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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 a variety of motivations 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 burnup credit 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 involves 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, both with and without cadmium (Cd) present. The ratios of the Singles, Doubles and Triples count rates obtained in each case are analyzed; known as the Cd ratio. The primary differences between the two measurements are the neutron energy spectrum and fluence in the spent fuel. By varying the thickness of the cadmium layer surrounding the spent fuel, a high and a low neutron-energy-measurement condition can be produced. The neutron detectors can be used to detect total neutrons (Singles) and/or Doubles and/or Triples. 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 attributed to a change in the fissile content of the sample.

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 principle motivations 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 for IAEA** - To verify the content of spent fuel without depending on unverified information from the facility, as requested by the IAEA. New spent fuel measurement techniques have the potential to allow the IAEA to recover continuity of knowledge and to possibly detect diversion.
- **Shipper/Receiver** – 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 of significant interest.
- **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. 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 an Electrochemical Processing Facility** – To determine the input accountability value at an electrochemical processing facility (e.g. future pyro-chemical processing facility). 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.
- **Burnup Credit for a Repository** – To fully understand the composition of the fuel in order to efficiently and safely pack spent fuel into a long-term repository. The composition of the fuel must be well understood in order to quantify how the repository will function over long time scales. The NDA of spent fuel can be part of a system that cost-effectively meets the burnup credit needs of a repository.

Adopted Approach

Thirteen techniques are being researched to form part of a 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), and Self-Interrogation Neutron Resonance Densitometry (SINRD).

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 an 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, 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].

It has been identified that no single technique can achieve the end goals of both quantifying the Pu mass of a spent fuel assembly and detecting the diversion of a significant number of rods. 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 Monte Carlo modeling and (2) researching the optimum way to combine the 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.

Spent Fuel Library

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

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

A fourth aspect will be investigated at a later stage:

- Diversion Scenarios (i.e. pin diversion)

The case with 4% initial enrichment, 30 GWd/te and 5 years cooled was deemed to be most relevant for safeguards applications from a diversion point of view.

Measurement Scenarios

In addition to the above parameters, measurements on the spent fuel assembly can be conducted in the following media:

- Water
- Borated Water
- Air

Instrument Calibration

The final instrument must be calibrated to enable inspectors to verify fissile mass. 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 burnup and cooling time.

PASSIVE NEUTRON ALBEDO REACTIVITY (PNAR)

Passive Neutron Albedo Reactivity (PNAR) is one technical approach to spent fuel Pu verification as outlined 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 background in spent fuel is dominated by ^{244}Cm. The PNAR technique uses this intrinsic neutron emission to self-interrogate the fissile material in the fuel itself. Both 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; both with and without Cadmium (Cd) surrounding the fuel assembly. The primary difference between the two measurements is the neutron energy spectrum and fluence in the spent fuel. This enables count rates to be measured for two different energy spectra in the fuel. In the presence of Cd, reflected neutrons below ~1eV 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.

Cadmium Ratio

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 centre of the detector assembly divided by the count rate obtained when Cd is present. The Singles, Doubles and Triples Cd ratios will scale with fissile content. Contributions from the spontaneous fission source neutrons will be cancelled out in this ratio.

Fissile Content

PNAR is used to quantify fissile content which is the weighted sum of ^{235}U, ^{239}Pu and ^{241}Pu. Using Cd ratios alone, PNAR cannot distinguish between contributions to the neutron fluence from 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.

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 model of the detector geometry developed for DDSI by Menlove, *et al.* [2].

