Discriminating Uranium Isotopes Based on Fission Signatures Induced by Delayed Neutrons

K. Ogren,* J. Nattress,† and I. Jovanovic‡
Department of Nuclear Engineering and Radiological Sciences,
University of Michigan, Ann Arbor, MI 48109 USA
(Dated: November 15, 2019)

The use of active interrogation (AI) to induce delayed neutron emission is a well-established technique for the characterization of special nuclear materials (SNM). Delayed neutrons have isotope-characteristic spectral and temporal signatures, which provide the basis for isotope identification. However, in bulk materials that contain an appreciable fissile (e.g., \(^{235}\)U or \(^{233}\)U) fraction, such as highly-enriched uranium (HEU), delayed neutrons have a high probability of inducing additional fissions. As a result, the overall delayed neutron signature consists of two distinct components: the “primary” delayed neutrons (emitted directly by fission fragments), and the “secondary prompt” fission neutrons produced in fission induced by primary delayed neutrons. These prompt products differ from “primary” delayed neutrons both in their energy spectra and in the presence of coincident radiation released by the parent fission event. The presence and relative quantity of prompt products from delayed fission depend on the cross-section of the material in the energy range of delayed neutrons, which may differ significantly between isotopes, thus providing an exploitable means for isotope differentiation. In this work, we demonstrate two experimental approaches for discriminating between \(^{235}\)U and \(^{238}\)U isotopes based on the measurement of delayed neutron-induced fission products. First, HEU and depleted uranium objects are differentiated through the detection of high-energy prompt neutrons from delayed fission using both recoil-based organic liquid scintillators and thermalization spectra from a custom-built capture-gated composite detector. Secondly, coincident radiation measurements are used as the basis for discrimination by comparing the overall rates and time evolution of fission events when delayed neutrons are present.

I. INTRODUCTION

Measurement methods that can provide detailed information on the composition of special nuclear materials (SNM) are integral to many nuclear security and non-proliferation applications. In particular, determination of the relative isotopic abundance of \(^{235}\)U and \(^{238}\)U plays a central role in international safeguards inspections, the production and accounting of nuclear fuel, and verification of proper storage and dismantlement of weapons materials [11]. Fission reactions generate a variety of unstable nuclei, which are typically neutron-rich and undergo \(\beta\)-delayed \(\beta\) decay. A fraction of the decaying nuclei also release their energy through neutron emission. The delay between the initial fission event and these secondary emitted neutrons depends on the chain decay kinetics, which are characteristic of the fission fragments, and ranges from a few hundreds of nanoseconds to tens of seconds. The \(\beta\)-delayed neutrons are commonly divided into a set of groups based primarily on similarities in their precursor half-lives [12]. The specific parameters for the groups depend on the isotope undergoing fission, as well as the type and energy of the fission-inducing particle. Despite the fact that delayed neutrons account for only a small fraction of the overall fission neutron yield, detection systems that utilize AI to intensify the emitted delayed neutron signal have been successfully used to detect and identify fis-
Because the delayed neutron groups for a particular fissionable isotope have a unique set of individual decay constants, each isotope possesses a characteristic aggregate temporal profile for delayed neutron emission, which can be used as the basis for identification. Previous studies have applied this principle to differentiate SNM samples by measuring the decay of the delayed neutron rate for short-lived [19] and long-lived groups [20–22]. In our own prior work, we have utilized both the buildup and decay time profiles of long-lived delayed neutron groups to perform isotopic discrimination and infer the enrichment level of uranium [23]. While the energy and timing characteristics of delayed neutrons have been determined by dedicated precision measurements, the observed delayed neutron signature for bulk materials can be complicated by additional interactions before the delayed neutrons escape the object. In the case of fissile materials such as $^{235}$U and $^{239}$Pu, delayed neutrons with an average energy of 250–450 keV can readily induce numerous additional fission events. This delayed neutron-induced fission is a basic concept in nuclear reactor kinetics, where it represents an important consideration in maintaining the desired state of criticality in a reactor. In the context of neutron energy spectrum, however, it leads to an overall delayed signal that is a superposition of two components: “primary” delayed neutrons, which are emitted directly from the decay of fission fragments, and the prompt fission neutrons, where the fission is induced by delayed neutrons. In fact, in order to measure the delayed neutron energy spectra with a high degree of accuracy, sample sizes have been restricted to small amounts (<10 g) of material with the express purpose of limiting distortions caused by fission multiplication [12, 24, 25].

Because the prompt fission products of the delayed neutron-induced fission events are emitted nearly instantaneously, they mimic the time distribution of the delayed neutrons, and do not significantly alter the measured neutron temporal profile (notwithstanding the potential differences in detector efficiency when measuring the time-evolving neutron spectrum). In contrast, the overall delayed energy spectrum is significantly changed by the introduction of prompt fission neutrons, which typically have much higher energies than delayed neutrons. Furthermore, the prompt neutrons from delayed fission are accompanied by additional coincident neutrons and γ rays. The determining factor in the relative abundance of prompt fission products in the delayed signal is the average fission cross-section of a particular material in the delayed neutron energy range. Because this cross-section may differ significantly between isotopes, as is the case for $^{235}$U and $^{238}$U, the detection of high-energy prompt neutrons and coincident radiation from fission in the delayed signature of SNM may provide the basis for isotopic identification.

