Active Neutron Interrogation to Detect Shielded Fissionable Material

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Abstract. Portable electronic neutron generators (ENGs) may be used to interrogate suspicious items to detect, characterize, and quantify the presence of fissionable material based upon the measurement of prompt and/or delayed emissions of neutrons and/or photons resulting from fission. The small size (<0.2 m^3), light weight (<12 kg), and low power consumption (<50 W) of modern ENGs makes them ideally suited for use in field situations, incorporated into systems carried by 2-3 individuals under rugged conditions. At Idaho National Laboratory we are investigating techniques and portable equipment for performing active neutron interrogation of moderate sized objects less than ~2-4 m^3 to detect shielded fissionable material. Our research in this area relies upon the use of pulsed deuterium-tritium ENGs and the measurement of die-away prompt fission neutrons and other neutron signatures in-between neutron pulses from the ENG and after the ENG is turned off.

1. Introduction

The detection of shielded highly enriched uranium (HEU) and Pu inside objects or in difficult-to-reach locations is a technical challenge that has a limited, and generally unsatisfactory, set of solutions. Passive techniques for detecting shielded special nuclear material (SNM) rely on the detection of radiation emitted from the SNM, and on the detection of indirect radiation generated in the vicinity of the SNM, such as prompt gamma rays generated from thermal neutron capture. With HEU the characteristic decay gamma ray at 0.186 MeV may be used for detection if there is minimal shielding but for most cases with shielding this signature is either absent or obscured by shielding. HEU which has spent time in nuclear reactor environments is typically isotopically contaminated with ^{232}U; the decay of ^{232}U generates 2.614 MeV gamma rays which are straightforward to detect even through moderate amounts of shielding. 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 measurements. Neutron emissions from HEU are also low in intensity, and are straightforward to shield to reach background levels that make detection difficult within reasonable time periods (minutes).[1]

Plutonium generally possesses a significant spontaneous neutron emission signature (~5.6×10^4 n/s/kg) and is often accompanied by an easily measureable yield of 2.22 MeV photons resulting from neutron capture in surrounding hydrogenous materials. Plutonium samples also present higher energy photons with energies >3 MeV due to the decay of spontaneous fission products. However, for plutonium both of these signatures may be masked within shipments of standard, legitimate 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.[2] Systems for detecting and verifying shielded SNM, beyond straightforward radiographic measurements of material density anomalies, must incorporate active interrogation probe radiation sources to generate unique fission signatures that can be measured outside of a shield.[3]

Active interrogation techniques using external radiation sources to interrogate objects to detect, identify, and characterize SNM 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 SNM hidden in cargo.[4-20] The most commonly used approach is to measure the net intensity of

neutrons produced in between pulses of the interrogating radiation source. The value of measuring the net intensity of 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.[16] Measurements of prompt signatures of fission resulting from active interrogation, including neutron multiplicity, have been described by researchers at Oak Ridge National Laboratory (ORNL) and Idaho National Laboratory and are the subject of ongoing research.[21-23] 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 very light shielding may be easily detected by measuring neutrons in-between pulses of an electronic neutron generator (ENG) in just a few seconds.[11]

At Idaho National Laboratory research is underway to investigate practical approaches for using active neutron interrogation to detect shielded fissionable material as a field-portable technique.[21-26] 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 studies to investigate different data collection techniques to identify shielded SNM.

2. Signatures and Characteristics

Nuclear emissions arising from induced fission are generally categorized as 'prompt' or 'delayed' but specific definitions for these two time regimes vary among researchers, depending upon the particular circumstances of their work. In the strictest sense prompt emissions would be considered those that occur during the actual separation, or scission, of a fissioning nucleus, prior to the distinct formation of fission products; a process which happens on the order of 10^{-14} seconds. [27,28] Delayed emissions would therefore be those originating from the subsequent decay of fission product nuclei. These definitions rely on the physical process of fission, regardless of the performance characteristics of detectors or radiation sources used to induce fission. With this understanding it is clear that prompt emissions could occur at any time following the turn-on of an external radiation field, even after the end of an external radiation pulse or burst if fission can still occur after the irradiation, for example, if neutrons are still present in an assembly. Similarly, following the onset of fission in a material delayed emissions begin almost immediately, even while new fission are occurring, while an external radiation source may be interrogating a material and after the end of a radiation burst or pulse. This is important because the characteristics of the prompt and delayed nuclear fission signatures are different, as shown in Table I. In particular it is worth noting the much higher intensity of the prompt emissions in contrast with the delayed emissions; noting the difference in energy between the prompt neutron emission and the delayed neutron emissions, and noting the difference in intensity between the delayed neutrons and the delayed photons.

