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Determining Fissile Content in PWR Spent Fuel Assemblies Using a Passive Neutron Albedo Reactivity with Fission Chambers Technique

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Abstract

State regulatory bodies and organizations such as the IAEA that are concerned with preventing the proliferation of nuclear weapons are interested in a means of quantifying the amount of plutonium in a given spent fuel assembly. The complexity of spent nuclear fuel makes the measurement of plutonium content challenging. There are a variety of techniques that can measure various properties of spent nuclear fuel including burnup, and mass of fissile content. No single technique can provide all desired information, necessitating an approach using multiple detector systems and types. This paper presents our analysis of the Passive Neutron Albedo Reactivity-Fission Chamber (PNAR-FC) detector system. PNAR-FC is a simplified version of the PNAR technique originally developed in 1997. This earlier research was performed with a high efficiency, ^{3}He-based system (PNAR-3He) with which multiplicty analysis was performed. With the PNAR technique a portion of the spent fuel assembly is wrapped in a 1 mm thick cadmium liner. Neutron count rates are measured both with and without the cadmium liner present. The ratio of the count rate with the cadmium liner to the count rate without the cadmium liner is calculated and called the cadmium ratio. In the PNAR-3He technique, multiplicity measurements were made and the cadmium ratio was shown to scale with the fissile content of the material being measured. PNAR-FC simplifies the PNAR technique by using only a few fission chambers instead of many ^{3}He tubes. Using a simplified PNAR-FC technique provides for a cheaper, lighter, and thus more portable detector system than was possible with the PNAR-3He system. The challenge with the PNAR-FC system are two-fold: 1) the change in the cadmium ratio is weaker as a afunction of the changing fissile content relative to multiplicity count rates, and 2) the efficiency for the fission chamber based system are poorer than for the ^{3}He based detectors. In this paper, we present our research on using the PNAR-FC detector system to quantify the fissile content of a spent nuclear fuel assembly.

1 Introduction

Quantifying the mass of plutonium in commercial spent nuclear fuel assemblies is of great interest to countries and organizations such as the IAEA that are concerned with preventing the proliferation of

nuclear weapons. A substantial amount of the world's plutonium is contained within the spent fuel assemblies of commercial nuclear reactors; it would be ideal if this plutonium could be easily quantified and monitored so that diversions of the plutonium from the spent nuclear fuel for clandestine activities could be identified.

It is possible to analyze and quantify the isotopics of spent nuclear fuel by taking the spent fuel assembly apart, dissolving its components and performing mass spectroscopy to identify and measure the masses of the many isotopes contained in the spent nuclear fuel. While possible, the technique is slow and expensive thus ruling out any possibility of performing this kind of analysis for safeguards purposes.

An ideal situation for quantifying the mass of plutonium in spent fuel assemblies would be a non-destructive assay (NDA) technique that could be used in a variety of surroundings (air, water, borated water), and was fast enough so as not to be a burden on the other operations in a reactor, spent fuel, or reprocessing facility.

Under the sponsorship of the NSI program, an effort is underway (1) to combine a few NDA techniques into one integrated design allowing a nuclear inspector to measure the amount of plutonium in a spent nuclear fuel assembly and determine whether plutonium has been removed from the assembly. This paper reports one of these techniques, Passive Neutron Albedo Reactivity with Fission Chambers.

2 Passive Neutron Albedo Reactivity Technique

In the PNAR technique, a radioactive, multiplying sample is placed in a sample chamber. The sample chamber is surrounded by neutron detectors embedded in a hydrogenous, moderating material. Typically the sample chamber is also flooded with water or borated water. A thin, removable cadmium liner is placed between the sample chamber and the hydrogenous material. Two measurements are made of the sample; one with the cadmium present and one with the liner removed. The ratio of the neutron count *without* the cadmium liner to the ratio of the neutron count *with* the cadmium liner is called the cadmium ratio;

$$R = \frac{\omega_o}{\omega}, \tag{1}$$

where ω_o is the neutron count without the cadmium liner and ω is the neutron count with the cadmium liner. Since the emission, transport, and detection of neutrons is statistical in nature we must quantify the statistical uncertainty (standard deviation) of the cadmium ratio. With statistical uncertainties in the total neutron counts (σ_{ω_o} and σ_ω) the standard deviation of the cadmium ratio is

$$\sigma_R = \left[\frac{1}{\omega^2}\sigma_{\omega_o}^2 + \frac{\omega_o^2}{\omega^4}\sigma_\omega^2 \right]^{(1/2)}. \tag{2}$$

