Nondestructive Determination of Plutonium Mass in Spent Fuel: Preliminary Modeling Results using the Passive Neutron Albedo Reactivity Technique - 10413

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ABSTRACT

There are various motives for quantifying plutonium (Pu) in spent fuel assemblies by means of nondestructive assay (NDA) including the following: strengthening the capability of the International Atomic Energy Agency (IAEA) to safeguard nuclear facilities; quantifying shipper/receiver difference; determining the input accountability value at pyrochemical processing facilities; providing quantitative input to burn-up credit assessments and final safeguards measurements at a long-term repository.

In order to determine Pu mass in spent fuel assemblies, thirteen NDA techniques were identified that provide information about the composition of an assembly. A key motivation of the present research is the realization that none of these techniques, in isolation, is capable of both (1) quantifying the Pu mass of an assembly and (2) detecting the diversion of a significant number of rods. It is therefore anticipated that a combination of techniques will be required. A 5 year effort funded by the Next Generation Safeguards Initiative (NGSI) of the U.S. DOE was recently started in pursuit of these goals. The first two years of the plan calls for researching all thirteen techniques using Monte Carlo modeling while the final three years involves fabricating hardware and measuring spent fuel. Here, we present the work in two main parts: (1) an overview of this NGSI effort describing the motivations and approach being taken; (2) The preliminary results for one of the NDA techniques - Passive Neutron Albedo Reactivity (PNAR).

The PNAR technique functions by using the intrinsic neutron emission of the fuel (primarily from the spontaneous fission of curium) to self-interrogate any fissile material present. Two separate measurements of the spent fuel are made, one with and one without a cadmium (Cd) layer surrounding the fuel assembly. The ratios of the Singles, Doubles and Triples count rates obtained in each case are analyzed; known as the Cd ratios. The primary differences between the two measurements are the neutron energy spectrum and fluence rate in the spent fuel, along with some impact on the detection efficiency which is a consequence of the change in the emerging spectrum. The presence of the Cd layer surrounding the spent fuel assembly hardens the neutron energy spectrum. If the geometry of the measurement situation is unchanged between the two measurements, the change in the Cd ratio between these two measurements can be related to the reactivity of the fuel assembly which in turn can be related to the fissile content.

INTRODUCTION – OVERVIEW OF THE NGSI SPENT FUEL EFFORT

A 5 year research effort is being sponsored by the Next Generation Safeguards Initiative (NGSI) of the US Department of Energy (DOE) with the overall goal of designing an integrated non-destructive assay (NDA) system to both (1) quantify the mass of elemental plutonium (Pu) in spent fuel and (2) detect the diversion of material from spent fuel assemblies.

Motivations for Quantifying the Mass of Pu in Spent Fuel

Research into the NDA of spent fuel is driven by safeguards needs. Five principal needs for quantifying the mass of Pu in spent fuel have been identified in Tobin, *et al*. [1] and can be summarized by the following:

- **Independent Verification as requested by the IAEA** - To verify the content of spent fuel without depending on information from the facility. New spent fuel measurement techniques have the potential to allow the IAEA to recover continuity of knowledge and to possibly detect diversion.

- **Shipper/Receiver** – Determining Pu mass at shipping and receiving sites to assure regulators that all of the nuclear material of interest leaving a nuclear facility actually arrives at another nuclear facility. Given the large stockpile of nuclear fuel at reactor sites around the world, it is clear that in the coming decades, spent fuel will need to be moved to either processing facilities or storage sites. Safeguarding this transportation is a vital part of the peaceful expansion of nuclear energy currently underway. Another key potential application of PNAR is “finger-printing”, as an alternative to quantitative Pu mass determination, to form the basis of shipper/receiver measurements. Finger-printing by the PNAR technique uses six primary NDA signatures: Singles, Doubles and Triples Cadmium (Cd) ratios to uniquely identify the fuel assembly, and confirm no changes have taken place during transport. Changes may indicate an attempt to divert material for example.

