Determining Plutonium in Spent Fuel with Nondestructive Assay Techniques

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Abstract:

There are a variety of motivations for quantifying plutonium in used (spent) fuel assemblies by means of nondestructive assay including the following: shipper/receiver difference, input accountability at reprocessing facilities and burnup credit at repositories or fuel storage facilities. Twelve NDA techniques were identified for providing fuel assembly composition information.[1] Unfortunately, none of these techniques, in isolation, is capable of determining the Pu mass in an assembly. However, it is expected that the Pu mass can be quantified by combining a few of the techniques. Determining which techniques to combine and estimating the expected performance of such a system is the purpose of a research effort recently begun. The research presented here is a complimentarily experimental effort. This paper will focus on experimental results of one of the twelve non-destructive assay techniques - passive neutron albedo reactivity. The passive neutron albedo reactivity techniques works by changing the multiplication that the pin experiences between two separate measurements. Since a single spent fuel pin has very little multiplication, this is a challenging measurement situation for the technique. Singles and Doubles neutron count rate were measured at Oak Ridge National Laboratory for three different burnup pins to test the capability of the passive neutron albedo reactivity technique.

Keywords: spent (used) fuel; nondestructive assay

1. Introduction

Although the majority of plutonium (Pu) in the world is stored in commercial spent (used) fuel assemblies, a measurement system for directly quantifying the Pu mass contained in these assemblies does not exist. The nondestructive assay systems in use today (Cerenkov Viewing Device,[2] Fork Detector[3] and Safeguards Mox Python Detector[4]) essentially measure indirect signatures from spent fuel such as gamma emission from fission fragments, or photons induced by radiation from fission fragment, or total neutron emission that pre-dominantly is emitted from curium. Calculation codes, known as burnup codes, can be used to infer plutonium mass from these measured signatures. In order to use burnup codes to predict the Pu mass in a particular assembly, input from the operator is required. From an international safeguards perspective, this input is undesirable given the regulatory requirement of independent verification.

Below, nine reasons for improving on the status quo are listed. These reasons are the motivation for designing a nondestructive assay (NDA) system that can quantify the Pu mass in spent fuel assemblies: (1) Provide regulators with the capability to independently verify the mass of plutonium at any site that has spent fuel. (2) Enable regulators and facilities to accurately quantify the Pu mass leaving one facility and arriving at another facility (“shipper/receiver difference”). (3) Provide confidence to the public that the shipment of spent fuel around the world is being undertaken in a rigorous manner; assure that material is not diverted during shipment. (4) Provide regulators with a tool for recovering continuity of knowledge at any site storing spent fuel. (5) Provide reactor operators with a tool enabling optimal reloading of reactor cores. (6) Provide regulators of once-through fuel cycle repositories the capability to optimally pack fuel both for transport, in a pool and into the repository ("burnup credit"). (7) Enable determination of the input accountability mass of an electro-chemical (pyro-chemical) processing facility. (8) Provide facility operators with a means for quantifying the Pu mass in spent fuel that is no longer considered "self-protecting." This is particularly relevant given that some regulatory agencies are considering changes to the level at which radioactive material is considered to be self-protecting. And (9) promote cost savings by facilitating assembly selection for reprocessing since facility operators need to combine assemblies to obtain optimal compositions in reprocessing solutions. The blending is presently based on reactor history and burnup codes. The inaccuracy of the status quo decreases plant operational efficiency.

For the purpose of measuring the Pu mass in spent fuel assemblies, 12 NDA techniques were identified.[1] The subject of this paper is one of those techniques – the passive neutron albedo reactivity technique (PNAR). The research presented here adds to recent publications on PNAR[5, 6, 7] in that it is the first time a single spent fuel pin was measured with the PNAR technique.

2. Passive neutron albedo reactivity

2.1. Basic concept

The PNAR technique functions by using the intrinsic neutron emission of the fuel (primarily from the spontaneous fission of curium) to self-interrogate the fissile material in the fuel itself.[5] Two separate measurements of the spent fuel are made, and the ratios of the count rates obtained are analyzed; this ratio is called the cadmium ratio. The primary difference between the two measurements is the neutron energy spectrum and fluence in the spent fuel. By varying the material around the spent fuel, a high and a low neutron-energy-measurement condition is produced. The PNAR technique can be used with total neutrons (singles) and/or doubles and/or triples; it is expected that doubles will produce the best result in the high count-rate regime.[6] If the geometry of the measurement situation is unchanged between two measurements, the change in the cadmium ratio between measurements of different pins or assemblies may be calibrated to a change in the fissile content of the pin or assembly.

