Using Electronic Neutron Generators in Active Interrogation to Detect Shielded Fissionable Material

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***Abstract*—Experiments have been performed at Idaho National Laboratory to study methodology and instrumentation for performing neutron active interrogation die-away analyses for the purpose of detecting shielded fissionable material. Here we report initial work using a portable DT electronic neutron generator with a ^{3}He neutron detector to detect shielded fissionable material including enriched uranium and reactor grade plutonium. Measurements have been taken of bare material as well as of material hidden within a large plywood cube. Results from this work have demonstrated the efficacy of the die-away neutron measurement technique for quickly detecting the presence of special nuclear material hidden within plywood shields by analyzing the time dependent neutron signals in-between neutron generator pulses. Using a DT electronic neutron generator operating at 300 Hz with a yield of approximately 0.36×10^8 neutrons per second, 2.2 kg of enriched uranium hidden within a 61 cm $\times$ 61 cm $\times$ 71 cm volume of plywood was positively detected with a measurement signal 2-sigma above the passive background within 1 second. Similarly, for a 500 second measurement period a lower detection limit approaching the gram level could be expected with the same simple set-up.**

***Index Terms*—Active interrogation, electronic neutron generator, special nuclear material.**

I. Introduction

THERE is a pressing need for a portable system capable of detecting the presence of shielded fissionable material, special nuclear material (SNM), in moderate sized containers. This capability is needed in order to have a rapidly deployable tool for field-response emergency situations to either a) perform primary field assays to identify the presence of SNM or b) serve as a secondary assessment tool to confirm or dismiss field assays of suspected SNM material. Techniques for performing passive screening to detect SNM have been described in the literature, a good survey of the topic has been presented by Fetter *et al.* [1] The principle materials of concern here are weapons grade uranium (WgU) and weapons grade plutonium (WgPu). WgU presents the most difficult detection challenge because it does not inherently emit either a large number of photons or neutrons; that is to say, its passive radiation emission signature is weak and difficult to measure. WgPu is generally a less difficult material to detect because it possess a naturally elevated neutron emission signature with an accompanying gamma-ray signature due to neutron interactions in surrounding media; also, its inherent gamma-ray signature is strong without shielding.

Gamma-ray spectrometers are the most commonly used tool for detecting SNM.[1] However, since these instruments are based upon low-energy gamma-ray spectroscopy they are generally ineffectual when searching for moderate quantities of SNM, especially WgU, hidden within mid- to high-Z shields such as steel or lead. For example, with less then 2 inches of lead the passive gamma-ray signatures from WgU (not reprocessed) are essentially unmeasurable [2]. Passive neutron detectors are also used for SNM detection, including systems using gross-neutron counting and systems examining neutron multiplicity characteristics [3]. Noting that 1 kg of WgU will generate roughly 1.6 neutrons per second [1], detection of WgU using passive neutron counting is obviously a difficult measurement to perform and moderate amounts of external shielding can be used to make detection in reasonable counting periods very challenging. Detection of modest amounts of WgPu, with an inherent neutron source intensity of approximately 56 000 neutrons per second per kilogram of material [1], is easier to perform using neutron detectors than photon detectors and more difficult to shield within modest sized-containers. Of course, even without any neutron shielding passive neutron measurement techniques are not easily and quickly able to provide a positive identification of SNM in some situations, e.g., confirmation of plutonium versus a legitimate shipment of a commercial neutron source.

In our current work at Idaho National Laboratory (INL) we are performing research to examine the use of active neutron interrogation to detect, characterize, and quantify hidden, shielded fissionable material. Using an external neutron radiation source to probe a suspect SNM-containing object produces additional SNM signatures, generally stronger and less ambiguous than the passive signatures, which can allow positive/negative determinations to be made quicker and with lower material detection limits than with passive screening alone [4]–[16]. The most commonly used active neutron interrogation techniques for SNM detection involve the use of electronic neutron generators (ENGs), which produce nearly monoenergetic fast neutrons at either 2.5 or 14.1 MeV, operating as pulsed neutron sources. For these measurements the ENGs generate short pulses of neutrons, ranging from 0.01 ms to 1 ms, and typically pulse at frequencies on the order of $\sim$100 Hz. During each ENG pulse neutrons enter the test object, or assembly, where they undergo

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[1]For example, the Identifinder from ICx Technologies, Arlington, VA; the Detective from Ortec, USA, Oak Ridge, TN; or the Falcon 5000 from Canberra, Meriden, CT (http://www.icx-radiation.com; http://www.ortec.com; http://www.canberra.com).

elastic and inelastic scattering and are eventually lost due to either absorption, leakage, or fission if SNM is present.