The PNAR technique uses intrinsic neutron emissions of the radioactive material to self-interrogate the fissile material. The source of neutrons in spent nuclear fuel comes from the spontaneous fission of ^{244}Cm (2) as well as some (α, n) reaction neutrons as well as the multiplication of these neutrons. In a light water reactor the probability of neutrons inducing fission in fissile isotopes is increased by moderating the neutrons through collisions with hydrogen.

In the PNAR technique, neutrons are also moderated to improve the probability of inducing fission. Some of the neutrons will be moderated by the water in the sample chamber while other neutrons will

escape the sample chamber, become moderated in the hydrogenous material surrounding the sample chamber, and then scatter back into the assembly and potentially induce fission. Cadmium has a large capture cross section for thermal neutrons, therefore the presence of the cadmium liner will absorb thermal neutrons leaving the sample chamber, and any neutrons that were thermalized in the hydrogenous material outside the sample chamber that are scattered back into the sample chamber.

The presence of the cadmium liner suppresses the thermal neutron flux and thus reduces induced fission and the multiplication of the sample. In essence, without the cadmium liner the radioactive material is being interrogated by a thermal neutron flux that isn't present when the cadmium liner is in the system.

2.1 PNAR with Fission Chambers

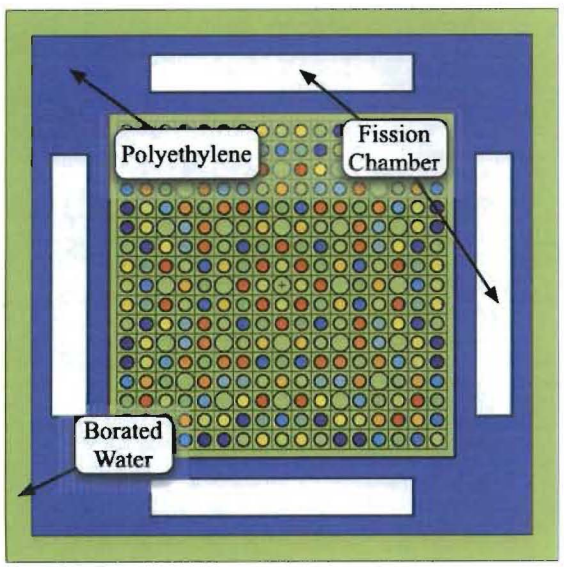
We have modified the PNAR (3) detector system design to measure spent nuclear fuel in commercial 17 x 17 PWR assemblies. The geometry of this system is depicted in Figure 1. In our modification we have made two major changes: reduced the number of neutron detectors to 4 and we are using fission chambers instead of ^{3}He tubes; the fission chambers have a radius of 1.26cm and a height of 17.22cm. This design is referred to as PNAR-FC (Passive Neutron Albedo Reactivity with Fission Chambers) and the system using ^{3}He tubes will be referred to as PNAR-3He. The fission chambers are placed perpendicularly to the long axis of the assembly and are embedded 5cm thick and 60cm high block of polyethylene. The detector system has been modified to be rectangular to accommodate the rectangular spent fuel assemblies. The full height of the assembly is modeled even though it is not shown in Figure 1c. The three figures (1a, 1b, and 1c) are not to scale.

