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Fissile Isotope Discrimination in Spent Fuel Assemblies by Analysis of the Correlated Neutron Signal

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Abstract:

The Next Generation Safeguards Initiative (NGSI) of the U. S. Department of Energy currently supports a substantial research effort focused on quantifying plutonium (Pu) mass in spent fuel using non-destructive assay (NDA) techniques. The Differential Die-Away Self-Interrogation (DDSI) technique, one of the several being investigated, has been shown to quantify the total fissile content in spent fuel. Unlike active-interrogation NDA methods, DDSI does not require an external neutron source, but rather, uses spontaneous fission neutrons within the spent fuel for self-interrogation. The essence of the technique lies in the time separation between the detection of spontaneous fission neutrons, captured in a signal-triggered early gate, and the capture of the dominant fraction of induced fission neutrons during a later gate, thus enabling the independent measurement of fissile and fertile mass in the sample. The time domain analysis of the doubles distribution also reveals a difference in the temporal fission response of ^{235}U compared to ^{239}Pu, due to their differing higher order induced fission moments. We have simulated the behaviour of a prototypical DDSI instrument using MCNPX to model the detector response for a library of PWR fuel assemblies. Also, by exploiting the recent advancements in the Particle Track (PTRAC) capability in MCNPX, it is possible to generate a capture time distribution with isotopic designation to partition the temporal signal from individual isotopes. Although PTRAC distributions from individual isotopes are not experimentally verifiable, the information they provide is used to optimize experimental acquisition gates for time-correlated counts to capture temporal differences from each isotope. Here, we explore the use of a previously established discrimination approach, the prompt-to-delayed response ratio, applied our generated spent fuel library. We then investigate the feasibility of using this novel time correlation analysis by comparison to the prompt-to-delayed approach.

Keywords: spent fuel; plutonium; nuclear safeguards; non-destructive assay

1. Introduction

Within the nuclear safeguards framework, there is great demand for the development of NDA instruments capable of measuring the fissile materials in Pu/U mixed fuels, such as spent fuel. The high gamma fields and large neutron backgrounds in spent fuel preclude the measurement of uranium and plutonium by conventional passive or active methods. For high burnup LWR fuel, there are three major fissile isotopes (^{235}U, ^{239}Pu, ^{241}Pu) which all contribute toward the detected signal. These isotopes vary with burnup, initial enrichment and cooling time. In order to quantify plutonium mass in spent fuel, we must be able to quantify the contribution of the individual fissile isotopes.

A few techniques capable of measuring ^{239}Pu and ^{235}U in mixed fuel have been investigated. One common approach involves the measurement of both the prompt and delayed fission neutron response. The prompt-to-delayed response ratio is then used to separate the ^{239}Pu and ^{235}U contributions. This approach has been demonstrated for fresh BWR rods [1] and fresh FBR, ATR, and LWR fuel rods, as well as irradiated LWR fuel pellets [2]. In this paper, we investigate the feasibility of using the prompt-to-delayed response ratio for the determination of ^{239}Pu and ^{235}U content in

simulated spent PWR assemblies. We use previously generated prompt and delayed neutron response data from the Differential Die-Away Self-Interrogation technique (DDSI) [3] and the Delayed Neutron [4] instrument, respectively, both of which have been studied as a part of the NGSI effort [5]. Other neutron techniques, such as the lead slowing down spectrometer (LSDS) and the Neutron Resonance Transmission Analysis (NRTA) may also provide this information. LSDS distinguishes the fissile isotopes based on cross-section differences at certain energies using time-energy correlation. This approach has been investigated for fresh and irradiated fuel rods [6], as well as for PWR spent fuel assemblies [7]. NRTA exploits the individual resonance absorption lines of the various fissile isotopes present in the fuel [8].

Here, we investigate the feasibility of using the correlated neutron signal obtained from the Differential Die-Away Self-Interrogation (DDSI) for the purpose of fissile isotope discrimination in PWR spent fuel assemblies. The DDSI instrument is shown in Figure 1. The technique uses the spontaneous fission neutrons from ^{244}Cm within the assembly as the "pulsed" neutron source. The time correlated neutrons from the spontaneous fission and the subsequent induced fissions are analyzed as a function of time after the trigger event to determine the spontaneous fission rate and the induced fission rate in the sample. Fissile mass is determined from the count rate acquired during the late gate. The dominant fraction of induced fission occurs later in time with neutrons that have been moderated and reflected toward the assembly. The doubles in the late gate, D(100-196μs), divided by the singles, S, (ratio denoted as *D(100-196)/S*) is used to determine the fissile content.

