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Verification of Plutonium content in Spent Fuel Assemblies Using Neutron Self-Interrogation

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ABSTRACT

The large amounts of plutonium in reactor spent fuel assemblies has led to increased research directed toward the measurement of the plutonium for safeguards verification. The high levels of fission product gamma-ray activity and curium neutron backgrounds have made the plutonium measurement difficult. We have developed a new technique that can directly measure both the ^{235}U concentration and the plutonium fissile concentration using the intrinsic neutron emission from the curium in the fuel assembly. The passive neutron albedo reactivity (PNAR) method has been described previously [1] where the curium neutrons are moderated in the surrounding water and reflect back into the fuel assembly to induce fissions in the fissile material in the assembly. The cadmium (Cd) ratio is used to separate the spontaneous fission source neutrons from the reflected thermal neutron fission reactions. This method can measure the sum of the ^{235}U and the plutonium fissile mass, but not the separate components. Our new differential die-away self-interrogation method (DDSI) can be used to separate the ^{235}U from the ^{239}Pu. The method has been applied to both fuel rods and full assemblies. For fuel rods, the epi-thermal neutron reflection method filters the reflected neutrons through thin Cd filters so that the reflected neutrons are from the epi-cadmium energy region. The neutron fission energy response in the epi-cadmium region is distinctly different for ^{235}U and ^{239}Pu. We are able to measure the difference between ^{235}U and ^{239}Pu by sampling the neutron induced fission rate as a function of time and multiplicity after the initial fission neutron is detected. We measure the neutron fission rate using list-mode data collection that stores the time correlations between all of the counts. The computer software can select from the data base the time correlations that include singles, doubles, and triples. The die-away time for the doubles distribution is distinctly different for ^{235}U and ^{239}Pu. The ^{239}Pu has a higher fission cross-section in the epi-cadmium neutron region and larger induced fission moments than ^{235}U, so the measured die-away time can provide the relative amounts of ^{239}Pu and ^{235}U. This paper will present the Monte Carlo simulations for the detector and sample configurations for both fuel pins and full fuel assemblies.

1. INTRODUCTION

The fissile content in special nuclear materials has been measured for more than three decades using the active neutron interrogation methods such as differential die-away analysis (DDA) [2], and ^{252}Cf shuffler neutron sources [3]. Un-irradiated product and waste samples have been the focus of the measurements. Traditionally the active assay techniques have used a pulsed neutron generator or a ^{252}Cf shuffler to create a burst of neutrons to interrogate the sample, and, between

the interrogation bursts, the neutron time distribution is measured to determine the neutron induced fission rate in the sample that is proportional to the fissile content.

The neutron self-interrogation method has three different technical approaches as illustrated in Fig.1. The passive-neutron albedo reactivity (PNAR) method is where the fission neutrons are reflected back into the fuel assembly to induce fission reactions in the fissile material. A cadmium (Cd) liner is then inserted around the perimeter of the fuel assembly to prevent the return of the thermal neutrons. The measured Cd ratio is proportional to the fissile content in the fuel. The Cd ratio is independent of the intensity of the interrogation source term that is primarily from ^{244}Cm and the neutron multiplication