One approach to producing these two energy conditions involves measuring the spent fuel with a thin layer of cadmium surrounding it and a second measurement without the cadmium around the fuel. The presence of the cadmium effectively eliminates all neutrons below 1 eV from reflecting back into the fuel from the detector walls, altering the in-leakage reactivity contribution. Hence, in the measurement made with no cadmium present, the fuel is interrogated by all the neutrons reflected back to the fuel. In contrast, when cadmium is present, the fuel is interrogated by only those reflected neutrons with energies above 1 eV. Effectively, how far the ratio deviates from ~1 indicates the impact thermal neutrons.

In order to illustrate this concept further, the results of an earlier publication[5] are reproduced in Fig. 1.

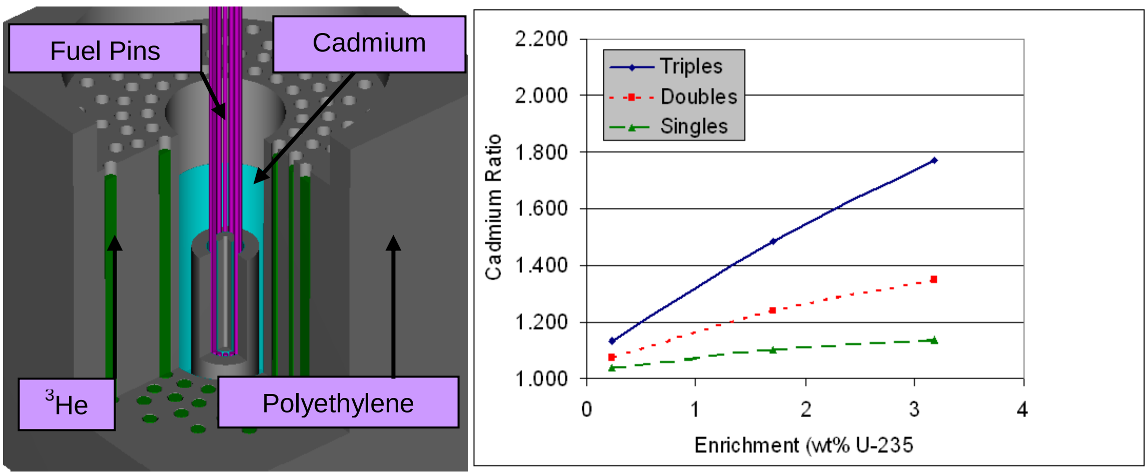

Fig.1. On the left an experimental setup used in a publication by Menlove and Beddingfield is illustrated. On the right, data obtained with the experimental setup is presented.

An interesting ramification of using the neutrons from the spent fuel to interrogate the spent fuel is that the statistics get better for the singles as the inherent neutron source in the spent fuel becomes more intense as compared to many other NDA techniques which need to detect a signal on top of the background.

It is noteworthy that the measurement of one pin is a very sub-optimal geometry for using the PNAR technique. This is due to the fundamental fact that the PNAR concept depends on the change in multiplication between two measurements. There is very little multiplication in one pin, hence the change in multiplication is very small.

2.2. Detector design

The detector utilized in these experiments was a modified Inventory Sample Neutron Coincidence Counter (INVS) detector (16 ^{3}He tubes, 4 atmospheres of pressure, 4 preamplifiers). The cross section and an exterior view of the detector are depicted in Fig. 2. The modification involved the following: (1) removing the polyethylene inside of the ^{3}He tubes, (2) fabricating three concentric inserts, from largest to smallest: lead, poly and cadmium, (3) covering the top and bottom of the detector with lead, (4) covering the entire detector in cadmium. The cadmium liner close to the fuel is removable in order to enable the PNAR technique. The reason the lead and the polyethylene layer are in the order they are, is to maximize the impact of removing the cadmium liner on the neutron energy spectrum in the pin. As modified, the detector is 30% efficient to californium neutrons emitted from the center of the chamber when the cadmium liner is in place. In order to keep the gamma dose to the ^{3}He tubes to an acceptable level, an iron shielding structure was fabricated around the detector; the detector inside the iron shield is depicted in Fig. 3.

