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Addressing Different Active Neutron Interrogation Signatures from Fissionable Material

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Abstract – **In a continuing effort to examine portable methods for implementing active neutron interrogation for detecting shielded fissionable material research is underway to investigate the utility of analyzing multiple time-correlated signatures. Time correlation refers here to the existence of unique characteristics of the fission interrogation signature related to the start and end of an irradiation, as well as signatures present in between individual pulses of an irradiating source. Traditional measurement approaches in this area have typically worked to detect die-away neutrons after the end of each pulse, neutrons in between pulses related to the decay of neutron emitting fission products, or neutrons or gamma rays related to the decay of neutron emitting fission products after the end of an irradiation exposure. In this paper we discus the potential weaknesses of assessing only one signature versus multiple signatures and make the assertion that multiple complimentary and orthogonal measurements should be used to bolster the performance of active interrogation systems, helping to minimize susceptibility to the weaknesses of individual signatures on their own. Recognizing that the problem of detection is a problem of low count rates, we are exploring methods to integrate commonly used signatures with rarely used signatures to improve detection capabilities for these measurements. In this paper we will discuss initial activity in this area with this approach together with observations of some of the strengths and weaknesses of using these different signatures.**

I. Introduction

The detection of shielded fissionable material inside objects or in difficult-to-reach locations is a technical challenge that has a limited and generally unsatisfactory set of solutions. Passive detection of shielded fissionable material relies on the detection of radiation emitted from the material and indirect radiation generated in the vicinity of the material. With highly enriched uranium (HEU), the characteristic 0.186 MeV gamma-ray line may be used for detection if there is minimal shielding but for cases with shielding this signature is absent or obscured. HEU which has spent time in a reactor (e.g., reprocessed naval reactor fuel) contains isotopic contamination with ^{232}U that produces 2.614 MeV gamma rays during its decay. However, pure HEU derived directly from isotopic enrichment does not contain this impurity and remains easy to shield and difficult to detect using passive photon interrogation. Neutron emissions from HEU generally categorized as weapons grade uranium (WGU) are low in intensity and are straightforward to shield to reach background levels.[1] Weapons grade plutonium (WGPu) generally possesses a significant spontaneous neutron emission signature ($\approx 5.6 \times 10^4$ n/s/kg) and is typically accompanied by an easily measureable yield of 2.22 MeV photons resulting from neutron capture in surrounding hydrogenous materials. Also, WGPu presents an inherent higher-energy photon signature with energies >3 MeV due to the decay of spontaneous fission products. However, these signatures may be masked within shipments of standard commercial neutron source instruments including ^{252}Cf sources used in bulk material analyzers, PuBe and AmBe radioisotope well-logging sources, and hand-held industrial moisture gauges.[1] Detection systems for detecting and verifying shielded fissionable material, beyond straightforward radiographic measurements of material density anomalies, must incorporate an active-interrogation probe radiation source to generate unique fission signatures that can be measured outside of a shield.[2]

Active interrogation techniques using external radiation sources to interrogate objects to detect, identify, and characterize fissionable material have been reported in the literature for many different applications ranging from the subsurface detection of uranium in mining exploration, to assaying fissile material in waste drums, to assaying plutonium content in spent nuclear fuel, to detecting shielded fissionable material hidden in cargo. Excellent survey reports on the subject have been prepared by Fetter et al., the JASON Study Group, the Defense Science Board, Gozani, Brodzinski et al., The Royal Society, Moss et al., and Sowerby and Tickner.[1 -8] Reviewing the work of Brodzinski et al. dealing with the measurement of signatures induced via fission (excluding radiography or nuclear resonance fluorescence) it is clear that many different permutations of irradiation source and detected signature have been explored. As if ordering from an *à la carte* menu of nuclear techniques work in this area usually involves the choice of a single probe source, a single signature, and a single detector type. Relatively few prior investigations have seriously examined multiple signatures within the context of the same measurement technique, nor have they looked at more than one of the time-correlated aspects of single types of measurements.