The reduction of the neutron detectors from 80 as used in the PSMC to just four (although fewer would also be possible), makes PNAR-FC a much cheaper and lighter and more portable system than the PSMC used for PNAR-3He system. The drawback is that fission chambers are not high efficiency detector like ^{3}He tubes. With a reduced efficiency we can no longer perform multiplicity measurements, but as will be demonstrated later, there is sufficient efficiency to make measurement times reasonable.

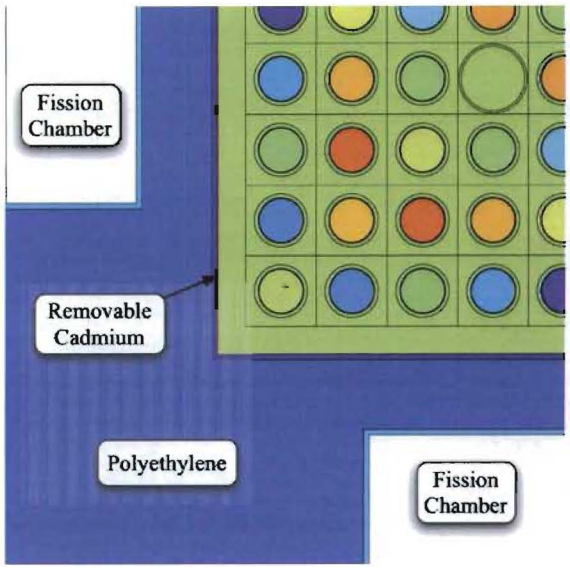
In the PNAR-FC design the detector system is open to the environment where the spent fuel assembly is located. Opening the detector system to the environment makes PNAR-FC applicable in a variety of environments. While we only present results from simulations in a borated water environment, we envision PNAR-FC being used in borated water, water and air environments.

2.2 Quantifying Fissile Content in a Spent Fuel Assembly

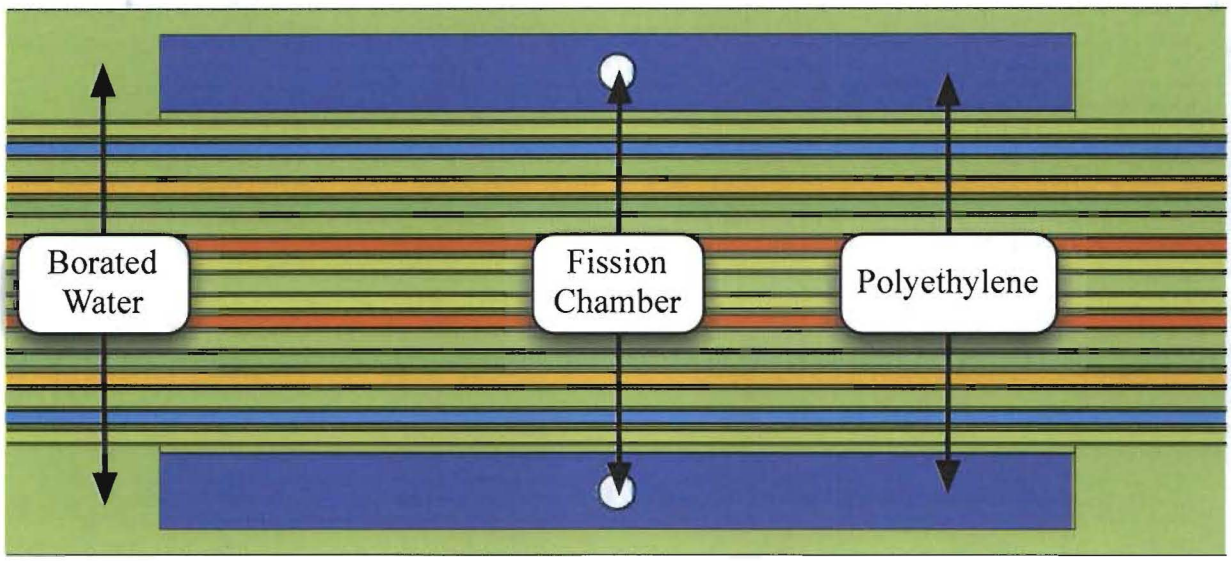
In their report, Menlove and Beddingfield (4) state that the cadmium ratio scales with the fissile content in the sample being measured. The samples they used were uranium oxide fuel pins containing either depleted uranium (0.2% ^{235}U) or 3.19% ^{235}U enriched uranium. They altered the fissile content by making varios combinations of depleted and enriched uranium fuel. The fissile content of these fuel pins is just the amount of ^{235}U in the fuel pins which is known from the enrichment. We have simulated a similar experiment using PNAR-FC for PWR assemblies with *fresh* uranium oxide fuel and found a similar trend in the cadmium ratio as a function of the fissile content (^{235}U enrichment) (3).



