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Early results utilizing high-energy fission product (gamma) rays to detect fissionable material in cargo

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Abstract

A concept for detecting the presence of special nuclear material (^{235}U or ^{239}Pu) concealed in intermodal cargo containers is described. It is based on interrogation with a pulsed beam of 7 MeV neutrons that produce fission events and their β-delayed neutron emission or β-delayed high-energy γ-radiation between beam pulses provide the detection signature. Fission product β-delayed γ-rays above 3 MeV are nearly ten times more abundant than β-delayed neutrons and are distinct from natural radioactivity and from nearly all of the induced activity in a normal cargo. Detector backgrounds and potential interferences with the fission signature radiation have been identified and quantified.

Introduction

An important goal in the US is the detection of nuclear weapons or special nuclear material (SNM) concealed in intermodal cargo containers. This must be done with high detection probability, low false alarm rates, and without impeding commerce, i.e. about one minute for an inspection. Our concept for inspection has been described before[1, 2] and its components are now being evaluated. While normal radiations emitted from plutonium may allow its detection, the majority of ^{235}U γ ray emission is at 186 keV, is readily attenuated by cargo, and thus not a reliable detection signature for passive detection. Delayed neutron detection following a neutron[3] or photon[4] beam pulse has been used successfully to detect lightly or unshielded SNM targets. While delayed neutrons can be easily distinguished from beam neutrons they have relatively low yield in fission, approximately 0.008 per fission in ^{239}Pu and 0.017 per fission in ^{235}U[5], and are rapidly attenuated in hydrogenous materials making that technique unreliable when challenged by thick hydrogenous cargo overburden. Below we propose detection of β–delayed high-energy γ radiation as a more robust signature characteristic of SNM.

Concept description

There are many high-yield fission products that have short half-lives and thus high specific activities. Some of them produce γ rays with energies exceeding 3 MeV. These high-energy β–delayed γ rays have nearly ten times greater yield than the delayed neutrons[1], and their high yield has been confirmed by recent experiments[6]. Delayed high-energy γ radiation is a characteristic of fission, distinct from normal radioactive background whose γ radiation lies below 2.6 MeV and from most, though not all, neutron activation products.

The architecture of our proposed system is illustrated schematically in Figure 1. A neutron generator is buried below ground and its output collimated into a fan at the surface illuminating the floor of the cargo container as it's scanned past. Detection of the delayed γ radiation occurs in two (or more) linear arrays of detectors along the sides, analogous to a car wash. The detectors can distinguish photons with energies above and below 3 MeV and they are loaded with Gd to provide detection of β-delayed neutrons. The fission products have a mixture of decay times ranging from 5-150 s and the dominant species depends on the length of neutron irradiation. For radiations in the range 10-30 sec the principal intensity decays with a half-life of approximately 25-55 s [1, 6] and is distinct from nearly all of the neutron activation products. High-energy γ radiation has nearly ten times higher yield than delayed neutrons, ~ 0.13 per fission[1], and several decades greater penetration in thick hydrogenous cargos[1]. The enhanced penetration of γ radiation in hydrogenous cargo compared to that for β-delayed neutrons is shown in Figure 2. For γ radiation in the range 3-5 MeV attenuation is due predominantly to Compton scattering and is thus small (~ 0.04 cm^2/g) and material independent, varying only with areal density. This facilitates reliable interrogation of SNM targets that may be buried behind ≤ 100 g/cm^2 of intervening material and greater thickness is usually prohibited by container weight limits.

Validation of concept

A Monte Carlo simulation code has been developed that generates the full range of fission products and generates β-delayed γ rays including the high-energy emissions[7]. This code has been utilized to study high-energy γ radiation signals from HEU targets with various cargo overburdens. The simulations are used to predict detection performance of this concept and to design experimental tests of it. Currently those predictions are being validated by measurements in the LLNL cargo-scanning lab.

In the cargo scanning facility a collimated 14 MeV neutron source at 4×10^{10} n/s output produces a neutron beam that irradiates a standard 20 ft cargo container through its floor, producing an unattenuated fast neutron flux 1.0 m above the floor at container centerline ~ 6×10^4 $n/cm^2/s$. The container is loaded with palletized cargos of plywood, aluminum sheets, or steel pipes; all at mean density $\rho \sim 0.4\text{-}0.6$ g/cm^3, and targets containing highly enriched uranium (HEU) are placed inside on its centerline. There will soon be capability to scan the loaded container past the interrogation system at speeds up to 40 ft/min. A 4-cell array of liquid scintillators (20 cm diameter, 200 cm long) outside the sidewall of the container record high-energy γ radiation (E > 3 MeV) and delayed neutrons. High-resolution γ ray spectra are recorded in a collimated HPGe spectrometer. All data are recorded in "event mode" for each detector so that off-line processing can be utilized to reconfigure data presentation and allow additional off-line analysis with varying parameter settings such as variations in energy discrimination.