The most commonly used approach is to measure the net intensity of neutrons produced in between pulses of the interrogating radiation source, seeking either prompt neutrons

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1 It is worth noting here that 1 microgram of ^{252}Cf emits the same number of neutrons per second as 50 kg of WGPu.

from follow-on fission in between source pulses in the "die-away" (DA) approach or neutrons from the decay of β-delayed fission products. The value of measuring the net intensity of β-delayed gamma rays has been shown by the Lawrence Livermore National Laboratory (LLNL) Nuclear Car Wash group and is currently under evaluation by industrial teams working in the field.[9] Active measurements of prompt signatures and multiplicity have been described by the Oak Ridge National Laboratory (ORNL) team and are the subject of ongoing research.[10,11] In terms of quantifiable results, the Nuclear Car Wash team has shown that, using a very intense neutron source and large liquid scintillator detection panels, a 5 kg ^{235}U sample hidden with shields of steel or wood in cargo containers can be detected in less than 30 seconds, based upon "after irradiation" data collection. Moss et al. have shown that kg quantities of HEU within light shielding may be easily detected by measuring β-delayed neutrons in between pulses in just a few seconds.[12]

A considerable effort has been expended recently to develop and test active neutron interrogation systems with the intention of measuring the DA neutron signal from a threat object and it deserves some elaboration.[13- 19] In DA measurements a pulsed radiation source is used to inject a population of neutrons into a test object (either directly using a neutron source or indirectly using bremsstrahlung). In between pulses of the source, measurements are made to evaluate the time-dependence of the decay of the epithermal and fast neutron population exiting the assembly (for example by using a ^{3}He detector wrapped in a thermal neutron shield such as cadmium). If fissionable material is present in the assembly, the residual neutrons from the pulse will continue to produce follow-on fission after the pulse, with the fission rate decreasing in time as the neutron population of the assembly decreases. If the decay rate is observed to be longer than in a known "empty" configuration then a declaration of "fissionable material present" can be made, since only fissionable material can explain the longer duration and lingering presence of high-energy neutrons in the test assembly after a pulse.

At Idaho National Laboratory research has been underway for three years to investigate instrumentation, approaches, and methods for using active neutron interrogation in situations requiring portable instrumentation to detect heavily shield fissionable material.[10,20-22] This work has included simulation and modeling studies to evaluate fission response signatures from different shield configurations, benchmark experiments to validate our modeling efforts, and exploratory experiments to investigate different data collection techniques to identify shielded fissionable material. In recent work we have presented testing results and have commented on the potential use of the DA neutron signature as a strong indicator for the presence of fissionable material.[22] Further study in this area, however, has shown some severe limitations of the utility of this approach if used singly as a method for detecting shielded fissionable material. Looking forward, we are now examining the integration of data from multiple fission signatures simultaneously, to augment the deficiencies of the DA neutron signature in order to develop a more effective and robust detection approach.

II. The Need to Assess Multiple Signatures (The Limitations of Differential Die Away)

Despite the likelihood that adversaries capable of acquiring fissionable material will have sufficient knowledge to understand active interrogation and its limitations, little consideration is given to the consequences of using neutron absorbers within shields in DA measurements. At INL we have examined active interrogations weaknesses in detail. In Fig. 1 data is presented showing the active neutron interrogation DA neutron signature from the irradiation of 9.4 kg of HEU within a 60 cm × 60 cm × 60 cm plywood box, measured using one low-efficiency, cadmium/boron-shielded, polyethylene-moderated ^{3}He detector. This general set-up has been described previously.[22] The figure presents the passive background neutron signal (gray), the detector's response to an active radiation field without the wood or HEU present (green), the detector's response when the HEU is hidden within the center of the wood box (blue), and the detector's response when 6.4 mm of borated rubber (25% natural boron by weight) is wrapped around the HEU in the center of the box (red). This data was collected in a high neutron background environment (about 20X the natural background in Idaho Falls, Idaho). **The borated rubber completely eliminates all signs of the die-away signature**. Fortunately, positive detection of shielded fissionable material can still be made based on the delayed-neutron signal strength in this case, as shown in the inset of Fig. 1.

