AIP Applied Physics Letters

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Citation: Applied Physics Letters **105**, 014102 (2014); doi: 10.1063/1.4887366
View online: http://dx.doi.org/10.1063/1.4887366
View Table of Contents: http://scitation.aip.org/content/aip/journal/apl/105/1?ver=pdfcov
Published by the AIP Publishing

Fission signal detection using helium-4 gas fast neutron scintillation detectors

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(Received 17 June 2014; accepted 24 June 2014; published online 8 July 2014)

We demonstrate the unambiguous detection of the fission neutron signal produced in natural uranium during active neutron interrogation using a deuterium-deuterium fusion neutron generator and a high pressure ^{4}He gas fast neutron scintillation detector. The energy deposition by individual neutrons is quantified, and energy discrimination is used to differentiate the induced fission neutrons from the mono-energetic interrogation neutrons. The detector can discriminate between different incident neutron energies using pulse height discrimination of the slow scintillation component of the elastic scattering interaction between a neutron and the ^{4}He atom. Energy histograms resulting from this data show the buildup of a detected fission neutron signal at higher energies. The detector is shown here to detect a unique fission neutron signal from a natural uranium sample during active interrogation with a (*d*, *d*) neutron generator. This signal path has a direct application to the detection of shielded nuclear material in cargo and air containers. It allows for continuous interrogation and detection while greatly minimizing the potential for false alarms.
 [http://dx.doi.org/10.1063/1.4887366]

Detection of the presence of shielded special nuclear material (SNM) hidden in transportable cargo is a global problem that has significant gaps in research and capabilities.[1] SNM can be identified by detecting gamma and neutron radiation produced by either spontaneous or induced fission. Passive detection systems have difficulty detecting SNM that is shielded or has a low spontaneous fission rate.[2] Active detection methods cause an additional signal from fission to be generated in SNM by interrogating it with radiation. This work focuses on the detection of SNM by actively interrogating material with neutrons and detecting the induced fission neutron signal. For this purpose, detecting neutrons has two benefits over gammas: (1) long attenuation lengths allowing the neutrons to penetrate through shielding material that would otherwise absorb gamma emissions and (2) low neutron background levels providing better specificity from neutron signal and reduced false positives.[3]

This work explores a method of scanning for nuclear material using ^{4}He scintillation detectors. The energy discrimination ability of ^{4}He detectors opens up a signal path that will allow the detection of the presence of SNM more quickly, accurately, and economically than existing systems. ^{4}He detectors are a recently developed gas-scintillator for fast neutron detection that allows for the direct measurement and analysis of individual neutron interactions.[4] We show that this detector and active interrogation method is able to detect SNM with a very low passive signal—natural uranium produces about 13.5 neutrons/(s kg).

Current SNM neutron detection systems rely on helium-3 (^{3}He) thermal neutron detectors or liquid scintillator detectors. The ^{3}He gas detectors are sensitive to thermal energy neutrons but do not provide incident neutron energy information. Active interrogation methods using ^{3}He detectors, such as differential die-away analysis, must rely on source pulsing schemes and differences in time behavior between interrogation and fission neutrons to detect a fission signal during interrogation.[5] Liquid scintillators rely on the recoil of hydrogen to detect fast neutrons; however, they are more sensitive to gamma interactions and cannot as easily differentiate between gamma and neutron interactions, which are crucial for SNM detection.[6] Work has been done to improve separation between gammas and neutrons using pulse shape discrimination.[7–9] Issues with internal moderation and multiple scattering interactions limit the capability of liquid scintillators to provide information on interacting neutron energy.[10]

The ^{4}He detector, developed and manufactured by Arktis Radiation Detectors, comprises a cylindrical volume filled with 120 bars of pure ^{4}He gas that has a density of 0.03 g/cm^3 and a photo-multiplier tube (PMT) optically coupled to each end (Fig. 1). The detectors operate at room temperature and the PMTs are powered by the