- **Self-protecting** – To quantify the Pu in spent fuel that is not considered “self-protecting”. Fuel is considered self-protecting by some regulatory bodies when the dose that the fuel emits is above a given level rendering it unattractive for certain diversion scenarios because the chemical separation would require specialist facilities. If the fuel is not self-protecting, then the Pu content of the fuel needs to be determined and the Pu mass recorded in the facility’s accounting system. This subject area is of particular interest to facilities that have research-reactor spent fuel or old light water reactor (LWR) fuel. It is also of interest to regulators considering changing the level at which fuel is considered self-protecting**.**

- **Input Accountability for future Electrochemical Processing Facilities** – It is not expected that an electrochemical reprocessing facility will have an input accountability tank, as is typical in an aqueous reprocessing facility. As such, one possible means of determining the input accountability value is to measure the Pu content in the spent fuel that arrives at the facility. Such a measurement could serve the dual purpose of forming part of a shipper/receiver accountability system.

- **Burn-up Credit** – To fully understand the composition of the fuel in order to efficiently and safely pack spent fuel into storage sites or a long-term repository. Spent nuclear fuel contains neutron poisons and a fissile content different than fresh fuel. Burn-up credit recognizes this so that worst case criticality assumptions traditionally made in storage ponds can be relaxed, increasing capacity. This is an important economic consideration to maintaining the existing fleet of nuclear power plants.

Adopted Approach

Thirteen techniques are being researched to form part of a comprehensive and integrated NDA system for spent fuel:

- Passive Neutron Albedo Reactivity (PNAR)
- Differential Die-Away Self-Interrogation (DDSI)
- Neutron Multiplicity (NM)
- Total Neutron (Gross Neutron) (TN)
- ^{252}Cf Interrogation with Prompt Neutron Detection (CIPN)
- Delayed Neutrons (DN)
- Differential Die-Away (DDA)
- Lead Slowing Down Spectrometer (LSDS)
- X-Ray Fluorescence (XRF)
- Delayed Gamma (DG)
- Nuclear Resonance Fluorescence (NRF)
- Passive Gamma (PG)
- Self-Interrogation Neutron Resonance Densitometry (SINRD)

Attributes of these techniques are described in a recent publication by Tobin, *et al*. [1]. Each of these thirteen techniques yields different information related to the composition of a discharged fuel assembly, for example; comparing two neutron techniques - PNAR measures total (Singles) and time correlated neutrons (Doubles and Triples) for two different fuel reactivity states enabling fissile content to be determined. To date, this technique can measure the sum of the ^{235}U and the Pu fissile mass (predominantly ^{239}Pu), but not the separate components. The method of DDSI, on the other hand, can be used to separate the ^{235}U from the ^{239}Pu [2].

No single technique can adequately quantify the Pu mass of a spent fuel assembly for the purposes identified earlier. It is therefore envisaged that a combination of techniques will be required, forming an integrated NDA system.

Research into an integrated NDA system will be conducted over two phases:

Phase I: Modeling (2 years)

The first two years of the research effort involves both (1) evaluating the performance of each technique individually for the NDA of spent fuel using analysis and simulation such as Monte Carlo modeling and (2) researching the optimum way to combine the promising techniques, followed by evaluating the overall performance of an integrated NDA system for quantifying the mass of elemental Pu in spent fuel assemblies and the ability to detect a range of diversion scenarios.

Phase II: Measurements (3 years)

The final three years involves fabricating hardware and measuring spent fuel to demonstrate capability predicted in the earlier evaluation and design phase.

Spent Fuel Library

Phase I modeling is based on a library of 17 x 17 Westinghouse PWR spent fuel assemblies [3]. 64 spent fuel assemblies have been modeled with the following range of parameters:

- Initial Enrichment: 2, 3, 4, 5 %
- Burn-up: 15, 30, 45, 60 GWd/tU
- Cooling Time: 1, 5, 20, 80 years

A fourth aspect will be investigated at a later stage:

- Diversion Scenarios (e.g. pin diversion)

The case with 4% initial enrichment, 30 GWd/tU and 5 years cooled was selected as one case of interest, relevant for safeguards applications from a diversion point of view.