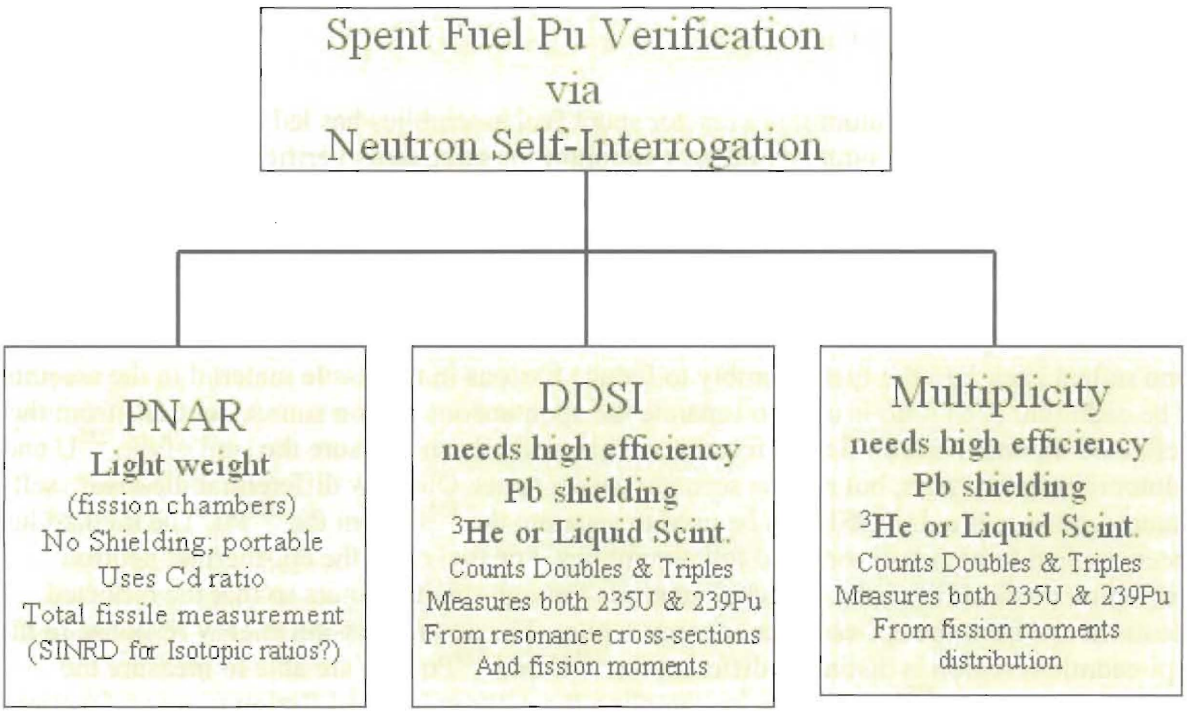

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Fig 1 Diagram illustrating the relationship between the PNAR , the DDSI, and the multiplication methods for spent fuel self-interrogation.

More recently, we have developed a new differential die-away self-interrogation (DDSI) method, [4] that uses time correlation neutron counting with triggering on the fission events in the assay sample. Thus, the need for the external neutron source is eliminated, significantly reducing the size, cost, and complexity of the system. Following each neutron event that is measured, the neutron time distribution is analyzed to determine the early and later neutron distributions from the spontaneous and induced fissions in the sample.

The early and late time windows that are used in the DDSI method are illustrated in Fig.2. The early gate interval is used to calculate the spontaneous fission rate, primarily ^{244}Cm, and the neutron multiplication (M). The later die-away neutron time distribution is populated by resonance energy neutron fissions and thermal-neutron fissions, and it provides a measurement

of the fissile content in the sample. The new DDSI method provides a significant improvement in the precision and sensitivity compared to the PNAR method.

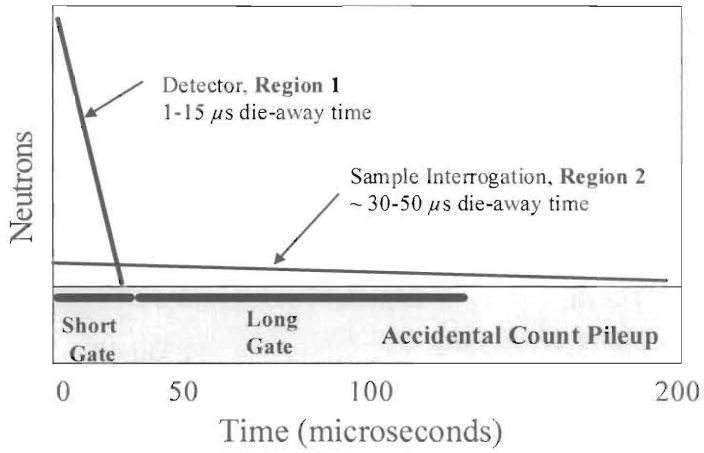


Fig. 2. Illustration of the neutron life-time in two spatial regions of the detector. Region 1 is the exterior part of the instrument where neutrons are detected and region 2 is the sample region where moderated neutrons interrogate the sample.