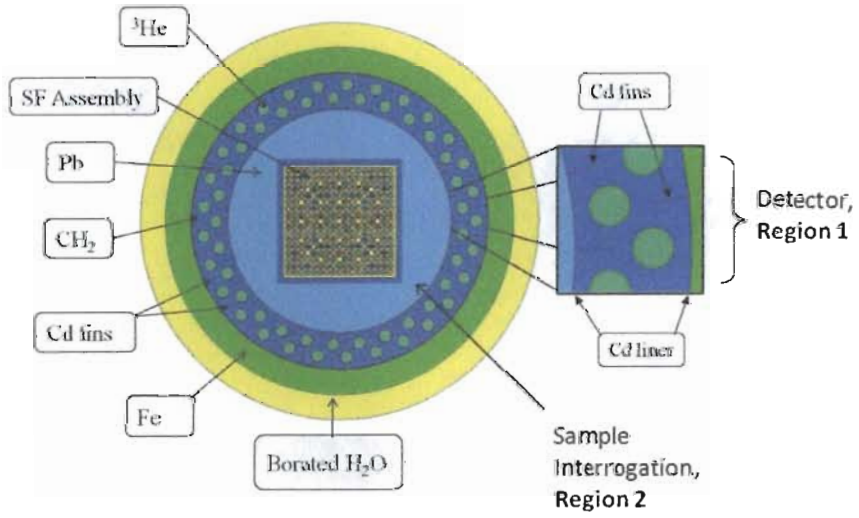

Figure 1: A horizontal cross-section of the DDSI detector configuration for MCNPX simulations is shown.

The DDSI technique has been used to quantify the fissile content in a range of possible PWR spent fuel assemblies. A comprehensive assembly library was developed [9] to provide a standard measurement sample set for the several NDA techniques being investigated in this research effort. The entire library contains 64 PWR spent fuel assemblies with burnup values of 15, 30, 45 and 60 GWd/tU; initial enrichment values are 2, 3, 4, 5 wt% ^{235}U, and cooling times are 1, 5, 20, and 80 years. For the prompt-to-delayed analysis presented in section 2, only the 5-yr cooled assemblies are considered (sixteen cases). For the time correlation analysis presented in section 3, we consider only eleven assemblies, a subset of the sixteen cases mentioned above, where the improbable cases of 2% and 3% both at 45 GWd/tU and 60 GWd/tU, and 4% at 60 GWd/tU have been removed.

2. Fissile isotope discrimination using the prompt-to-delayed response ratio

Active interrogation NDA techniques, which measure the prompt induced fission response, as well as the delayed fission response, have been used to verify both the uranium and plutonium content of spent LWR fuel. The prompt/delayed-neutron ratio is used to "separate" the contribution from ^{235}U and ^{239}Pu. The reason why the prompt/delayed-neutron ratio depends on the U/Pu ratio is due to the fact that ^{235}U and ^{239}Pu have very differing delayed-neutron yields, i.e. the delayed neutron yield of ^{235}U is

2.56 times larger than that of ^{239}Pu. The prompt-delayed method has not been used in the presence of ^{241}Pu, which is present in high burn-up fuel. This concern will be addressed later.

In general, pulsed-neutron or shuffled isotopic sources are used to interrogate the fuel, and the prompt-neutron measurement time is chosen to capture the fissile content contribution, and the delayed-neutron measurement, which is taken while the source is "off". The total fissile content response can be captured using techniques, such as DDSI, Differential Die-Away Analysis (DDA)[10], ^{252}Cf Interrogation Prompt Neutron (CIPN) [11], and other techniques which have been investigated under this research effort.

In practice, only one detector configuration should be used to measure both the prompt and the delayed signal; however, we will use the prompt signal from DDSI and the delayed signal from a separate delayed neutron (DN) instrument by Blanc et. al. [4], since the data is already available; even though the DDSI and DN are not hardware-compatible. Here we apply the prompt-delayed technique for our simulated assemblies in order to: (1) ensure that the physics of fissile isotope discrimination have been captured in our simulations, and (2) to use the prompt-delayed discrimination as a basis for comparison for the time-correlated discrimination, developed in the subsequent section.