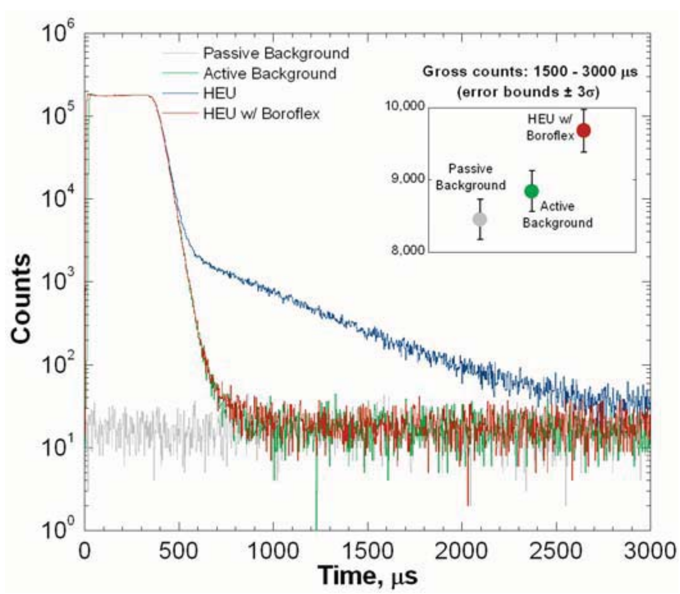

Fig. 1 Comparison of different neutron DA signatures and the impact of using a borated rubber absorber shield. The threat object was 9.4 kg of HEU in a wood box. The inset shows the gross neutron count intensity over a 600-s data collection period from 1500 to 3000 μs after the start of each neutron generator pulse. The error bars are the 3-σ Poisson counting uncertainties.

Recognizing that weaknesses exist when relying on a single signature it is clearly more beneficial to employ multiple signatures simultaneously, preferably using distinctly different signatures (neutrons vs. photons, prompt vs. delayed, steady-state vs. varying) in order to avoid common-mode vulnerabilities. The different types of interrogation signatures available from fissionable material have been presented elsewhere; condensed and arranged with regard to timing with an interrogation pulse, these parameters have been collected symbolically in Fig. 2.[5,23,24] Both complementary and orthogonal fission signatures that can be used. However, while the coupling of individual fission signatures (e.g. neutron die away) with other signatures (transmission and back-scatter x rays, prompt gamma-ray neutron activation analysis (PGNAA) for explosives detention, etc.) has been examined and described in the literature a comprehensive exploitation of multiple fission signatures simultaneously in one implementation is an area of focus that has yet to be fully explored.

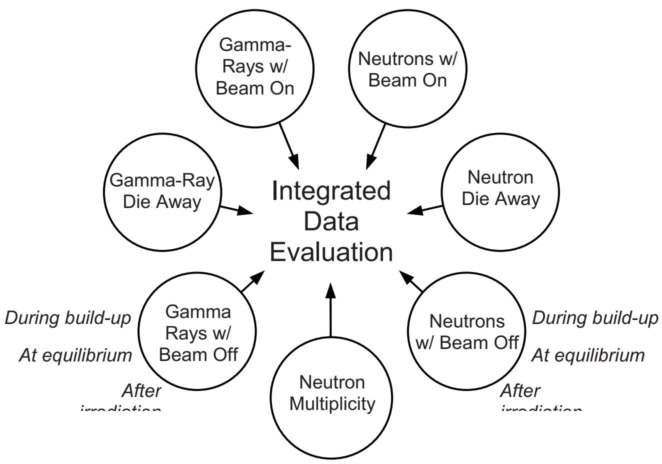

Fig. 2 Multiple signatures can be used during one active interrogation measurement to improve our ability to detect shielded fissionable material but very few examinations look at more than one at a time.

III. Comprehensive Signature Collection

To conduct robust active interrogation measurements as many signatures as possible from Fig. 2 must be used.

Considering this it is useful to refer to the partial list of measurable signatures presented in Table I. This list presents many types of measurements that may be made during active interrogation. For ease of reference the 16 measurements of Table I are also illustrated schematically in Fig. 3 where the timing of the different signatures with respect the active interrogation pulses is illustrated.