The accidental pileup events are the dominant contribution to the counting statistical uncertainty, and the primary challenge of the DDSI method is to reduce this uncertainty by careful design of the detector and self-interrogation regions.

The early and late time distributions are analyzed for singles, doubles, and triples counts to further separate the induced fission events from the spontaneous fission trigger events. The higher multiplicity events are strongly weighted to the induced fissions because the induced fissions are counted in the same time gate as the spontaneous fission event, and the effective ν (prompt neutrons per fission) increases from 2.72 for ^{244}Cm to ~ 4.6 for the combination of spontaneous fission and induced fissions. This paper presents the MCNPX [5] calculations of the detector systems that could be applied for both fuel rods and fuel assembly measurements.

2. DETECTOR AND MODERATOR DESIGN FOR A SINGLE FUEL ROD

Our initial MCNPX design simulation was for a single spent fuel rod as illustrated in Fig. 3a.We configured an annulus of 36 ^{3}He tubes with 10 atm pressure that surrounded the rod in the central position. Because of the high gamma-ray background from the fission products in the spent fuel, we introduced a 40 mm thick lead shield between the fuel pin and the ^{3}He detectors. The simulations showed that the efficiency could be significantly improved with only a small increase in the die-away time by using a new moderator configuration with Cd fins. The fin design cuts off the longer die-away neutrons that were migrating to the ^{3}He tubes from the more distant positions in the HDPE moderator. The overall length of the detector is 500 mm, and there is a 50-mm-thick annulus of iron on the outside of the system to reflect fast neutrons back into the detectors and to shield the detectors from other spent fuel rods that could be inside the measurement hot cell .

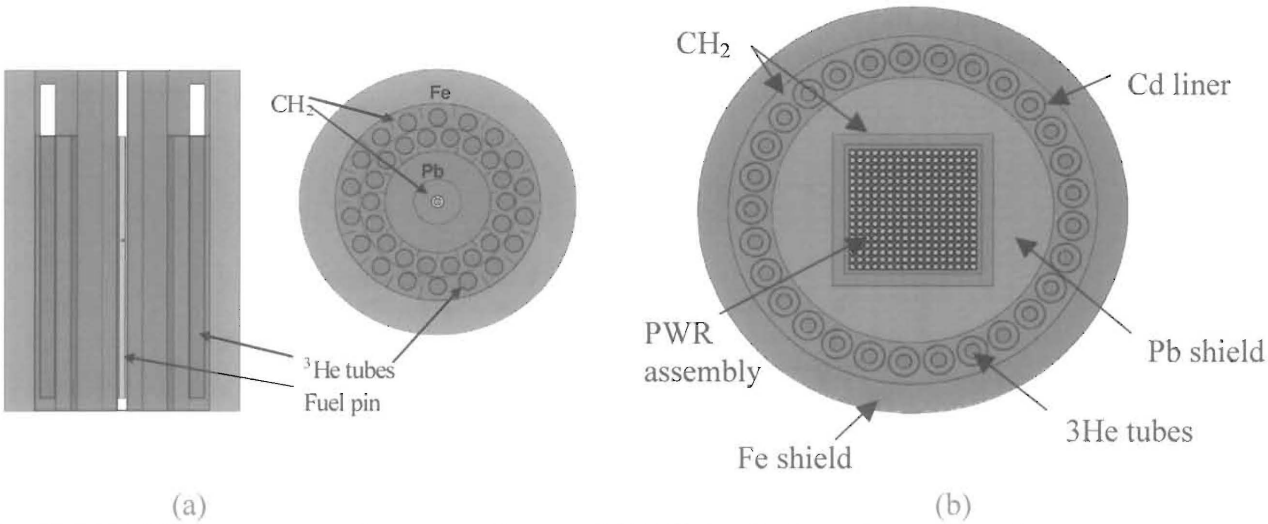


Fig. 3.(a) DDSI detector configuration for the MCNPX simulations for single rod measurements and (b) the fuel assembly moderator and detector geometry.

