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FISSILE AND FERTILE NUCLEAR MATERIAL MEASUREMENTS USING A NEW DIFFERENTIAL DIE-AWAY SELF-INTERROGATION TECHNIQUE

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ABSTRACT

This paper presents a new technique for the measurement of fissile and fertile nuclear materials in spent fuel and plutonium laden materials such as mixed oxide (MOX) fuel. The technique, called differential die-away self-interrogation, is similar to traditional differential die-away analysis, but it does not require a pulsed neutron generator or pulsed beam accelerator, and it can measure the fertile mass in addition to the fissile mass. The new method uses the spontaneous fission neutrons from ²⁴⁴Cm in spent fuel and ²⁴⁰Pu effective neutrons in MOX as the "pulsed" neutron source with an average of ~ 2.7 neutrons per pulse. The time correlated neutrons from the spontaneous fission and the subsequent induced fissions are analyzed as a function of time to determine the spontaneous fission rate, the induced fast-neutron fissions, and the induced thermal-neutron fissions. The fissile mass is determined from the induced thermal-neutron fissions that are produced by reflected thermal neutrons that originated from the spontaneous fission reaction. The sensitivity of the fissile mass measurement is enhanced by the use of two measurements, with and without a cadmium liner between the sample and the hydrogenous moderator. The fertile mass is determined from the multiplicity analysis of the neutrons detected soon after the initial triggering neutron is detected. The method obtains good sensitivity by the optimal design of two different neutron die-away regions: a short die-away for the neutron detector region and a longer die-away for the sample interrogation region.

Keywords: spent fuel measurements, plutonium measurements, differential die-away, nuclear safeguards, neutron detectors

INTRODUCTION

The fissile content in special nuclear materials has been measured for more than two decades using the differential die-away analysis (DDA) method [1,2]. The focus of the measurements has been for waste assay and portal monitoring. Traditionally the DDA technique uses a pulsed neutron generator or an accelerator to create a burst of neutrons to interrogate the sample. The pulsing frequency is typically ~100 per second and, between the pulses, the neutron time distribution is measured to determine the thermal-neutron induced fission rate in the sample that is proportional to the fissile content.

The new differential die-away self-interrogation (DDSI) method, described in this paper, uses time correlation neutron counting with triggering on the spontaneous fission events in the assay sample. Thus the need for the external pulse neutron source is eliminated, significantly reducing

the size, cost, and complexity of the system. After each spontaneous fission event, the neutron time distribution is analyzed to determine the fast and slow neutron distributions from the spontaneous and induced fissions in the sample, respectively. The time correlated fast-neutron distribution provides a measure of the spontaneous fission rate from the fertile isotopes as well as the induced fast-neutron fission rate in the sample. Figure 1 illustrates the early and late time windows that are used in the DDSI method. The early gate interval is used to calculate the spontaneous fission rate and the fast-neutron multiplication (M). The late die-away neutron time distribution is populated by thermal-neutron fissions, and it provides a measure of the fissile content in the sample. The signal to background ratio for the induced fissions is further increased by performing the measurement both with and without a cadmium (Cd) sleeve around the sample. This Cd ratio method, referred to as passive neutron albedo reactivity (PNAR) [3], has been used in the past for fissile mass measurements. The new DDSI method.



Fig. 1. 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 interior region where reflected 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 traditional DDA method does not share this problem because the neutron generator pulses are far enough apart for the neutron die-away to reach equilibrium between pulses.

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, and the effective v (prompt neutrons per fission) increases from 2.72 for ²⁴⁴Cm to ~4.6 for the combination of spontaneous fission and induced fissions. The Cd ratios for the singles, doubles, and triples provide three separate measurements for the fissile content in the sample. This paper presents the design of the detector package and the self-interrogation region for the application for the new DDSI method.

We have simulated detector and moderator configurations using Monte Carlo MCNPX [4] calculations to optimize the design of the detectors and moderators for the DDSI method. The results of the experiments and an optimization study for a spent fuel rod and a mixed oxide (MOX) plutonium (Pu) rod are presented in this paper.