Here, we present two experimental methods for disambiguating the genesis of delayed neutrons as a means for discriminating $^{235}$U and $^{238}$U. In the first approach, recoil-based organic liquid scintillators and a custom-built capture-gated composite detector are used to perform spectroscopic measurements of delayed neutrons, and highly-enriched uranium (HEU) is successfully differentiated from depleted uranium (DU) based on the presence of high-energy prompt neutrons in its delayed signature. While the measurement of high-energy prompt neutrons from delayed neutron-induced fission has previously been proposed as a method for discriminating uranium isotopes [26], to the best of our knowledge, this work represents the first time that such energy information has been specifically targeted and extracted from the overall delayed signature as a means for isotopic identification. In the second approach, we demonstrate the first use of coincidence counting to observe the contribution of delayed neutron-induced fission to the overall fission rate and successfully differentiate HEU from DU on this basis.

II. MATERIALS & METHODS

Experimental measurements were performed at the Device Assembly Facility (DAF), Nevada National Security Site, using highly-enriched uranium (HEU) and depleted uranium (DU) test objects of similar mass. A set of concentric HEU hemispheres known as the Rocky Flats shells was used as the HEU object. The Rocky Flats shells have a bulk density of 18.664 g/cm³ and an isotopic content of 93.16% $^{235}$U, 5.35% $^{238}$U, and less than 2% of other isotopes of uranium [27]. Shells 01–24 were assembled into a spherical object with an approximate mass of 13.8 kg. Detailed information on the masses and dimensions of the individual shells can be found in Ref. [27]. The depleted uranium object also consisted of a set of hemispherical shells arranged to form a sphere. The mass of the DU object was 12.8 kg, which was the closest approximation to the HEU object mass that could be achieved with the available shell configurations.

Each uranium object was interrogated with 14.1-MeV neutrons produced by a Thermo Scientific P211 DT neutron generator, with an approximate isotropic yield of $10^8$ n/s. The objects were placed at a distance of 13 cm from the generator, as measured from the center of the object to the center of the target plane in the generator tube. The DT generator was operated at a pulse rate of 100 Hz with a pulse width of approximately 10 µs, which was consistent across all measurements.

In each measurement, the uranium object was surrounded by an array of detectors, which included one two-inch NaI(Tl) detector, two three-inch Eljen EJ309 organic liquid scintillators [28], and one custom-built heterogeneous composite scintillator. The composite detector is a larger version of the prototype described in Ref. [29], and consists of an array of $1 \times 1 \times 76$ mm³ GS20 lithium glass square rods embedded in a cylindrical matrix of scintillating polyvinyl toluene (PVT) with a height and diameter of 12.7 cm. The GS20 glass is 6.6% lithium...
by weight, and is enriched to approximately 95% $^6$Li. A diagram of the composite detector design is shown in Fig. 1.

![Diagram of the composite detector design](image)

The principal detection mechanism for the composite detector is neutron capture by $^6$Li, which has a reaction $Q$-value of 4.8 MeV and releases a triton and an alpha particle. The short range of the heavy charged particle products means that they generally deposit most of their energy in the lithium-doped glass, which possesses very different scintillation properties from the PVT plastic. As a result, neutron capture events are easily distinguishable by both a characteristic pulse shape and the characteristic $Q$-value of the reaction. The PVT matrix surrounding the lithium glass rods serves a dual purpose. Not only does it increase the capture efficiency of the detector by moderating the incident neutrons, but the scintillation response of the PVT to proton recoils in the neutron thermalization process provides a signal whose magnitude is correlated to the incident neutron energy [30]. By exploiting the time coincidence between a capture pulse and the preceding thermalization pulse, spectroscopic neutron energy analysis can be performed [31].

For each measurement, the composite detector was placed vertically at a distance of 21 cm from the central axis of the PVT cylinder to the center of the uranium object. The NaI(Tl) and EJ309 scintillators were placed at different locations around the object, each at a distance of 11 cm from the center of the object to the front face of the detector. Fig. 2 shows the experimental setup used for measurement of both uranium objects.

Each detector was powered using a CAEN DT5533 high-voltage power supply, and the output was digitized using a CAEN DT5730 14-bit, 500-MHz desktop waveform digitizer with digital pulse processing-pulse shape discrimination (DPP-PSD) firmware. Data acquisition and storage was performed using CAEN Multi-Parameter Spectroscopy Software (CoMPASS) [32]. For each waveform, short-gate ($Q_{\text{short}}$) and long-gate ($Q_{\text{long}}$) charge integrals were recorded to provide the basis for pulse shape discrimination. The integration boundary parameters were defined relative to $t_{\text{start}}$, the start of the waveform determined by the leading edge trigger time in the digitizer. $Q_{\text{short}}$ was integrated from $t_{\text{start}} - t_{\text{offset}}$ to $t_{\text{start}} + t_{\text{short}}$, and $Q_{\text{long}}$ was integrated from $t_{\text{start}} - t_{\text{offset}}$ to $t_{\text{start}} + t_{\text{long}}$, where $t_{\text{offset}}$ is the offset time prior to the start of the waveform, and $t_{\text{short}}$ and $t_{\text{long}}$ are the endpoints for the short-gate and long-gate integration windows, respectively. The integration parameters were optimized for each detector prior to the experiment, and are presented in Table I.