Somewhat broader definitions for prompt and delayed emissions are occasionally defined, however. In some cases the time resolution of a particular detector used to monitor the fission process, ranging from durations of a few nanoseconds to several tens of microseconds, is used to bound the definition of prompt.[27] In other cases researchers choose to define prompt emissions in relation to the timing structure of their irradiation source, calling all emission generated while the source is on as prompt.[29,30] Sometimes this form of definition is extended to include an arbitrary period of time following the end of an irradiation pulse to also be prompt.[29] Often researchers write that they are measuring 'delayed' neutrons or gamma rays in a generic sense, applying the label to any

measurements acquired in-between pulses of a probing radiation, or to measurements acquired after the end of an irradiation.[11,16,20,31-34]

TABLE I NEUTRON INDUCED FISSION SIGNATURES[1] [27,35,36]

Parameter				^{235}U	^{238}U	^{239}Pu
	σ	Fission cross-section	barns	2734 (thermal) 1.287 (2 MeV) 2.0839 (14.1 MeV)	0.534 (2 MeV) 1.1516 (14.1 MeV)	3204 (thermal) 1.975 (2 MeV) 2.4094 (14.1 MeV)
Prompt Emissions	$\bar{\nu}_{n,prompt}$	Average prompt neutron yield (multiplicity)[2]	prompt neutrons per fission	2.43 (thermal) 2.57 (fission spec.) 4.6 (~14 MeV)	2.79 (fission spec.) 4.5 (~ 14 MeV)	2.87 (thermal) 3.09 (fission spec.) 4.9 (~ 14 MeV)
	$\bar{E}_{n,prompt}$	Average prompt neutron energy[3]	MeV	1.935 (thermal) 2.03 (14 MeV)[4]	1.99 (14 MeV)[4]	2.010 (thermal) 2.19 (14 MeV)[4]
	$\bar{\nu}_{\gamma,prompt}$	Average prompt photon yield	prompt photons per fission	6.60 ± 0.2 (thermal)[5]	7 – 8	7.06 ± 0.2 (thermal)[5]
	$\bar{E}_{\gamma,prompt}$	Average prompt photon energy[6]	MeV	0.97 ± 0.04 (thermal)[5]	~ 1	0.95 ± 0.04 (thermal)[5]
Beta-Delayed Emissions	$\bar{\nu}_{n,delayed}$	Average delayed neutron yield[2]	delayed neutrons per fission	0.0158 (thermal) 0.0165 (1.45 MeV)	0.0412 (3.01 MeV)	0.0061 (thermal) 0.0063 (1.58 MeV)
	$\bar{E}_{n,delayed}$	Average delayed neutron energy	MeV	0.43	0.49	0.40
	$\bar{\nu}_{\gamma,delayed}$	Average delayed photon yield[7]	delayed photons per fission	0.613 *short period* 3.31 *long period*	1.42 *short period* 5.50 *long period*	0.608 *short period* 3.26 *long period*
	$\bar{E}_{\gamma,delayed}$	Average delayed photon energy[8]	MeV	0.96	0.92	0.98

A schematic representation of a typical pulsed active interrogation irradiation sequence showing this convention for labelling the prompt and delayed signatures is shown Figure 1. Also shown in this figure is the general shape of a die-away signature. Die-away signatures originate as the neutron population within an assembly being interrogated decays over time following each interrogating radiation pulse; new thermal-neutron-induced fission events continue to take place at a smoothly decreasing rate after the end of each pulse as thermal neutrons are absorbed within the assembly and