In low-Z assemblies the source neutrons are quickly down-scattered to thermal energies within tenths of a millisecond but then slowly leak out within tens of milliseconds (depending upon material composition and assembly size). In high-Z assemblies the fast neutrons injected from the ENG tend to escape the assembly rather than losing energy and staying inside; in these systems the fast neutron and thermal neutron population decay times are much faster. In an unshielded SNM-containing assembly (e.g., bare U in an empty box) the residual neutron population following the ENG pulse immediately decays to very low levels which are influenced by the surrounding environment including proximity to and composition of the floor, walls and ceiling if present. In either case, as the initial neutron population decreases in energy, and while the thermal neutron population is resident in the assembly, fission will occur in SNM if present. The time-dynamics of the fission rate in an assembly are situation specific, influenced positively over time as the mean energy of the neutron population decreases towards energies where the fission cross-sections become higher, but influenced negatively over time due to neutron absorption and leakage out of the assembly, which reduce the overall neutron population. Fast neutron induced fission occurs in these cases also but the resultant prompt neutron production and delayed-neutron production from fast fission are low and usually important only for bare, unshielded cases where thermal fission is minimal.

One category of active neutron interrogation relies on the measurement of radiation emissions from fission products created in an assembly [4]–[7], [9], [10], [14]. These measurements can include the detection of gamma rays and/or neutrons and the technique has proven useful in many situations. One challenge with this technique is that the intensity of these post-fission 'delayed' signatures is not high, at least in comparison with the gamma-ray and neutron signatures originating directly from fission. A strong-point for measuring the delayed radiation signatures is that competing background signals are usually very low.

Another category of measurements, die-away analysis, involves the detection of neutrons shortly after the neutron pulse as the fast neutrons down-scatter in energy and then as the thermal neutron population in the assembly decays [6]–[8], [11][12][13], [15], [16]. During this period fission is still occurring and detectors suited for measuring these fission signatures directly can be used. The intensity of these prompt fission signatures is much stronger than those from the fission products used in the delayed measurements. A challenge in performing die-away assessments is that the detectors used for these measurements must be simultaneously sensitive to the prompt signatures while remaining insensitive to the generic signatures emanating from the test object from benign neutron scattering, capture, and leakage. For neutron detection a detector that is insensitive to thermal neutrons, such as a cadmium shielded helium-3 proportional counter or a liquid scintillator, may be used to accomplish this. For gamma-ray detection a spectrometer or a high-threshold counter may be used.

While fission is occurring in an assembly more sophisticated measurements analyzing neutron multiplicity may also be performed. These measurements are highly fission specific and have very high signal to noise relationships; however, the overall sensitivity for these types of measurements is less than in basic neutron counting when comparable sized detector systems are used. Measurements may also be performed during the ENG pulse, which are sometimes called prompt or in-beam measurements, but these are challenging because of interferences from the ENG and because scattering inside the object are not easily accounted for.

Active interrogation may also be performed using high-energy bremsstrahlung, where x rays exceeding the photofission thresholds for SNM are used to induce fission in an assembly rather than neutron-induced fission. It is worth noting, however, that photoneutrons ejected from non-SNM materials often make an important contribution to the total fission signature once they've down scattered to thermal energies. Photon active interrogation is a powerful technique and can often complement neutron interrogation in situations where neutron penetration into an assembly is low, and vice versa. However, the large size and complexity of the electron accelerators needed to generate adequate radiation fields precludes their use for portable applications.

This paper presents results from experiments conducted at INL using an ENG to perform active interrogation of lightly-shielded fissionable material. INL maintains an inventory of SNM, including uranium of varying enrichments and plutonium with varying ^{239}Pu isotopic purity, in multiple forms including metal plates and rods, alloyed metals, and oxides. The work took place inside INL's zero-power physics reactor (ZPPR) facility which allows for the safe and secure use of SNM in a controlled environment. The facility includes a large area suitable for active interrogation research using both ENGs and high-energy x-ray machines. It should be noted that the passive neutron background inside this facility, 24.1 ± 0.2 counts per second (cps) for the detectors used here, is significantly higher than Idaho Falls' natural neutron background of 0.984 ± 0.032 cps with the same detectors.