![Experimental setup during measurement of the HEU object](image)

### TABLE I: Waveform integration parameters.

| Detector | $t_{\text{offset}}$ (ns) | $t_{\text{short}}$ (ns) | $t_{\text{long}}$ (ns) |
|----------|--------------------------|------------------------|------------------------|
| Composite | 24 | 36 | 376 |
| EJ309 - 1 | 24 | 36 | 376 |
| EJ309 - 2 | 50 | 50 | 350 |
| NaI(Tl) | 24 | 176 | 776 |

The neutron generator was operated in a series of on/off cycles, during which the induced delayed neutron signatures of the HEU and DU objects were recorded. In each cycle, the generator was turned on for one minute, then off for one minute. Each object was interrogated over a period of about 2.5 hours ($\sim$70 cycles), and the data collected during the off cycles was aggregated to form the overall delayed signal. Passive measurements of the HEU and DU objects were also recorded, for 3 minutes and 10 minutes, respectively. The detectors were calibrated using $^{137}$Cs and AmBe sources.

### III. SIMULATION

To estimate the expected contrast in the delayed neutron emission spectra from bulk samples of HEU and DU, Monte Carlo simulations were conducted using the MCNPX-PoliMi particle transport code [33]. Spherical HEU and DU objects of the approximate size and mass of the experimentally measured assemblies were interrogated with 14.1-MeV neutrons, and the time of generation and initial energy of emitted neutrons were recorded upon their arrival in an organic liquid scintillator.
prompt and delayed neutron spectra were separated using a simple time threshold, where neutrons arriving at the detector within 10 microseconds of the generation of the source particle were vetoed. For an average source-detector distance of 20 cm, prompt neutrons would have to have energy lower than about 0.5 keV to arrive after the veto time, and portion of the Watt distribution that falls within this range is less than 0.1%. Within the delayed neutron data, primary delayed neutrons and prompt neutrons from delayed fission were differentiated by tracking their individual histories in the MCNPX-PoliMi collision file output. Based on the particle and generation numbers of each delayed neutron, the corresponding parent fission event was located within the fission chain in the uranium object. If the time delay between the parent fission and detection of the delayed neutron was greater than 10 μs, the event was categorized as a primary delayed neutron. Otherwise, it was considered to be a prompt neutron from delayed fission. This analysis method allowed for estimation of the relative contribution of each type of delayed neutron to the overall delayed spectrum emitted by each object. For HEU, prompt neutrons from delayed fission account for about 65% of the delayed neutron signature, with the remaining 35% contributed by primary delayed neutrons. In contrast, primary delayed neutrons made up over 98% of the emitted signal for DU.

In addition to the experimental object materials, simulations of the expected relative proportion of primary delayed neutrons and prompt neutrons from delayed fission were performed for low-enriched uranium (LEU) to explore the potential for finer determination of the enrichment level based on the induced neutron spectrum. Enrichment levels of 5% and 20% were chosen for simulation, as they represent common benchmarks on the uranium enrichment scale (reactor fuel and the LEU enrichment limit, respectively). Table II shows a summary of the simulated neutron spectrum breakdown for each material.

| Material (Enrichment) | Primary Delayed | Prompt |
|-----------------------|-----------------|--------|
| DU (0.02%)            | 98.5%           | 1.5%   |
| LEU (5%)              | 86%             | 14%    |
| LEU (20%)             | 72%             | 28%    |
| HEU (93%)             | 35%             | 65%    |

While higher-energy prompt neutrons make up only a small fraction of the delayed neutron spectrum for DU, they form a significant part of the delayed signal for 5%-enriched LEU. As a result, it may be possible to distinguish LEU from unenriched uranium based on the presence of higher-energy neutrons in the delayed neutron signature. Furthermore, the relative contribution from prompt neutrons doubles as enrichment increases from 5% to 20%, and more than doubles again for weapons-grade enrichment levels (>90%). Such separation suggests that the proportion of higher-energy delayed neutrons may serve as an observable for estimating the enrichment level of uranium-containing materials.

For each material, the fractional contributions of primary delayed neutrons and prompt neutrons from delayed fission were used to approximate the delayed neutron energy spectra, which were then used to simulate the expected response in the composite detector using the Geant4 framework [34]. While MCNPX is better suited for simulating the production of delayed neutrons, Geant4 does not have the same geometrical limitations, making it the more convenient choice for modeling the complex structure of the composite detector. To simulate the light output response to thermalization recoils in the PVT, the detector was bombarded with neutrons with energies sampled from the delayed [12] and prompt [35] energy spectra in accordance with their relative proportion for each isotope. The light output produced by neutron elastic scatters on protons in the detector was modeled using a similar method to the one described in Ref. [36], with a polynomial function of the form

\[ L = aE - b[1 - \exp(-cE)], \]

where \( L \) is the light output, \( E \) is the energy deposited on the proton, and \( a, b, \) and \( c \) are fitting parameters. The light output contribution from scatters on carbon nuclei was assumed to be approximately 2% of the energy deposited. A Gaussian broadening function was parameterized and applied to the calculated light output according to the method outlined in Ref. [37]. Fig. 3 shows the simulated delayed neutron energy spectra for bulk HEU and DU, as well as the expected light output response of the composite detector.

The simulated delayed energy spectra and corresponding detector response show very significant differences for each material based on the presence of delayed fission events, especially at higher energies. This suggests that \(^{235}\text{U}\) and \(^{238}\text{U}\) should be readily distinguishable based on the presence of high-energy neutrons in the measured delayed signal.