A typical spent fuel rod would have a ^{244}Cm neutron emission rate of approximately 5E+05 neutrons per second from the section of the rod that is inside the detector. The multiplicity counting uncertainty gets large for high counting rates because of accidental counts in the coincidence gates, so it is desirable to keep the die-away time short to reduce the accidental pileup in the counting gates. The liquid scintillator option described in Section 5.0 helps to alleviate the problem because of the short coincidence gates.

Sample Interrogation Region Design

The next target of the design was to configure the materials and thickness in the sample region to provide a maximum induced fission rate from the reflected thermal neutrons without a significant reduction in the detector efficiency. The optimum interrogation configuration for a rod was concentric annuli of 1-mm Cd, 25-mm HDPE followed by the annulus of lead and the rest of the detector configuration shown in Fig. 3a. The inner 1-mm annuli (sleeve) of Cd was removable to provide the Cd ratio measurement. The die-away time in the fuel rod region should be longer than the die-away time in the detector region to provide separation of the fast neutron pulse (spontaneous fission) and the subsequent induced fissions in the fissile material. The design shown in Fig. 3a has a die-away time of ~ 14 μs in Region 1 containing the ^{3}He tubes. In the sample interrogation zone (Region 2), the die-away time was ~35 μs. For the liquid scintillator, the die-away time for the detector region reduces to less than 1 μs.

3. TIME GATE ANALYSIS

There are two time regions of interest, as illustrated in Fig. 1. The first is the early time zone which is 1-20 μs in duration to encompass most of the spontaneous fission events. The second time zone of interest intended to quantify the induced fissions has a die-away time of ~8-128 μs. The time gates for the data evaluation are set to provide good separation for the early spontaneous fission events and the later induced fission events. The lengths of the spontaneous fission early gate and the induced fission late gate are influenced by the neutron source strength.

For spent fuel rods at 40 GWD/tU, the curium neutron source yield of ~5E+05 neutrons/s is expected within the detector geometry (~500 mm long). If the signal data is collected in the list mode that time stamps each neutron count, then the DDSI data evaluation can select the optimum time gates for the early gate (spontaneous fissions) and the late gate (induced fissions) to provide the minimum statistical uncertainty. For the single fuel rod, the statistical error for the induced fission doubles rate was 2.4 % in 1,000 s, and the Cd ratio was 1.15 for the doubles. We also simulated the case for a full PWR fuel assembly that is described in the next section.