II. The Experiment

Experiments were performed to measure the time dependent die-away neutron signature from a variety of test items in a bare configuration and when shielded with wood. A list of the test items is presented in Table I, they included enriched uranium (Item 1), depleted uranium (Item 2), a mixture of reactor grade plutonium and depleted uranium (Item 3), and tungsten (Item 4). The items were all in the form of multiple metal plates with dimensions of approximately 0.3 cm $\times$ 5.1 cm $\times$ 10.2 cm (0.125 in $\times$ 2 in $\times$ 4 in), the SNM plates were clad in a very thin shell of stainless steel. All test items were further encased within a rectangular protective aluminum canister, or "clamshell," measuring 69.8 cm $\times$ 18.7 cm $\times$ 28.6 cm (2.75 in $\times$ 7.375 in $\times$ 11.25 in) in size with a walls approximately 0.6 cm thick. Plates were stacked upright, side by side in the center of the clamshells so that their longest dimension was aligned with and parallel to the clamshell's 18.7 cm dimension and their second largest dimension was aligned with and parallel to the 69.8 cm dimension. In all cases the flat, large base of the clamshell remained aligned parallel with the floor.

TABLE I
MATERIAL COMPOSITION OF THE TEST ITEMS (KG)

Component	*ITEM 1* *Enriched Uranium*	*ITEM 2* *Depleted Uranium*	*ITEM 3* *Reactor Grade Plutonium & Depleted Uranium*	*ITEM 4* *Tungsten*
W	0	0	0	2.0
^{235}U	2.011	0.004	0.008	0
^{238}U	0.145	1.965	3.862	0
^{239}Pu	0	0	1.438	0
^{240}Pu	0	0	0.545	0

Measurements were taken with the clamshells unshielded and supported by a steel table, or hidden within a wood cube having a square base measuring 61 cm × 61 cm, with a height of 71 cm. When sitting on the steel support table the material inside the clamshell was approximately 85 cm above the floor. The wood cube was comprised of eight separate wood cubes made from standard plywood, each measuring 30.5 cm (12 in) on a side and an adjustable wood drawer measuring 61 cm × 61 cm square with a height of 10 cm. Four plywood cubes were placed on the floor, then the wood drawer was placed on top of these, and then four more plywood cubes were placed on top of the drawer. The wood drawer was filled with movable planks of wood arranged in a way so that a hidden void large enough to hold a clamshell was made within the drawer. The planks could be rearranged to relocate the void to different positions within the drawer. For the experiments reported here the void was arranged at the front of the box (closest to the neutron generator), in the middle of the box, and at the rear of the box (farthest away from the neutron generator.) The set-up has the versatility to 'hide' the clamshell in 9 locations inside the wood drawer; positioning the wood drawer on the floor, in the middle, or on top of the plywood allows a total 27 void locations. Photos of a clamshell sitting on the steel support table and of a clamshell inside the wood cube are shown in Fig. 1.