DETECTOR AND MODERATOR DESIGN

Preliminary Moderator Measurements

For spent fuel neutron measurements, the gamma-ray background level is very high ($\sim 1E+04$ R/h), and it is necessary to use gamma shielding so that the neutron detector is insensitive to gamma rays. In this publication, ³He gas tubes have primarily been used, but liquid scintillator detectors were also evaluated for applications in lower gamma fields. For the DDSI method, we want to have a neutron detector with both high efficiency and a fast die-away time. Higher ³He gas pressures and relatively thin high density polyethylene (HDPE) moderators are used to obtain good results.

A set of experiments was performed to compare the efficiency and die-away times for different ³He gas pressures for a relatively thin (1.0-cm-thick) HDPE moderator. The ³He tubes were placed in an HDPE moderator that was 300 mm in diameter and 400 mm tall. The 25.4-mmdiameter tubes were in a central 50-mm-diameter hole with a 12-mm-thick HDPE annular sleeve surrounding the tube and a 1-mm-thick Cd sleeve surrounding the HDPE sleeve. Thus, the thermal neutrons that were created outside of the ³He tube annulus were absorbed by the Cd before they could reach the ³He tube and the neutrons detected in the ³He tube were from the 12mm-thick annular sleeve . A small ²⁵²Cf source was positioned a few centimeters away from the ³He tube to set up the time-correlated neutron flux in the tube. Table 1 lists the measured efficiencies (normalized at 4 atm) and measured die-away times together with the MCNPX simulations. Figure 2 shows the normalized singles and doubles efficiencies, and die-away times for the different tube gas pressures. The efficiency increase with ³He gas pressure is more pronounced than it would be for a fully moderated ³He tube system. The reduction in die-away time as the gas pressure increases is a result of counting more of the epithermal neutrons.

pressure. ³ He Gas Pressure (1 inch diameter)	Relative Efficiency (12 mm poly)	Die-away Time (microseconds)	MCNPX Calculated Relative Efficiency	MCNPX Normalized Die-away Time	
4 atm	1.00	17.6	1.00	17.6	
6 atm	1.20	15.3	1.164	15.1	
7.5 atm	1.30	14.1	1.254	13.9	
10 atm			1.369	12.8	

Table 1 Relative neutron efficiency and die-away time as a function of ³He gas



Fig. 2. Simulated efficiency and die-away times as a function of 3 He gas pressure for a 25.4-mmdiameter tube with normalization at 4 atm gas pressure.

Neutron Detector Design

For the DDSI design applied to spent fuel rods, we assembled an annulus of 36 ³He tubes with 10 atm pressure and surrounded the sample in the central position, as illustrated in Fig. 3. Each ³He tube was initially surrounded by an annulus of HDPE and a Cd sleeve to provide the short die-away time. However, the simulations showed that the efficiency could be significantly improved with only a small sacrifice 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 ³He tubes from the more distant positions in the HDPE moderator. The apparent loss of efficiency from the fins is actually a benefit for coincidence counting because the neutrons with longer die-away times put the neutrons outside of the short gate. Figure 4 shows the MCNPX simulation for the detector and sample regions. 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. Detailed material configuration illustrating the Cd fins used to remove the long die-away tail in the detector region.



Fig. 4. DDSI detector configuration for the MCNPX simulations.

Typical spent fuel rods have a ²⁴⁴Cm neutron emission rate of approximately one-half million neutrons per second in the ~500-mm-long 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 optimum design needs to provide a high efficiency and short die-away time.

Sample Interrogation Region Design

The next target of the design was to configure the materials and thicknesses in the sample region to provide a maximum induced fission rate from the reflected thermal neutrons without a significant reduction in the ³He detector efficiency. Also, for the spent fuel application, we needed shielding to provide about a 4 orders of magnitude reduction in the fission product gamma dose at the ³He tube location. The initial design used HDPE and Pb of different thicknesses. About 40 mm of Pb was required for the gamma shielding, and a 25-mm-thick annulus of HDPE surrounding the fuel rod gave the best results. The Pb annulus was also expected to increase the thermal-neutron lifetime in the interrogation region. The MCNPX simulations showed that the HDPE dominated the thermal-neutron lifetime, and the Pb contribution was negligible for the small diameter of the fuel rod.