### IV. EXPERIMENTAL RESULTS & DISCUSSION

A significant advantage of the composite detector is that it provides strong discrimination of neutron-capture events on \(^{6}\text{Li}\), which can then be used to identify potential preceding thermalization pulses in the detector and extract spectroscopic energy information from the incident neutrons. Fig. 4 shows the pulse-shape parameter (PSP) and light output distribution for the AmBe calibration measurement in the composite detector, where the PSP is defined as

\[ PSP = (Q_{\text{long}} - Q_{\text{short}}) / Q_{\text{long}}. \]
The parameter space located around $PSP = 0.55$ and light output of about 0.32 MeVee corresponds to neutron capture events. A 3-$\sigma$ cut was established in two dimensions around this region, and any events falling within the cut were classified as neutron captures in the subsequent measurements.

The delayed neutron energy spectra for HEU and DU were compared by analyzing the capture-gated thermalization light output response in the composite detector. For each neutron capture event, the previously recorded pulse was examined to determine if it could have been caused by thermalization of the fast neutron in the PVT prior to capture. Because the type of PVT used in the composite detector is not PSD-capable, there is only one recoil region corresponding to both neutron and gamma-ray interactions. A Gaussian fit to this region established a mean $PSP$ value of 0.0865, and pulses exhibiting deviation from the mean greater than 3-$\sigma$ were rejected. Geant4 simulations were also used to determine the time scale of neutron thermalization in the composite detector. Both prompt and delayed incident neutron energy spectra were modeled; the results indicate that 99% of captures occur within 76 $\mu$s of the initial scattering interaction in the detector, and that incident neutron energy has little effect on the shape of the time distribution of thermalization pulses. As such, candidate thermalization pulses that occur more than 76 $\mu$s before the subsequent capture event were also rejected from the thermalization light output distribution. Fig. 5 shows the resulting capture-gated thermalization light output distributions for HEU and DU.

FIG. 3: (a) Simulated delayed neutron energy spectra for bulk HEU and DU, based on proportional contribution of prompt products of delayed fission, and (b) simulated response of the composite detector to overall HEU and DU delayed energy spectra. In both cases, counting results are based on simulation of 250,000 source particles.

FIG. 4: $PSP$ and light output distribution in the composite detector when exposed to an AmBe calibration source. Neutron captures were identified using a 3-$\sigma$ cut around the island feature centered at $PSP = 0.55$ and light output of 0.32 MeVee.

FIG. 5: Comparison of thermalization light output distributions for HEU and DU. The count rate for DU above 400 keVee is consistent with the measured rate of gamma-ray accidentals within the thermalization time window.
While some increase in the delayed neutron counting rate is expected for HEU relative to DU due to the greater mass of the HEU object and increased fission cross section for 14.1-MeV neutrons, the higher overall rate is also consistent with increased multiplication caused by delayed neutrons. However, the marked increase in high-light-output events for HEU indicates a significant difference in the overall energy spectrum, which can be explained by the presence of higher-energy prompt neutrons from delayed fission.

Examination of the time distribution of thermalization pulses can provide additional insight into the observed light output spectra. While the simulated light output response for delayed neutrons from $^{238}\text{U}$ is largely restricted to below 400 keVee, the experimentally measured distribution extends to higher light outputs. Since the delayed neutrons do not have enough energy to produce higher light output pulses in the detector (and a large fraction cannot even produce a pulse above a 100 keVee detection threshold), this suggests either that there is a higher rate of delayed fission than predicted by tabulated nuclear data, or that the thermalization light output distribution is dominated by gamma-ray accidentals from background or passive emission by the DU object. If accidentals dominate, then the distribution $I(t)$ of time differences between two adjacent pulses is governed by the general expression

$$I(t) = r \exp(-rt), \tag{3}$$

where $t$ is the time between two pulses, and $r$ is the rate of accidentals. In the scenario where there are very few true thermalization events, Eq. (3) predicts that the time distribution should exhibit a simple exponential decay shape. Fig. 6 shows the experimental thermalization time distributions for HEU and DU, which have been fitted with an exponential function representing the expected contribution of background accidentals.

In both cases, the exponential function was fitted to the time window beyond 90 ns, where the contribution of true neutron thermalization events is negligible (less than 1% of the total distribution). The model is then extended back over the range of possible neutron thermalization times. In the case of DU, the exponential function is consistent with the entire distribution, suggesting that few, if any, of the pulses preceding neutron capture events are caused by thermalization of delayed neutrons. In contrast, the time distribution for HEU departs significantly from a simple exponential shape in the neutron thermalization window, and the point where this deviation becomes noticeable is near the maximum thermalization time of 76 $\mu$s predicted by simulation. After subtracting the background exponential fit, 98.5% of thermalization pulses occur within the 76 $\mu$s time window, in close agreement with the simulated result of 99%. Furthermore, the slope of the exponential fit ($-0.02358$) is the same for each data set, which suggests a common cause of accidentals, such as gamma-ray background.