2.1. The two-component linear system

The two-component linear system, as defined by Equation 1, is used as a starting point for ^{235}U and ^{239}Pu discrimination, meaning the presence of ^{241}Pu is ignored. The prompt and delayed neutron response, P and D respectively, are shown in Equation 1, as suggested by Matsuda et. al.[2],

$$P = a_1 N_5 + a_2 N_9 \qquad \text{Eq. 1a}$$

$$D = b_1 N_5 + b_2 N_9 \qquad \text{Eq. 1b}$$

The N_5 and N_9 values are the masses of ^{235}U and ^{239}Pu, respectively, in the interrogated section of the fuel assembly, while the a_1 and a_2 coefficients weigh the prompt neutron response per unit mass of ^{235}U and ^{239}Pu respectively. Likewise the b coefficients weigh the delayed neutron response for each isotope. Note that the P value in Eq. 1a will be represented by the DDSI ratio, i.e. the D(100-196μs)/S ratio, normalized to the assembly of least fissile content. Likewise, the D values in Eq. 1b are normalized to the least fissile assembly; therefore the D/P ratio is dimensionless. Note that the P and D values are obtained from separate instruments with different interrogating source strengths, but the normalized values are independent of the source strengths. The normalized D/P ratio shown in Figures 2 and 3 merely captures the relative change of the D/P ratio among the sixteen assemblies and does not show the actual delayed-to-prompt ratio for a given assembly. We use Eq. 1, expressed in the matrix algebra form of Eq. 2a, to define a discrimination ratio, DR, a dimensionless parameter used to quantify the discrimination ability of the two-component system; where $X=\begin{pmatrix} P \\ D \end{pmatrix}$ and $N=\begin{pmatrix} N_5 \\ N_9 \end{pmatrix}$. This system will only have a solution if the determinant of the coefficient matrix is not equal to zero, i.e. if the coefficient matrix has an inverse, as shown in Eq. 2b.

$$X = \begin{bmatrix} a_1 & a_2 \\ b_1 & b_2 \end{bmatrix} \cdot N \qquad \text{Eq. 2a}$$

$$N = \begin{bmatrix} a_1 & a_2 \\ b_2 & b_2 \end{bmatrix}^{-1} \cdot X = \frac{1}{(a_1 b_2 - a_2 b_1)} \begin{bmatrix} b_2 & -a_2 \\ -b_1 & a_1 \end{bmatrix} \cdot \begin{pmatrix} P \\ D \end{pmatrix} \qquad \text{Eq. 2b}$$

Therefore if $a_1b_2=a_2b_1$, there is no solution and hence no discrimination; so if DR is defined as (a_1b_2/a_2b_1), there is no discrimination if DR is equal to unity. DR should be much smaller than unity, with the smaller DR value translating to better fissile isotope discrimination. For DDSI [12], the average a_1=0.46; a_2=1 (w.r.t. to ^{239}Pu) and for DN [13], the average b_1=1.54 and b_2=1, so a prompt-delayed system using DDSI and DN has a DR=0.30. The DR values for other prompt-delayed integrated systems are shown in Table 1.

Integrated System	Discrimination Ratio (DR)
DDSI+DN	0.30
CIPN+DN	0.34
DDA+DN	0.35

Table 1: DR values for ^{239}Pu and ^{235}U separation for prompt-delayed integrated NDA systems.

Figure 2 shows the D/P ratio for the sixteen assembly cases (listed in Table 2) versus the $^{239}Pu/(^{239}Pu+^{235}U)$ mass ratio using the DDSI and DN data.