The optimum interrogation configuration was concentric annuli of 1-mm Cd, 25-mm HDPE, and 0.1-mm Cd, followed by the annulus of lead and the rest of the detector configuration shown in Fig. 4. 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. 4 has a die-away time of $\sim 14 \ \mu s$ in Region 1 bythe ³He tubes and a sample die-away time of $\sim 30 \ \mu s$ in Region 2.

TIME GATE ANALYSIS

There are two time regions of interest, as illustrated in Fig. 1. The first is the early time zone which is 20 μ s in duration to encompass most of the spontaneous fission events (die-away time of 14 μ s). Note that the early (short?) time zone starts after a 1.5 μ s predelay that is necessary for ³He tube and amplifier recovery. The second time zone of interest intended to quantify the induced fissions has a die-away time of ~30 μ . The second time zone extended between 21.5 μ s and 41.5 μ s for the fuel rod geometry. The selection of the time gates depends on the accidental pileup that is a continuum under both of the first two gates. For high source strengths, it is desirable to keep the signal gates short to reduce the accidental pileup counts. The accidental rate can be calculated from the totals rate squared multiplied by the gate length.

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 \sim 5E+05 neutrons/s is expected within the detector geometry (500 mm long). On the other hand, for MOX rods, the primary neutron source is from Pu, and the neutron yield is about 2 orders of magnitude smaller. Thus, the statistical counting error will be better for MOX rods than for spent fuel.

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. The detector configuration shown in Fig. 3 was simulated to determine the singles, doubles, and triples counting rates as a function of gate lengths. The early gate length that is used to determine the spontaneous fission rate has a minimum uncertainty for a gate length of ~20 μ s.

The early gate, used to determine the induced thermal-neutron fission rate, was varied from 16 μ s to 64 μ s, with different times intervals to minimize the statistical error in the doubles rate. The optimum gate, considering both the statistical error and the Cd ratio, was from 21 μ s to 41 μ s. 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.

RESULTS

Spent Fuel Rod

Using the configuration shown in Fig. 4 with an efficiency of 29.1% and a die-away time of 14 μ s in Region 1, the ²³⁵U and the ²³⁹Pu concentrations in the fuel rod were varied from 0 to 5%; note the two isotopes are not mixed. The induced fission signal was obtained from the late coincidence gate (21.5 μ s to 41.5 μ s). 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, and the doubles and triples rates increases the Cd ratio above the singles rate because of the increase in the effective ν (prompt neutrons per fission that is included in our gate interval. Figure 5 shows the Cd ratio for the singles and doubles as a function of ²³⁵U and ²³⁹Pu loading. The non-linear shape of the curves is the result of fissile material self-shielding of the thermal neutrons that interrogate the rod. The Cd ratio for the triples is approximately two times larger than that for the doubles, but we did not include it on the graph because of the large statistical error for the triples rate for spent fuel rods.



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.

One of the DDSI applications is to measure the fissile content in spent fuel rods. After a few years of cooling, the spontaneous fission interrogation source for spent fuel rods is dominated by ²⁴⁴Cm; however, the DDSI technique works the same if ²⁴²Cm is still present in the rod. The early time-gate doubles rate measures the ²⁴⁴Cm mass, and the late time gate, combined with the Cd ratio, measures the fissile mass that is a mixture of ²³⁵U, ²³⁹Pu, and ²⁴¹Pu. For typical spent fuel, the ²³⁵U is ~1% and the Pu fissile is about 0.6%. The ratio of these isotopes can be estimated from the burnup (BU) codes [5,6]. The ²⁴⁴Cm is a strong function of the BU, and the ²⁴⁴Cm is measured in the DDSI early gate so the approximate BU can be determined. The ratios of the fissile isotopes can then be determined from the BU calculation or possible inclusion of results from another nondestructive assay instrument.