Additionally, the overall rate of recoil pulses recorded by the composite detector between generator runs for DU is consistent with the assertion that the events in the DU thermalization spectrum above 400 keVee are caused by gamma-ray accidentals in the thermalization window. When the generator is turned off following interrogation, the observed rate of recoil pulses is $\sim 2.1 \times 10^4 s^{-1}$. At this rate, the average time between events is 48 $\mu$s, and the probability of observing a random recoil pulse within the 76 $\mu$s time window preceding a capture pulse is 79.5%. In the experimental results, 74.0% of recorded capture events are accompanied by a preceding pulse within 76 $\mu$s.

The observed rate of events within the thermalization window is close to the prediction if all thermalization candidate pulses were truly from gamma-ray accidentals, but it is still somewhat too low. This may be explained by the effects of delayed neutron contributions to the recoil PSP region and pileup pulses in the neutron capture region. Specifically, if the recoil rate contains significant contributions from delayed neutrons, the rate of events in the thermalization window will be higher than expected for random gamma-ray accidentals because neutron re-
coils are much more likely to be followed by a subsequent capture event. Furthermore, misclassified pileup pulses in the capture PSP region are less likely to be preceded by a neutron recoil event. This results in a situation where the random gamma-ray rate, and thus the probability of an event within the thermalization time window, is slightly overestimated.

To correct for the effects of delayed neutron recoils on the estimation of the gamma-ray accidental rate, a 400 keVee light output threshold was applied, effectively removing neutron contributions from the spectrum. The threshold was determined based on the simulated light output response of the composite detector to delayed neutrons, as shown in Fig. 3. The overall rate of recoil events above the threshold was $5.6 \times 10^3 \, \text{s}^{-1}$, resulting in an average time between events of 177 $\mu$s and a 34.9% probability of observing a random event within the 76 $\mu$s thermalization window. In the experimental data, 33.9% of capture events were accompanied by a preceding recoil event within 76 $\mu$s, which strongly suggests that the rate of thermalization events in the DU light output spectrum (Fig. 5) above 400 keVee is fully accounted for by gamma-ray accidentals. Table III provides a summary of the recoil rates, probability of gamma-ray accidentals in the thermalization window, and comparison to experimental data, both with and without application of the 400 keVee light output threshold.

TABLE III: Summary of overall recoil rates in the composite detector and probability of gamma-ray accidentals within the 76 $\mu$s thermalization window. Predicted probabilities based on the overall rate are compared to experimental results.

| Event Type | All Events | Events Above 400 keVee |
|------------|------------|-----------------------|
| Recoil Rate ($\text{s}^{-1}$) | $2.1 \times 10^4$ | $5.6 \times 10^3$ |
| Average Time Between Events ($\mu$s) | 48 | 177 |
| Probability of Event Within 76 $\mu$s (%) | 79.5 | 34.9 |
| % of Captures with Preceding Event Within 76 $\mu$s | 74.0 | 33.9 |

Recoil-based organic liquid scintillators are poorly suited to detecting lower-energy primary delayed neutrons because those neutrons are unlikely to produce a response above a detection threshold of $\sim$100 keVee. However, higher-energy prompt neutrons from delayed fission can be easily detected, as they are far more likely to produce a pulse above the threshold. Fig. 7 shows the PSP and light output distributions measured by the EJ309 detector for HEU and DU, respectively. In the HEU data, a fast neutron recoil region around $\text{PSP}=0.28$ is readily apparent. However, this feature is entirely absent from the DU distribution. This stark contrast provides convincing evidence that the presence or absence of high-energy fission neutrons in the delayed neutrons spectrum can be used to perform isotopic discrimination.

Coincidence measurements were also used to detect the presence of delayed neutron-induced fission events. Coincidence time distributions were recorded for HEU and DU using two different detector pairings: composite-EJ309 and composite-NaI(Tl). In each case, the coincidence event rate recorded during the neutron generator off cycles was compared with the rate observed during passive measurement for each uranium object. Example coincidence time distributions measured during the delayed neutron window are shown in Fig. 8 for the composite-EJ309 and composite-NaI(Tl) detector pairings, respectively.

A summary of the measured coincidence rates for each detector pairing and scenario is presented in Table IV. Because both samples contain $^{238}\text{U}$, which undergoes spontaneous fission, the comparison between HEU and DU is not as simple as noting the presence or absence of coincident radiation from fission. While the relative change in the total coincidence rate after interrogation (when delayed neutrons are present) is much greater for HEU, suggesting that much of the change is due to delayed neutron-induced fission events, this interpretation must be weighted against a number of complicating fac-
FIG. 8: Coincidence time distributions for HEU measured during the delayed neutron time window based on (a) composite-EJ309 and
(b) composite-NaI(Tl) detector pairings.

First, the measured passive coincidence rate for each
material is much too high to attribute to spontaneous
fission alone. Based on the material compositions, a spe-
cific activity of 12.44 MBq/kg, and a spontaneous fission
probability of $5.4 \times 10^{-7}$ per decay, the expected sponta-
neous fission rate for the DU and HEU objects would be
87 fission/s and 5 fissions/s, respectively. MCNPX sim-
ulations of the experimental configuration showed that
the probability of a fission event producing observable
coincident pulses in the composite and EJ309 detectors
was 0.3% for HEU and 0.053% for DU. For coincidence
pulses in the composite and NaI(Tl) detectors, the proba-
bilities were 0.08% and 0.0006% for HEU and DU, respec-
tively. Table V shows the expected spontaneous fission
(SF) tagging rates for each object based on the calculated
spontaneous fission rates and simulated tagging proba-
bilities. In both cases, the expected tagging rates are
several orders of magnitude lower than the experimen-
tally observed coincidence rates. Further investigation
of the coincidence pulses showed that they were consist-
tent with true recoil events and not caused by spurious