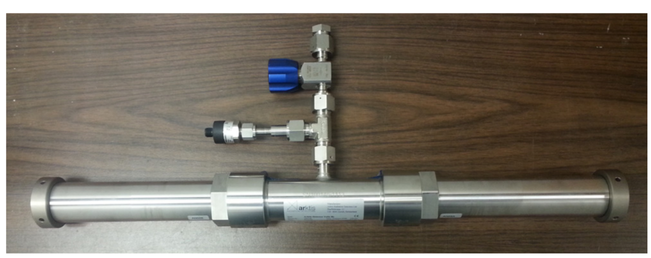

FIG. 1. A single ^{4}He detector. The central tube is filled with 120 bars of ^{4}He gas and is the active detection volume. Photo-multiplier tubes are optically attached to either end. The perpendicular port is for filling and verifying gas pressure.

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instrumentation. The ^{4}He gas is both the interaction and scintillation medium. It detects neutron interactions by measuring the scintillation light produced by recoiling ^{4}He nuclei. The inside of the detector volume is coated with a wave-shifter to shift the scintillation light from ≈80 nm to an optimal wavelength for detection by the PMTs of 450 nm. The low electron density of ^{4}He, 5.3×10^{21} cm^{-3} at 120 bars, greatly reduces the probability of gamma interactions.[11] It is capable of rejecting a gamma dose of 100 μSv/h at a power of 10^{-7} and maintaining a 5% detection efficiency for fission neutrons.[12]

A fast neutron that enters the detector volume and undergoes an elastic scattering interaction will transfer a portion of its kinetic energy to the ^{4}He nucleus, which is stripped of its electrons and becomes an alpha particle. This alpha particle is accelerated through the gas and interacts with other helium atoms by excitation or ionization. These excitations result in the production of helium excimer states, which decay and emit scintillation light that is counted by the PMTs at either end of the detector volume.

This elastic scattering interaction produces scintillation light with two components. The fast component of the scintillation light occurs within several nanoseconds of the interaction and is created by the initial energy transfer interaction between the neutron and the ^{4}He nucleus. This component is a large and brief light pulse lasting approximately 5 ns and is used by the electronics as an interaction trigger. The slow component of scintillation begins immediately after the fast component, lasts several μs, and is the product of excited ^{4}He atoms releasing gained energy in the detector resulting in scintillation light. The light produced is counted by both PMTs and the instrumentation correlates and digitizes each interaction event. The total amount of light produced in the slow component is proportional to the amount of energy transferred by the neutron to the ^{4}He atom.[11]

Fig. 2 shows the fast and slow component of a single neutron interaction as digitized by the detector instrumentation. The integral of the slow scintillation component is proportional to the energy deposited in the detector. Gamma radiation primarily interacts with electrons in the gas, and the resulting recoil electron deposits less energy over a longer distance in the detector, allowing for simple pulse shape discrimination of gamma interactions.

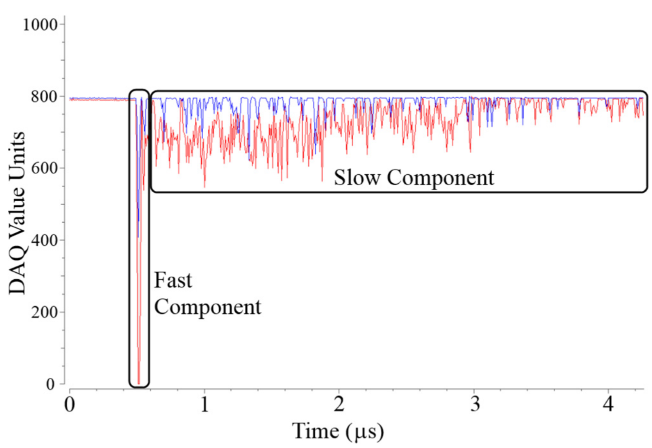

FIG. 2. A graph of the detector and instrumentation output for a single neutron event. There are two traces shown, one for each PMT.