4. FUEL ASSEMBLY MEASUREMENT SIMULATIONS

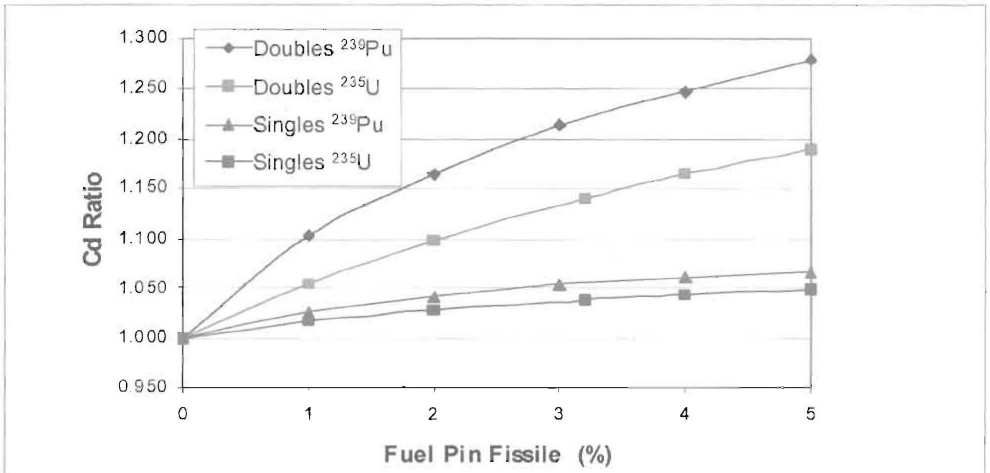
The measurement of full fuel assemblies is more important for safeguards verification than the measurement of single rods, and we have extended the PNAR and DDSI methods to the fuel assembly geometry. Figure 3b illustrates the MCNPX configuration for a full PWR fuel assembly. The assemblies were calculated for three cases, 1) borated water (2200 ppm B), 2 pure water, and 3) air. Because of page limitations, we will only cover the borated water case in this paper. The neutron source term for a typical fuel assembly is ~ 1E+08 neutrons per second from the ^{244}Cm depending on the initial enrichment and the burnup level. There is an additional factor of about two increase from neutron multiplication in the water. However, the detector geometry sees only about 1/6 of the neutrons depending on the detector length. For our fuel assembly simulations, we have used a detector active length of 400 mm with 29 3He tubes at 6 atm gas pressure. We have surrounded each detector tube with 14 mm of HDPE wrapped in a 0.5 mm thick Cd sleeve. The resulting efficiency is ~ 3% and the die-away time is ~ 18 us.

The PNAR and DDSI techniques work much better for fuel assemblies than single rods because of the high neutron multiplication in the assemblies and the large solid angle for the reflected neutrons returning to the assembly.

4. RESULTS

Spent Fuel Rods

Using the configuration shown in Fig 3 for single fuel rods with an efficiency of 29 % and a die-away time of 14 μs in Region 1, the ^{235}U and the ^{239}Pu concentrations in the fuel rod were varied from 0 to 5%; The induced fission signal was obtained from the late coincidence gate (21.5 μs to 41.5 μs). This gate also contains the residual tail of the prompt spontaneous fission neutrons that drive the interrogation. However, for the Cd ratio, the spontaneous fission neutron intensity cancels, and the ratio is proportional to the induced fission rate in the fissile content. We are using multiplicity counting that includes singles, doubles and triples neutrons., The doubles rates increases the Cd ratio above the singles rate because of the increase in the effective ν and the higher fission moments that are included in our gate interval. Figure 5 shows the Cd ratio for the singles and doubles as a function of ^{235}U and ^{239}Pu loading. The non-linear shape of the curves is the result of fissile material self-shielding of the thermal neutrons that interrogate the rod.



.Fig. 5 Response curves for the singles and doubles rates as a function of the ^{235}U and ^{239}Pu concentration in the fuel rod, using the MCNPX simulations.

In addition to the PNAR analysis, we have analyzed the data for the DDSI method using boron loaded liquid scintillator detectors The early time-gate doubles rate measures the ^{244}Cm mass, and the late time gate, measures the fissile mass that is a mixture of ^{235}U, ^{239}Pu, and ^{241}Pu. Figure 6 shows the ratio of the late gate divided by the early gate as a function of fissile concentration. We see that the signal to background ratio has increased by more than an order of magnitude compared with the Cd ratio For typical spent fuel, the ^{235}U is ~1% and the Pu fissile is about 0.6% of the heavy metal mass. The ^{244}Cm is a strong function of the burn up (BU), and the ^{244}Cm is measured in the DDSI early gate so the approximate BU can be determined. For the typical spent fuel rod, about half of the induced fission response comes from the ^{239}Pu and half comes from the ^{235}U. The doubles response per gram for ^{239}Pu is higher than that for ^{235}U because of the higher thermal-neutron cross section and the larger induced fission multiplicity distributions [6].