Measurement Scenarios of Interest

Spent fuel assemblies may be stored in the following media in fuel cycle facilities:

- Water
- Borated Water
- Air

The performance of an integrated NDA system will be studied for each of these listed media configurations. For PNAR, the neutron energy spectrum and therefore fluence will change between a measurement in air, water and borated water. A measurement in water will increase multiplication within the spent fuel assembly. Borated water introduces a thermal neutron absorber to the measurement. A measurement in air reduces the multiplication in the spent fuel assembly, compared to water, because the spectrum in the fuel assembly is comparatively harder

so the overlap with the product of the fission cross-section and neutron yield is less and the escape probability is higher.

Instrument Calibration

The final instrument must be calibrated to enable inspectors to verify fissile mass. Calibration involves developing relationships between the observable quantities and the properties of the fuel assembly of interest. Simulations allow a broad range of conditions to be studied and are invaluable for helping construct the calibration relations. Validation and normalization against actual characterized fuel assemblies is equally important to establish confidence in the calibration procedure. The research goal for each technique is therefore to obtain a full calibration curve to quantify the fissile content of spent fuel assemblies (sum of the masses of fissile isotopes) from the detector response. One approach is to relate contributions from each fissile isotope in terms of an effective mass of ^{239}Pu and assign factors to weight each isotope accordingly. Weighting factors or co-efficients will be functions of burn-up and will be mode specific.

PASSIVE NEUTRON ALBEDO REACTIVITY (PNAR)

Passive Neutron Albedo Reactivity (PNAR) is a concept for spent fuel Pu verification as proposed by Menlove, *et al*. [2]. The basis of this technique is outlined by Tobin, *et al*. [1]. The NDA signature for PNAR is fissile content.

The neutron production rate in commercial spent fuel cooled > 2 years is dominated by ^{244}Cm spontaneous fission. The PNAR technique uses this intrinsic neutron emission to self-interrogate the fissile material in the fuel itself. Both the total neutron count rate (Singles) and count rates derived from time-correlated neutrons (Doubles and Triples) may be detected. Two separate measurements of the fuel are made; with and without Cadmium (Cd) surrounding the fuel assembly. The primary difference between the two measurements is the neutron energy spectrum and fluence rate in the spent fuel. This enables count rates to be measured for two different energy spectra in the fuel.

In the presence of Cd, neutrons below ~1eV reflected from the albedo structure surrounding the fuel assembly are removed from the flux. Without Cd, low energy neutrons are present and can interrogate the fuel. In other words, a high and a low-energy-measurement condition can be produced. The high absorption cross-section at low energies of the fissile isotopes means a greater number of fission events are induced in the fuel in the case without Cd. Additionally a change to the detection efficiency of neutrons born in the fuel assembly will take place due to the change in geometry and absorption properties.

Cadmium Ratio

Each mode may require individual optimization of counting parameters to obtain the best counting precision. The Cadmium (Cd) Ratio is defined as the detector response or count rate (Singles, Doubles or Triples) when no Cd liner is present at the center of the detector assembly divided by the count rate obtained when Cd is present. The Singles, Doubles and Triples Cd ratios vary with fissile content.

Fissile Content

The PNAR concept is to quantify fissile content by responding to a weighted sum of ^{235}U, ^{239}Pu and ^{241}Pu. PNAR cannot distinguish between contributions to the neutron fluence from the individual isotopes. In future work, it is hoped more information can be gained from the ratios of Doubles/Singles Cd ratios and potentially Triples/ Doubles Cd ratios. Currently we are looking at the potential of a prototypical instrument via Monte Carlo modeling in order to assess the sensitivity (response) to the variables of interest in spent fuel (such as burn-up and the compositional changes that brings about).

DETECTOR GEOMETRY

Research on the techniques of both PNAR and neutron multiplicity counting is being conducted alongside that on the technique of Differential Die-Away Self-Interrogation (DDSI). Figure 1 depicts the MCNPX [4] model of the detector geometry developed for DDSI by Menlove, *et al*. [2].