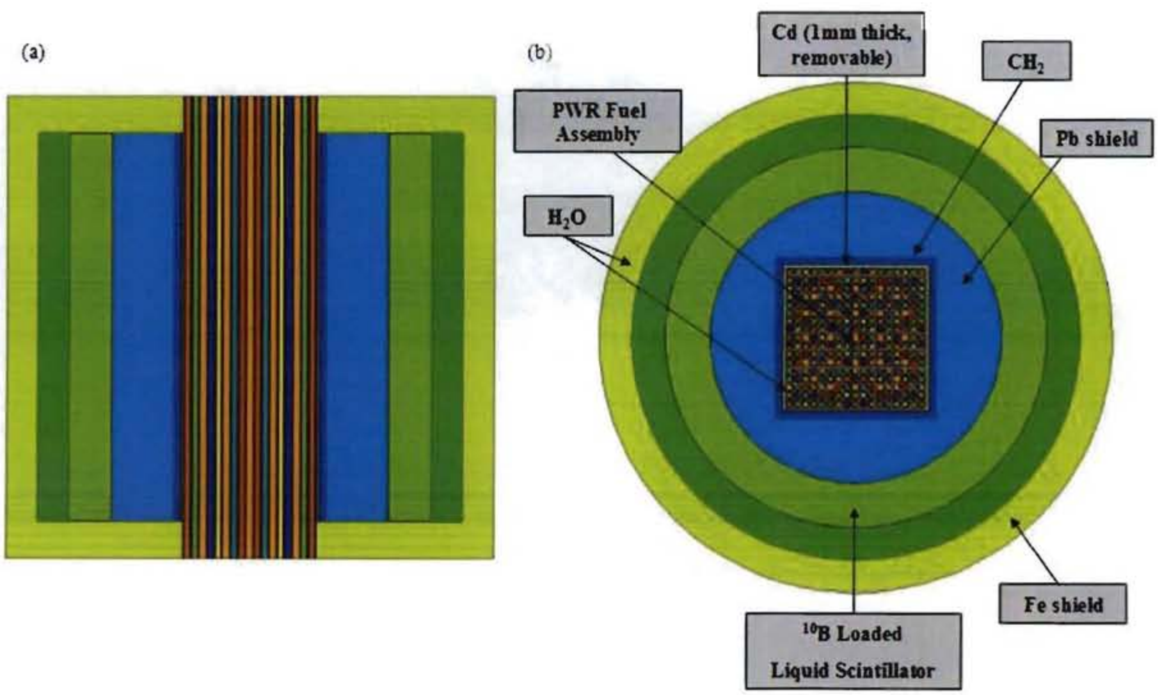

Figure 1. (a) Vertical cross-section of the detector and spent fuel assembly configuration. (b) Horizontal cross-section of the detector and spent fuel assembly configuration.

This geometry was 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 is ~ 5 x 10^5 ns^{-1} [2]. It is therefore favorable to use a detector with a short die-away time to allow counter operation at shorter coincidence gate widths hence reducing accidental pileup. A boron loaded liquid scintillator was chosen for preliminary work on PNAR due to the fast die-away characteristics, compared to ^3He. 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 detector assembly from the high gamma-ray emission from the spent fuel assembly.

The PWR spent fuel assembly itself can be seen in figure 1 at the centre of the detector assembly.

SIMULATIONS USING MCNPX

Sixteen base cases were simulated using MCNPX. An initial enrichment of 4% was used in each case, with the full range of burnups and cooling times previously defined. Simulations were conducted both with and without Cd at the centre of the detector assembly in order to derive Cd ratio values in each case.

Neutron capture events were tallied in the liquid scintillator detector using the coincidence capture tally in MCNPX. Tallies were repeated for the 16 base cases using full fuel assemblies with 4% initial enrichment. The following range of burnups was used: 15, 30, 45 and 60 GWd/te, together with the following range of cooling times: 1, 5, 20, 80 years cooled. Initial values of 4.5 μs and 64 μs were used for the pre-delay and coincidence gate width, respectively.

Single Rods vs. Full Fuel Assemblies

Due to the high neutron multiplication, the technique is able to penetrate the whole assembly and is therefore applicable to the measurement of both full assemblies and single rods. 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. 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.

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 burnup and cooling time to better understand the detector response with characteristics of the spent fuel assembly.

Spent Fuel Assemblies

Figure 2 shows the Singles, Doubles and Triples Cd ratios for full fuel assemblies as a function of burnup. The Cd ratios are a measure of fissile content in the fuel and therefore the PNAR response is 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. neutron multiplication. The dominant contribution to the decrease in the Cd ratios with burnup is thought to be the decrease in the concentration of fissile ^{235}U in the fuel assembly with burnup, which is shown in figure 3.