[1] Values followed by parenthesis indicate quantities associated with fission induced by neutrons with an energy or energy-spectrum spectrum shown in the parenthesis.
[2] Neutron yields vary depending upon the energy of the neutron that induces fission.
[3] An increase of ~ 4% is expected for these average fission neutron energies in going from thermal-neutron-induced fission to fission-spectrum-induced fission (where the average neutron inducing fission is ~ 2 MeV).
[4] From Reference [35].
[5] From Reference [36].
[6] Less than 2% of prompt fission photons have energies greater than 2 MeV.
[7] Delayed photon yields determined for short ($0.2 < t < 0.5$ sec) and long ($0.2 < t < 45$ sec) counting periods.
[8] Delayed photon energies are averaged over a $0.2 < t < 45$ sec time period. Less than 1.8 % of delayed fission photons are > 2.3 MeV in this time period.

leak out of the assembly. Die-away measurements are frequently used as the basis for studies of how to develop instruments to detect shielded special nuclear material.[5,9,12-15,24,26] Measurement approaches based upon the detection of die-away signatures alone can be problematic, however, due to possible complications arising from the presence of neutron absorbers in the test assembly being interrogated.[9]

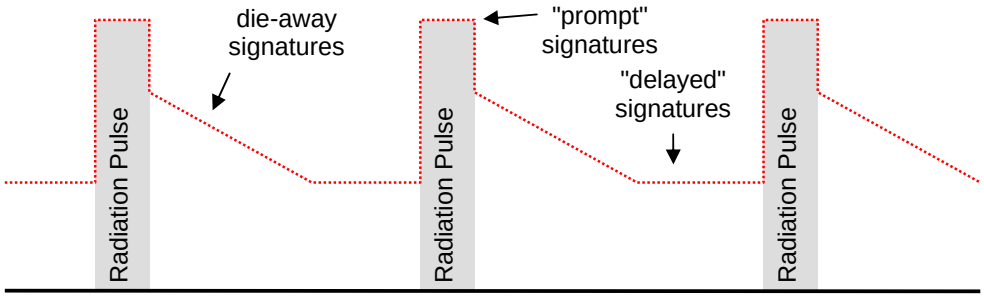

Figure 1 Representative active interrogation timing sequence. The dotted line (red) indicates the measured values that would be observed using a gamma-ray or neutron detector near an SNM-bearing assembly being irradiated.

Following the nomenclature of the prompt and delayed signature labels shown in Figure 1 many researchers have written of measurements that detect "prompt" emissions or "delayed" emissions, according to the timing structure of their detection apparatus. Unfortunately, in some cases though these general terms do not correctly reflect the signatures actually being measured. To understand this difference it is useful to consider traditional nuclear reactor physics concepts for subcritical reactors, including the effective multiplication factor $\boldsymbol{k_{eff}}$ and the subcritical multiplication, $\boldsymbol{M}$.[37] The effective multiplication factor is a ratio indicating the average fraction of new neutrons that will be produced within a multiplying nuclear assembly (accounting for absorption, capture, and fission) following the introduction of an initial neutron into the assembly, such as from a nearby ENG. For example, if a particular assembly has an effective multiplication factor of 0.8 then if 100 neutrons enter the assembly (the zeroth generation of neutrons), while most will escape or be captured, some will cause fission (yielding on average ~2.6 neutrons per fission) to create a so-called "first" generation of 80 new neutrons. These 80 neutrons will again be subjected to the effects of the assembly with $\boldsymbol{k_{eff}}$ = 0.8 to yield a second generation of 64 neutrons, followed by 51, and so on. Summing the zeroth generation plus all of the daughter generations a total of 500 neutrons will have been in the system including those initially from the ENG and then those subsequently produced in the system from fission. Subcritical multiplication is defined as the ratio of the total number of new fission neutrons produced within an assembly to the original number of neutrons injected into the system, Eq. 1.

$$\boldsymbol{M} = \frac{\boldsymbol{1}}{\boldsymbol{1 - k_{eff}}} \qquad \text{Eq. 1}$$