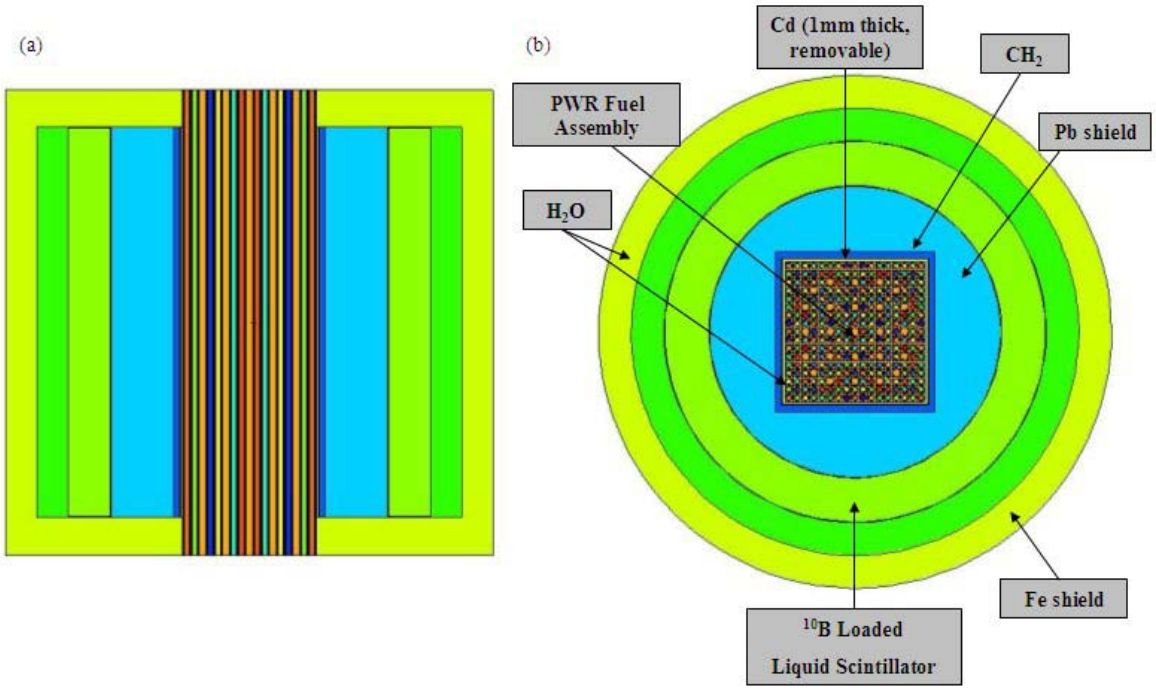

Figure 1. (a) Vertical cross-section of the detector and spent fuel assembly configuration. (b) Horizontal cross-section of the detector and spent fuel assembly configuration.

This geometry was also chosen for initial research into PNAR. The neutron emission rate from ^{244}Cm from the section of a typical single spent fuel rod positioned within the detector assembly (400mm active length) is ~ 5 x 10^5 ns^{-1} [2]. It is favorable to use a detector with a short die-away time so that short coincidence gate widths can be used to reduce accidental pileup without loss of signal. A boron loaded liquid scintillator (BC-523A, ~5% ^{10}B) was chosen for preliminary work

on PNAR due to the fast die-away characteristics, compared to moderated ^{3}He proportional counters. The benefit comes about because the scintillator acts as its own moderator and so once neutrons have been slowed they are available for absorption in the boron immediately, whereas in ^{3}He proportional counter based systems the ^{3}He tubes sample the flux established in the moderator surrounding them. Additionally, the signal forming process in the scintillator is much faster than in gas filled proportional counters. Later work will include a comparison of these two types of detector for spent fuel measurements using PNAR.

A removable Cd sleeve surrounds the fuel assembly, enabling measurements to be conducted with and without Cd. The lead (Pb) shield is in place to shield the liquid scintillator from the intense gamma-ray emission from the spent fuel assembly. The PWR spent fuel assembly itself can be seen in Figure 1 at the center of the detector assembly.

SIMULATIONS USING MCNPX

Neutron capture events via ^{10}B(n, α) reactions were tallied in the liquid scintillator detector using the coincidence capture tally in MCNPX. Sixteen base cases of full fuel assemblies were simulated. An initial ^{235}U enrichment of 4% was used in each case, with the following range of burn-ups: 15, 30, 45 and 60 GWd/tU and cooling times: 1, 5, 20, 80 years cooled. Simulations were conducted both with and without Cd at the center of the detector assembly in order to derive Cd ratio values in each case. Initial values of 4.5 μs and 64 μs were used for the pre-delay and coincidence gate width, respectively, which are in line with ^{3}He based systems but will likely need optimizing for the boron loaded liquid scintillator.