TABLE I
MEASURABLE ACTIVE INTERROGATION SIGNATURES OF FISSION

1. Measure neutron intensity during irradiation pulses
2. Measure gamma-ray intensity during irradiation pulses
3. Measure neutron multiplicity during irradiation pulses
4. Measure fast-neutron intensity and signal decay rate during interpulse die-away regions (if present)
5. Measure gamma-ray intensity and decay rate during interpulse die-away regions (if present)
6. Measure neutron multiplicity in interpulse die-away regions (if present)
7. Measure fast-neutron intensity during the interpulse β-delayed emission regions (equilibrium is not necessarily established and is not considered)
8. Measure gamma-ray intensity during the interpulse β-delayed emission regions (equilibrium is not necessarily established and is not considered)
9. Measure neutron intensity after the AI source is turned off (comparison with β-delayed neutron intensity during build-up or at equilibrium is not considered)
10. Measure gamma-ray intensity after the AI source is turned off (comparison with β-delayed gamma-ray intensity during build-up or at equilibrium is not considered)
11. Measure the rate of buildup of the β-delayed neutron intensity during the buildup to equilibrium
12. Measure the rate of buildup of the β-delayed gamma-ray intensity during the buildup to equilibrium
13. Wait until equilibrium (~120 seconds), then measure the β -delayed neutron intensity (compare with non-equilibrium measurements)
14. Wait until equilibrium (~120 seconds), then measure the β-delayed gamma-ray intensity (compare with non-equilibrium measurements)
15. Measure the rate of decay of the β-delayed neutron intensity after the AI source is turned off (compare with observations during buildup and at equilibrium)
16. Measure the rate of decay of the β-delayed gamma-ray intensity after the AI source is turned off (compare with observations during buildup and at equilibrium)

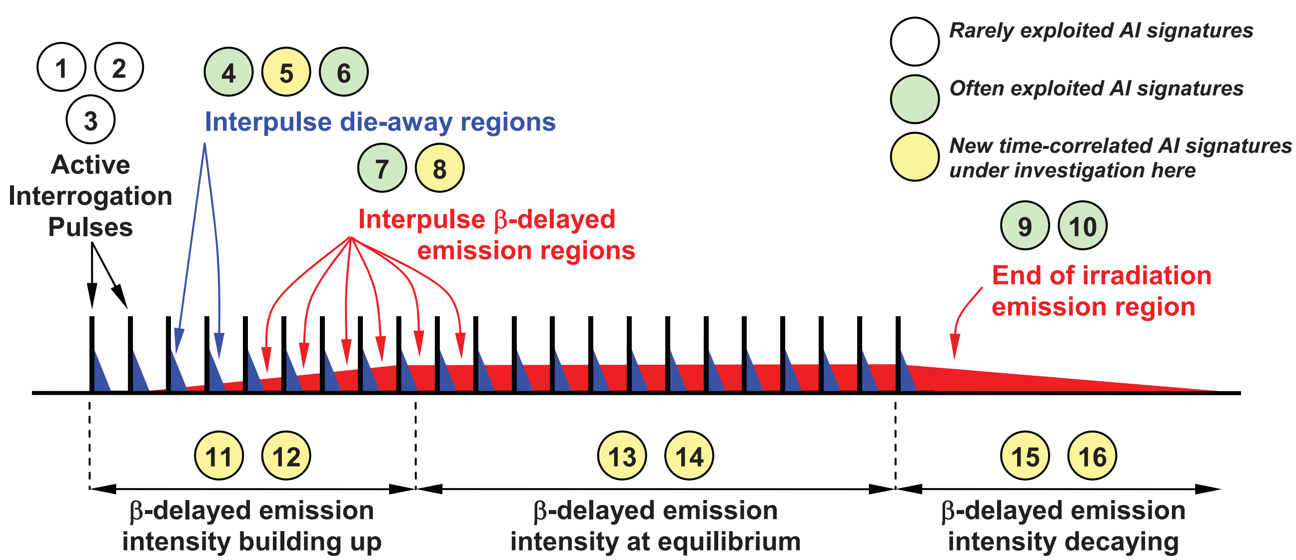

Fig. 3 A representation of the time-dependent active interrogation signatures of fission.

Often the terms "prompt" and "delayed" are used when discussing active interrogation and fission signatures but there has been some confusion with nomenclature in some cases. In a strict definition related to the fission process, prompt phenomena are those events which occur near simultaneously $\mathcal{O}(10^{-10}$ s) with a fission event, while delayed phenomena are events that occur after this.[24,25] These definitions are important for active interrogation because the measurable signatures of prompt fission are quite different from those of β-delayed fission products in energy, intensity, and multiplicity. Unfortunately other definitions are encountered in practice, including "prompt is with the source on, delayed is with the source off." Considering actual measurements and the formal definition, prompt and delayed emission often occur simultaneously. Prompt emissions can obviously occur while the beam is on but they can also occur in between pulses if a high latent neutron population exists in an inspection area (if die-away follow-on fission occurs). Similarly, delayed emissions begin immediately after prompt fission, even while an active interrogation pulses is occurring, although at an intensity less than the prompt emissions until die-away process/fissioning is over and only the decay of β-delayed fission products remains to generate signatures.