| HEU | DU  |
|-----|-----|
| Passive Rate (s$^{-1}$) | 55.87 ± 0.70 | 50.30 ± 0.34 |
| Active Rate (s$^{-1}$) | 89.17 ± 0.25 | 58.90 ± 0.16 |
| % Change | 60% | 17% |

| HEU | DU  |
|-----|-----|
| Passive Rate (s$^{-1}$) | 1.70 ± 0.25 | 2.62 ± 0.14 |
| Active Rate (s$^{-1}$) | 3.78 ± 0.10 | 2.91 ± 0.07 |
| % Change | 122% | 11% |

| HEU | DU  |
|-----|-----|
| SF Rate (s$^{-1}$) | 5 | 87 |
| Tagging Probability | 0.30% | 0.053% |
| Tagged Event Rate (s$^{-1}$) | 0.015 | 0.046 |
| SF Rate (s$^{-1}$) | 5 | 87 |
| Tagging Probability | 0.08% | 0.006% |
| Tagged Event Rate (s$^{-1}$) | 0.004 | 0.0052 |

TABLE IV: Measured coincidence rates for HEU and
DU for composite-EJ309 and composite-NaI(Tl)
detector pairings. For each material, the coincidence
rate observed during delayed signal measurements was
compared to the coincidence rate for passive
measurements.

TABLE V: Simulated spontaneous fission (SF) rates,
tagging probability, and expected tagged event rates for
HEU and DU using each detector pairing.
During the experiment means that thermal neutron induced fission may also have been a factor.

With the goal of eliminating events that were not caused by fission, the coincidence rates were reexamined while only accepting neutron recoil pulses from the EJ309 detector. Fig. 9 shows the resulting coincidence time distributions for HEU and DU after interrogation with the neutron generator. Notably, the neutron-based coincidence time distribution for HEU is much broader than the corresponding distribution for DU and previous distributions where all event types were accepted. This is due to differences in the time of flight for different neutron energies, and perhaps to some extent by the fact that coincident pulses may be produced by radiation from different generations in the fission chain reaction. The broadening of the HEU coincidence distribution is also accurately reflected in the MCNPX model, which has a full-width at half maximum (FWHM) of 15.7 ns, compared to 16.1 ns for the experimental data. A comparison of the simulated and experimentally measured coincidence peaks for HEU is shown in Fig. 10.

Table VI presents a summary of the simulated and experimentally measured coincidence rates when the neutron recoil criterion is applied. When coincidence events are required to contain at least one neutron interaction, the experimentally measured passive rates are much closer to those predicted by simulation. In the case of DU, the measured passive rate is within 2-\(\sigma\) of the simulated rate. After interrogation, the coincidence rate for DU increases by more than a factor of 2, even though delayed neutrons are not expected to cause an increase in coincidences due to fission, but this is most likely due to an increased number of pileup events in the neutron recoil region for EJ309, as Fig. 7(b) suggests.

**TABLE VI: Simulated and experimental fission tagging rates for HEU and DU using the composite-EJ309 detector pairing, with only neutron events accepted for the EJ309 detector.**

|            | HEU       | DU       |
|------------|-----------|----------|
| SF Rate \(s^{-1}\) | 5         | 87       |
| Tagging Probability | 0.25%     | 0.03%    |
| Tagged Event Rate \(s^{-1}\) | 0.012     | 0.026    |

**Experiment**

|            | HEU       | DU       |
|------------|-----------|----------|
| Passive Rate \(s^{-1}\) | 0.442 ± 0.054 | 0.035 ± 0.007 |
| Active Rate \(s^{-1}\) | 9.14 ± 0.07 | 0.079 ± 0.005 |
| % Change    | 1968%     | 126%     |

While the passive coincidence rate for HEU is still quite high relative to expectation, it is much closer than when all events are considered. Given the presence of other neutron sources in the room, it is also reasonable to assume that some of the discrepancy is accounted for by additional fissions caused by thermal neutrons. Most notably, the change in the HEU coincidence rate between the passive and active measurements is very significant, increasing by more than a factor of 20. This is consistent with the expectation that delayed neutrons will induce
additional fission at a much greater rate in HEU than DU, providing the basis for discrimination.

Furthermore, the time evolution of the rate of coincidence events from fission can provide valuable information on the $^{235}$U content of the test material. Because the delayed neutron groups for each uranium isotope constitute a unique set of decay time constants, the overall delayed neutron time emission profile can be used to discriminate between isotopes and infer enrichment. Delayed neutron-induced fission events occur on the same timescale as their delayed neutron precursors, so coincidence-based measurements of the rate of delayed fission events should exhibit the same temporal shape predicted for delayed neutron emission. Fig. 11 shows the time distribution of coincidence events for HEU in the period after the neutron generator has been turned off.