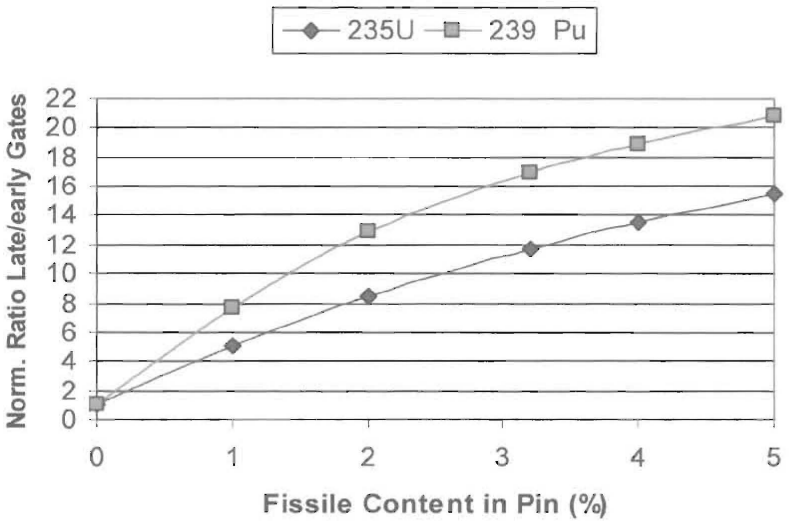


Fig. 6. The self-interrogation response for the normalized ratio of the late gate divided by the early gate as a function of the fissile concentration in a ^{239}Pu fuel rod using a liquid scintillator detector.

Figure 7 shows the MCNPX simulation results for a single fuel rod containing either all ^{239}Pu or all ^{235}U and we see that the die-away time ratio for ^{239}Pu /^{235}U is 0.83 with no Cd filter. This ratio would be unity if the fission cross-section shapes were the same for the two isotopes. The signature ratio increases to 1.21 when a 0.2 mm thick Cd filter is placed around the sample. For this case, the signature ratio to separate the two isotopes is a factor of 1.45. If the epi-cadmium neutron cross sections were not different for the two fissile isotopes, the die-away time ratio would be unity for all of the filter cases.

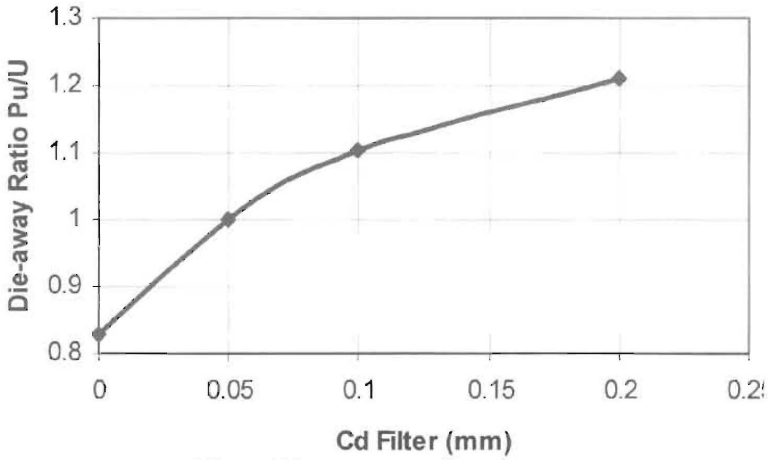


Fig 7. The die-away time ratio of $^{239}Pu/^{235}U$ as a function of the Cd filter thickness surrounding the fuel pin.

Spent Fuel Assemblies

The spent fuel assemblies provide a larger Cd ratio and DDSI signal than for the fuel rods because of the higher neutron multiplication in the fuel assemblies. The Cd ratios for the singles and doubles rates are shown in Fig 8 for the fuel assembly geometry.