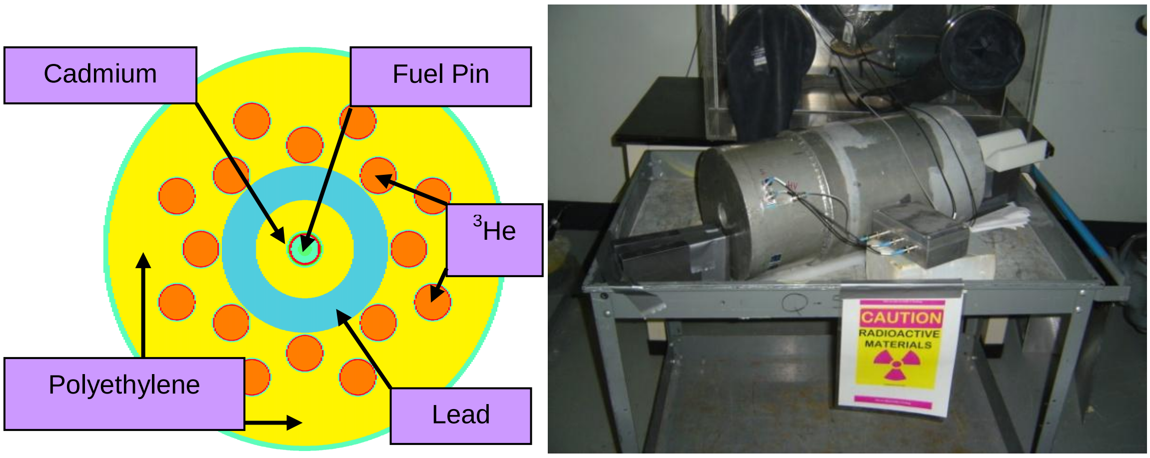

Fig. 2. On the left, the cross section of the detector is illustrated. On the right, an external view of the detector is depicted.

The detector system was designed to allow full length fuel pins to be measured as well as pin segments. In order to measure full length pins, the iron shielding depicted in Fig. 3 was designed to fit over the mechanical fuel drive system used to manipulate full length pins at Oak Ridge National Laboratory (ORNL). The two support structures depicted in Fig. 3 were designed to center the fuel for the measurement of partial length fuel pins. The support was designed so that the Cd liner could be moved in and out of the detector without moving the fuel.

Fig. 3. The iron shielding and fuel pin support structures are depicted.

2.3. Experimental operation

2.3.1 Measurement opportunities

There were three measurement opportunities: May and July of 2008 and January of 2009.

2.3.2 Data acquisition

During the May 2008 measurements, the signal from the detector went to two Advanced Multiplicity Shift Registers (AMSR). The difference between the two AMSRs was only the duration of the gate; one was set to 64 μs while the other was 128 μs. In the later two experimental campaigns, the signal was split as well. However, in order to enable more versatile data analysis, one signal went to an AMSR with a 128 μs gate while the other signal went to a list mode data acquisition system.

The following settings were used for all the data presented in this paper unless stated otherwise: predelay of 4.5 μs, 128 μs gate, 180 ns multiplicity deadtime, deadtime coefficient A of 0.72, deadtime coefficient B of 0.13, doubles gate fraction of 0.65. These values were determined from measurements of ^{252}Cf sources of variable strength and experience with similar detectors.

2.3.3 Description of spent fuel

The fuel measured for this research was at ORNL as part of research being performed for other sponsors. For this reason, the fuel was cut up into segments. The segments used in this publication are listed in Table 1; these segments were between 60 and 75 cm long. The fuel segment for which data is depicted were at least 60 cm from the end of the pin to assure that each pin segment had a relatively uniform axial burnup and to assure that the burnup of the pin segment was close to the average burnup of the assembly.[8, 9] The details of the fissile content calculation are given in section 3.1

Pin Segment	Reactor	Burnup (GWd/tU)	Initial Enrichment	Pin Location	Fissile Content (g/cm)
591D	Surry-2	36.0	3.11%	H9	0.186
616B	Three Mile Island - 1	50.9	4.00%	D5	0.193
651B	North Anna	68.4	4.12%	B16	0.147
652B	North Anna	71.6	4.12%	D5	0.147

Table 1, the pin segment labelling system used by those cutting the pins is listed along with the reactor from which the fuel came, the approximate burnup, and the initial enrichment. The pin location refers to the location of the pin within an assembly.

In order to protect the cladding, each fuel pin segment was placed inside of a steal cylinder. One consequence of this measurement situation is that the fuel had two possible means by which to move within the steel cylinder. The pellets could move within the cladding. Also the cladding could move within the protective steel cylinder.