![FIG. 11: Time distribution of delayed coincidence events in HEU, measured using the composite-EJ309 detector pairing. Only neutron recoil pulses were accepted from the EJ309 detector. The fit is based on tabular nuclear data and parameterized with only a scaling factor (C) and constant background term (B).](image)

The composite-EJ309 detector pairing was used again, but while accepting only neutron recoil events from the EJ309 detector. The experimental data are fitted with a parameterized model based on a six-group superposition of delayed neutrons, whose decay constants are obtained from tabular nuclear data; the procedure is described in depth in Ref. [23]. The experimental results show close agreement with the model ($\chi^2 = 200.2/198$), which confirms that the coincidence events are caused by delayed neutron-induced fission and suggests that discrimination based on the delayed neutron time emission profile can also be performed using fast neutron measurements, provided that the material is fissionable by lower-energy delayed neutrons. The coincidence-based approach presented here could supplement the methods described in Ref. [23], providing an additional point of distinction between isotopes.

In conclusion, we have demonstrated two measurement methods for differentiating the components of delayed neutron signals in bulk samples of SNM based on their origin. Through fission radiation coincidence counting and spectroscopic neutron energy measurements, we have shown a significant refinement in the ability to capture information on delayed neutron-induced fission as a means for performing isotopic identification. For fissionable materials with large differences in fission cross-section at typical delayed neutron energies, such as $^{235}$U and $^{238}$U, we show that these types of measurements are sufficient to successfully perform isotopic discrimination. Further refinement of the capture-gated neutron spectroscopy technique to detect small changes in the delayed neutron energy signature would provide even greater precision in differentiating materials. The measurement approaches presented here have the potential to complement existing delayed neutron analysis techniques, and when employed in concert with methods that focus on time-dependent signatures, they may lead to even greater accuracy in SNM characterization.

**ACKNOWLEDGMENTS**

The authors would like to thank J. Mattingly of North Carolina State University and J. Hutchinson of Los Alamos National Laboratory for their assistance in organizing and executing the experimental campaign at the DAF. This work was supported by the U.S. Department of Homeland Security under Grant Award No. 2014-DN-077-ARI078-02 and 2015-DN-077-ARI096 and by the Consortium for Verification Technology and Consortium for Monitoring, Verification and Technology under U.S. Department of Energy National Nuclear Security Administration award numbers DE-NA0002534 and DE-NA0003920, respectively. The research of J. Nattress was performed under appointment to the Nuclear Nonproliferation International Safeguards Fellowship Program sponsored by the National Nuclear Security Administration’s Office of International Safeguards (NA-241).

[1] D. Reilly, N. Ensslin, H. Smith Jr., and S. Kreiner, Passive Nondestructive Assay of Nuclear Materials (Los Alamos National Laboratory, 1991).

[2] R. Berndt, E. Franke, and P. Morretou, $^{235}$U enrichment or UF$_6$ mass determination on UF$_6$ cylinders with non-destructive analysis methods, *Nuclear Instruments and Methods in Physics Research A* 612, 309 (2010).
[3] R. C. Runkle, A. Bernstein, and P. E. Vanier, Securing special nuclear material: Recent advances in neutron detection and their role in nonproliferation, Journal of Applied Physics 108 (2010), 10.1063/1.3503495.

[4] R. C. Runkle, D. L. Chichester, and S. J. Thompson, Rattling nucleons: New developments in active interrogation of special nuclear material, Nuclear Instruments and Methods in Physics Research, A 663, 75 (2012).

[5] K. H. Beckurts, Measurements with a Pulsed Neutron Source, Nuclear Science and Engineering 2, 516 (1957).

[6] J. T. Caldwell and W. E. Kunz, Experimental Evaluation of the Differential Die-Away Pulsed-Neutron Technique for the Fissile Assay of Hot Irradiated Fuel Waste, Tech. Rep. No. LA-UR-82-788, Los Alamos National Laboratory (1982).

[7] K. A. Jordan and T. Gozani, Pulsed neutron differential die away analysis for detection of nuclear materials, Nuclear Instruments and Methods in Physics Research, B 261, 365 (2007).

[8] R. B. Roberts, R. C. Meyer, and P. Wang, Further observations on the splitting of uranium and thorium, Phys. Rev. 55, 510 (1939).

[9] R. B. Roberts, L. R. Hafstad, R. C. Meyer, and P. Wang, The delayed neutron emission which accompanies fission of uranium and thorium, Phys. Rev. 55, 664 (1939).

[10] N. Bohr and A. Wheeler, The mechanism of nuclear fission, Phys. Rev. 56, 426 (1939).

[11] S. Amiel, Analytical Applications of Delayed Neutron Emission in Fissionable Elements, Analytical Chemistry 34, 1683 (1962).

[12] G. R. Keepin, T. F. Wimett, and R. K. Zeigler, Delayed neutrons from fissionable isotopes of uranium, plutonium, and thorium, Physical Review 6, 1 (1957).

[13] G. R. Keepin, Nuclear safeguards research and development, Tech. Rep. No. LA-4368-MS, Los Alamos National Laboratory (1969).

[14] J. L. Jones, K. J. Haskell, J. M. Hoggan, D. R. Norman, and W. Y. Yoon, in AIP Conference Proceedings, Vol. 680 (2003) pp. 947–950.

[15] D. Slaughter, M. Accatino, A. Bernstein, J. Candy, A. Doughan, J. Hall, A. Loshak, D. Manatt, A. Meyer, B. Pohl, S. Prussin, R. Walling, and D. Weirup, Detection of special nuclear material in cargo containers using neutron interrogation, Tech. Rep. No. UCRL-ID-155315, Lawrence Livermore National Laboratory (2003), 10.2172/15005260.