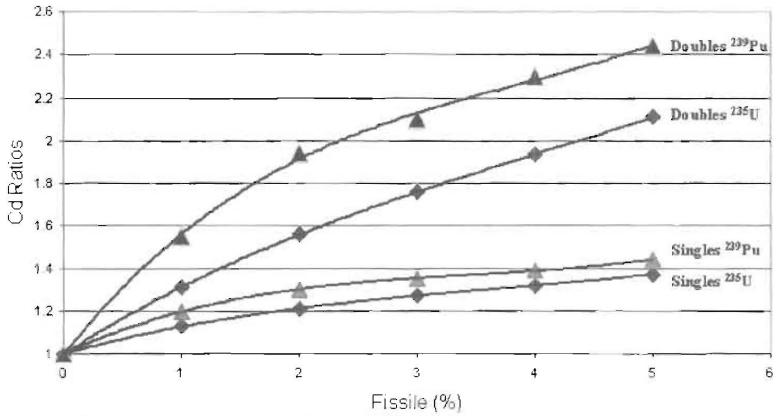


Fig. 8. Fuel assembly Cd ratios for singles and doubles rates as a function of fissile concentration.

4. METHOD FOR THE DETERMINATION OF BOTH ^{235}U AND ^{239}Pu

The normal application of the PNAR technique measures the total fissile mass, and for spent fuel it gives the sum of the ^{235}U mass and the Pu fissile mass. To determine the Pu mass separately, it is necessary to measure the ratio of the ^{239}Pu to the ^{235}U. With the DDSI method we have additional information that makes it possible to separate the ^{235}U and the ^{239}Pu. The separation requires the measurement of both singles and doubles rates. The triples rate can also be used to increase the ratio signature. There are two physics signatures that make the determination of the ^{235}U /^{239}Pu ratio possible. One method is based on the difference in the fission multiplicity distributions (ν) , $\nu(\nu\text{-}1)$,and , $\nu(\nu\text{-}1)(\nu\text{-}2)$ for ^{235}U and ^{239}Pu. The singles rate is a function of the first moment, and the doubles and triples rates are a function of the higher moments.

The second method to separate the two fissile components is based on the difference in the neutron fission resonance cross-section structure as a function of energy. Our DDSI technique provides reaction rate versus time and energy, and we can use the time bins in the doubles rate to determine the ratio of ^{235}U to ^{239}Pu .

For the case of a single fuel pin, we have determined the ^{239}Pu fraction using the DDSI method and the liquid scintillator detector described in the next section.

5. PERFORMANCE OF DDSI USING PLASTIC OR LIQUID SCINTILLATORS

The statistical performance of the DDSI method is significantly improved when the ^{3}He detectors are replaced by liquid or plastic scintillator detectors because of their short die-away times and fast data collection electronics. This allows a more complete separation of the early die-away detector region and the late die-away sample region. The sacrifice, when compared to ^{3}He tubes, is increased gamma-ray sensitivity and reduced stability for the scintillator system. A commercial liquid scintillator system (BC-523A) was simulated with the MCNPX code, and the^{3}He detector region was replaced by the ^{10}B-loaded liquid scintillator. The scintillator was loaded with 5% ^{10}B to provide a strong light signal when the epi-thermal neutron was absorbed. For the BC-523A system with an annulus thickness of 68 mm surrounding the lead shield, the calculated efficiency was ~40%, and the die-away time was 0.6 μs. The DDSI efficiency and sensitivity for measuring the induced fission events is significantly improved. The 0.6 μs die-away time of the scintillator provides excellent separation of the early and late time gates.

We have performed MCNPX calculations for the full fuel assembly using the liquid scintillator detector to replace the ^{3}He tubes. Figure 9 shows the induced fissions in the late gate interval divided by the singles rate to cancel the interrogation source strength. We see that the normalized induced fission rate ratios track the fissile content with a large signal to background ratio of ~ 20.

