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Signatures of fissile materials: high-energy γ rays following fission

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Abstract

The spectral and temporal dependence of β-delayed γ rays in the range $E \geqslant 3.0$ MeV from thermal–neutron-induced fission are shown to be characteristic signatures of ^{235}U and ^{239}Pu. For the interrogation of large sea-going cargo containers filled with hydrogenous materials, β-delayed γ rays can offer an increase in sensitivity for the detection of these and other fissile materials by as much as 10^3–10^4 compared to the detection of β-delayed neutrons.

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Since September 11, 2001, an increased urgency has been associated with the development of new and improved means for the detection and prevention of the clandestine transport of nuclear weapons materials and other materials for producing weapons of mass destruction. A particularly difficult problem is posed by highly enriched ^{235}U and ^{239}Pu that might be hidden in large sea-going cargo containers, which may be filled with masses approaching 27 metric tons and which might represent areal densities of more than 50 g cm^{-2} through which an identifying signal must penetrate to reach a detector. Passive detection methods [1] based on measurements of neutrons and/or photons are either inapplicable or impractical in many such cases. Traditional methods of radiography are unlikely to provide a unique signature of highly enriched ^{235}U or ^{239}Pu. Active interrogation with neutrons or high-energy photons in a variety of forms [2,3] currently depends upon the observation of β-delayed neutrons following

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induced fission to provide a unique signature for fissile material. However, the shielding provided by a thick hydrogenous cargo could be so large that this method will fail or will have a very low detection sensitivity.

In this letter we describe a method that provides unequivocal signatures of ^{235}U and ^{239}Pu that can provide high sensitivity in the presence of thick hydrogenous and other cargos. Our method is based on the relatively high intensity of γ rays with $E_\gamma \geqslant 3.0$ MeV that are emitted from short-lived fission fragments [4,5]. These β-delayed γ rays have yields that are roughly an order of magnitude larger than the corresponding β-delayed neutron intensities from the thermal fission of ^{235}U and ^{239}Pu. They are likely to be transmitted through thick hydrogenous material with 10^2–10^3 times the probability likely for β-delayed neutrons and thus offer an increase of as much as 10^3–10^4 times the sensitivity that can be achieved with delayed neutrons. Their energies lie above interferences from normal environmental radioactivity. Finally, the energy spectra and time dependencies for emission of the β-delayed γ rays provide unique signatures for ^{235}U and ^{239}Pu.

To demonstrate the main properties of the high-energy delayed γ rays, we studied the γ-ray spectra following thermal neutron-induced fission of ^{235}U and ^{239}Pu. Neutrons were produced by bombarding a 1-in. thick water-cooled Be target with 16-MeV deuterons from the Lawrence Berkeley National Laboratory's 88-in. Cyclotron. Neutrons were then moderated using a 15 cm cube of steel surrounded by up to 45 cm of polyethylene. The steel cube was located immediately downstream of the Be target. A pneumatic transfer system shuttled targets between an irradiation location inside the polyethylene and a remote shielded counting station with a transit time of 2–3 s. The thermal neutron flux at the irradiation site was about 1.5×10^6 cm^{-2} s^{-1}. We irradiated ^{235}U (93% isotopic content), ^{239}Pu (95% isotopic content), and, as representative of the characteristics of some cargo loadings, wood, polyethylene, aluminum, sandstone, and steel. In each case, targets were repeatedly subjected to cycles of 30-s irradiations followed by 30-s counting periods, during which 10 sequential 3.0-s γ-ray spectra were acquired. Counting began 3 s after the end of irradiation. γ rays were detected with an 80% relative efficiency coaxial HPGe detector and with a 30-cm × 30-cm × 10-cm plastic scintillator. Data were acquired and sorted using ORTEC PC-based electronics and software.

Fig. 1 shows γ-ray spectra for $E \geqslant 1.0$ MeV acquired with the HPGe detector from irradiation of 0.568 g of ^{239}Pu and 115 g of steel. The temporal behavior of detected high-energy events is shown in the inset. Both the energy and temporal distributions of the high-energy γ rays from thermal fission of ^{235}U are quite similar to those shown for ^{239}Pu but their intensity per fission is about a factor of 3 larger. Also, results similar to those shown for steel were found from the irradiation of wood, polyethylene, aluminum and sandstone in terms of the most important characteristic, i.e., no spectrum indicated the presence of γ rays with energies exceeding 3.0 MeV. From the steel target, we observed a small number of lower-energy γ rays produced by the decays of much longer-lived nuclides such as ^{56}Mn ($t_{1/2} =$ 2.58 h). To the contrary, the spectrum from ^{239}Pu is indicative of a fairly intense γ-ray emission at $E \geqslant 3.0$ MeV that extends to at least 5.5–6.0 MeV.