The amount of energy deposited in the detector by an interacting neutron depends on its initial energy and scattering angle. The maximum elastic energy transfer from a neutron to ^{4}He is 64% based on scattering kinematics. The actual energy transfer is a function of maximum energy based on the elastic scattering angle that has a non-linear probability distribution over the values in this range. The elastic scattering cross section for ^{4}He over the incident neutron range of interest varies from 1.0 to 7.5 b between the range of 350 keV and 10 MeV, peaking at 1.1 MeV.[13]

This neutron interaction mechanism gives the ^{4}He detector significant energy information of the incident neutron spectrum. Once a response matrix is derived over the range of energies, unfolding algorithms can be used to convert the detected spectrum to the incident spectrum.[14] Determining the response matrix for these detectors is an ongoing area of research, and has not yet been determined; therefore, a threshold method is used in this work to differentiate between fission and interrogation neutrons.

The interrogation neutrons for this experiment are produced by a deuterium-deuterium (*d*, *d*) neutron generator. Mono-energetic neutrons are produced isotropically with an energy of 2.45 MeV by the fusion of deuterium atoms. The generator used in this work is capable of producing 1×10^9 n/s. These neutrons are used to actively interrogate and cause fission in fissionable material.

A majority of the neutrons that are produced per fission have energy between 0.1 and 10 MeV with an energy distribution that follows a Watt spectrum. Of these, 26% are emitted with energy greater than 2.45 MeV, the energy of the interrogation neutrons. Adjusting for the probability of angle of scatter with a ^{4}He atom, 3.2% of fission neutrons have the potential to deposit more energy in the detector than the interrogation neutrons thus making them differentiable by energy discrimination.

The nuclear material used are cylinders of natural uranium encased in aluminum with a 2.85 cm radius and 21 cm length with a weight of 2 kg. This gives a mass of 1970 g of natural uranium of which 1956.2 g is ^{238}U and 13.8 g is ^{235}U.

For the first experiment, the detector was placed 380 cm from the (*d*, *d*) generator and the generator was run for 60 min. Two natural uranium slugs were added for the second experiment, one placed directly above and one below the detector (Fig. 3). Again the generator was run for 60 min. The average generator output for both experiments was 1.25×10^8 neutrons per second. The detector recorded over 2 million events for each experiment and the first 2 million events from each are used for analysis. Fig. 4 shows the results of these experiments.

The top subfigure of Fig. 4 is an energy histogram of the generator only data (dark) and an energy histogram of the detector response with natural uranium (white). The unique higher energy signal from fission neutrons is apparent at the higher energy levels. The middle sub-figure shows the total number of counted events above a given cutoff (the value on the x axis) and is the total signal above an energy cut. This shows that summing the number of counts above a given energy cut gives a clear and significant indication of the

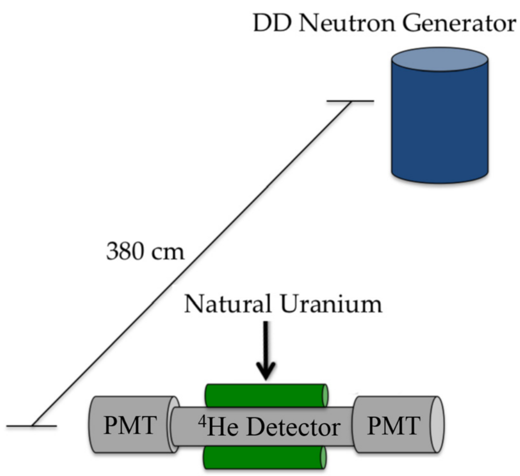

FIG. 3. Schematic of the irradiation setup with the natural uranium present.

presence of nuclear material. The bottom sub-figure shows the ratio between the total number of counts with energies greater than the x axis value with and without natural uranium. This shows that there is an optimal cutoff for determining where to look for higher energy neutron events from the detector. The fission to interrogation signal ratio is significant and shows a unique identification of the fission neutrons produced by the fission of the natural uranium.