(a) XY slice.



(b) XY slice close-up.



(c) YZ slice.

Figure 1: Geometry of PNAR-FC detector system. The blue region surrounding the assembly is polyethylene, the white cylinders embedded in the polyethylene are the fission chambers. Thin purple region surrounding the assembly (only visible in Figure (b)) is the removable cadmium liner. The bright green color represents the material surrounding the assembly. For this paper, the surrounding material is borated water at 2200 ppm. Note the three figures are not to scale.

In spent nuclear fuel assemblies determining the fissile content is much more ambiguous than using known uranium enrichments. Two issues make this definition challenging: what fissile isotopes should be included in the definition and how much of those isotopes is in the spent fuel assembly. We have borrowed a concept from neutron coincidence counting (5, Chapter 16), *effective mass*. In our calculations we have defined the ^{239}Pu effective mass as

$$^{239}\mathrm{Pu}_{\mathrm{e}} = C_1 M_{U235} + M_{Pu239} + C_2 M_{Pu240}, \tag{3}$$

where M_x is the mass in grams of isotope x in the assembly and C_1 and C_2 are parameters defined as

$$C_1 = \frac{\text{Net neutron contribution from induced fission in } ^{235}\text{U per gram of } ^{235}\text{U}}{\text{Net neutron contribution from induced fission in } ^{239}\text{Pu per gram of } ^{239}\text{Pu}}, \tag{4a}$$

$$C_2 = \frac{\text{Net neutron contribution from induced fission in } ^{240}\text{Pu per gram of } ^{240}\text{Pu}}{\text{Net neutron contribution from induced fission in } ^{239}\text{Pu per gram of } ^{239}\text{Pu}}. \tag{4b}$$

With the definitions in this manner, the units of $^{239}\mathrm{Pu}_{\mathrm{e}}$are in grams of ^{239}Pu. Alternatively C_1 and C_2 can be written as

$$C_i = \frac{\left(\nu^{(i)} - 1\right)\sigma_f^{(i)} - \sigma_a^{(i)}}{\left(\nu^{(\mathrm{Pu239})} - 1\right)\sigma_f^{(\mathrm{Pu239})} - \sigma_a^{(\mathrm{Pu239})}}, \tag{5}$$

where ν, σ_f, and σ_a are all energy dependent. Using the definition of ^{239}Pu effective mass in Equation (3) we have a method of quantifying the fissile content of our spent fuel assemblies.

3 Numerical Results

To simulate the response of the PNAR-FC detector system we have modeled the detector system with a range of spent fuel assemblies surrounded by borated water. The spent fuel assembly models have initial enrichments of 2, 3, 4, or 5%; burnups of 15, 30, 45, or 60 GWd; and cooling times of 1, 5, 20, or 60 years. The combination of all of these parameters yields 64 assembly models and represent the range of spent fuel assemblies that exist in spent fuel pools around the world. These assembly models were created using Monte Carlo burnup calculations (6) and were developed for the NGSI effort of quantifying plutonium content in spent nuclear fuel assemblies.