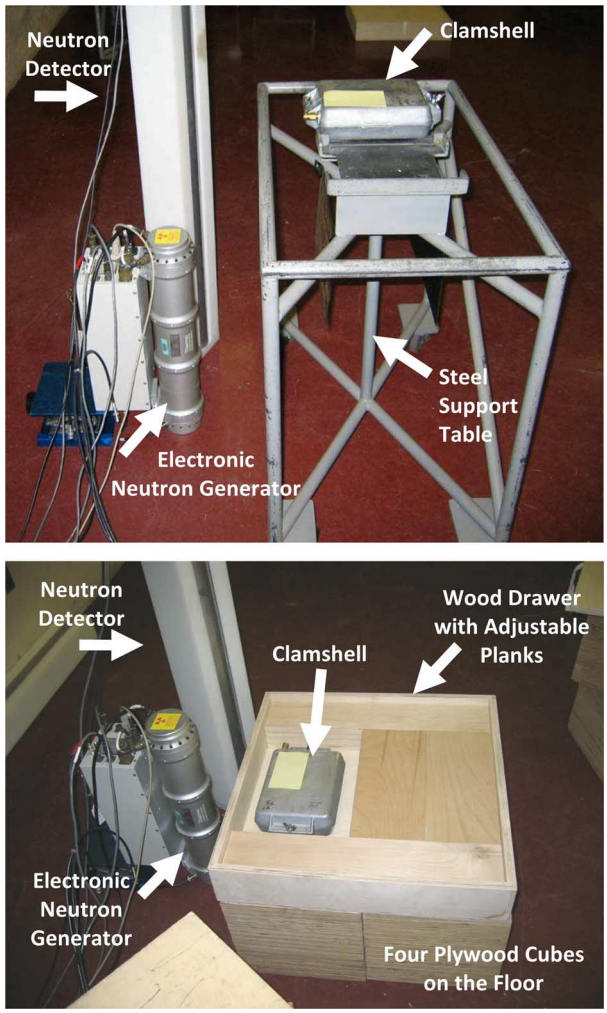

Fig. 1. (Top) Experimental arrangement for testing material in a bare clamshell, showing the relative orientation of the neutron generator, detector, and clamshell. (Bottom) Experimental arrangement for testing material in the hidden void of the wood cube, showing the relative orientation of the neutron generator, detector, and clamshell. For the experiments a wood lid was placed on top of the drawer, which was then covered with four additional wood cubes. In this image the clamshell is shown at the front of the box, closest to the neutron generator.

An MP-320 electronic neutron generator (Thermo Scientific, Colorado Springs, CO, USA) producing 14.1 MeV neutrons via the deuterium-tritium fusion reaction was used as the neutron source for these experiments [17]. It was operated with an average neutron yield of $(0.36 \pm 0.04) \times 10^8$ neutrons per second. Although capable of producing a higher neutron yield a lower output setting was used for these experiments to avoid saturating the neutron detector. The ENG was pulsed at a frequency of 300 Hz with a pulse width of 0.333 ms (a 10% duty cycle). The ENG was placed directly on a concrete floor, neutron emission from the generator occurred approximately 14 cm above the floor. When irradiating the bare clamshell the ENG was approximately 61 cm (24 in) in front of the materials inside; when irradiating the wood cube the ENG was approximately 15.2 cm (6 in) in front of the box.

A 10 atm ^{3}He proportional counter with a diameter of 2.54 cm (1 in) and an active length of 76.2 cm (30 in) was used in these experiments [18]. The detector was immediately surrounded by 2.54 cm (1 in) of plastic and then surrounded by cadmium and boron to absorb thermal neutrons. It has an intrinsic efficiency for fission neutrons from a ^{252}Cf source of approximately 5.9%. The detector generates a TTL (transistor-transistor logic) data signal output when a neutron detection event occurs. The detector was oriented perpendicular to the ground, as shown in Fig. 1, 15.2 cm (6 in) in front of the box, 30.5 cm (12 in) to the side of the ENG.

A multi-channel scaler (MCS) counting unit operated using a personal computer was used to collect data for 500 second periods. The start time for the MCS was taken directly from the MP-320 ENG as a TTL output. It took roughly 15 seconds for the ENG to reach the selected neutron yield when turned on; data collection was started as soon as the ENG reached this level, thus delayed neutron equilibrium was not achieved prior to the collection of data. Long-period drift in the output of the neutron generator which may have occurred during the day was not accounted for. Passive background data (BKD) was collected with and without the Pu (which had a high natural neutron activity), with and without the wood, using the MCS but with an

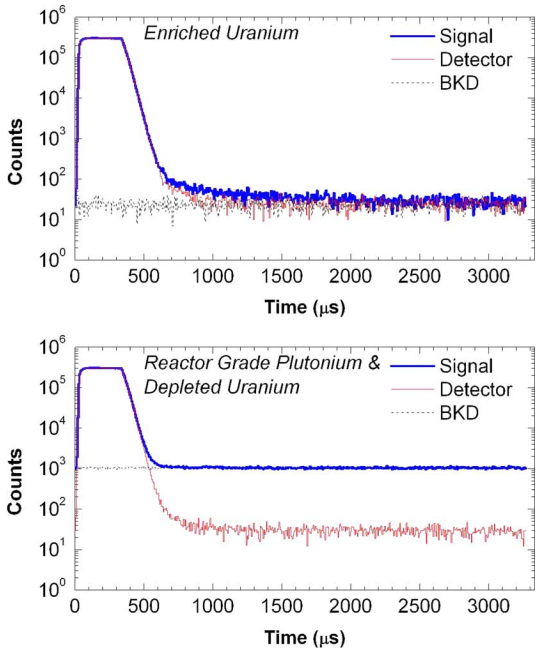

Fig. 2. Neutron die-away time spectra for enriched uranium (top) and the reactor grade plutonium/depleted uranium mixture (bottom) on the steel support table.