2.3.4 Experimental procedure

Given operational constraints, it was not possible to use the mechanical structures designed to center the fuel pin in the detector. This situation means that the fuel had to be moved between each measurement. The intent had been that only the Cd liner was moved between a "*with Cd*" and "*without Cd*" comparison. This movement means that the change in the count rate between two such measurements was due to statistical variation, change in the fissile content as well as a change in positioning. In order to minimize the possibility of the fuel moving inside the cladding or support structure, the fuel was tilted before putting it into the detector to cause any mobile fuel to go to the end of the pin.

Since the fuel measured was in the form of pin segments and since the segments were inside of a protective cylinder and since it was possible for the fuel might move inside the protective cylinder, the location of the fuel was not certain. In order to determine the location of the fuel with the neutron detector, the fuel was pushed through the detector in 5 cm intervals so that the center of the pin could be found.

The first fuel measured was the 70 GWd/tU fuel and had the most intense gamma dose. It was so intense that it was necessary to lower the bias voltage of the ^{3}He tubes to prevent gamma pile up from registering as neutron counts. The detector was 30% efficient for a voltage of 1680 V. For the operating voltage of 1560 V, it was 19% efficient.

3. PNAR Experimental data and analysis

3.1. Data quality check

In order to gain confidence that the detector was operating properly and to confirm that the neutron intensity scaled with burnup as expected, the neutron intensity detected is graphed in Fig. 4 as a function of burnup, or more precisely, as a function of exposure. It is expected that the singles (or total) neutron count rate will scale as the burnup raised to the third or fourth power for burnups above 10 GWd/tU.[10] This scaling is due to the fact that ^{244}Cm is produced at this rate as well as to the fact that ^{244}Cm is responsible for over 95% of the spontaneous fission neutrons for fuel that is cooled for more than 4 years. Slight corrections were made for cooling time and variations in the fuel cross sectional area among the three fuel types. Note that the statistical error bars are much smaller than the width of the data points; a typical uncertainty for the singles count rate was 0.05% as determined by analyzing the scatter in the measured count rate. The scatter was determined by breaking every count time into numerous subintervals. No correction was made for variations in initial enrichment or multiplication since the impact of these factors does not prevent the overall purpose of Fig. 4 which is to gain confidence in the singles neutrons data relative to the documented burnup of each pin.

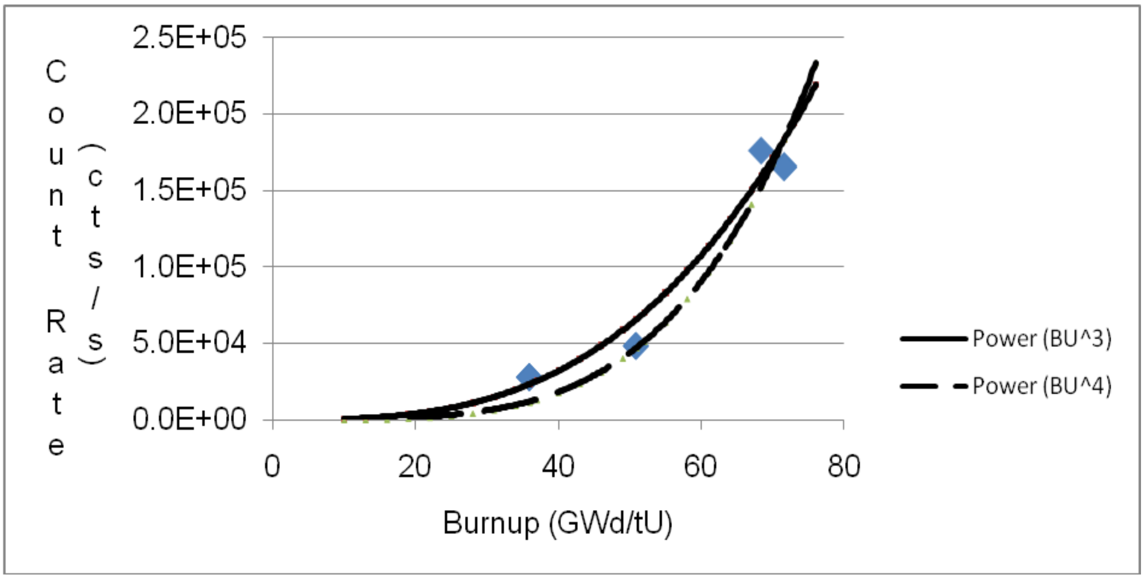

Fig. 4, Singles count rate as a function of burnup. The four data points are for 4 different pins. The two curves indicate the count rate if this rate varied with the burnup to either the third (solid line) or fourth (dashed line) power.