With this background now consider a pulsed active neutron interrogation measurement with a 25-kg assembly of highly enriched uranium (HEU) enriched to 93% ^{235}U, without any external neutron reflecting materials present, that has an effective multiplication factor of 0.8 and a subcritical multiplication $\boldsymbol{M}$ of 5.[37] During each neutron pulse many neutrons will injected into the systems, some of these will cause fission in the uranium. At the end of each interrogation pulse the neutron population in the assembly from the pulse will quickly die-away, at that time the only sources of neutrons emanating from the assembly will be neutrons produced from the decay of delayed-neutron emitting fission products created earlier and neutrons produced from new fission events in the assembly initiated by these delayed neutrons. For the assembly described here 80% of all the

neutrons in the assembly during the interpulse region, often called the delayed neutron region, will be prompt neutrons from fission while only 20% will actually be delayed neutrons.

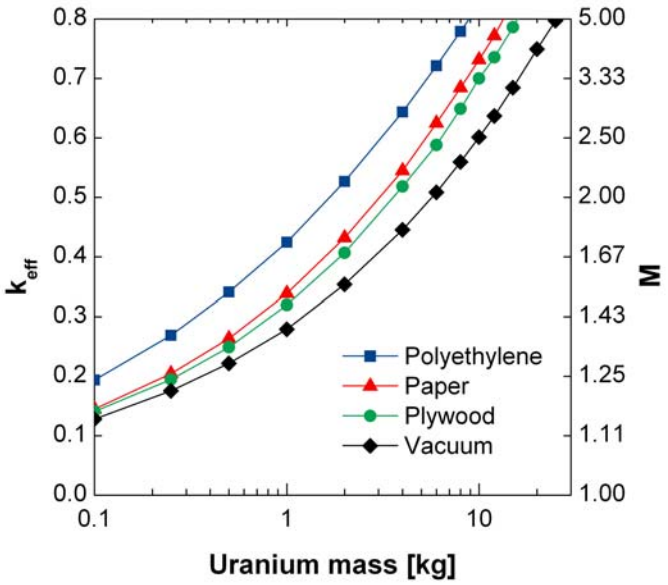

Figure 2 Effective multiplication factor and subcritical multiplication for different mass uranium spheres centered within 1 m^3 cubes of different packing materials and for bare HEU (vacuum).

The implications of this observation are important. In some active interrogation system the measurement objective is to be able to detect very small quantities of fissionable material, in the range of 1 gram or less; such as in systems intended to assay transuranic material content within waste drums and crates.[5] In these cases k_{eff} <<1 and measurements carried out in-between interrogation pulses of after the end of an irradiation cycle will indeed only detect radiation products from the delayed decay of fission products. In other situations, however, there is interest in detecting larger quantities of fissionable material.[38] For measurement scenarios considering fissionable material quantities in the 1 – 10 kg range k_{eff} can be found to range from ~0.25 to greater than 0.8, depending upon the type and quantity of nearby neutron reflecting materials. In these cases prompt fission neutron emission signatures can be expected to contribute significantly to the total observable signatures resulting from active interrogation. Illustrating this point, simple estimates of k_{eff} and M for different quantities of HEU have been calculated using the MCNP5 simulation code for different types of commonly found materials including polyethylene, paper, plywood, as well as bare material (vacuum), and are shown in Figure 2.[39] For this example the bare HEU case presents a lower bounding limit for the HEU assembly's effective multiplication factor while polyethylene presents a useful upper bounding limit for an HEU assembly's effective multiplication factor.

3. Active Neutron Interrogation Scoping Studies at INL

A photograph of the set-up used in one of our active neutron interrogation experiments is shown in Figure 3, where 9.4 kg of HEU has been placed within a wood-filled box; typical details about the fissionable materials, shield materials, ENG, and neutron detectors used in our experiments have been described previously.[26] Our measurements are usually performed within shielded test facilities in Idaho Falls (without special nuclear materials) or at INL's Materials and Fuels Complex.[25,40] In most cases the primary measurement protocol involves an interrogation of between 100 and 500 seconds with the ENG operating in pulsed mode from 300 – 1000 Hz.

Figure 3 Representative photo of the set-up used for active neutron interrogation experiments at INL.