The effective fissile content in the spent fuel can be represented by the ²³⁹Pu effective for the doubles rate that is defined as

239
Pu effective = 239 Pu + a 241 Pu + b 235 U,

where

a = the thermal-neutron fission cross-section ratios of $(^{241}Pu/^{239}Pu)$ * the second moments ratios of $(^{241}Pu/^{239}Pu) = (1,000/746)$ * (6.70/6.773) = 1.326, and b = the thermal-neutron cross-section ratios of $(^{235}U/^{239}Pu)$ * the second moments ratios of $(^{235}U/^{239}Pu) = (584/746)$ * (4.636/6.773) = 0.535.

The 239 Pu effective = 1.268 % for the simulated fuel rod.

We can see that 0.535 g of 239 Pu gives the same doubles signal level as 1 g of 235 U; thus 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.

Table 2 lists the singles, doubles, and triples rates for fuel rods with ²³⁵U and ²³⁹Pu contents varying from zero to 5% for the design basis detector configuration shown in Fig. 3. The singles counting rate for the case with zero fissile mass and the Cd liner is 115,826 cps for a neutron source emission rate of 400,000 n/s. The Cd ratio for the doubles rate varies from 0.993 to 1.181 over the same ²³⁵U range. The triples rates have the highest Cd ratios; however, the statistical uncertainty for the triples is large for the high counting rates of spent fuel because of the accidental rate pileup.

The doubles response per gram for ²³⁹Pu is higher than that for ²³⁵U because of the higher thermal-neutron cross section and the higher multiplicity distribution [7].

²³⁵ U	S (Cd)	D (Cd)	T (Cd)	S (no Cd)	D (no Cd)	T (no Cd)			
%	cps	cps	cps	cps	cps	cps	Cd ratio S	Cd ratio D	Cd ratio T
0	115826	4984	96	116404	4949	. 114.0	1.005	0.993	1.192
1	115900	4997	96	118339	5231	164.6	1.021	1.047	1.722
2	115982	5008	96	119739	5455	207.4	1.032	1.089	2.160
3.2	116066	5020	98	121024	5683	254.8	1.043	1.132	2.600
4	116113	5024	98	121757	5813	282.4	1.049	1.157	2.876
5	116172	5033	99	122493	5946	305.4	1.054	1.181	3.091
²³⁹ Pu									
%									
0	115826	4984	96	116404	4949	114.0	1.005	0.993	1.192
1	116041	5022	99	119676	5497	208.8	1.031	1.095	2.118
2	116252	5060	101	121729	5849	271.6	1.047	1.156	2.689
3	116425	5094	102	123222	6142	326	1.058	1.206	3.184
4	116618	5126	104	124298	6345	378	1.066	1.238	3.628
5	116860	5161	106	125267	6551	398	1.072	1.269	3.762
²³⁹ Pu-e									
1.268	116063	5002	93	120335	5606	215.6	1.037	1.121	2.328

Table 2. Singles (S), doubles (D), and triples (T) counting rates and Cd ratios as a function of fissile content for a gate of 21.5 to 41.5 ms and a source emission rate of 4E+05 n/s.

The induced fission content can be measured by the difference between the D (no Cd) and the D (Cd liner) (net D) after dividing by the source emission rate. Figure 6 shows a plot of the net S/S(Cd) and net D/D(Cd) data as a function of 235 U and the 239 Pu effective (fissile content). We see that the 239 Pu provides a larger induced fission signal than that for the 235 U.



Fig. 6. Plot of the data for net D/D(Cd) for the doubles rates as a function of the ²³⁵U and ²³⁹Pu concentrations in the fuel rod, using the MCNPX simulations.

Figure 7 shows the statistical uncertainty in the doubles rates for the assumed 1,000 s counting interval, using the late gate of 21 μ s to 41 μ s. BC-523A refers to a liquid scintillator material discussed in the next section. The uncertainty in the early gate (20 μ s) for the spontaneous fission rate is very small (~0.15%), whereas the uncertainty for the induced fission rate varies from 1% to 7%, depending on fissile composition. The uncertainty for the 239 Pu is typically about half the 235 U uncertainty. For the spent fuel rod, the uncertainty was 2.8%.