external pulse generator set to 300 Hz in place of the ENG TTL pulse signal. At the beginning of the day an active background (Detector) response of the detector with the ENG pulsing but without the steel test stand or wood, was taken for use with measurements of the enriched uranium. A similar data set was taken at mid-day for use with the depleted uranium, reactor grade plutonium/depleted uranium mixture, and tungsten test items. This active background signal is plotted as "detector" in the plots below.

III. Results

Neutron die-away time spectra from the four test items are presented in Figs. 2 through 5 below. In each figure the raw data from the MCS are shown for 500 seconds of collection a) with the ENG on when the sample is present, *Signal* (blue), b) data collected from the detector when the ENG was on but neither material nor the steel test stand nor any wood was present, *Detector* (red), and c) for passive background data taken in the area, *BKD* (black).

As a simple estimate to evaluate the sensitivity of the die-away measurements for the materials and geometry used here, the net number of counts (signal minus passive background) was determined over the period from 1000 μs to 3270 μs for each case. This data is presented in Table II. The start time of 1000 μs was arbitrarily chosen as a conservative time at which all transient signals from neutron scattering in the object, the room, and the detector have returned to background levels.

IV. Discussion

The neutron die-away signal intensity from the enriched uranium used here is very strong. It is interesting to compare the data for the enriched uranium with the delayed-neutron data presented by Moss *et al.* using test samples of a 5 kg sphere of