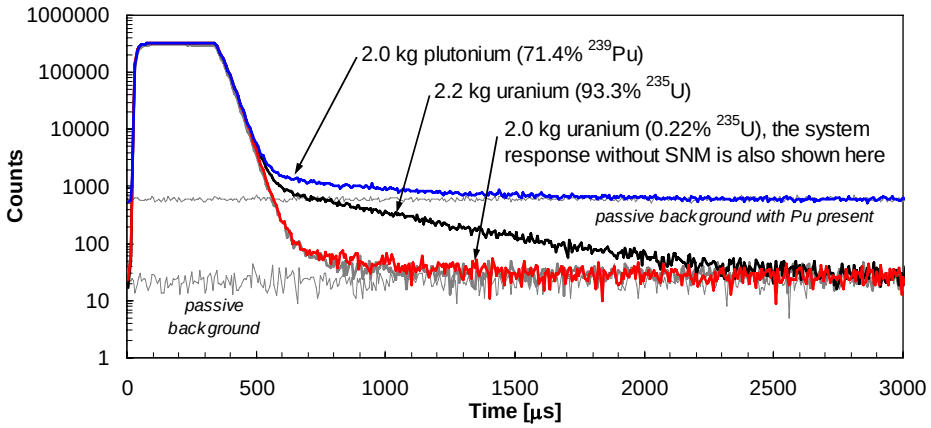

Figure 4 Die away data from HEU, depleted uranium, and Pu within the wood cube. Note that the response of the detectors to an interrogation measurement without material (dark grey) is very difficult to discern from the die-away signal of the depleted uranium (red).

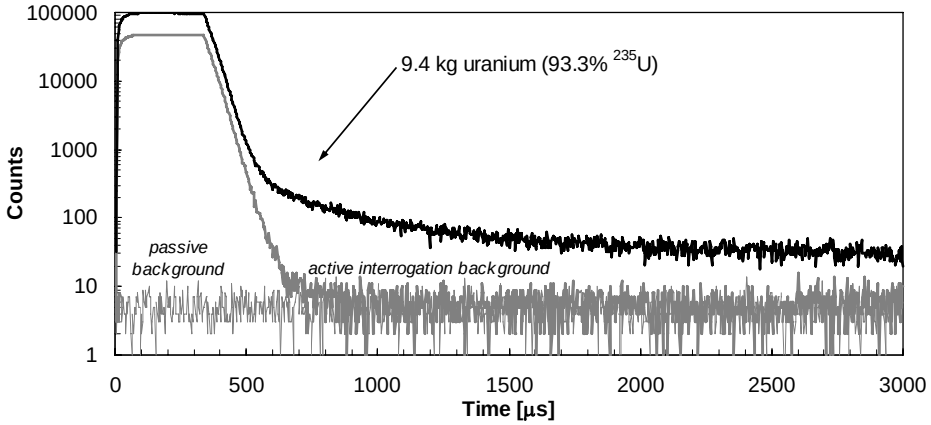

Figure 5 Die away data from 9.4 kg of bare HEU.

Neutron count data is time-synchronized with each ENG pulse, collected using a commercial multi-channel scalar counting system, and recorded via laptop computer. The detectors used in these experiments consist of 10-atm. ^{3}He tubes embedded within polyethylene sleeves 2.54 to 5.08 cm thick. The detectors are covered in thermal-neutron absorbing materials to prevent the detections of low-energy room-scattered neutrons; it typically takes about 400 microseconds for these neutron detectors to settle to background counting rates after the end of a neutron pulse. Examples of the die away data collected from this system are shown in Figure 4 and Figure 5. The data in Figure 4 were collected using a set-up like the one shown Figure 3 using separate samples of HEU, plutonium, and depleted uranium. For both the HEU and Pu measurements there are very strong die-away signatures (consisting of prompt and delayed neutron emissions) indicative of the presence of fissionable material. With the depleted uranium no clear die-away signature is apparent but a statistical evaluation of the net counts in the time span from 1000 to 3270 microseconds does indicate a small fissionable material signature (618 ± 137 counts), roughly scaling with the HEU data in consideration of the mass of ^{235}U still present within the 2.0 kg of depleted uranium (0.0044 g). Data is shown in Figure 5 for the measurement of a bare assembly of 9.4 kg of HEU supported on a metal cart roughly 33 cm above a concrete floor. Here a die-away signal can still be observed which is likely due to a loose neutronic coupling between the HEU and the floor (for a discussion of this effect see, for example, Reference 12.) In the 2500 to 3000 microsecond time period in these measurements, after the end of the die-away signal, a nearly constant but elevated neutron count rate was observed. In some cases researchers refer to this signal as the delayed neutron signal. However, it follows from the discussion presented earlier that, with the large mass of HEU used in these measurement, these neutrons are not purely originating with the beta-decay of neutron-emitting fission products but that a non-negligible fraction are the result of prompt fission taking place within the subcritical HEU assembly.