The relationship between the burnup and the singles count rate in Fig. 4 agrees roughly with the expected power scaling. Note that the burnup values used were the values declared by the reactor operators. There was no effort made to correct for variations in the burnup within an assembly. Furthermore, since the fuel came from three different reactors, it is not clear how consistent the techniques for determining burnup were, nor if the timing of assemblies in the reactor were the same. If greater accuracy in burnup is needed, detailed burnup modelling could be done.

The PNAR technique measures fissile content. It does this by changing the energy spectrum of the neutrons reflected back into the fuel. Hence, the PNAR technique is measuring the change in multiplication between two measurements – provided no other factors changed. Another indicator of multiplication is the ratio of doubles to singles count rates for a given measurement. It is interesting to look at this independent measure of multiplication to gain confidence in the data set. In Fig. 5, the ratio of the doubles to singles is given for 14 separate measurements of four separate pins.

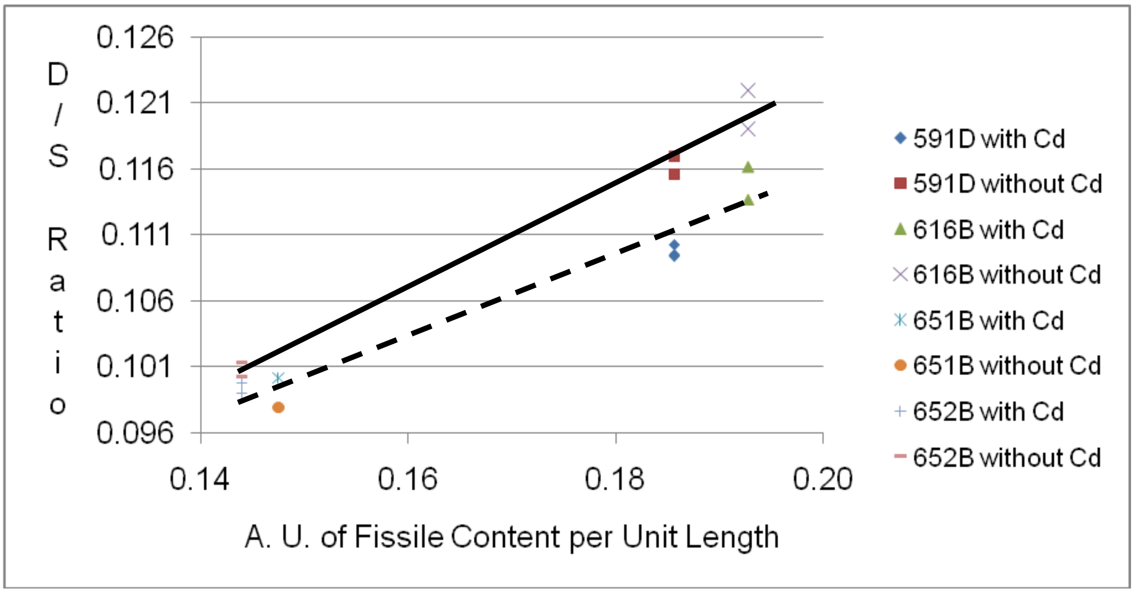

Fig. 5, the ratio of the doubles to singles count rate as a function of fissile content is illustrated for 4 different pins. Each pin was measured with a Cd liner about it (*with Cd*) and with the Cd liner removed (*without Cd*).

The fissile content per unit length illustrated in Fig. 5 was determined by starting with the declared uranium and plutonium fissile mass and isotopic concentrations for ^{235}U, ^{239}Pu and ^{241}Pu. The declared mass of each fissile isotope was multiplied by the respective thermal cross sections (586 b, 748 b, 1,013 b, respectively) to approximate the fission rate of each isotope (assuming the same neutron flux to each isotope). The thermal cross section is what is of interest with PNAR since the count rate from the non-thermal portion of the spectrum is nearly identical in both the numerator and the denominator of the Cd ratio. In order to weight the fissile content by the number of neutrons produced per isotope, the normalized fission rate was multiplied by the number of neutrons produced per fission (2.41, 2.88, 2.80, respectively) to get the fissile content of a pin. Finally, the total fissile content was divided by the length of each pin to obtain the fissile content per unit length. The lines in Fig. 5 were inserted to roughly separate data points when the Cd liner was not present (solid line) from the cases when the Cd liner was present (dashed line).