Single Rods vs. Full Fuel Assemblies

Due to the high neutron multiplication, which involves the creation of fast (fission) neutrons, the technique is able to penetrate the whole assembly and is therefore applicable to the measurement of both full assemblies and single rods since the multiplication of a fuel assembly is much greater. The performance of the technique is expected to improve for the measurement of full fuel assemblies, compared with measuring single rods. High neutron multiplication is expected to provide a larger difference in the Doubles and Triples Cd ratios as a function of fissile content. The measurement of full fuel assemblies also provides more useful quantitative information for the application of the technique to safeguards verification than the measurement of single rods, because in general a complete fuel assembly is an item which will remain intact and subject to inspection.

PRELIMINARY RESULTS

Here, preliminary simulation results for the PNAR technique are presented. The final metric will be fissile mass, however here Cd ratios are plotted as a function of burn-up and cooling time to better understand the detector response with gross characteristics of the spent fuel assembly. At this stage we are more concerned with proof of principle than deriving detailed calibration relationships. We also recognize that numerous practical challenges will have to be addressed before a PNAR instrument can be deployed.

Spent Fuel Assemblies

Figure 2 shows the Singles, Doubles and Triples Cd ratios for full fuel assemblies as a function of burn-up. The Cd ratios are a measure of fissile content in the fuel and therefore the PNAR response is related to the sum of the ^{235}U mass and the Pu fissile mass. Cd ratio values are greater than unity due to the contribution to the neutron emission rate from induced fissions within the fuel assembly i.e. enhanced neutron multiplication when the Cd is removed. There is an offset that comes about by the change in efficiency for neutrons born in the water filled fuel assembly when the Cd is removed (since neutrons below the Cd cutoff energy (~0.5 eV) can now emerge).