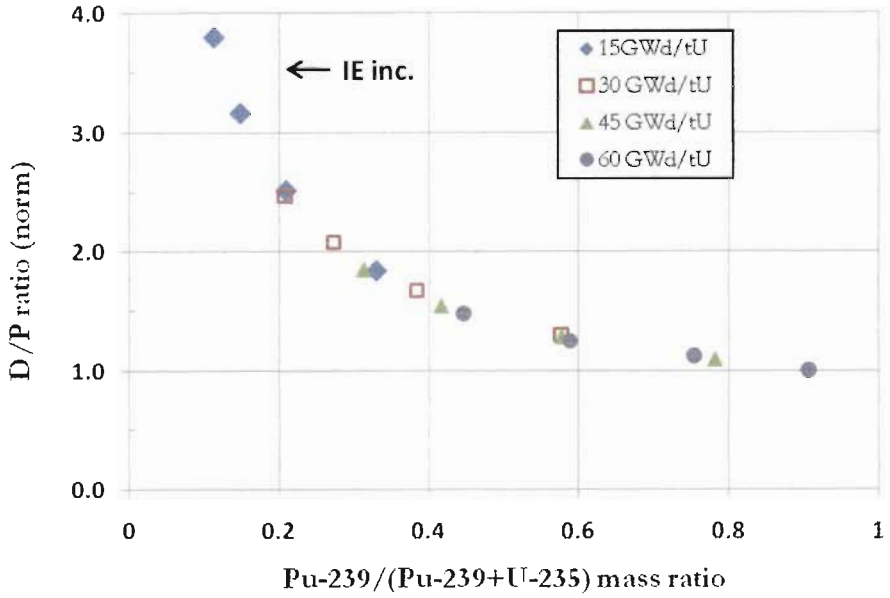

Figure 2: Correlation between the D/P ratios and the $^{239}Pu/(^{239}Pu+^{235}U)$ ratios for 16 assemblies

CT [yr]	BU [GWd/tU]	IE [wt% ^{235}U]	$^{239}Pu/(^{239}Pu+^{235}U)$
5	15	2	0.33
5	15	3	0.21
5	15	4	0.15
5	15	5	0.11
5	30	2	0.58
5	30	3	0.38
5	30	4	0.27
5	30	5	0.21
5	45	2	0.78
5	45	3	0.58
5	45	4	0.42
5	45	5	0.31
5	60	2	0.91
5	60	3	0.75
5	60	4	0.59
5	60	5	0.45

Table 2: Sixteen assembly cases used in analysis and corresponding ^{239}Pu mass ratio [row in red corresponds to maximum 239Pu mass fraction, while blue indicates least]

Note that there is a clear correlation between the D/P ratio and the $^{239}Pu/(^{239}Pu+^{235}U)$ ratio regardless of the initial enrichment and burn-up of the assemblies. In other words, the combination of the

simulated prompt neutron signal from DDSI, which captures the fissile content, and the simulated delayed neutron signal from the DN instrument, provides the necessary information for fissile isotope discrimination; the physics necessary for discrimination has been captured in our simulations. If the prompt and delayed signals are accurately scaled based on the interrogating neutron source strengths and the integrated system is calibrated using a standard sample, the correlation seen in Figure 2 can be used to determine the ^{239}Pu and ^{235} U mass ratio quantitatively. The correlation, however, does not appear to be linear, as one would expect given the defining relation stated in Eq. 1. Figure 3 shows this deviation from linearity, with the greatest deviation occurring at low burn-up highly enriched fuel, where the ^{235}U is greatest component of the fissile content. The linear response is calculated using Eq.1 with the weighting coefficients tallied from the MCNPX simulation. The "detected" response is also obtained from the simulated prompt and delayed counts arriving at the detector. This discrepancy shows that the linear two-system model, although it shows the negative correlation between the D/P ratio and the ^{239}Pu mass fraction, does not adequately characterize the system, with the ^{241}Pu contribution being ignored.

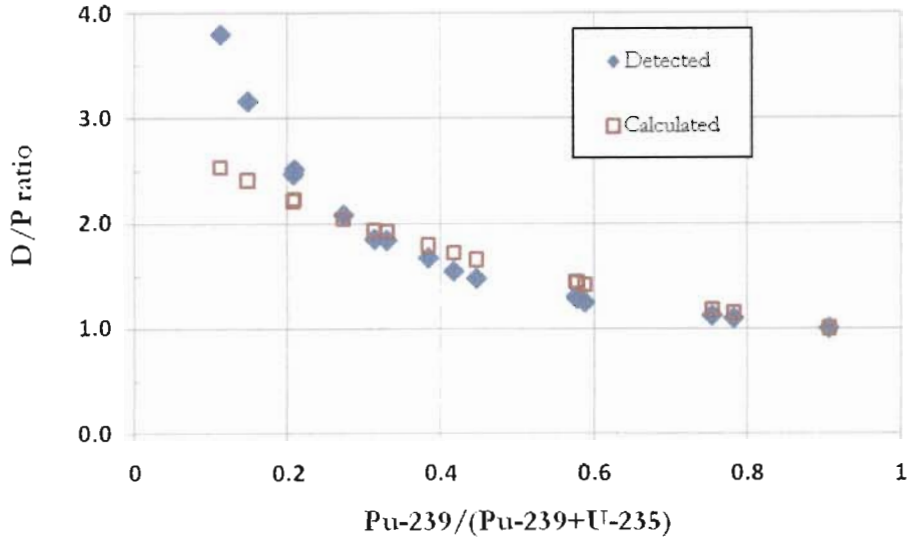

Figure 3: Detected and calculated correlation between the D/P ratios and the ^{239}Pu/(^{239}Pu+^{235}U) ratios for 16 assemblies