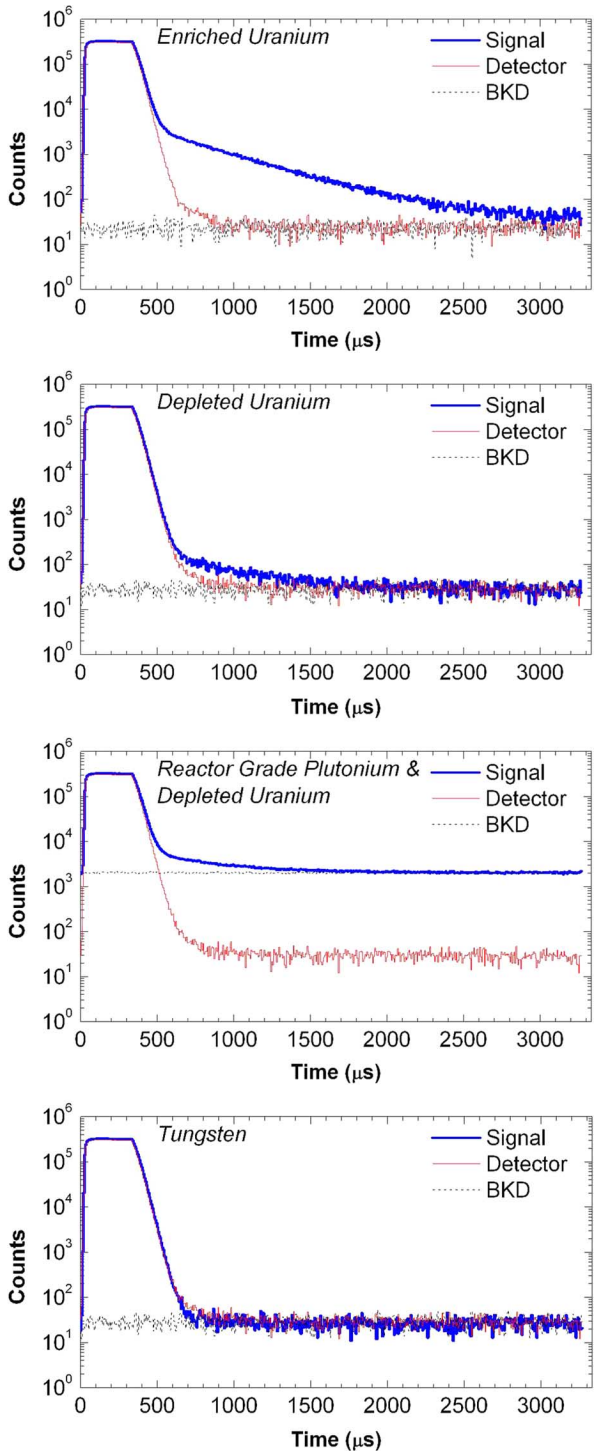

Fig. 3. Neutron die-away time spectra for enriched uranium (top), depleted uranium (second from top), reactor grade plutonium/depleted uranium mixture (second from bottom), and tungsten (bottom) located in the front of the wood shield cube.

similarly enriched uranium in a bare configuration, and when shielded by iron, lead, and polyethylene [10]. In particular it is worth noting that *the neutron generator used in this reference is liquid-insulated (fluorinert FC-77), with several liters of fluid used in place of the low-density sulfur hexafluoride gas used in the MP-320, which may have slightly moderated the out going neutron spectrum and improved the induced fission*

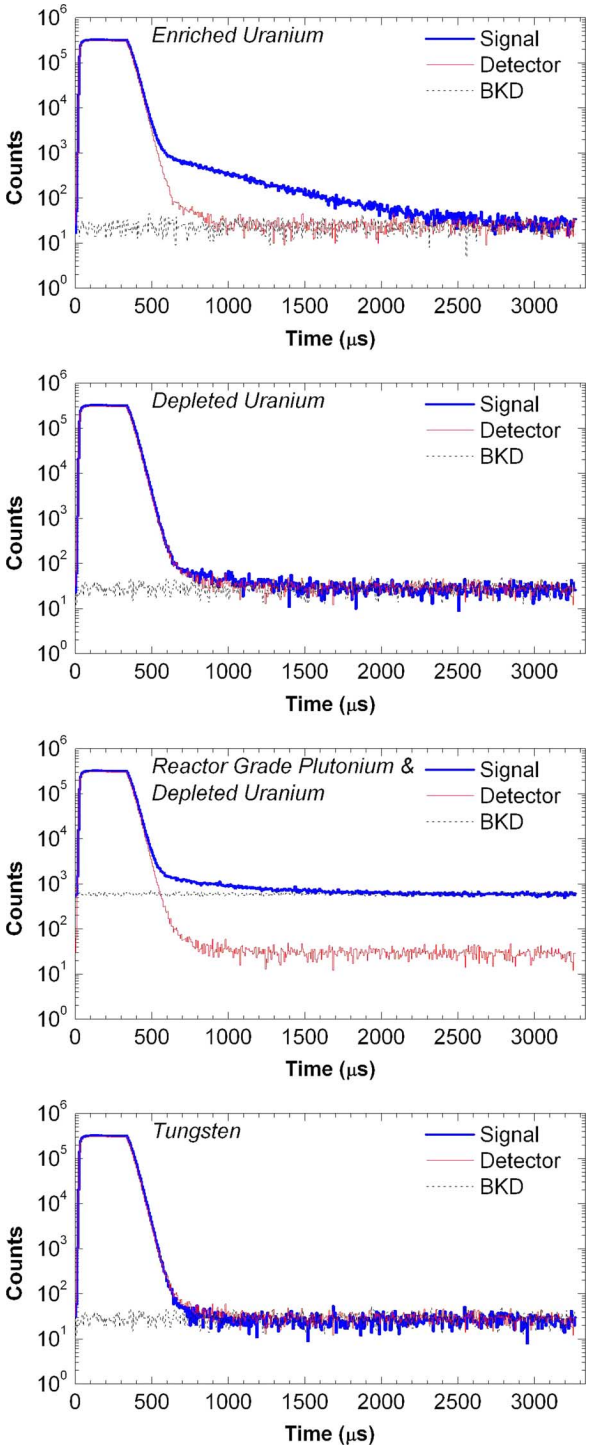

Fig. 4. Neutron die-away time spectra for enriched uranium (top), depleted uranium (second from top), reactor grade plutonium/depleted uranium mixture (second from bottom), and tungsten (bottom) located in the middle of the wood shield cube.