In the first experiments a cargo of plywood 4x8x6 ft high was installed over the neutron source without the cargo container present and high-energy γ radiation recorded in the four liquid scintillators located on the side at R=200 cm from the HEU target. A cylindrical target of 380 g 93% enriched HEU oxide was placed near the cargo centerline within the wood cargo, a distance 61 cm above the floor so that the entering neutron beam passes through $\sim$ 40 g/cm^2 of wood and exiting γ radiation traverses 61 cm of wood (40 g/cm^2) reaching the detectors. Data obtained in these experiments relied upon a 30 sec irradiation followed by 100 sec counting interval. To improve confidence in the data the irradiations were repeated over 59 cycles of irradiation and counting. A second set of measurements used the same radiation/counting cycle with the HEU target removed, and it was called "Active-background" to distinguish it from the normal cosmic ray background recorded in the detectors and from natural radioactivity in the environment which these detectors do not see due to their high energy threshold. For data comparisons the cosmic background was subtracted and the Active-Background (target-out) was re-scaled to the same neutron fluence as the foreground (target-in) data. Data obtained are shown in the decay curve in Figure 3.

Two lines are shown in Figure 3, one with half-life 7.13 s characteristic of ^{16}N. This activation product has been identified by high resolution γ ray spectroscopy[1, 2] where its principal emission, a 6.1 MeV γ ray, is produced in 68% of its β-decays and is prominent in the spectrum at early times. The other line is a much longer half life consistent with several of the major β-delayed fission products that produce high-energy γ rays as observed previously and whose half-lives are in the range 25-55 s. The ^{16}N activity is a significant interference for the first 40 sec of counting following 14 MeV neutron interrogations. Longer-lived fission product radiation dominates at later times with intensity roughly ten times above the interfering radiation for this configuration.

The fission product half life can be understood in terms of the fission products whose high-energy γ–ray yield are highest in ^{235}U fission, i.e. 55-s ^{86}Br, 55 s-^{87}Br, 156-s ^{90}Rb, 258-s ^{90m}Rb and 58-s ^{91}Rb[1]. Further, some of the intensity attributed to ^{16}N must be due to the decay of a number of shorter-lived fission products such as 4.35-s ^{89}Br, 4.49-s ^{92}Rb, and others. That intensity can be fruitfully utilized for improved detection when the ^{16}N interference is eliminated.

The ^{16}N is due to oxygen activation via the $^{16}O(n,p)^{16}N$ reaction and has been noted before [1, 2]. Its reaction threshold is 10 MeV[6]. It is eliminated by utilizing a neutron beam energy below 10 MeV. An RFQ accelerator has been purchased for the cargo scanning lab to provide 100 μA of deuterons on a gas deuterium target, producing an unattenuated fast flux at container centerline ~$6x10^5$ n/cm^2/sec at E_n=7 MeV, a 10-fold increase over the present configuration.

Conclusions

A concept for detection of SNM hidden in cargo containers is proposed and some of its elements have been studied. The method is based on 7-MeV neutron interrogation to produce fission followed by detection of very abundant, distinctive and penetrating β-delayed high-energy γ radiation and/or delayed neutrons from decay of the fission products. The entire contents of the container are irradiated as the container passes over a source buried below ground and aimed vertically. The container then passes through a linear array of neutron and γ ray detectors where both delayed neutrons and delayed γ rays are detected. Simulations and measurements carried out thus far indicate that the high-energy β-delayed fission product γ radiation detection is up to 10^4 more sensitive than β-delayed neutron detection when challenged by thick hydrogenous intervening cargo.