2.2. ^{241}Pu Correction

Among the sixteen assemblies simulated, the ^{241}Pu content can be as high as 20% of the total fissile content, so Eq. 1 does not adequately describe the prompt and delayed fission response. It is not possible to quantify isotope discrimination ability in a three-component system, so the analysis presented above cannot be simply extended by adding a third term, for ^{241}Pu, to Eq. 1a and 1b. Also of note is the fact that ^{241}Pu has a delayed neutron fraction similar to that of ^{235}U (0.0154 vs. 0.0158 respectively), so as the ^{241}Pu increases, the discrimination between ^{235}U and Pu will diminish. One method of correcting for the presence of ^{241}Pu is to use an iterative scheme, as proposed by Menlove et. al.[14], where a two-component system with ^{239}Pu and ^{235}U is initially assumed (no ^{241}Pu present). The D/P ratio is used to obtain a first guess of the ^{239}Pu to ^{235}U ratio. Based on this first guess, isotopic correlation would then be used to estimate ^{241}Pu content. This estimate of ^{241}Pu mass is then used to correct the ^{239}Pu and ^{235}U ratio, which will then be used to update the ^{241}Pu mass, until convergence occurs for all three components. It is clear that either isotopic correlation or information from another NDA measurement is needed to de-convolute the contribution of all three major fissile isotopes i.e. three observables are needed to solve for three unknowns. This will be the subject of future work.

3. Fissile isotope discrimination using the correlated neutron signal

The detected induced fission response as a function of time depends on the relative concentrations of ^{239}Pu and ^{235}U. Table 3 shows the induced fission moments of these isotopes for the sake of comparison. Note that ^{239}Pu has increasingly larger second and third induced fission moments than

^{235}U, meaning the time correlations of the induced fission neutrons can give the relative concentrations of the two isotopes.

Isotope	ν	$\nu(\nu-1)$	$\nu(\nu-1)(\nu-2)$
^{235}U	2.406	4.626	6.862
^{239}Pu	2.879	6.733	12.630
Ratio $^{239}Pu/^{235}U$	1.197	1.455	1.84

Table 3: Prompt fission neutron multiplicity moments for ^{239}Pu and ^{235}U

The two isotopes also differ in the energy-dependence of their fission cross-section in the epi-cadmium region, with ^{239}Pu fission occurring preferentially over ^{235}U fission at the 0.3 eV resonance energy. For the single fuel pin assay, this cross-section difference at a specific energy results in a time signature in the response, as the neutron must first slow down to that energy (time-energy correlation). This signature is useful for the assay of a single fuel pin, which is compact with low multiplication, but for the assay of a large 16x16 assembly submerged in water, the high multiplication smears this distinct time feature, and the increased size of the sample diminishes the energy resolution.

We can sample the induced fission rate as a function of time after the first detected neutron using list-mode data collection. A measure of the die-away time of these correlated distributions should depend on the relative concentrations of ^{239}Pu and ^{235}U. The doubles and triples distribution for ^{239}Pu should peak earlier and have a shorter die-away time than that of ^{235}U. This would be observable experimentally if two separate samples with only either of the isotopes present (no mixture-pure sample), but this cannot be readily observed in a mixed sample since we do not know the isotopic origin of the detected fission neutrons. A recent modification to the particle track (PTRAC) capability in MCNPX [15] breaks down the isotopic contributions such that this effect can be observed, and furthermore used to optimize gates to determine the relative concentrations of the two isotopes. Figure 4 shows the results of the PTRAC analysis of a simulated 45GWd/tU, 4 wt% ^{235}U, and 5-yr cooled assembly.

