0.00066 49.9%	12 0.00099 74.3%
13 0.008 69%	14 0.0084 72.1%	15 0.0095 81.2%	16 0.0117 100%		13 0.00087 65.9%	14 0.00087 65.4%	15 0.00099 74.5%	16 0.00133 100%
Total time – 0.120					Total Region 1 – 0.012			
Fissions in fuel – Region 2					Fissions in Fuel – Region 3			
1 0.00014 24.6%	2 0.00014 24%	3 0.00019 31.7%	4 0.00034 57.3%		1 0.00019 30.5%	2 0.00018 30.2%	3 0.00023 38.2%	4 0.00037 60.4%
5 0.00014 23%	6 0.00011 19.5%	7 0.00016 26.6%	8 0.00032 54.2%		5 0.00019 30.7%	6 0.00017 28.4%	7 0.00022 35.3%	8 0.00037 60.7%
9 0.00019 32.8%	10 0.00016 27.3%	11 0.0002 33.9%	12 0.00038 64.7%		9 0.00023 37.9%	10 0.00022 35.9%	11 0.0027 43.6%	12 0.00043 70%
13 0.00033 56.5%	14 0.00032 53.8%	15 0.00039 65.9%	16 0.00059 100%		13 0.00037 60.2%	14 0.00037 60.3%	15 0.00043 70.4%	16 0.00061 100%
Total Region 1 – 0.0041					Total Region 2 – 0.0048			

In Table VI, the detector response to all fissions in the fuel, shows a different result than expected after looking at the fissions in the fuel due to interrogation neutrons only in Table V. The interrogation flux is overcome, depending on the amount of ^{239}Pu present and which resonance region is examined, by the larger number of fission neutrons creating secondary fissions in the fuel. The increase in the detector response is 3.4 % for fissions integrated over the total time, 9.3% in resonance region 1, 13.7% in resonance region 2, and 7.4% in resonance region 3. This indicates that secondary fissions are most important in resonance region 2.

Table VI. Detector response per source particle to each fuel cell and as a percent of cell 4 for 30% ^{239}Pu fuel. The response is integrated over the total collection time, and resonance regions 1, 2, and 3. Results are x10^{-6}.

Detector Response x10^{-6} –Total time					Detector Response x10^{-6} –Region 1				
Det1	Det2	Det3	Det4	Det5	Det1	Det2	Det3	Det4	Det5
1 0.082 15.8%	2 0.264 51.0%	3 0.48 92.7%	4 0.518 100%		1 0.0071 13.5%	2 0.236 44.5%	3 0.0434 82.0%	4 0.0529 100%	
5 0.053 10.2%	6 0.11 21.2%	7 0.171 33.1%	8 0.188 36.4%		5 0.0046 8.8%	6 0.0093 17.6%	7 0.0149 28.1%	8 0.018 33.9%	
9 0.031 5.9%	10 0.053 10.2%	11 0.073 14.0%	12 0.086 16.6%		9 0.0027 5.1%	10 0.0045 8.6%	11 0.0066 12.5%	12 0.0082 15.6%	
13 0.016 3.1%	14 0.026 5.1%	15 0.034 6.5%	16 0.042 8.1%		13 0.0016 3.1%	14 0.0025 4.6%	15 0.0034 6.4%	16 0.0044 8.2%	
Total time response – 4.453 x10^{-6}					Total Reg 1 response -0.415 x10^{-6}				
Detector Response x10^{-6} – Region 2					Detector Response x10^{-6} – Region 3				
Det1	Det2	Det3	Det4	Det5	Det1	Det2	Det3	Det4	Det5
1 0.0024 12.3%	2 0.0074 38.3%	3 0.015 77.2%	4 0.0194 100%		1 0.0028 13.0%	2 0.0089 41.5%	3 0.0186 87.1%	4 0.0214 100%	
5 0.0015 7.8%	6 0.0029 14.8%	7 0.0045 23.3%	8 0.0071 36.6%		5 0.0018 8.4%	6 0.0037 17.5%	7 0.0064 30.1%	8 0.0076 35.5%	
9 0.0009 4.9%	10 0.0015 7.6%	11 0.0021 10.6%	12 0.0032 16.4%		9 0.0011 5.3%	10 0.002 9.1%	11 0.0026 12.4%	12 0.0036 17.0%	
13 0.0006 3.1%	14 0.0009 4.8%	15 0.0012 6.2%	16 0.0019 9.7%		13 0.0007 3.2%	14 0.0011 5.1%	15 0.0014 6.7%	16 0.002 9.2%	
Total Reg 1 response -0.145 x10^{-6}					Total Reg 2 response – 0.172 x10^{-6}				