In Fig. 5, the lowest fissile content was from the highest burnup cases of ~70 GWd/tU (pins 651B and 652B). In order to discern the data separately from each of these pins, a slight shift in fissile content was made in the graphing. The middle fissile content pin was for the lowest burnup case up of 36 GWd/tU (pin 591D). The highest fissile content was for pin 616B with a burnup of 51 GWd/tU. The one sigma variation in the D/S ratios was dominated by the statistics of the doubles for which the data points as graphed are ~2 sigma (+/- 1 sigma) in width. In the case of 616B (no Cd liner, 10 minute live count time divided into 20 s subintervals), the D/S ratio determined from averaging the three separate measurements of this pin was 0.1210 +/- 0.0003 (Singles = 52,569 cts/s +/- 11.16, Doubles = 6,361 cts/s +/- 16.94).

The primary conclusion from Fig. 5 is that it is possible to detect an increase in multiplication. For 3 of the 4 pins, or 6 of the 7 measurement pairs, the D/S ratio increased with the removal of the Cd liner. The lines that have been inserted were done so to indicate the expected separation between data points when the Cd liner was in place and those when the Cd liner was not present. This is expected since the multiplication should be greater without the Cd liner in place. Pin 651B is the exception. The variation in the S/D for repeat measurements was a little greater than statistics would predict.

Only with pin 651B was the D/S ratio greater with the Cd liner in place than with it absent. It is expected that this result is due to more than just repositioning the pin in the detector. The two data points for this pin are separated by 4 sigma. Based on the other data depicted in Fig. 5, it is expected that the two D/S ratios for pin 651B would have been reversed and separated by a few sigma. Instead they are separated by 4 sigma in the opposite direction. Perhaps the fuel moved within the steal support structure or some other error was made. Note that the D/S ratio is different from the Cd ratio in that both D and S are measured at the same time, hence there is no positioning uncertainty. Yet when comparing two D/S values, there is a positioning uncertainty. It is relevant to note that as a nuclei that fissions moves out of the detector, the probability of producing a doubles count falls off more rapidly than the singles count since the doubles count rate varies as efficiency squared while the singles count rate falls off as the efficiency.

Note that the D/S ratio data in Fig. 5 could be used to quantify the fissile content. If used in this way, one would not need a movable cadmium liner. This approach appears to have greater dispersion and sensitivity than the singles or doubles Cd ratio data for the presently implemented detector. Since the results *without Cd* give a great dispersion, not using a Cd liner would be a preferable means of using this approach.

3.2. Passive neutron albedo reactivity results

In Fig. 6 and Fig. 7 the cadmium ratio as a function of fissile content per unit length is depicted for singles and double count rates, respectively. The data for four pins is depicted. The multiple data points for each pin were determined by taking all possible ratios of *without Cd*/*with Cd*. For example, there are 6 data points for pin 616B since it was measured 3 times *without Cd* and 2 times *with Cd*. Since the goal of presenting the data this way is to indicate the degree of scatter in the data due to positioning, data points were only used if the pins were moved before measurements.