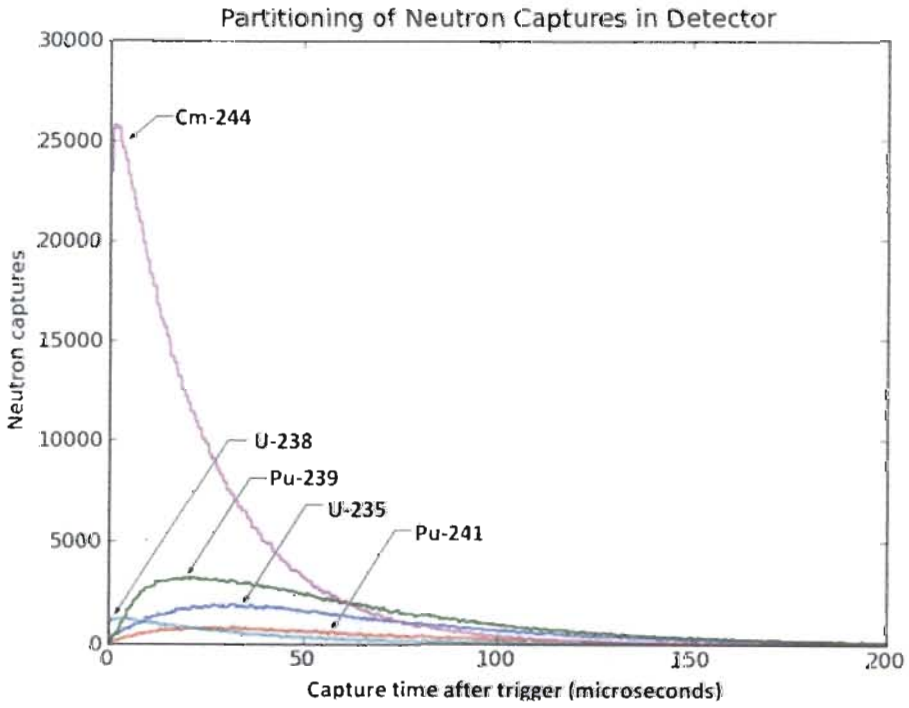

Figure 4: The enhanced PTRAC Capture file capability labels the isotopic origin of each detected neutron. Neutrons are captured in ^{3}He detectors in the DDSI geometry surrounding a 45GWd/tU, 4% IE, and 5-year cooled PWR assembly.

The total detected induced fission neutron response is determined by the time response of the three major fissile isotopes, ^{239}Pu, ^{235}U, and ^{241}Pu, The shape of the total induced fission time response will depend on the relative concentrations of these isotopes. The PTRAC capability can also sum the total induced fission time response, as shown in Figure 5. Here, the normalized induced fission distributions are shown for assemblies with the smallest and largest ^{239}Pu fraction across the 11 assemblies listed in Table 4. The curves are normalized to only show shape differences in the distributions. The smallest and largest ^{239}Pu fraction cases define an envelope of variation of the induced fission response as seen in Figure 5. These two distributions contain valuable information for doubles gate optimization in DDSI to capture this shape difference and hence the relative concentration of the two isotopes.

CT [yr]	BU [GWd/tU]	IE [wt% ^{235}U]	^{238}Pu/(^{239}Pu+^{235}U)
5	15	2	0.33
5	15	3	0.21
5	15	4	0.15
5	15	5	0.11
5	30	2	0.58
5	30	3	0.38
5	30	4	0.27
5	30	5	0.21
5	45	4	0.42
5	45	5	0.31
5	60	5	0.45

Table 4: Sixteen assembly cases used in analysis and corresponding ^{239}Pu mass ratio [row in red corresponds to maximum mass ratio, while blue indicates the least]