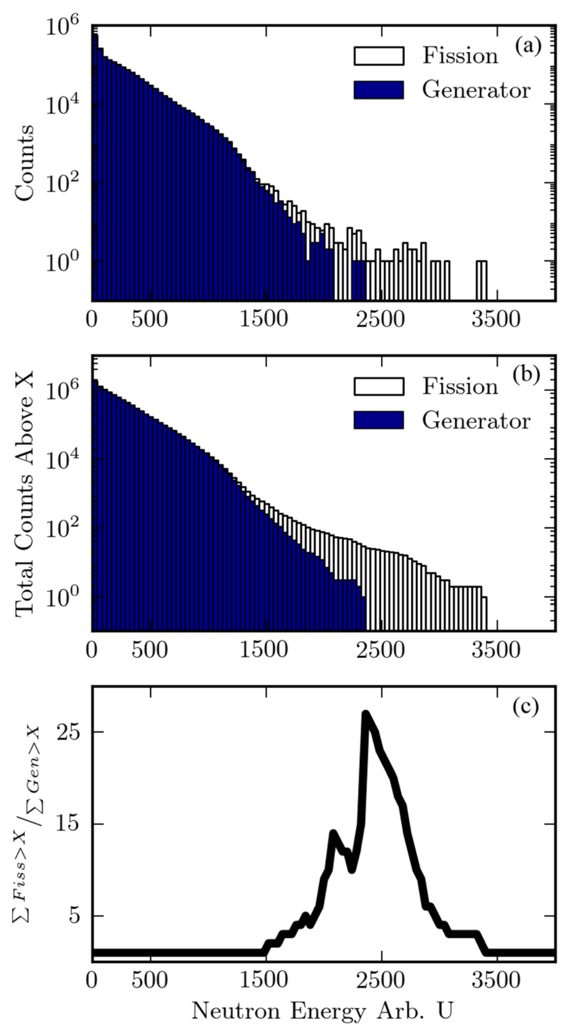

FIG. 4. A histogram (a), total counts above cut (b), and ratio between generator only and fission signals above cut (c) of the detector response to a (*d*, *d*) generator with (light) and without (dark) the presence of natural uranium.

Monte Carlo calculations estimate that 1.5 million fission neutrons are produced in each natural uranium slug during the 60 min interrogation time. Adjusting for detector efficiency, geometric constraints, and minimum required energy deposition, approximately 150 fission neutrons per slug should be uniquely distinguishable from the interrogation neutrons. The results show that only 72 neutrons are exclusively above the cutoff, which is one quarter of the expected amount with two slugs. The difference can be attributed to pileup in the detector that extends the background signal up to higher energies. Pileup events occur when a neutron interacts during the 4 μs that the instrumentation is counting the slow scintillation response from a previous interaction.

The ability of a ^{4}He gas scintillation fast neutron detector and instrumentation to detect the presence of fissionable material during active neutron interrogation is shown using natural uranium and a deuterium-deuterium (*d*, *d*) neutron generator. The neutron energy discrimination capability of the detector is found to be able to uniquely distinguish a significant portion of the induced fission neutron signal from the interrogation signal during interrogation. The results suggest this detector, analysis, and signal path could fill a critical gap in nuclear material detection and security because of its excellent gamma ray rejection efficiency and neutron energy discrimination capability. The capability to uniquely identify neutrons from fission is a powerful tool and has the potential to be used in several nuclear security and safeguard applications.[15]

Most importantly, and contrary to similar methods that rely on the detection of lower-energy thermal neutrons or gammas that are easily attenuated by shielding materials, this SNM detection method is focused on the detection of higher-energy neutrons. These neutrons are naturally more penetrating and therefore negate the effectiveness of a majority of common radiation shielding materials. The significant quantity of available signal, combined with the natural ineffectiveness of shielding materials at the target energies, illustrates the potential of a ^{4}He detection system across a wide range of the proposed threat space, producing a quicker and more accurate assessment of a container's contents.

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