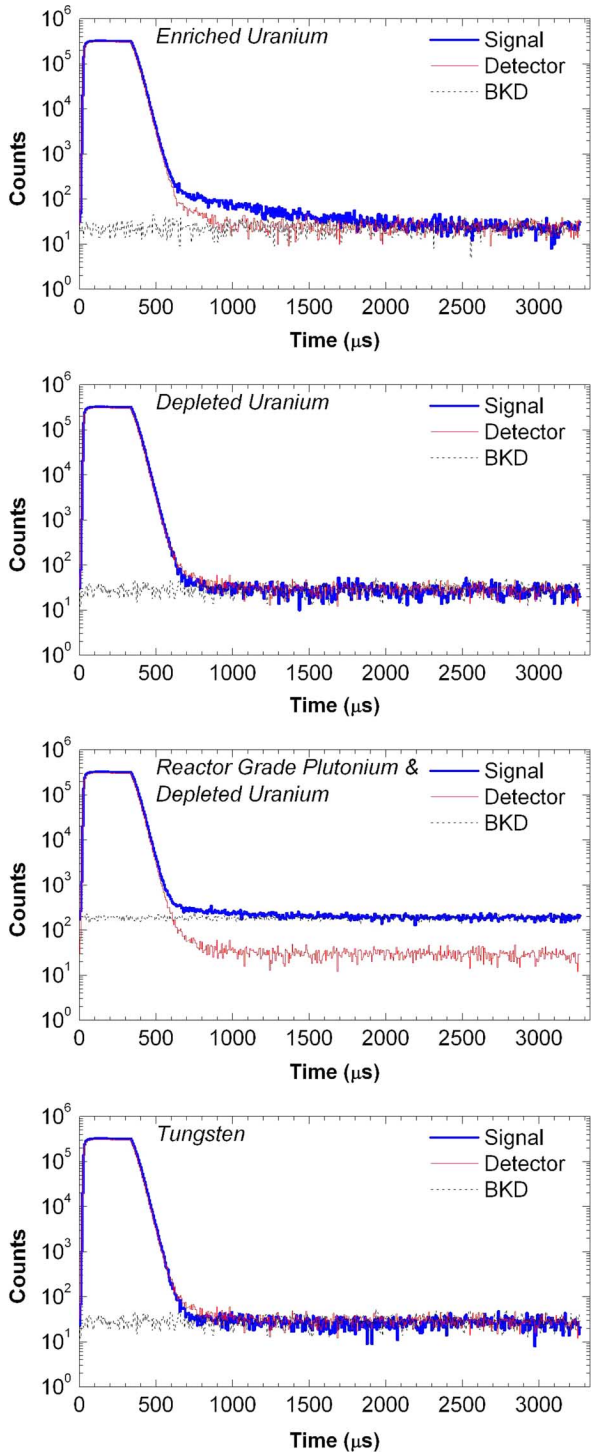

Fig. 5. Neutron die-away time spectra for enriched uranium (top), depleted uranium (second from top), reactor grade plutonium/depleted uranium mixture (second from bottom), and tungsten (bottom) located in the rear of the wood shield cube.

rate versus that from an unmoderated spectrum. For those measurements the ENG to test item spacing was 100 cm (compared with approximately 50 cm for the tests reported here) and the test item to detector spacing was also 100 cm (again compared with approximately 50 cm for the tests reported here.) The detector used in the reference demonstrated a 20% intrinsic efficiency versus the 5.9% intrinsic efficiency of the detector used here, it also presented a larger solid angle than in the case shown here. Making some simple $1/\mathrm{r}^2$ estimates for ENG/test item/detector spacing, accounting for fissile material mass, neutron generator yields (0.5×10^7 neutrons per second in the reference), and detector efficiencies, and neglecting detector solid angle differences, the measurement count rate date for the set-up presented here could be scaled, very roughly, by a factor of