The data of Fig. 1 includes both prompt and delayed phenomena. Consider the case where 9.4 kg of HEU was shielded with borated rubber, at times greater than 1000 µs β-delayed emissions are the majority of the measurable signature. As seen in the inset of this figure, after the 600-s measurement period for this experiment a clear β-delayed neutron signature was observed well above the background signature. However, other signatures were present and might have also been measurable including possibly prompt signatures during the neutron pulses and β-delayed gamma rays in between pulses. Even with borated rubber, if a faster responding neutron sensor had been used such as a liquid scintillator a DA signal might still have been observed. If more signatures, prompt and delayed, are used in collaboration active interrogation measurement times may be reduced, shielded interrogation volumes and separation distances may be increased, and material detection limits may be reduced.

Due to the simplicity of use of ^{3}He-based proportional counters neutron detection is most frequently used in active interrogation. One simple method of collecting additional data from active interrogation is to look for the characteristic six-group decay of the β-delayed neutrons from fission.[24,26] An example of data from this type of measurement is shown in Fig. 4 for neutrons measured with a polyethylene-moderated, cadmium/boron-shielded ^{3}He detector following the irradiation of 9.4 kg of HEU with a neutron generator. The HEU was again in a wood shield but in this case it did not have a borated-rubber absorber; data was collected in 5-s second time bins following a 120-s irradiation (to ensure all of the β-delayed fission products reached equilibrium activity levels). A measurement system capable of collecting the data of Fig. 1 can easily be used to collect this post-irradiation data also. Using β-delayed neutron decay-rate data such as this to help confirm interpulse steady-state β-delayed neutron data provides a robust tool for confidence building. Within our active interrogation research program we collect both of these signatures together with several others, such as the inverse of the data in Fig. 4 dealing with the build-up of the β-delayed neutron signal, a useful confirmatory signature used in our work and that has been proposed by others too.[27] β-delayed gamma rays in these different time regimes are another complimentary and orthogonal signature for this type of measurement.[9] Taken as a whole, looking for all of these signatures in a single measurement evolution has the potential to increase the performance of active interrogation systems.

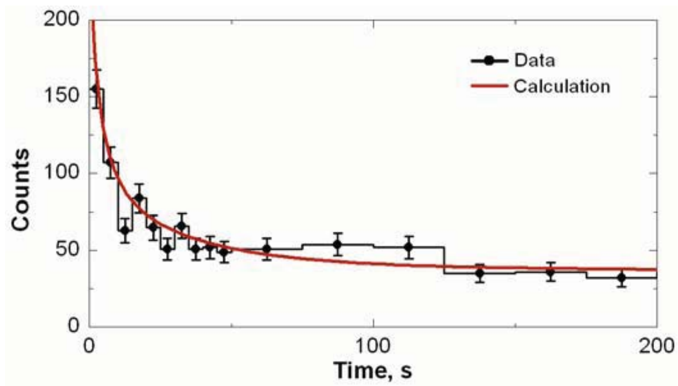

Fig. 4 Post-irradiation active neutron interrogation signature corresponding to the decay of β-delayed neutrons (the solid line is an estimated fit to the data using the six-group decay data of reference 26).

IV. Summary

A key challenge for detecting fissionable material is addressing the statistics of low signal count rate phenomenon.[28 -34] When a strong signal exists it is comparatively easy to declare it as an indicator of fissionable material; it is the situations with low signal count rates that are of primary interest. (During prompt measurements of fissionable material emission, either during an irradiation pulse or in the die-away region, absolute count rates may be very high; however, the active interrogation detection challenge is to identify small fissionable material signals within high count rate environments.) The assumption is made that if passive screening is able to make a positive confirmation of fissionable material (e.g., passive neutron multiplicity counting) then active interrogation isn't needed, and that active interrogation will be primarily used as a second-level screening tool in the most challenging measurement conditions (to probe for fissionable material where passive signatures have not been found or to confirm fissionable material identifications based on radiography). To solve this challenging problem it is important to make use of all available signature data. In some cases one or more of the data signatures may not be available (for example, uranium presented in a non-multiplying form factor will not generate a large active multiplicity signature in the interpulse regions) but collecting and comparing multiple data sets creates a robust approach, less vulnerable to obfuscation or deception.

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