[16] I. Jovanovic and A. S. Erickson, Active Interrogation in Nuclear Security Science, Technology, and Systems (Springer Verlag, 2018).

[17] D. L. Chichester and E. H. Seabury, in IEEE Nuclear Science Symposium Conference Record (NSS/MIC) (2009) pp. 956–960.

[18] M. Jovanovic, Detection of special nuclear material from delayed neutron emission induced by a dual-particle monoenergetic source, Applied Physics Letters 108, 264102 (2016).

[19] M. T. Kimlaw and A. W. Hunt, Fissionable isotope identification using the time dependence of delayed neutron emission, Nuclear Instruments and Methods in Physics Research, Section A: Accelerators, Spectrometers, Detectors and Associated Equipment 562, 1081 (2006).

[20] X. Li, R. Henkelmann, and F. Baumgärtner, Rapid determination of uranium and plutonium content in mixtures through measurement of the intensity-time curve of delayed neutrons, Nuclear Instruments and Methods in Physics Research B 215, 246 (2004).

[21] W. L. Myers, C. A. Goulding, and C. L. Hollas, Determination of the 235 U Enrichment of Bulk Uranium Samples Using Delayed Neutrons, Tech. Rep. No. LA-UR-06-3984, Los Alamos National Laboratory (2006).

[22] M. T. Sellers, D. G. Kelly, and E. C. Corcoran, An automated delayed neutron counting system for mass determinations of special nuclear materials, Journal of Radioanalytical and Nuclear Chemistry 291, 281 (2012).

[23] J. Nattress, K. Ogren, A. Foster, A. Meddeb, Z. Ounaies, and I. Jovanovic, Discriminating Uranium Isotopes Using the Time-Emission Profiles of Long-Lived Delayed Neutrons, Physical Review Applied 10 (2018), 10.1103/PhysRevApplied.10.024049.

[24] S. Shalev and J. M. Cuttler, The Energy Distribution of Delayed Fission Neutrons, Nuclear Science and Engineering 51, 52 (1973).

[25] R. Batchelor and H. R. M. Hyder, The Energy of Delayed Neutrons from Fission, Journal of Nuclear Energy 3, 7 (1956).

[26] C. L. Morris, K. Chung, S. Greene, G. Hogan, M. Makela, F. Mariam, E. C. Milner, M. Murray, A. Saunders, R. Spaulding, Z. Wang, L. Waters, and F. Wysocki, Active Interrogation Using Energetic Protons, Tech. Rep. LA-UR-10-04680, Los Alamos National Laboratory (2010).

[27] R. E. Rothe, Extrapolated Experimental Critical Parameters of Unreflected and Steel-Reflected Massive Enriched Uranium Metal Spherical and Hemispherical Assemblies, Tech. Rep INEEL/EXT-97-01401, Lockheed Idaho Technologies Co., Idaho National Engineering and Environmental Lab. (1997).

[28] NEUTRON / GAMMA PSD LIQUID SCINTILLATOR EJ301, EJ309 (Eljen Technology, 1300 W. Broadway, Sweetwater, TX 79556, 2016).

[29] M. Mayer, J. Nattress, V. Kukharev, A. Foster, A. Meddeb, C. Trivelpiece, Z. Ounaies, and I. Jovanovic, Development and characterization of a neutron detector based on a lithium glass-polymer composite, Nuclear Instruments and Methods in Physics Research A 785, 117 (2015).

[30] T. Shi, J. Nattress, M. Mayer, M. W. Lin, and I. Jovanovic, Neutron spectroscopy by thermalization light yield measurement in a composite heterogeneous scintillator, Nuclear Instruments and Methods in Physics Research, A 839, 86 (2016).

[31] J. Nattress, M. Mayer, A. Foster, A. Barhoumi Meddeb, C. Trivelpiece, Z. Ounaies, and I. Jovanovic, Capture-gated Spectroscopic Measurements of Monoenergetic Neutrons with a Composite Scintillation Detector, IEEE Transactions on Nuclear Science 63, 1227 (2016).

[32] User Manual - UM5960 CoMPASS Multiparametric DAQ Software for Physics Applications, (2018).

[33] S. A. Pozzi, S. D. Clarke, W. J. Walsh, E. C. Miller, J. L. Dolan, M. Flaska, B. M. Wieger, A. Enqvist, E. Padovani, J. K. Mattingly, D. L. Chichester, and P. Peerman, MCNPX-PoliMi for nuclear nonproliferation applications, Nuclear Instruments and Methods in Physics Research A 694, 119 (2012).

[34] S. Agostinelli et al., Geant4 simulation toolkit, Nuclear Instruments and Methods in Physics Research, A 506, 250 (2003).
[35] L. Cranberg, G. Frye, N. Nereson, and L. Rosen, Fission Neutron Spectrum of U235, Physical Review 103, 662 (1956).

[36] A. Enqvist, C. C. Lawrence, B. M. Wieger, S. A. Pozzi, and T. N. Massey, Neutron light output response and resolution functions in EJ-309 liquid scintillation detectors, Nuclear Instruments and Methods in Physics Research, A 715, 79 (2013).

[37] J. Nattress and I. Jovanovic, Response and calibration of organic scintillators for gamma-ray spectroscopy up to 15-MeV range, Nuclear Instruments and Methods in Physics Research, A 871, 1 (2017).