For each spent nuclear fuel assembly model two simulations were performed using MCNPX with the PNAR-FC detector system; one with the cadmium liner and one without the cadmium liner. The source of neutrons for these results are from the spontaneous fission neutron emission from the spontaneous fission emitters in each pin of the modeled assembly. The number of fissions in the fission chamber were tallied in as well as the number of fission neutrons and absorptions in the fuel pins. Using these tallies and their reported uncertainties we were able to estimate the cadmium ratio as measured by the PNAR-FC and the parameters C_1 and C_2 for each model. In addition, we have tallied the fissions and absorptions in the fuel pins enabling us to estimate a ^{239}Pu effective mass described by Equations (3) and (5).

In Figure 2 the results of MCNPX simulations using the 64 spent fuel assemblies surrounded by borated water with and without cadmium are shown. The cadmium ratio is shown as a function of

the ^{239}Pu effective mass. The assembly parameters of burnup and initial enrichment are delineated in by the color and symbol of the data point. The cooling time dependence, while not differentiated by a difference in appearance can still be viewed from the Figure. Of the three isotopes included in Equation (3) only ^{241}Pu has a half-life comparable to the time scales of 1–80 years (14.4 year half-life for ^{241}Pu compared to half-lives of 7.04×10^8 years and 2.410×10^4 years for ^{235}U and ^{239}Pu respectively). ^{241}Pu is the only isotope in the ^{239}Pu effective mass equation that appreciably decays in 1–80 years, therefore the change in ^{239}Pu effective mass for a change in cooling time is a reduction in the mass of ^{241}Pu and a decrease in the ^{239}Pu effective mass.

