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Conference on Accelerator Applications in Research and Industry

Daren R. Norman
James L. Jones
Brandon W. Blackburn
Kevin J. Haskell
James T. Johnson
Scott M. Watson
Alan W. Hunt
Randy Spaulding
Frank Harmon

August 2006

The INL is a
U.S. Department of Energy
National Laboratory
operated by
Battelle Energy Alliance

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Time-dependent delayed signatures from energetic photon interrogations

Daren R. Norman ^{a,*}, James L. Jones ^a, Brandon W. Blackburn ^a, Kevin J. Haskell ^a,
James T. Johnson ^a, Scott M. Watson ^a, Alan W. Hunt ^b,
Randy Spaulding ^b, Frank Harmon ^b

^a Idaho National Laboratory, P.O. Box 1625, Idaho Falls, ID 83415-2802, United States

^b Idaho State University, Pocatello, ID, United States

Abstract

Pulsed photonuclear interrogation environments generated by 8–24 MeV electron linac are rich with time-dependent, material-specific, radiation signatures. Nitrogen-based explosives and nuclear materials can be detected by exploiting these signatures in different delayed-time regions. Numerical and experimental results presented in this paper show the unique time and energy dependence of these signatures. It is shown that appropriate delayed-time windows are essential to acquire material-specific signatures in pulsed photonuclear assessment environments. These developments demonstrate that pulsed, high-energy, photon-inspection environments can be exploited for time-dependent, material-specific signatures through the proper operation of specialized detectors and detection methods.

Keywords: Photonuclear; Nuclear materials detection; Explosives detection; Gamma-ray spectroscopy; HPGe detector; Time-dependent signatures

1. Introduction

Pulsed photonuclear assessment (PPA) technologies are being developed by Idaho National Laboratory (INL) and the Idaho Accelerator Center (IAC) to address advanced cargo inspection and material identification needs [1–3]. The high-energy photons are generated by bremsstrahlung from 8 to 24 MeV electron linacs. For most commercial-off-the-shelf (COTS) spectroscopy systems, operating in the PPA environment is difficult. A major operational problem is the recovery from the linac's photon pulse, a very high flux of photons that last for a few microseconds and the accompanying strong radio frequency (RF) fields. Earlier PPA related research and development efforts have

provided High Purity Germanium (HPGe) gamma-ray spectroscopy detection systems capable of operating in these intense environments [4].

Use of these modified HPGe detectors shows the rich, material-specific, gamma-ray signatures generated in the PPA environment. However, during previous testing, the detection sensitivity has changed drastically, with small changes in the experimental configurations. To understand this sensitivity, both numerical models and experiments were used. The results of the models and experiments indicate that there is strong time dependence relative to the interrogating photon pulse for gamma-ray signatures. An experimentally-based graphic of the time-dependent HPGe detector spectrum is shown in Fig. 1. The time axis in Fig. 1 is relative to the photon pulse from the linac, and the energy axis has key energies identified. The high-energy portion of the spectrum dies away within a few 100 μ s after the pulse. This die away can be seen in the upper right side

* Corresponding author. Tel.: +1 208 526 3953; fax: +1 208 526 5208.
E-mail address: Daren.Norman@inl.gov (D.R. Norman).

Fig. 1. Representation of time and energy dependent response of a HPGe detector in a PPA environment with ammonium nitrate-fuel oil explosive surrogate, a dashed box highlighting the long-delayed region (4–8 ms) used for nuclear material detection.

of Fig. 1, indicated by the lack of counts following the pulse. Soon after the pulse the spectrum is dominated by neutron capture, gamma-ray (n, γ) and scatter reactions. Therefore, to detect material-specific signatures based on the (n, γ) reactions, like nitrogen-rich explosives, counting soon after the pulse is essential. Conversely, for increased detection sensitivity for radioactive nuclei and isomers, like fission fragments, it is necessary to wait for the (n, γ) and scatter reactions to die away. This long-delayed region is highlighted in Fig. 1 with a dashed line which is about 4–8 ms after the pulse. In this delayed-time region, the highest energy material-specific signature is the ^{60}Co , which was used as a constant calibration source.

2. Neutron capture gamma-ray signatures

Assessments using nitrogen-based explosives and the time-dependent nature of the characteristic neutron cap-

ture gamma-rays (n, γ) emissions were evaluated. To better understand the time dependence, a numerical model was developed using MCNPX [5]. The model predicted the time dependence of the ^{14}N (n, γ) reactions in the ammonium nitrate-fuel oil (ANFO) surrogate and the surrounding air, Fig. 2 shows the time-dependent numerical predictions for ^{14}N (n, γ) reactions from the ANFO drum and surrounding air. The results of the ANFO drum show that within the first millisecond the capture reaction rate drops about five orders of magnitude. This was the first indication of strong time dependence. The numerical predictions focused the testing on quickly recovering from the photon pulse so the ^{14}N (n, γ) reaction gamma-rays could be detected. The model also indicates that the majority of the ^{14}N (n, γ) reactions before about 500 μs are from the ANFO drum and not the surrounding air. However, after about 1 ms, the surrounding air provided more ^{14}N (n, γ) reactions than the ANFO drum. The configuration

Fig. 2. Numerical predictions using MCNPX showing the time-dependent capture gamma-ray response for 100 kg of ANFO in a 30-gal drum and the surrounding air.

Fig. 3. HPGe spectrum of a 30-gal drum of ANFO surrogate with 10.829 MeV nitrogen peak acquired between linac pulses.

modeled contained 100 kg of ANFO surrogate in a 30-gal drum. The 30-gal drum was centered in a 20 MeV linac beam at 3 m from the tungsten electron-to-photon converter, and the 3-m sphere of air surrounding the drum.