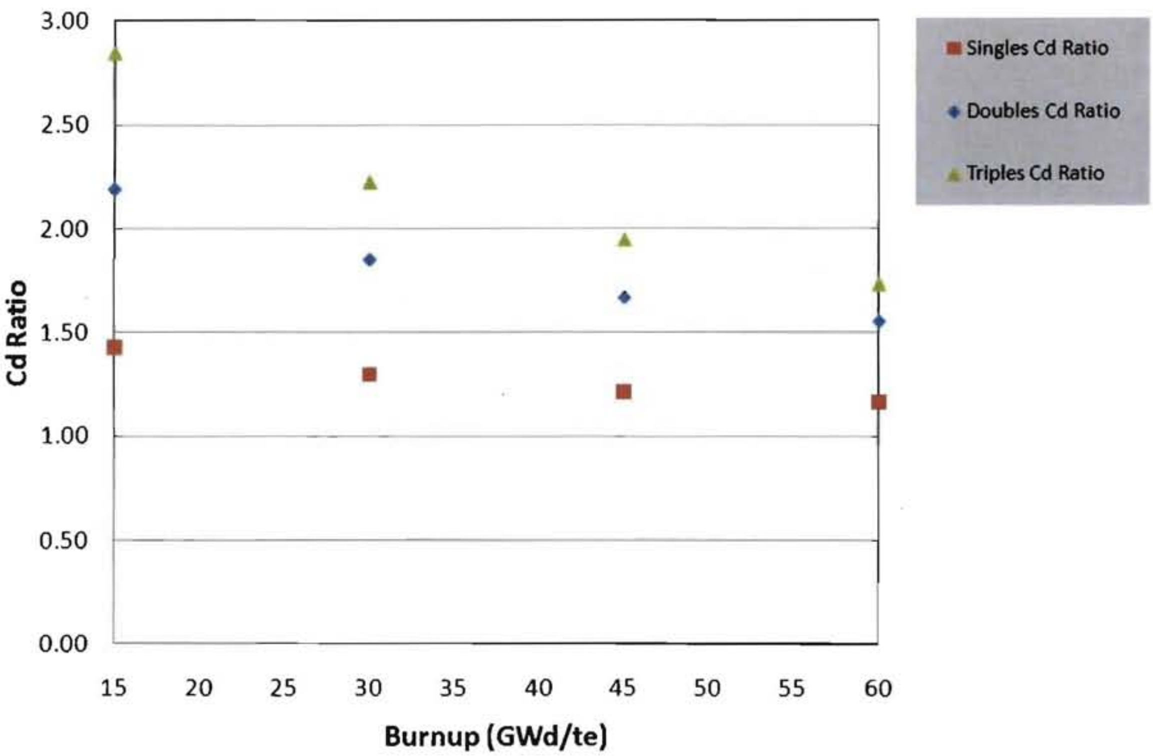

Figure 2. Fuel assembly Singles, Doubles and Triples Cd Ratios as a function of burnup, for the case using 4% initial enrichment, 5 years cooled and measured in water.

Cd ratio values are large due to multiplication in the spent fuel assembly. The differences in the y-direction between the Triples and Doubles, and the Doubles and Singles, decrease with burnup. This decrease is driven by the decrease in multiplication in the fuel assembly with burnup, which is shown in figure 5.

Figure 3 shows the mass of the fissile isotope ^{235}U in the modeled fuel assembly as a function of burnup. Figure 4 shows the mass of all Pu isotopes as a function of burnup.