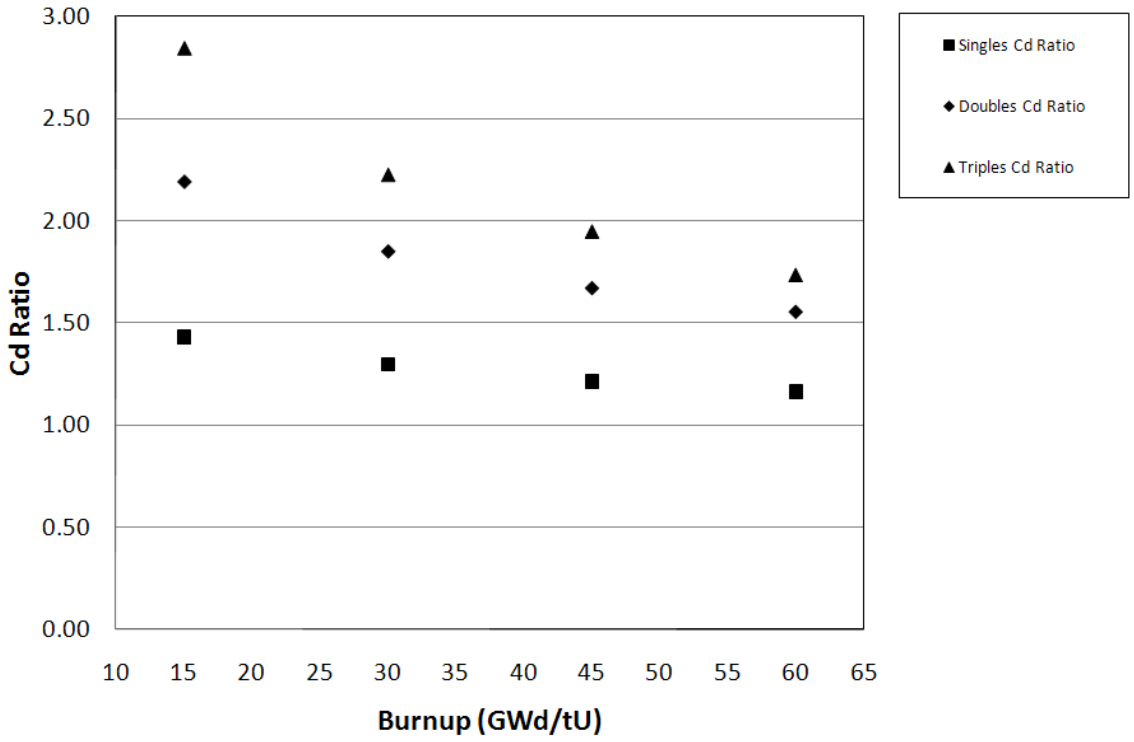

Figure 2. Fuel assembly Singles, Doubles and Triples Cd Ratios as a function of burn-up, for the case using 4% initial ^{235}U enrichment, 5 years cooled and measured in water. Uncertainties are being investigated, but preliminary results indicate the trends shown.

The dominant contribution to the decrease in the Cd ratios with burn-up is thought to be the decrease in the mass of fissile ^{235}U in the fuel assembly with burn-up. Table 1 below shows the masses of the fissile isotopes of interest in the fuel assembly modeled as a function of burn-up.

	^{235}U Mass (g)	^{239}Pu Mass (g)	^{241}Pu Mass (g)
15 GWd/tU	11840	1985	198.1
30 GWd/tU	7058	2597	545.2
45 GWd/tU	3855	2712	785.9
60 GWd/tU	1903	2663	900.2

Table 1. U and Pu isotopic mass for fissile isotopes of interest as a function of burn-up at 4% initial ^{235}U enrichment.

Singles, Doubles and Triples ratios all trend downwards with burn-up reflecting the decrease in reactivity with exposure. Intuitively one expects Singles rates to be least sensitive to multiplication and Triples to be the most sensitive, qualified by the data in Figure 2.

Below is Figure 3 which shows the neutron multiplication, M, both with and without the Cd sleeve surrounding the spent fuel assembly, as a function of burn-up. The decrease in the difference between the Triples Cd ratios and Doubles Cd ratios, and between the Doubles Cd ratios and Singles Cd ratios can be attributed to the decrease in multiplication within the spent fuel assembly. Multiplication is sensitive to the isotopic composition of the fuel and is therefore sensitive to the information of importance to our stated safeguards goals.

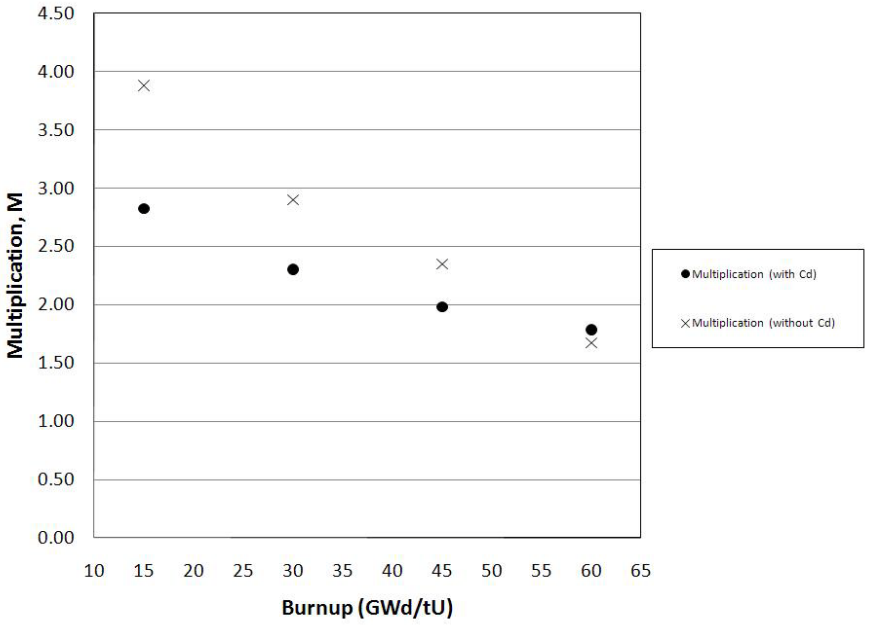

Figure 3. Neutron Multiplication (with and without Cd) as a function of burn-up.