Gamma-ray spectra of the ^{14}N (n, γ) reaction were collected using a modified HPGe detector. The ANFO surrogate was moved 50 cm from the detector, which was outside of the primary beam. However, the collected gamma-ray spectra showed the predicted time dependence. Fig. 3 highlights one of the acquired spectra with the first major peak at 10.833 MeV [6], which is the full energy peak for the ^{14}N (n, γ) reaction. The full energy peak contained 12 counts, and the single escape peak contained 25 counts. Below the nitrogen peaks, in Fig. 3, is the full energy peak for iron at 9.298 MeV. The spectrum shown in Fig. 3 was collected with an INL-modified pre-amp on an 18% Princeton Gamma Tech HPGe detector. The detector was encased in 20 cm of borated polyethylene (B-Poly) and a 2.5 cm collimator, with 0.635 cm of lead and 20 cm of B-Poly over the opening of the detector. The spectrum was collected for 2000 s with the accelerator operated at 60 Hz and very low beam current.

Assessments of the time-dependent nature of several spectra were evaluated with both gated windows and list-mode acquisition systems. As predicted with the numerical model, the (n, γ) response is very time-dependent. Fig. 1 was developed using the time-dependent list mode counts from the HPGe detector. In this graphic the 1.333-MeV ^{60}Co source and the 10-MeV ^{14}N reactions and their time dependence are clearly visible. The linac was operating at 60 Hz, and this data collection was for 30 min. As both the numerical model and the experimental results show the (n, γ) reactions in the PPA environment are extremely time-dependent.

Therefore, for effective material (n, γ) signature acquisitions, recovery within 100 μs is necessary. Small changes in testing configurations can result in detector recovery times ranging from tens to hundreds of microseconds, which makes a huge difference in the ability to detect these fast die-away signatures. However, for materials-specific signatures from radioactive nuclei and isomers such as those

emitted from nuclear material fission fragments, the recovery time is not as critical, because the detection of longer-lived components requires that the (n, γ) signatures have time to die away.

3. Nuclear material fission fragment gamma-ray signatures

The real-time measurements of characteristic fission fragments in PPA environments have proven to be difficult. While it has been proposed by many that the delayed gamma-ray emissions above 3 or 4 MeV could be used to indicate the presence of nuclear materials, these measurements are made after the interrogating source is turned off [7]. As Fig. 1 shows, soon after the interrogating pulse, gamma-ray energies are well in excess of 5 MeV. Therefore, simply using the energies above 3 or 4 MeV would not work well for this PPA technique. These high-energy (n, γ) also introduce a major noise signal to the low-energy (<5 MeV) spectroscopy needed for nuclear material fission fragment detection. However, if the HPGe detector acquisitions are delayed several milliseconds after the interrogating pulse as shown by the dashed box region in Fig. 1, it becomes possible to measure the fission fragment signatures between pulses.

An INL modified HPGe detector gated to operate from 4 to 8 ms was used to obtain spectra from the nuclear-material fission-fragments. Fig. 4 shows HPGe spectra for 300 s collection time (150-s active time because of gating) during linac operations with and without nuclear material. When nuclear material was present the two spectra clearly separate below about 5 MeV. By comparing these two spectra at 2 MeV, it can be seen that the counts per channel with nuclear material is an order-of-magnitude higher than without nuclear material. In Fig. 4 there are several high-energy peaks causing the divergence of these two spectra. Additionally, a well-defined peak is highlighted between the ^{60}Co calibration source peaks and the ^{28}Al activation peak at about 1.43 MeV (channel width 6.2 eV). Fig. 5 shows an increased energy resolution of the energies between 1.15 and 1.8 MeV. With the increased energy resolution, the 1.43 MeV peak shown in Fig. 4 splits

Fig. 4. Gamma-ray spectra acquired between linac pulses from HPGe detector with and without nuclear material. The ^{94}Sr peak is highlighted, and the peak is only present with nuclear material.

Fig. 5. Higher resolution spectrum with ^{94}Sr and ^{138}Cs fission fragments identified.

into two separate peaks; one at 1.428 and another at 1.436 MeV. The fission fragments that produced these two peaks are ^{94}Sr (1.4277 MeV, 75 s $T_{1/2}$), ^{138}Cs (1.43586 MeV, 33.41 m $T_{1/2}$), and $^{138\text{m}}\text{Cs}$ (1.4360 MeV, 2.9 m $T_{1/2}$). Even in this small 650 keV section of the spectrum after only 150 s acquisition many fission fragments are identifiable. The purpose of this paper is to show the rich material-specific signatures gamma-ray emissions provided by the PPA environment and their time dependence relative to the photon pulse. Therefore, a complete list of identified fission fragments is not provided.

The nuclear material experiments were conducted with the INL Varitron (2–12 MeV linac) operated at 10 MeV, 125 Hz, and an average beam current of 3.6 μA . The experiment used a 1.2-kg depleted uranium (DU) plate of

nuclear material placed 1 m from the photon-to-electron converter, and the HPGe detector was placed 70 cm below the DU on beam centerline. The HPGe had 10–15 cm of lead collimation towards the DU and with an open back.

4. Conclusion

It has been demonstrated that modified HPGe detectors operated properly can detect material-specific gamma-ray signatures in PPA environments. To detect neutron capture gamma-ray signatures, fast recovery after the photon pulse is necessary. For detecting radioactive nuclei and isomers, longer delays after the photon pulse are needed. Even though PPA environments are rich with material-specific signatures, existing COTS detection systems and

photonuclear sources have not been developed to exploit these signatures. As these detection technologies develop they could complement high-energy cargo inspection systems.

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