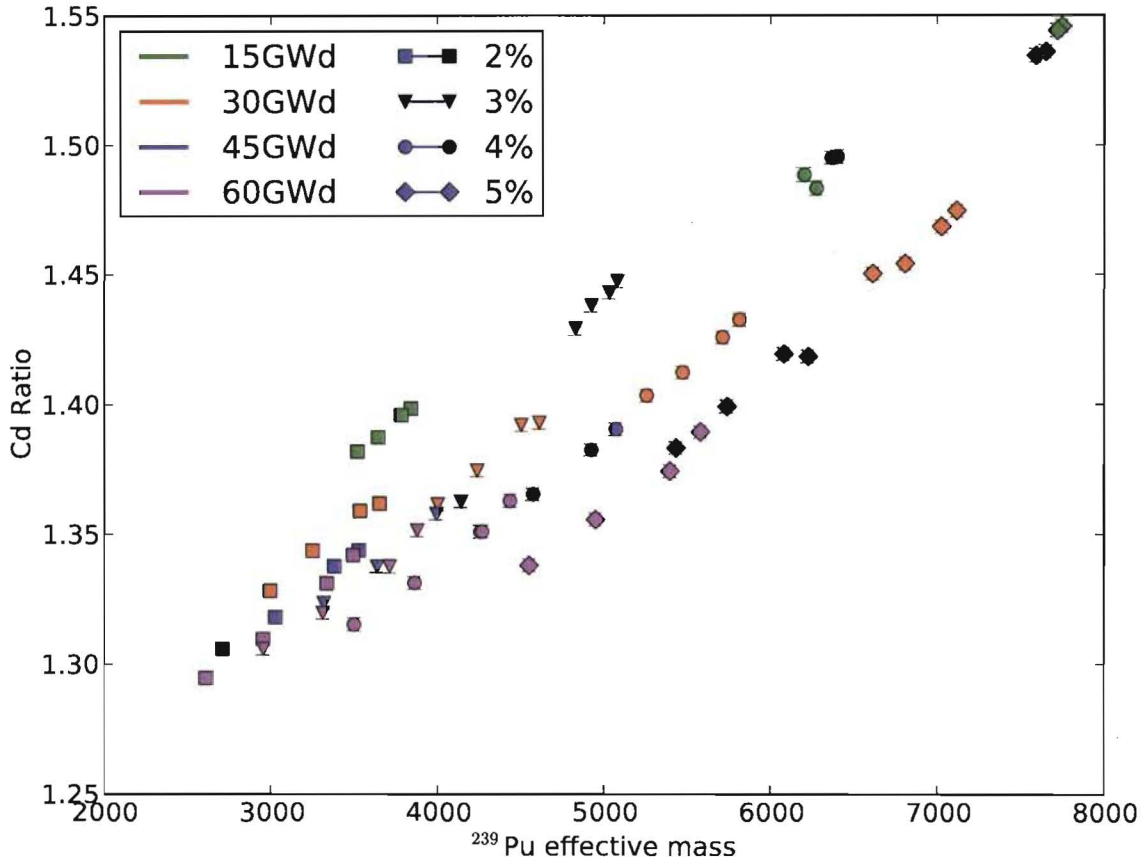


Figure 2: Cadmium ratio as a function of ^{239}Pu effective mass. The colors: green, red, blue and purple represent burnups of 15, 30, 45 and 60 GWd respectively. Symbols: square, triangle, circle, diamond represent initial uranium enrichments of 2, 3, 4 and 5% respectively. Cooling times of 1, 5, 20, and 80 years can be identified by knowing that the ^{239}Pu effective mass decreases with an increase in cooling time.

In Figure 2 we see there are some definite trends with respect to the burnup, initial enrichment, and cooling time parameters. We can see a decrease in the cadmium ratio as burnup increases. The decrease in the cadmium ratio is due to a combination of the increase in the neutron absorbers with burnup as well as the consumption of ^{235}U. We see a decrease in the cadmium ratio as a function

of both initial enrichment and cooling time. The cadmium ratio is a ratio of two measurements, therefore, to reduce the cadmium ratio the measurement made without the cadmium liner must be reduced more than the measurement made with the cadmium liner other wise the ratio would remain the same. Since the difference between the two scenarios is an interrogating thermal flux without the cadmium liner, we see that a decrease in the cadmium ratio is caused by a reduction primarily in the thermal multiplication of the system.

3.1 Limits of Detectability

The error bars in Figure 2 are the standard deviation of the estimate of the cadmium ratio. They represent an uncertainty of 0.2% in the cadmium ratio. Note the errorbars are the statistical uncertainties of the cadmium ratio estimated from the uncertainties of tallies of MCNPX simulations. The uncertainties in the MCNPX tallies are not the same as the standard deviation of the number of detector counts in real-world measurements, but are estimates of the statistical uncertainty in the tally itself. The precision of real-world measurements are limited by the statistical uncertainty in the counts measured by the detectors, but are also influenced by systematic errors.

3.1.1 Counting Statistics

In 2004, Menlove et al. (7) demonstrated that detection precision (for an ENMC using typical electronics for safeguards neutron detection) is limited by electronic stability; the minimum relative uncertainty for a single measurement is 0.0145%. This number was obtained after measuring for 28 hours in a laboratory setting; real-world precision is expected to be $\sim 0.05\%$ (8). We know that the relative uncertainty of a measurement of the number of counts in a detector is $1/\sqrt{N}$ with N being the number of counts in the detector. Setting this equal to 0.05% we find $N = 4 \times 10^6$ counts.

We have calculated the rate of spontaneous fission neutrons for a 45 GWd, 4% initial enrichment, with 5 year cooling time is 3.92×10^8 neutrons per second. The results of tallying the number of fissions in our fission chambers in MCNPX is 2.78×10^{-4} counts *per source neutron* without the cadmium liner and 2.02×10^{-4} counts per source neutron with the cadmium liner. The tally results are multiplied by the spontaneous fission activity of the assembly to determine the count rate in the fission chambers during a measurement. Table 1 shows the tally value per source neutron, the count rate, and the number of second required to measure 4×10^6 counts. From this we claim that with measurements of less than one minute we can reach the precision limit of the electronics of our detector system.

Cadmium	Counts per Source Neutron (10^{-4})	Counts per Second (10^4)	Seconds Required
with	2.017	7.907	50.6
without	2.789	10.93	36.7

Table 1: MCNPX tally results for number of counts *per source neutron* measured in PNAR-FC fission chambers and number of second required to reach precision limit of electronics.