Acknowledgements

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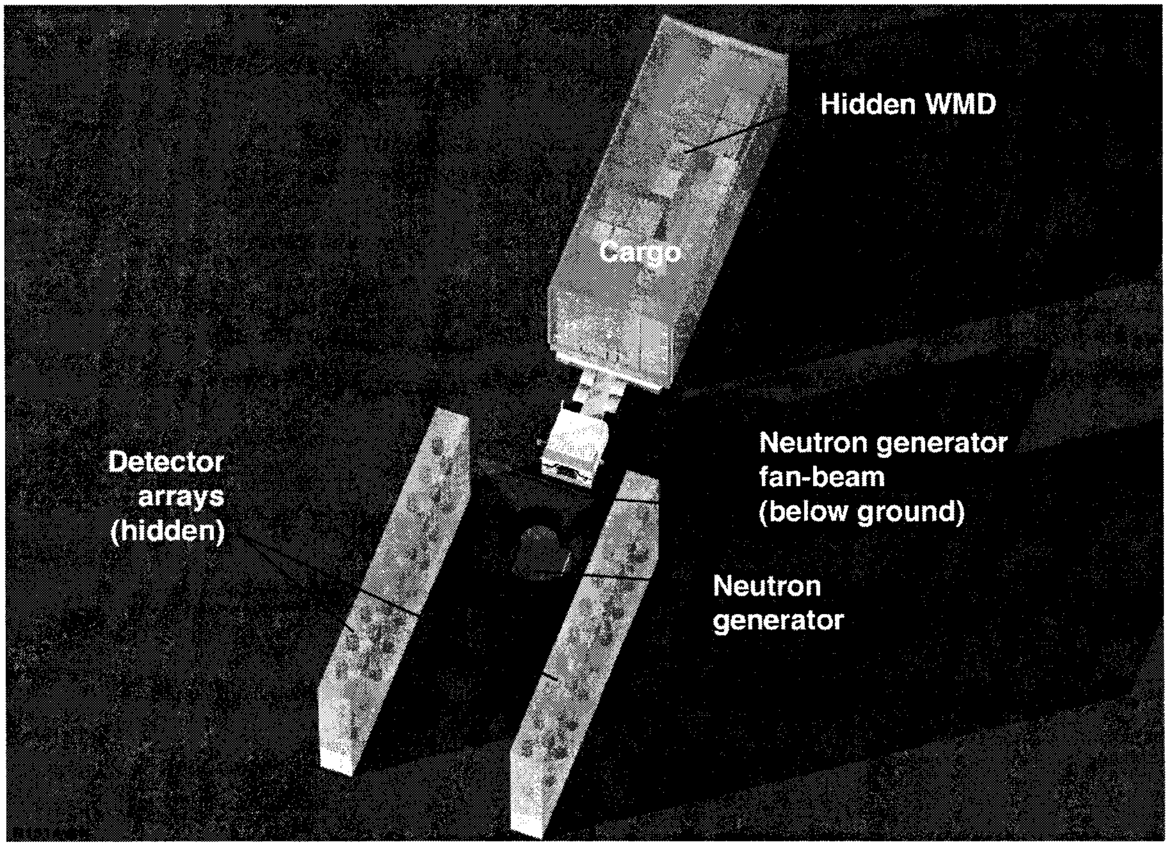

Figure 1 *Schematic of the "nuclear car wash" showing a below-ground collimated neutron source irradiating an intermodal cargo container from below, and two linear detector arrays to detect subsequent fission product β-delayed γ-rays.*