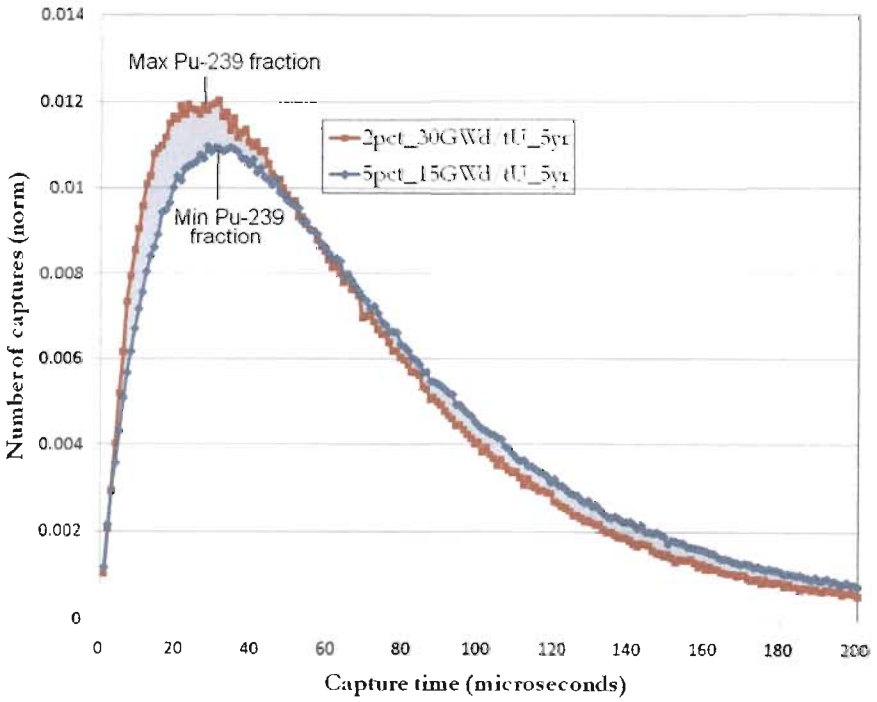

Figure 5: Normalized Induced fission distribution envelope defined by the maximum and minimum ^{239}Pu fraction assemblies obtained from the PTRAC capture file in MCNPX simulation (for 11 assemblies considered)

As seen from Figure 5, the ^{239}Pu fraction affects the rise time and the die-away time of the neutron capture distributions, obtained using the PTRAC capture file in MCNPX. The shape of the capture

distributions in Figure 5 can be characterized by the R-parameter, defined in Equation 3 and shown in Figure 6. Here, R is defined as the ratio sum of the captures in the "tail" to the total number of captures over all time, in essence the ratio of the tail integral to the total integral. The R-parameter was chosen to characterize the shape of the capture distribution borrowing from a common pulse shape discrimination method in detector physics.

$$R = \frac{Tail}{Total} \qquad \text{Eq. 3}$$

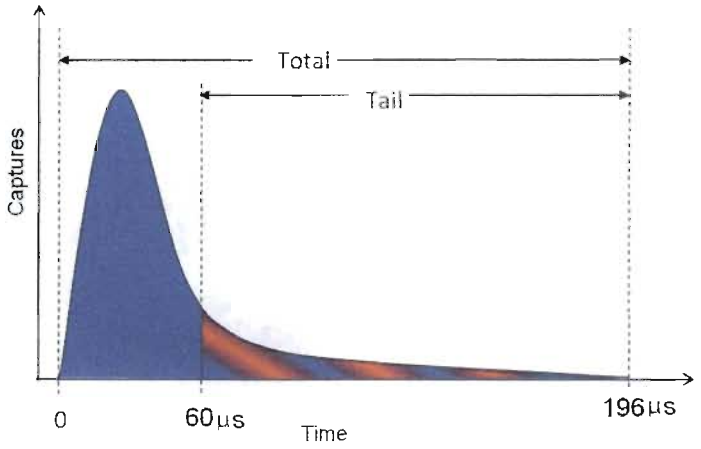

Figure 6: R-parameter used to characterize shape of the capture distribution as a function of time with a 4 μs gate width

A similar characterization can be performed using the factorial moment distribution, proportional to the doubles, from the F8 Capture tally capability of MCNPX. Figure 7 shows the dependence of the factorial moment (and hence the doubles) as a function of pre-delay after the trigger event for the limiting cases shown in Table 4, A similar shape difference, as that seen in the capture distribution is observed.

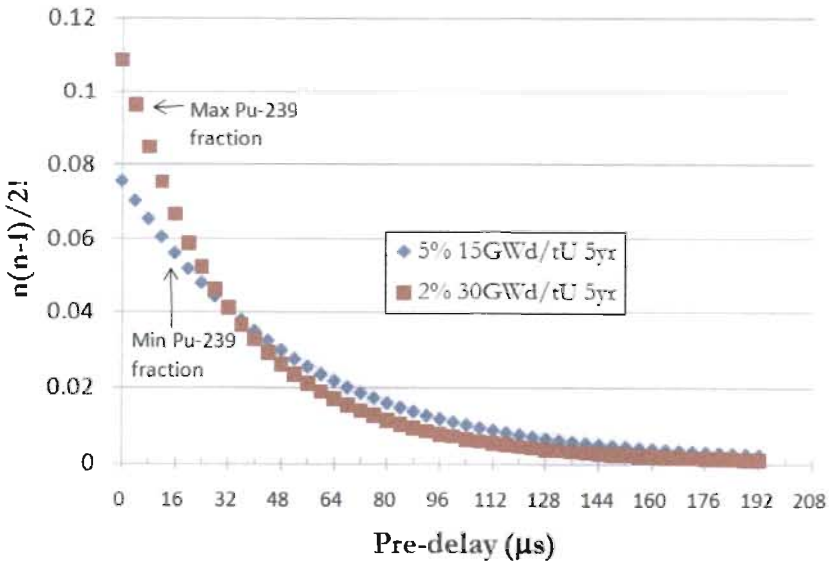

Figure 7: Normalized factorial moment distribution for limiting cases with max and min ^{239}Pu fraction with a gate width of 4μs