Fig. 7. Statistical uncertainty versus fuel rod fissile loading for the 3He tube configuration and the BC-523A detector assuming a 1000 s measurement time.

Fresh MOX Rods

A potential application of the DDSI is to measure the Pu in fresh MOX fuel rods. These rods are presently measured with a combination of neutron coincidence counting, to determine the ²⁴⁰Pu effective, together with high resolution gamma spectroscopy (HRGS), to determine the Pu isotopic ratios [8]. The DDSI provides the same measurement capability for the ²⁴⁰Pu effective as the previous methods [8,9], but in addition, it measures the Pu fissile content in the late gate. The DDSI measures 3 parameters of interest:

- 1. The ²⁴⁰Pu effective,
- 2. The fast-neutron multiplication M, and
- 3. The thermal-neutron induced fissions fissile mass.

The spontaneous fission neutrons have an average energy of about 1 MeV, and they induce fission reactions as they escape from the fuel rod. The resulting fast-neutron multiplication is small (M = 1.01), and it is measured via the doubles/singles ratio in the early coincidence gate [9].

The induced thermal-neutron fissions are measured via the Cd ratio doubles in the late coincidence gate. Thus, for inspection verification purposes, we have measured both the Pu non-fissile mass (²⁴⁰Pu effective) in the early gate and the Pu fissile mass in the late gate. This combination of measurements can provide the total Pu consistency check for International Atomic Energy Agency verification purposes without the need for an HRGS measurement.

A typical MOX fuel rod contains ~7% Pu, and the statistical precision for the ²⁴⁰Pu effective content is ~0.11%. On the other hand, the induced fissions that are measured in the late gate have a larger statistical uncertainty. For the induced fission ²³⁹Pu effective, the precision is ~1% for the 1,000 s measurement.

PERFORMANCE OF DDSI USING PLASTIC OR LIQUID SCINTILLATORS

The statistical performance of the DDSI method is significantly improved when the ³He 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 ³He tubes, is increased gamma-ray sensitivity and reduced stability. A commercial liquid scintillator system (BC-523A) was simulated with the MCNPX code, and in it, the detector region was replaced by the ¹⁰B-loaded liquid scintillator. The scintillator was loaded with 5% ¹⁰B to provide a strong light signal when the thermal neutron was absorbed. This thermal-neutron capture event provided good energy separation from gamma-ray events. Because the neutron absorber was intimately mixed with the hydrogenous scintillator, the die-away time was significantly reduced, and the fast electronics allowed the pre-delay time to be negligible.

For the BC-523A system with an annulus thickness of 68 mm surrounding the lead shield, the calculated efficiency is ~40%, and the die-away time is 0.6 μ s. The DDSI efficiency and sensitivity for measuring the induced fission events is significantly improved. The statistical uncertainty is shown in Fig. 7 for a 1,000 s measurement. The 0.6 μ s die-away time of the scintillator allows excellent separation of the early and late time gates, so the doubles Cd ratio increases to ~12. For fresh MOX fuel with the high ²³⁹Pu content, it is possible to measure both the spontaneous fission rate (²⁴⁰Pu effective) and the induced fission rate (²³⁹Pu effective) without performing the Cd ratio measurement. Figure 8 shows the induced fissions in the gate interval from 6 μ s to 40 μ s divided by the neutron doubles rate in the early gate (0 to 1 μ s) to

cancel the interrogation source strength. We see that the normalized induced fission rate tracks the fissile content, as shown in Fig. 8.



Fig. 8. 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 239Pu fuel rod.

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 and ²⁴⁰Pu effective from Pu and MOX 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 (from 1-20 μ s) for the spontaneous fission rate and a late gate (from ~20–40 μ s) 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 2cm 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 ³He tubes were filled with 10 atm of gas pressure to increase the detection efficiency and to decrease the die-away time. The ratio of the efficiency to the die-away time is increased by a factor of \sim 2 in going from 4 atm to 10 atm fill pressure. The location and thickness of the Cd

liners plays a key role in the DDSI design optimization. There are Cd liners at fixed locations around the ³He tubes and removable Cd around the sample. It is important that the ³He tubes be surrounded by approximately a uniform thickness of HDPE that is inside the fixed Cd liners so that the early gate has a fast die-away time without a slow component.