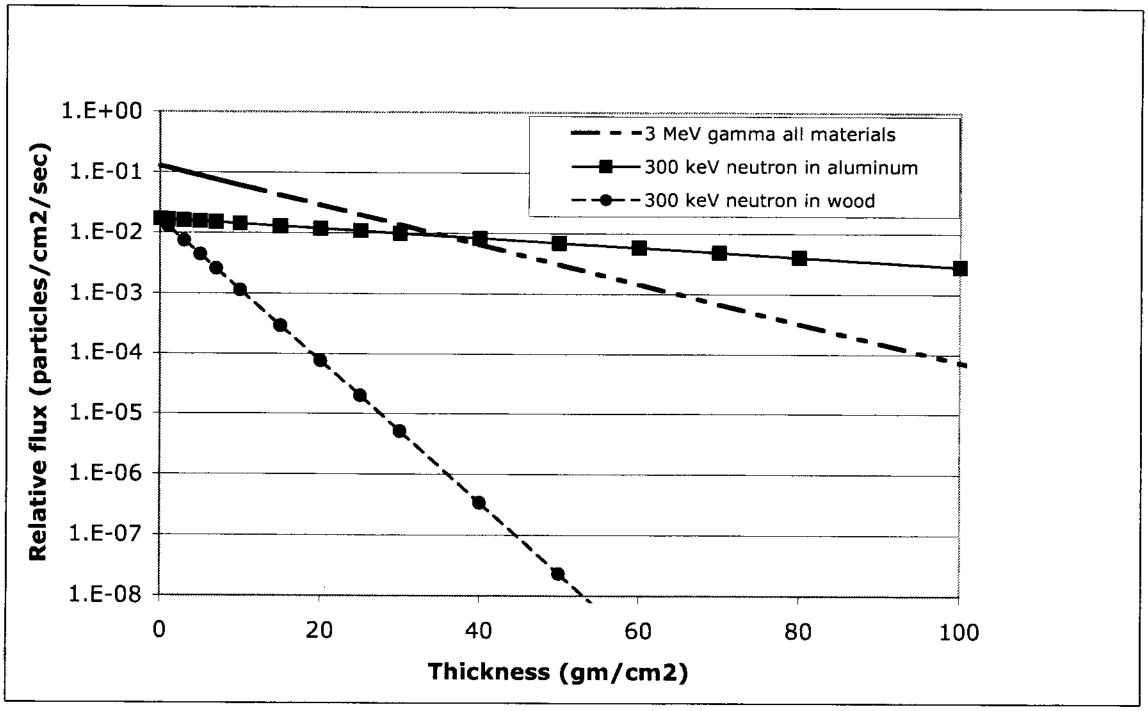

Figure 2. Relative γ ray and delayed neutron fluxes after attenuation in aluminum or wood

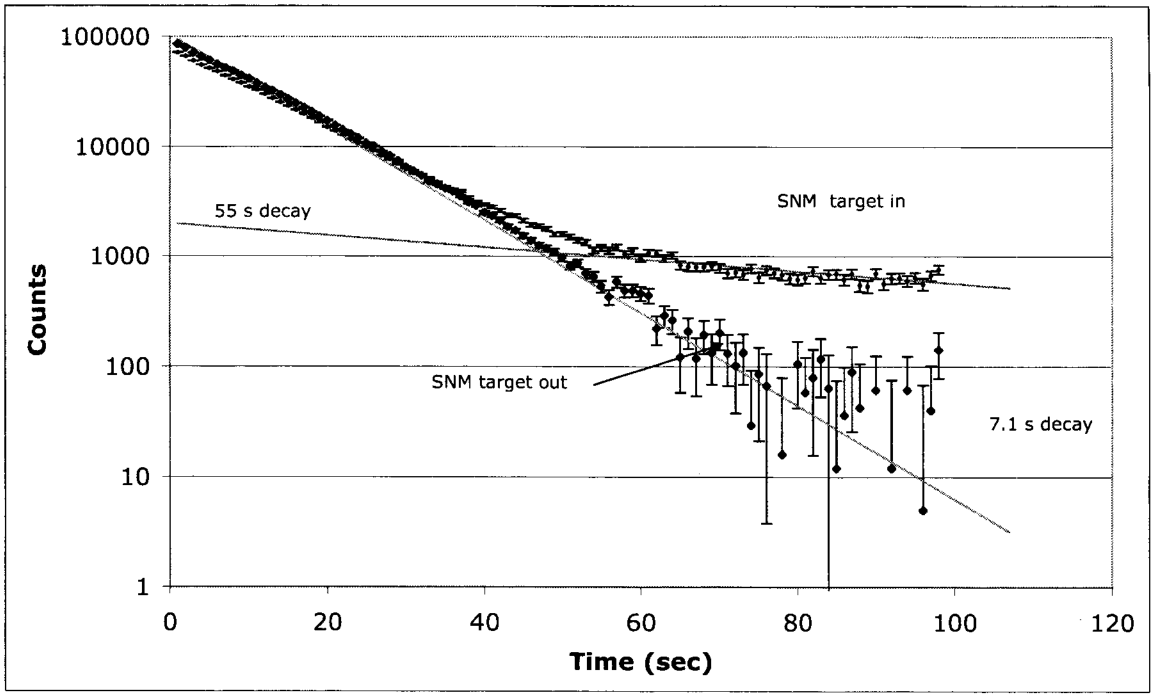

Figure 3. Decay of γ radiation at energies above 3 MeV for a 130-s interrogation cycle. Two data sets are shown: solid circles are foreground, i.e. target present, and diamonds are background, i.e. target removed but interrogation otherwise identical. Lines are not fits but intended to guide the eye. They correspond to 7.1 and 55 sec half-lives.

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