TABLE II
NET DIE-AWAY NEUTRON COUNTS (ACTIVE SIGNAL MINUS PASSIVE BACKGROUND) COLLECTED OVER 500 SECONDS

Case	*ITEM 1* *Enriched Uranium*	*ITEM 2* *Depleted Uranium*	*ITEM 3* *Reactor Grade Plutonium & Depleted Uranium*	*ITEM 4* *Tungsten*
No Shield	3,309 ± 142		665 ± 862	
Wood – Front	68,302 ± 292	3,826 ± 149	63,683 ± 1,228	327 ± 137
Wood – Middle	22,885 ± 200	1,439 ± 141	19,858 ± 670	123 ± 136
Wood – Rear	3,880 ± 143	618 ± 137	4,449 ± 371	317 ± 137

$(2^2 \times 2^2 \times (2.011/5) \times (0.36/0.5) \times (5.9/20))^{-1} = 0.732$ to allow comparisons with the Moss *et al.* data. Using this as a guide the advantage of performing die-away measurements over pure delayed-neutron measurements is seen.

For the bare uranium used here the scaled die-away signal is (6.6 cps × 0.732) 4.8 cps versus 2.2 cps from the Moss delayed-neutron data. At first glance the die-away signal would appear to be not much better than the delayed signal. However, the presence of pre-moderating material (insulating liquid) in the ENG used for delayed neutron measurement is likely serving to significantly boost this signal over what would be expected in an unmoderated situation. A more comparable situation may exist for the shielded uranium case presented here with enriched uranium in the middle of the wood block, (45.8 cps × 0.732) 33.5 cps, and the case where Moss *et al.* used 6.35 cm of polyethylene (1.7 cps). In this case both interrogating neutron fields undergo significant moderation prior to interacting with the uranium and the die-away signal is much larger than the delayed neutron signal. Based upon these observations, it is clear that a) a modest amount of local moderating material near the generator (in the Moss *et al.* data) makes a significant improvement in the measured material signature and b) under similar conditions the die-away signal can provide a more intense signature than the delayed neutron signal.

The net neutron count intensity values presented in Table II were determined starting from $t = 1000\ \mu$s. Examining Figs. 2 through 5 it is clear that this sacrifices a significant amount of valuable data. When a more careful evaluation of the detectors response is used up to twice as many counts can be recognized in some cases, particularly for the higher count-rate scenarios. It has been suggested previously that using a thermal neutron sensitive neutron detector, in addition to the fast-neutron sensitive detector, can allow for a better determination of the count start time by examining the assembly's thermal neutron die-away characteristics [13]. Considering this concept in light of the data shown here it seems that this approach would indeed prove useful for improving die-away measurements of unknown objects.

One apparent discrepancy in the data is the repeatable but slightly high net neutron intensity observed for the non-fissionable tungsten test object. An explanation for this may be due to a small change in the intensity of the background between when the tungsten data was taken and when the background data used with the tungsten data was taken. Recognizing that the background neutron rate in the ZPPR area is elevated, several background measurements were taken over the day. The background rate at the end of the day was observed to be 26% higher than at the start of the day, which was due to the presence of the reactor grade plutonium during the afternoon measurements (when data with the depleted uranium, reactor grade plutonium and depleted uranium mixture, and tungsten were taken), which was not present during the morning when data with the enriched uranium was assessed. Although the Pu was on the far side of the room for the afternoon measurements, it is possible that when the wood was removed from the test area to take the afternoon background measurement data it was put in a position where it affected the local neutron background rate from the Pu. For reference, the on-contact neutron dose rate from the clamshell filled with reactor grade plutonium and depleted uranium was 0.30 mSv/hr (30 mrem/hr).

V. SUMMARY

Examining the results of Table II it is clear that the die-away neutron measurement technique is an exceptionally powerful method for detecting the presence of shielded special nuclear material. For the cases studied here, using a low/mid-Z shield such as wood, the presence of kilogram quantities of fissionable material including both enriched uranium and a mixture of reactor grade plutonium and depleted uranium could be easily confirmed within these small shield volumes in just a few seconds. Conversely, minimum detection limits approaching the gram level could likely be achieved in similar shield objects for comparable counting times to those used here. Detection of uranium hidden within this type of wooden shield would be very difficult or impossible using standard passive screening techniques [2]. Similarly, while the strong neutron emission rate from the reactor grade plutonium used in these experiments would be easily detectable using passive neutron screening, the active interrogation measurements demonstrated here would provide a useful secondary-confirmation for screening a container declared as a legitimate shipment of a commercial neutron source, helping to eliminate any concern that the shipment was being used to mask a significant undeclared quantity of fissionable material.

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