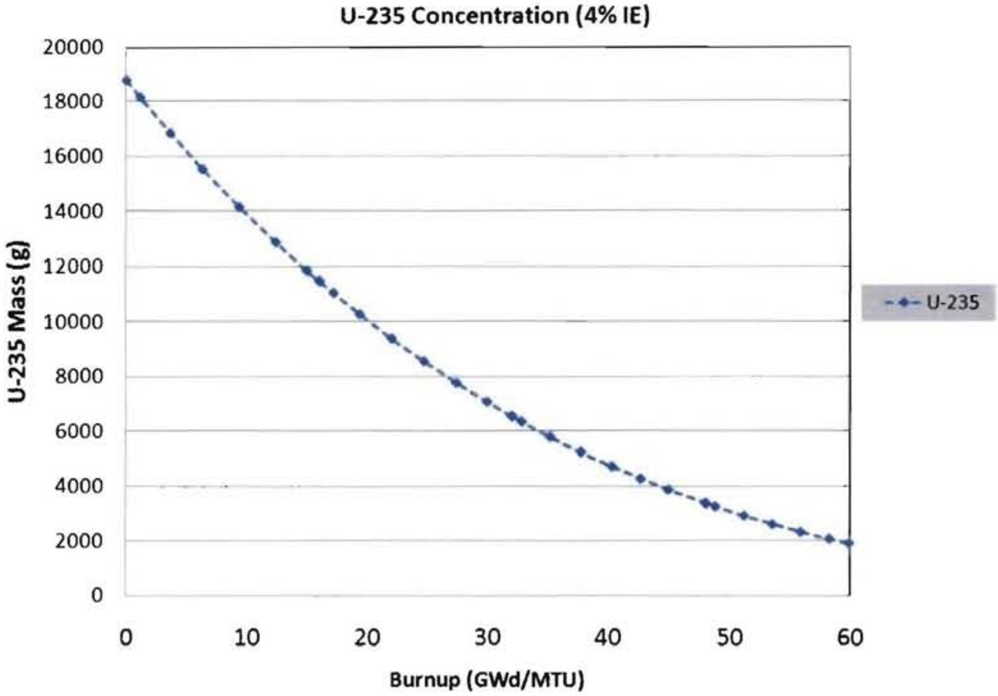

Figure 3. ^{235}U concentration as a function of burnup, for the fuel assembly modeled with 4% initial enrichment. Plot from calculations performed at LANL by N. Sandoval.

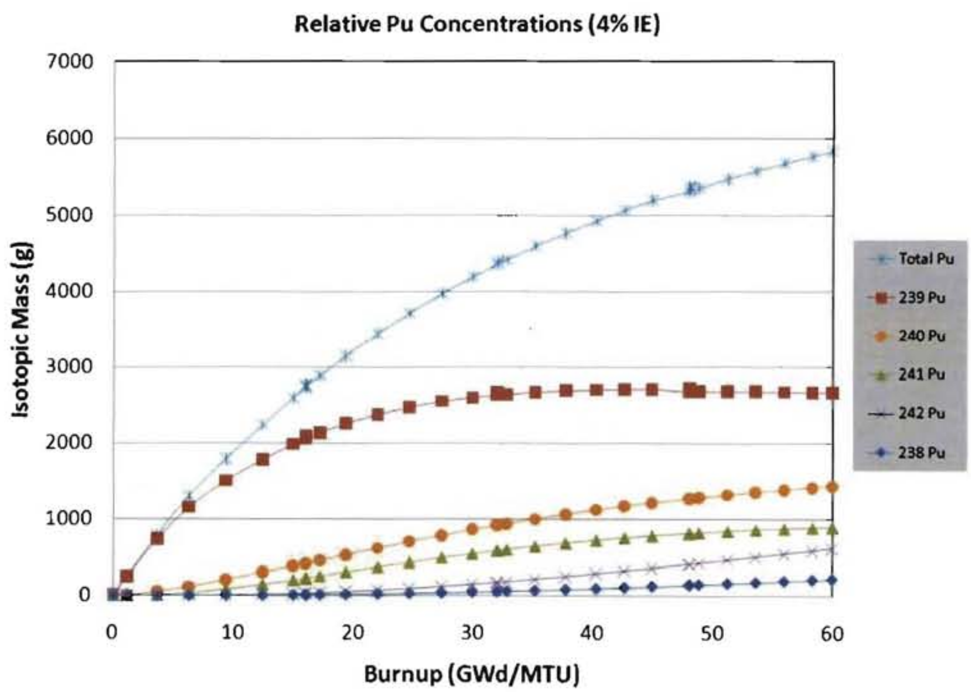

Figure 4. Relative Pu Concentrations as a function of burnup, for the fuel assembly modeled with 4% initial enrichment. Plot from calculations performed at LANL by N. Sandoval.

Figure 5 shows the neutron multiplication, both with and without the Cd sleeve surrounding the spent fuel assembly, as a function of burnup. 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.