3.1.2 Systematic Uncertainties

Systematic errors will be a more significant source of measurement uncertainty than counting statistics. The space between the spent fuel assembly is modeled as a 0.5 cm gap filled with borated water. This gap is necessary to account for slight variations in the dimensions of the assembly, but adds some variability in the position of the assembly with respect to the PNAR-FC detector system.

To estimate the systematic uncertainty we have simulated the response of the detectors by moving the assembly within the polyethylene sleeve. We ran three simulations of a spent fuel assembly; one centered simulation and two off-center:

1. assembly in center of polyethylene sleeve (centered),
2. assembly moved 0.5 cm up from center of polyethylene sleeve (up), and
3. assembly moved 0.5 cm up and 0.5 cm right from center of polyethylene sleeve (corner).

The movement "up" refers to the positive y-axis and "right" refers to the positive x-axis; the long axis of the assembly is parallel to the z-axis. For these three simulations, the assembly used has 45 GWd burnup, 4% initial enrichment, and 5 year cooling time parameters.

The two perturbations represent the maximum change that can occur. Due to the symmetry of the detector system, movement in any other fashion will either be equivalent to the above perturbations or the change in the cadmium ratio will be less. The result of these perturbations are shown in Table 2.

Simulation	Cadmium Ratio (0.0004)	Difference (0.0006)	Relative Difference (0.0004)
Centered	1.3833	0.0	0.0
Up	1.3851	0.0019	0.0014
Corner	1.3874	0.0041	0.0030

Table 2: Results of simulating systematic uncertainties in placement of the spent fuel assembly with-ing the PNAR-FC detector system. The number in parentheses represents the statistical uncertainty in the results in that column.

From the results of the perturbations we see that the biggest change to the cadmium ratio occurs when the assembly is moved to the corner of the polyethylene sleeve as expected. The difference in this instance to the normal simulation is 0.3% and 0.14% when the assembly is just moved up. While 0.3% is larger than the errorbars shown in Figure 2 we don't think that a systematic error of this magnitude alone will eliminate PNAR-FC from being effective as a tool for quantifying plutonium content in spent fuel assemblies. Also this can be eliminated by engineering a system that mechanically moves the polyethylene blocks so there is physical contact with the assembly.

The estimate of the limiting factors are either much smaller than the displayed errorbars in Figure 2 as in the case for counting statistics, or are approximately the same size as for systematic uncertainties of imprecise placement of the assembly in the detector. We think that the trends shown in Figure 2 will not be overshadowed by the uncertainties of the real-world measurements and therefore, we can use those trends to determine the response of our detector.

4 Future Work

Much work remains to be done to accurately quantify the plutonium mass using PNAR-FC detector system. While the data in Figure 2 appear to trend nicely, understanding what the data represents is a more complicated tasks. Ideally, a safeguards inspector would take a measurement and be able to read from a graph or figure what the associated ^{239}Pu effective mass would be. With our current scenario, this isn't possible.

Similar efforts (9, 10) for other detector systems have tried to make corrections to the count rates and ^{239}Pu effective mass to push all the data onto a single curve. The hope would then be to write an analytical formula for this curve based upon given parameters of burnup and cooling time and calibration data. Then when an inspector records a count rate, the ^{239}Pu effective mass could be calculated directly. While this approach can work, it is proving to be more complicated than originally expected, no doubt a result of the complexity of spent nuclear fuel.

We are working on other techniques to simplify working with the array of data associated with the gamut of potential spent nuclear fuel assemblies. On idea being considered is the use of multiple calibration curves. While using multiple calibration curves adds somewhat to the complexity, we anticipate that we will not have to employ artificial correction factors to make the data easily understandable and immediately applicable to the safeguards inspector.

Another component not considered here is the sensitivity of the PNAR-FC detector system to potential diversion of material. If PNAR-FC is insensitive to diversion scenarios then it is not very useful for safeguards inspectors as the very thing they are trying to prevent would be undetectable with this system. Further work still needs to be done to quantify the PNAR-FC sensitivity to diversion scenarios.

5 Acknowledgements

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