Below is Figure 4 which shows the Singles, Doubles and Triples Cd ratios as a function of cooling time.

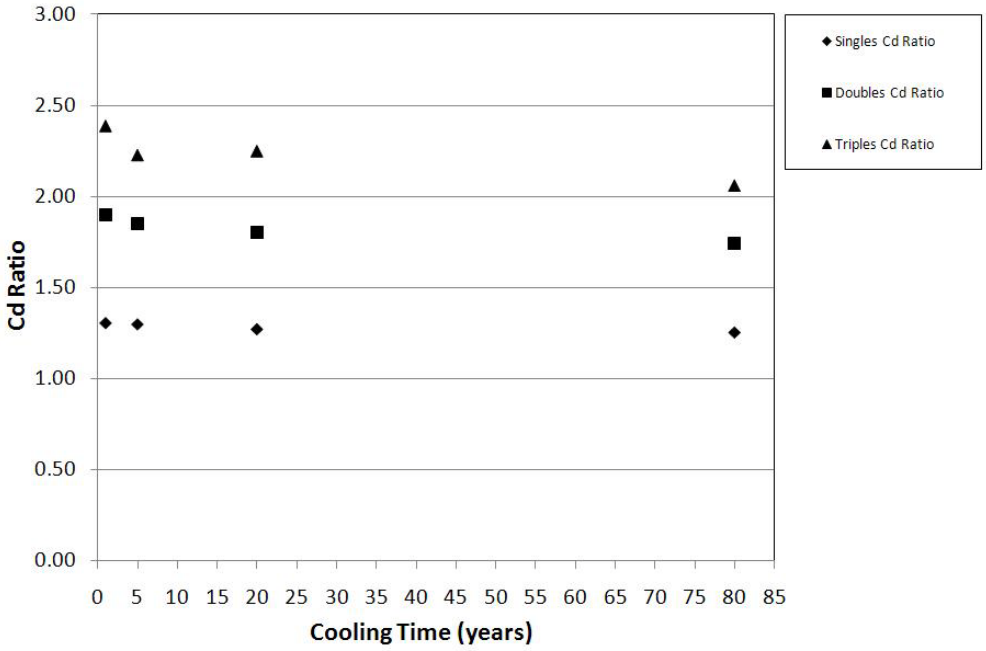

Figure 4. Singles, Doubles and Triples Cd Ratios as a function of cooling time, for a full spent fuel assembly with 4% initial enrichment, 30 GWd/tU burn-up, measured in water.

The mass of ^{241}Pu present in the spent fuel assembly decreases with cooling time (half life ~ 14.3 years). However, the mass of the fission product ^{155}Gd present within the spent fuel assembly increases with cooling time and this effect is dominant.

PHYSICS ASPECTS FOR FURTHER RESEARCH

Performing actual measurements requires a feasible detector and the optimization of the set up. In this work, a boron-loaded liquid scintillator has been assumed. Establishing the optimum gate width over which to perform shift register analysis will be an important aspect since this controls the rate of accidental correlations (Doubles and Triples) on the Cd ratio values over the selected range of both burn-up and cooling time since the neutron emission rate will vary. Factoring realistic detector behavior will be important. This work will include a full gate width optimization study to determine the trade-off between maximizing the discrimination ratio or Cd ratio in this case and the uncertainty in this ratio due to accidental correlations. From this work, conclusions will be drawn as to whether Triples counting is a useful signature for the assay of spent fuel. Accidental pile-up may compromise precision unless the detector can cope with the dynamic range.

Simulations will be used to quantify the variation in the Cd ratios with fissile content. Initial studies will be conducted using simple fuel and varying the quantity of each fissile isotope individually. All calculations performed using a liquid scintillator detector will be repeated using ^{3}He by way of comparison; since ^{3}He proportional counter based systems are familiar in the safeguards arena and can serve as a convenient benchmark of our models.

ACKNOWLEDGEMENTS

The authors would like to acknowledge DOE-NE and DOE-NA for funding this research. The authors would also like to thank Stephen Croft (LANL) for his valuable time and input.

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