The R-parameter can be re-defined by Eq. 4 to be used for Figure 7.

$$R = \frac{D(60-196)}{D(0-196)} \qquad \text{Eq. 4}$$

By inspection of Figure 5, the tail start time was chosen as 60 μs and the overall time extended to 196 μs. In conventional doubles counting, the tail gate can be optimized to detect this change in shape of the doubles distribution, and hence the fissile Pu/U ratio.

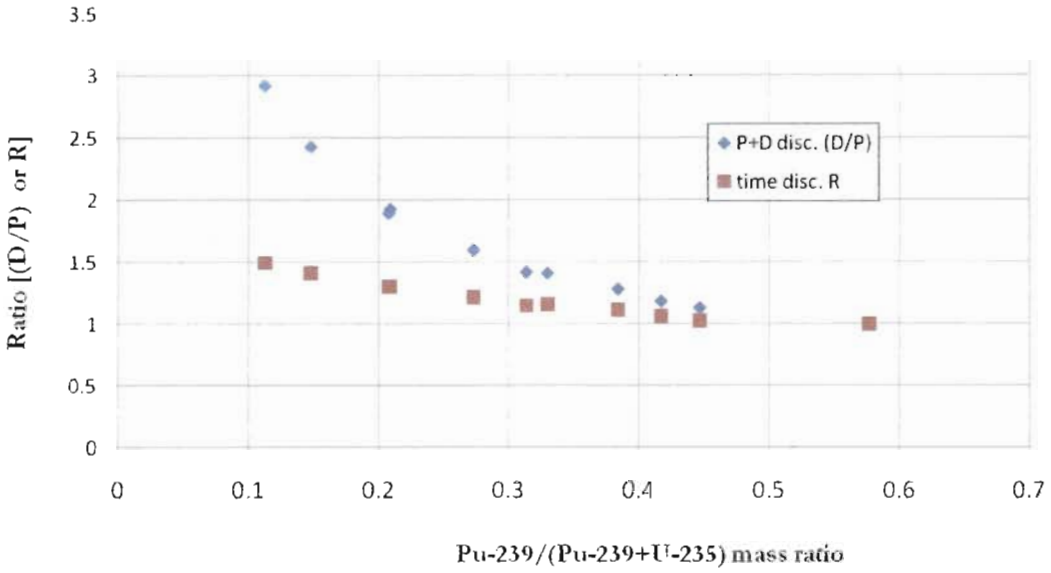

Figure 8: R shape parameter and the D/P ratio for 11 assembly cases as a function of fissile Pu/U mass ratio

Figure 8 co-plots the normalized D/P ratio and the R values (from Eq. 4) against the $^{239}Pu/(^{239}Pu + ^{235}U)$ mass ratio for the eleven assemblies being considered. Both the D/P ratio and the R values show a negative correlation with the ^{239}Pu mass fraction. As the ^{239}Pu fraction increases, the prompt response increases slightly but the delayed response decreases, hence the D/P ratio should decrease with an increasing ^{239}Pu fraction. For the time correlated discrimination, as the ^{239}Pu fraction increases, less of the counts are in the "tail" so R decreases. The normalized D/P ratio exhibits a greater variation over the mass ratio range (1 to ~3), i.e greater sensitivity to the ^{239}Pu fraction variation compared to the normalized R-values (1 to ~1.5).

3. Summary

The R-parameter does in fact capture the shape, or the temporal response of the induced fission response, giving the relative change in the ^{239}Pu mass fraction present in an assembly. A well-characterized sample must be used to calibrate the system in order to measure actual ^{239}Pu mass fractions. Also, the influence of presence of ^{241}Pu is not considered here for the time-correlated discrimination approach. This study is intended to summarize the initial concept of applying the time-correlated neutron signal to discriminate between ^{239}Pu and ^{235}U, however the gate optimization will be unique for different detector configurations. Although the discrimination ratio, R, is not as large as the D/P ratio among the eleven assemblies, it may provide enough information for discrimination without requiring a large, external interrogation source needed by the delayed neutron measurement.

4. Acknowledgements

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