A typical spent fuel rod corresponding to a pressurized water reactor with a BU of 40 MWd/tU would have a fissile concentration of ~1% 235 U and about 0.5% Pu fissile. For this of type fuel rod, the 244 Cm source singles counting rate for the present DDSI detector would be ~125,000 cps and, assuming a 1,000 s measurement, the statistical error for the spontaneous fission rate would be ~0.15%; for the induced fissile doubles rate, the statistical error would be ~2.8%. For spent fuel measurements, the systematic errors related to calibration would likely be larger than the statistical uncertainty.

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 give a larger Cd ratio but have a larger statistical error. For the design basis spent fuel rod, the detector design shown in Fig. 4 provided a Cd ratio of 1.04 for singles, 1.15 for doubles, and 2.4 for triples rates analysis. The statistical uncertainty, as a function of fissile content in the fuel rods, are shown in Fig. 6.

For an unirradiated MOX fuel rod, the counting rates are much lower, and that reduces the uncertainty for the triples rates. The statistical error for the prompt neutron ²⁴⁰Pu effective measurement is ~0.15% for the doubles. For the induced fissions in the fissile component, the statistical uncertainty for the doubles is ~1.0% in a measurement of 1,000 s.

The use of a liquid scintillator in place of the ³He 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. Future studies will evaluate the scintillator detectors and the capability of ¹⁰B, ⁶Li, or Gd loadings to reduce the gamma-ray sensitivity.

The fast die-away time of the ¹⁰B-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 (\sim 1 µs) doubles, the source strength cancels, and the result provides the fissile content without the necessity for a Cd ratio measurement as shown in Fig. 8.

Future simulations using the MCNPX code will extend the self-interrogation concept to spent fuel assemblies where the neutron source term is about 2 orders of magnitude higher than the present case for a fuel rod. The dead-time considerations will require a reduced detector length and fast die-away time for the neutron detectors.

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REFERENCES

- 1. J. T. Caldwell et al., "Apparatus and method for quantitative assay of generic transuranic wastes from nuclear reactors," US Patent No. 363,979, (1982).
- Kelly A. Jordan and Tashi Gozani, "Detection of ²³⁵U in hydrogenous cargo with differential die-away analysis and optimized neutron detectors," *Nucl. Instrum. Methods Phys Res A* 579 1, 388–390 (2007).
- H.O. Menlove and D.H. Beddingfield, "Passive neutron reactivity measurement technique," Los Alamos National Laboratory document LA-UR-97-2651 (May 1997).
- 4. *MCNPXTM USER'S MANUAL Version 2.5.0*, J.F. Pelowitz, Ed., Los Alamos National Laboratory report LA-CP-05-0369 (2005).
- ORIGEN Code 2. SCALE: A Modular Code System for Performing Standardized Computer Analyses for Licensing Evaluations, ORNL/TM-2005/39, Version 5, Vols. I-III, April 2005. Available from Radiation Safety Information Computational Center at Oak Ridge National Laboratory as CCC-725. Origen-arp.
- W.B. Wilson, et al, "Accelerator Transmutation at Los Alamos with CINDER 90", Proceedings of the Workshop on Simulation of Accelerator Radiation Environments, Jan 11-15, Santa Fe, New Mexico, (1993).
- 7. D. Reilly, et al,"Passive Nondestructive Assay of Nuclear Materials", United States Nuclear Regulatory Commission, NUREG/CR-5550, LA-UR-90-732, (March 1991).
- 8. M.S. Krick and J.E. Swansen, "Neutron multiplicity multiplication measurements," *Nucl. Instrum. Methods* **219 38**, 393 (1984).
- 9. N. Ensslin et al., "Application guide to neutron multiplicity counting," Los Alamos National Laboratory report LA-13422-M (1998).