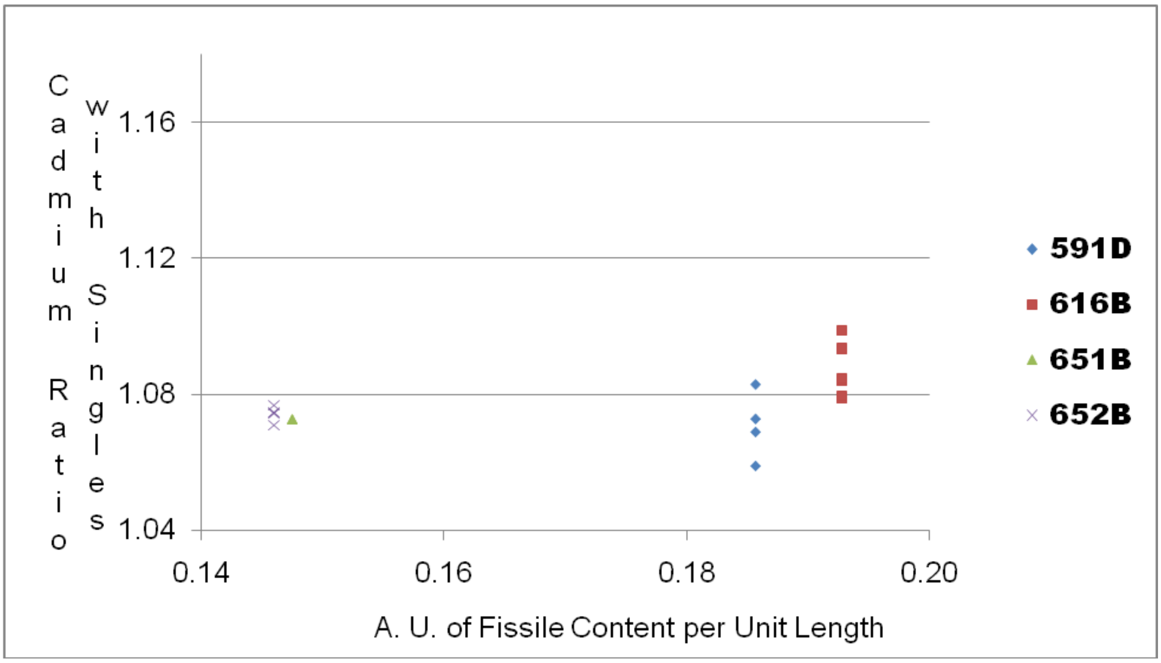

Fig. 6, Cd ratio as a function of fissile content for single (total) neutron counting is illustrated

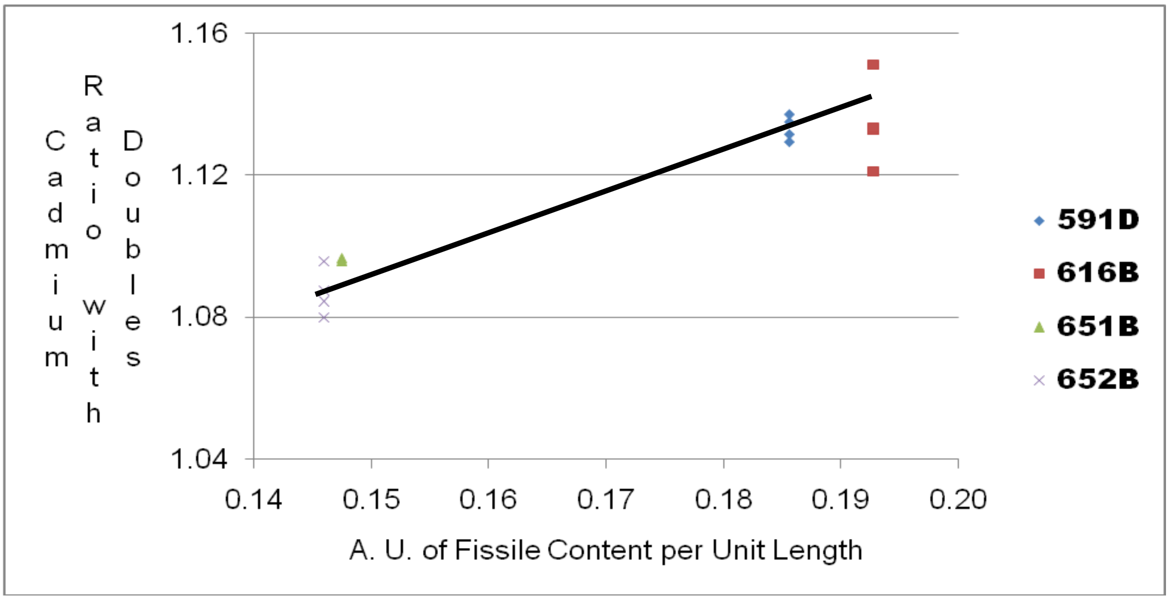

Fig. 7, Cd ratio as a function of fissile content for double neutron counting is illustrated.

Before interpreting Fig. 6 and Fig. 7, it is important to discuss the uncertainties involved. For the singles count rates data of Fig 6, the one sigma uncertainty determined from the scatter in the count rate varied from 0.01% to 0.06%. For the poorest statistical case (pin 591D), the singles ratios was 1.0728 +/- 0.0009. Hence, each data point as graphed in Fig. 6 is ~3 sigma wide. For the doubles rates in Fig. 7, the one sigma uncertainty determined from the scatter in the count rate ranged from 0.2% to 0.6%. For the poorest statistical case (pin 651B), the doubles ratios was 1.097 +/- 0.008. Hence, each data point is ~0.3 sigma wide.