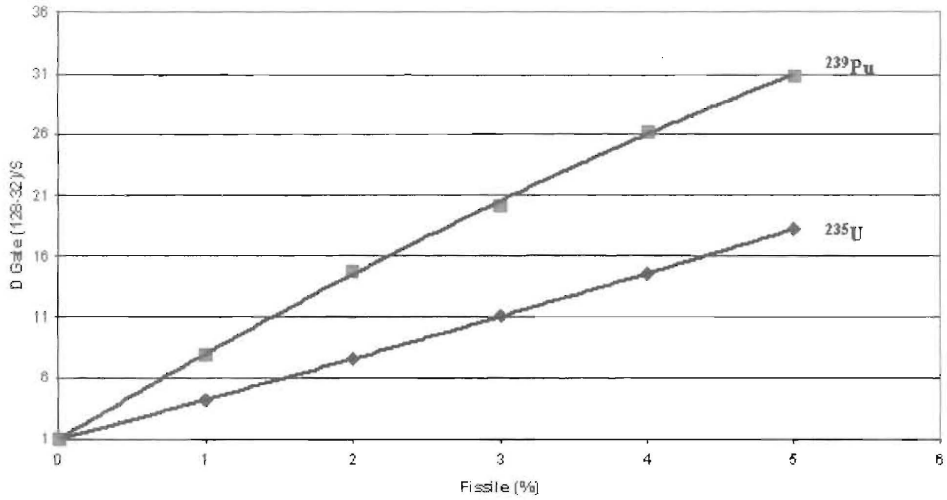


Fig. 9 The doubles/singles ratio for a full fuel assembly as a function of the fissile fraction of the heavy metal.

6. SUMMARY

The DDSI method provides an active-neutron interrogation for the fissile mass measurement as well as a passive-neutron coincidence measurement for the spontaneous fission rate. The spontaneous fission events are primarily from curium for spent fuel The spontaneous fission neutrons provide the neutron "pulse" that initiates the list-mode time analysis of the neutron pulses. The fissile material that is intimately mixed with the fertile material provides the induced fission neutrons that are displaced in time from the spontaneous fission events. For the DDSI design concept, we have optimized two regions of interest: (1) the detector region, with high efficiency and a short die-away time, and (2) the sample interrogation region, with a high thermal-neutron albedo and a longer die-away time. The data are collected in the list mode and analyzed with an early coincidence time gate for the spontaneous fission rate and a late gate for the induced fission events in the fissile mass. The optimum gate lengths are dependent on the sample size in that for a small sample such as a fuel rod, the thermal-neutron albedo is localized to approximately a 2-cm radius surrounding the rod, and neutrons that become thermal energy at greater radii have only a small probability of returning to the rod. For larger samples, such as a fuel assembly, the thermal neutrons from more distant locations have a reasonable solid angle for returning to the assembly.

The list-mode data collection makes it possible to analyze the data for different levels of time correlated multiplicity and for different time gate intervals. The higher levels of multiplicity increase the signature ratio.

The use of a liquid scintillator in place of the ^{3}He tubes significantly reduces the statistical uncertainty for the DDSI method because the die-away time is much shorter for the scintillator. This permits a very short gate for the spontaneous fission trigger events for the time analysis. The drawbacks of the scintillator detectors are less stability and higher gamma- ray sensitivity.

The fast die-away time of the ^{10}B-loaded scintillator detectors provides the possibility of complete time separation between the early and late time gates. In this case, the DDSI concept can be deployed without doing a Cd ratio measurement. When the late gate doubles are normalized by the early gate (~1 μs) doubles, the source strength cancels, and the result provides the fissile content without the necessity for a Cd ratio measurement.

. The results for the full assemblies have much better statistical precision than for single rods because of the high multiplication. Also, the multiplication is greater in the central regions of the fuel assembly than for the perimeter resulting in good penetrability for rod removal scenarios. The separation of the fissile components including ^{235}U, ^{239}Pu, and ^{241}Pu makes use of both the resonance structure differences in the isotopes as well as the higher fission moments distribution for plutonium compared with ^{235}U. Future work will evaluate the penetrability of the method for rod removal scenarios, and the time gates and multiplicity ratios will be optimized for the minimum error in the separation of the fissile isotopes.

7. ACKNOWLEDGEMENTS

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