9 FITTING THE DATA

A simple linear fitting program was written to examine the ability to determine the quantity of ^{239}Pu in the fuel from collected data and fitting to benchmark calculations. The baseline measurements included the detector response to a fresh fuel assembly with the fissile material comprised of 100% ^{235}U, an assembly with the fissile material 100% ^{239}Pu, and the background of the ^{235}U detectors with the fuel pins voided (later determined to be negligible). The background was subtracted from the detector response for the ^{235}U and ^{239}Pu calculations to provide the baseline reaction rates for fitting purposes. A calculation was completed with an assembly of 10% ^{239}Pu and 90% ^{235}U, the background subtracted, and a fitting program was used to fit the 10% ^{239}Pu calculation to a linear combination of the 100% ^{235}U and 100% ^{239}Pu assembly results. The fitted result was 34% ^{239}Pu. The same calculations were made using only the resonance 1, 2, or 3 region data and are summarized in Table VII. The difference in the fuel and detector response result is caused by the self shielding of the interrogation flux and the

quantity of secondary fissions. The correlation of the quantity of fissile materials in the fuel, the type of fissile material, self shielding, secondary fissions and detector response is a complicated nonlinear relationship.

This brings into question of to what benchmark data will the measurements be fitted? The current calculations simply used fuel assemblies with 100% ^{235}U or 100% ^{239}Pu as the fissile material. This caused errors due to differences in the self shielding and secondary fissions in the assembly. Simply placing a probe detector into the fuel may not reflect the proper result due to the varied response in each cell. The relationship of flux in the cells and detector response to the quantity of ^{239}Pu in the fuel makes determining the expected response extremely complicated.

Table VII. Calculated quantity of ^{239}Pu in fuel assembly based on results from the total collection time, and resonance regions 1, 2 and 3.

Actual Quantity of ^{239}Pu	10% ^{239}Pu	30% ^{239}Pu
	Calculated Quantity ^{239}Pu	
Total	34%	66%
Res 1	21%	40%
Res 2	54%	53%
Res 3	60%	47%

It is recommended that future analysis includes a plot of the dependence of fissions in the fuel due to interrogation flux only, the total fissions in the fuel, and detector response as a function of ^{239}Pu quantities. This will be beneficial in the understanding of the behavior of the assembly.

Past measurements on mock spent fuel assemblies have determined ^{239}Pu quantities within 1% accuracy [7] when using least squares fitting. However, the density and size of the assembly was considerably smaller than the current calculations. The mock assembly consisted of 16 fuel pins in a 15 cm square assembly, and the calculated self shielding was only about 16% for this assembly verses up to 80% in the AP1000 assembly. A previous calculation on a 19 pin circular fuel assembly [9] was completed using a wide range of spent fuels with fissile content of up to 70%. The least squares fitting method produced an average error of 20% for ^{235}U and 43% for ^{239}Pu content. A neural network method was also investigated with errors of only 5%. These results, along with the present calculations, indicate that measurements of assemblies can become inaccurate when the assembly becomes too large, or the fissile content is a large percentage of the fuel. However, assembly measurements can be useful to find missing fuel content when an expected result is known.

Single pin measurements have been made in the past [10] which have determined Plutonium quantities with an accuracy of better than 0.2% if each rod was measured for 7 minutes. This may be the best method because, due to the nonlinearity of the assembly measurement system, accuracy will not meet 0.2% even with excellent statistics from a very time consuming run.

10 CONCLUSIONS

The AP1000 assembly assay system was modeled to simulate an actual measurement in RPI's LSDS. There have been no calculations or measurements for an assembly of this size to this point. The results indicate that self shielding of interrogation neutrons (up to 80%) is a serious issue in a larger assembly. This is further complicated by the variance in shielding with

incident neutron energy. The resonance regions which create the most absorption in the fuel also create the most secondary fissions in the fuel. The fission response is also position dependent. The more ^{239}Pu that is present in the fuel, the stronger the self shielding and secondary fissions in the fuel. This creates a nonlinear response at the detectors which greatly complicates fitting the data to the correct solution. Errors using only a linear fit of the ^{239}Pu quantity in these calculations were greater than 100%. This only means that the response needs to be studied further in order to create a suitable fitting procedure. It is recommended that additional calculations be made to fit the data to probe chambers within the fuel. Also, fissions due to interrogation neutrons should be compared to the total fissions in the fuel to better understand the fuel response.

It is still uncertain whether a large assembly such as the AP1000 assembly modeled in these calculations can be accurately assayed. Effort must be made to optimize the calculations and determine the best methods for fitting the data to benchmarks and what those benchmarks need to be. Also, neural networks methods of fitting may be the optimum method and should be investigated as well as other iterative approaches. Calculations should be completed with an array of ^{238}U threshold detectors around the entire perimeter of the fuel. The economics of single pin assay methods should be reexamined due to the uncertainties in the assembly calculations even with long measurement times.

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