For the singles Cd ratio depicted in Fig. 6, a positive slope with fissile content may exist but the spread in the data for each individual pin is so large that it is rather unlikely that the singles Cd ratio is of much use for quantifying the fissile content in a pin for the experimental setup as it was implemented. It is expected that centering the pin in a more reproducible way and using longer fuel that could not possible move around would improve the singled Cd ratio. It is expected this poorer performance of singles relative to doubles is due to the following two factors: (1) the dispersion in the singles Cd ratio as a function of fissile content is less as depicted in Fig. 1. As a result, the singles Cd ratio will be more sensitive to positioning changes. (2) Variation in the location of the pin in the axial direction of the detector will produce a greater variation in the singles Cd ratio than the doubles Cd ratio since the doubles count rate falls off more rapidly along the axis as you move out of the detector (i.e. the neutrons coming from outside the active region of the detector will be "filtered" out more effectively by doubles counting than singles). This is because the doubles count rate varies as efficiency squared while the singles count rate fall off as the efficiency.

For the doubles Cd ratio depicted in Fig. 7, a positive slope is clearly present; the current detector as used can detect a change in fissile content for the range of commercial fuel measured. Both improved positioning of the fuel and longer fuel that does not have the possibility of moving will likely improve the results further. However, for the present detector and experimental procedure, the dispersion in the data points for an individual pin is such that a given Cd ratio value corresponds to a wide range of fissile content. To quantify this point, for the three pins that were measured multiple times, the spread in the doubles Cd ratio between the extreme points depicted in Fig. 7 was equivalent to a 2, 3, and 4 sigma variation for pins 591D, 652B, and 616B, respectively. Hence, statistical variation is significant for the doubles Cd ratio even if the uncertainties from experimental procedure were improved.

Counting longer will not improve the results much for the current system. The overnight measurements and several hour measurements were not much better than the ten minute measurements. Drifting in the singles count rate over time was evident in the overnight measurements, because the reduced high voltage was below the plateau for the ^{3}He tubes.

4. Future work

Future work can take one of two paths: (1) research that could be done with the present detector system and (2) research that builds on the lessons learned with the current detector with the focus of designing a new more sensitive detector.

Regarding use of the present system, one could measure full fuel pins. This would minimize the uncertainty due to positioning. Another option is to analyze the list mode data collected to date and possible acquire new list mode data to see if novel analytical techniques can improve the sensitivity of the current detector.[11]

Regarding research that builds on the lessons learned from this work. This path has been developed in the document by Menlove et al.[11] The challenge of applying PNAR in the very non-ideal situation of a single pin resulted in the development of a novel analysis approach and detector design that uses list mode data acquisition to obtain greater fissile content sensitivity. This new detector has a long neutron lifetime near the fuel and a short neutron lifetime near the ^{3}He detectors where the fuel could be either individual pins or full assemblies. The name for this new approach is differential die-away self-interrogation (DDSI). Both DDSI and PNAR are expected to produce significantly better results for a full fuel assembly as compared to single pins because of the increase in multiplication.

5. Summary

A passive neutron detector was modified to enable spent fuel pins to be measured using the passive neutron albedo reactivity technique. This technique uses a movable cadmium liner to change the multiplication in the fuel between two measurements so that the fissile content of the fuel can be quantified by using the inherent neutron emission of the fuel itself to interrogate the fuel. The measurements were made at ORNL with fuel that ranged in burnup from 36 to 70 GWd/tU. From the ratio of the doubles to single count rates, it was clear that a change in multiplication is detected. Both the ratio of the doubles to singles count rates and the doubles Cd ratio allow the fissile content to be quantified experimentally with the ratio of the doubles to singles count rates providing the best results for the detector as designed and implemented. The singles Cd ratio did not provide a good indicator of fissile content. This is thought to be due to the greater sensitivity of this ratio to systematic uncertainties in the experimental procedure. In the context of future work, measuring full length pins would allow some of the experimental uncertainties (moving the fuel between *with Cd* and *without Cd* measurements, uncertainty in the location of fuel, centering of fuel) to be minimized. A novel detector design and analysis approach called differential die-away self-interrogation was conceived in the course of this work that is expected to improve the sensitivity of a passive neutron detector for quantifying the fissile content of either spent fuel pins or assemblies.

6. Acknowledgements

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