It is also clear, as expected [4], that the high-energy intensity is spread over a relatively large number of lines rather than concentrated in only a few. Thus, a simple and sensitive method to identify fissile material is to integrate the total number of events in a wide energy interval, regardless of whether the events represent full- or partial-energy depositions. The results from this type of analysis for the energy intervals 3–4 MeV and 4–8 MeV are shown in the inset of Fig. 1. The integrated numbers of events from irradiated ^{235}U and ^{239}Pu decay with a short effective half-life of approximately 25 s, whereas those from all other materials tested showed much longer decay times. The two features—large numbers of γ rays with energies above 3.0 MeV and a short effective half-life—are unique signatures of ^{235}U and ^{239}Pu.

Because of the high density of γ-ray lines produced by the decay of fission fragments, a practical system for interrogating large objects does not require high-resolution detectors. In fact, we obtained essentially the same results shown

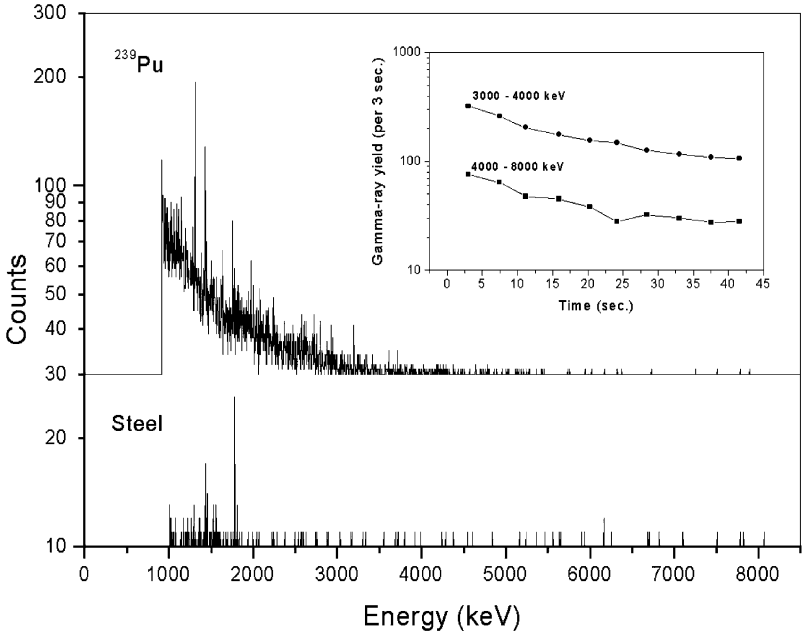

Fig. 1. γ-ray spectra observed in the HPGe detector in 30 s of live time following the neutron irradiation of 0.568 g of ^{239}Pu and of 115 g of steel. In order to display these two spectra on the same plot, offsets of 30 and 10 counts per channel were added to the data obtained from the ^{239}Pu and steel targets, respectively. Inset: Background-corrected decay curves for gamma rays in the energy intervals 3000–4000 keV and 4000–8000 keV observed from the ^{239}Pu target. Similar results were obtained from a ^{235}U target.

in the inset of Fig. 1 with the low-resolution plastic scintillator described above. This may be particularly significant because such scintillators are sufficiently low cost such that one can conceive of a large array of such devices surrounding a cargo container to provide a large solid angle for detecting photons. To demonstrate that such a system might yield practical results in reasonable times, we have estimated the response of such an array following a 30-s irradiation of a cargo container with a source producing 10^{11} 14-MeV neutrons s^{-1} when the cargo is assumed to be wood with a 5-cm (radius) sphere of ^{239}Pu located at its center. We find that a 30-s count would result in about 350 *detected* γ-ray events above 3 MeV (for ^{235}U the detected events would be approximately 1000). This suggests that with the currently available technology, it is plausible that an entire cargo container could be scanned for ^{235}U and ^{239}Pu in approximately 1 min. Possible interferences from activities induced by other materials are few and can be negated substantially by an appropriate choice of the interrogating source [6]. Combined with a radiographic imaging system, the method we propose could prove attractive for rapid identification of ^{235}U and ^{239}Pu and other fissile materials in a wide range of applications.

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