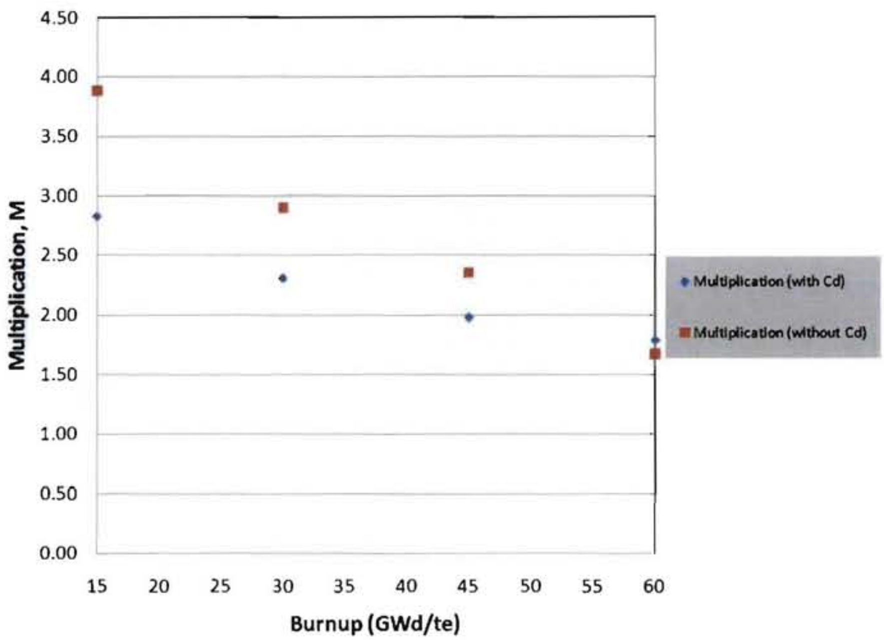

Figure 5. Neutron Multiplication (both with and without Cd) as a function of burnup.

Figure 6 shows the Singles, Doubles and Triples Cd ratios as a function of cooling time.

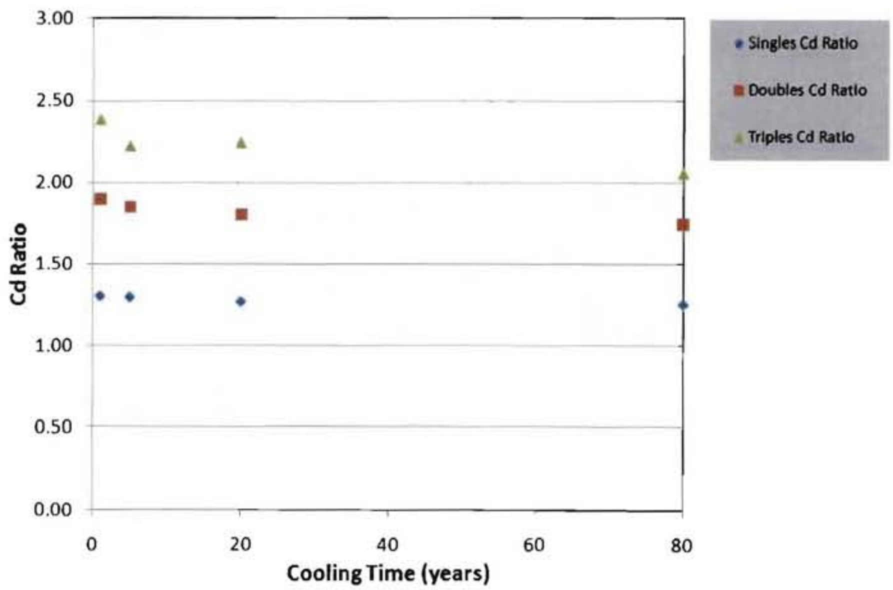

Figure 6. Singles, Doubles and Triples Cd Ratios as a function of cooling time, for a full spent fuel assembly with 4% initial enrichment, 30 GWd/te burnup, measured in water.

PHYSICS ASPECTS FOR FURTHER RESEARCH

Establishing the optimum gate width over which to perform shift register analysis will be an important aspect of this initial study. It is necessary to consider the effect of accidental correlations (Doubles and Triples) on the Cd ratio values over the selected range of both burnup and cooling time since the neutron emission rate will vary. Accidental Doubles scale with the width of the coincidence gate width, whereas currently no analytical form for the accidentals Triples exists. 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.

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.

ACKNOWLEDGEMENTS

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