To study the time-decay structure of the delayed neutron signal associated with our active neutron interrogation measurements we occasionally switch our operational mode to collect data after the ENG is turned off. In these cases we typically operate the ENG for a minimum of 120 seconds in order to reach delayed-neutron fission product equilibrium within the assembly, data collection is then aggregated into 5-second time bins for 100 measurement periods. An example of this type of data is presented in Figure 6. In this data a second, confirmatory indication exists to complement the interpulse observations and substantiate the declaration that fissionable material is present. Relating this observation to the well-known six-group delayed neutron structure for ^{235}U we see strong evidence of the quickly decaying component associated with the 2.23 second delayed-neutron group (relative abundance of 0.407.)[41] In the future we intend to study this fissionable material active interrogation signature and other signatures in greater detail by using a more sensitive neutron detector and improving the timing resolution in our post-irradiation time-decay analyses.

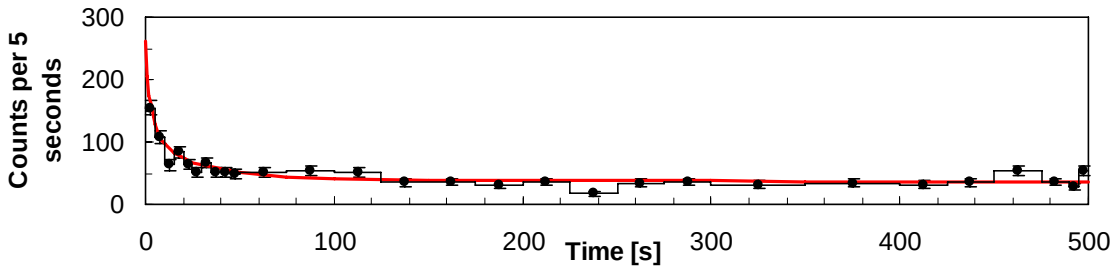

Figure 6 Post-irradiation active neutron interrogation signature corresponding to the decay of beta-delayed neutron emitting fission products (the solid line is an estimated fit to the data using the six-component delayed neutron group parameters from Reference 41).

4. Summary

Using an external neutron source to probe a suspect object, active neutron interrogation has the potential of serving as a useful technique for detecting shielded fissionable material. The technique has been well-developed for nuclear safeguards measurements for assaying waste drums and stored materials but also has a role to play beyond application for these well-constrained problems. Recent innovations in the design and engineering of small, compact-accelerator electronic neutron generators now make it feasible to consider deploying portable active neutron interrogation systems for field use.[42] Most prior work in this area has focused on the measurement of either die-away neutron signatures or delayed neutron signatures in-between pulses of an ENG, some prior work has also considered the detection of prompt emissions during the ENG pulses and the detection of delayed gamma rays after the end of an ENG irradiation. Unfortunately, in many cases the nomenclature used in these types of measurements can be misleading, especially for SNM masses exceeding a few kilograms when subcritical neutron multiplications effects become important and both prompt and delayed emission signatures may be present simultaneously at comparable levels. At Idaho National Laboratory a comprehensive program of research is underway to study theoretical and practical question related to the design and implementation of portable active neutron interrogation systems. Included in our work are examinations to study how to optimize the design of equipment used in active neutron interrogation and to develop new methods for integrating data from multiple SNM signatures into an automated detection/decision evaluation algorithm. Experiments are performed using portable electronic neutron generators, multiple types of radiation detectors, and significant quantities of special nuclear material, including the use of shield materials.

5. References

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