INL/CON-06-11167
PREPRINT

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Conference on Accelerator Applications in Research and Industry

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August 2006

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PACS CODES: 25.20.-x, 32/80.-t

Abstract Reference ID#: 319

Abstract

The Pulsed Photonuclear Assessment (PPA) technique, which has demonstrated the ability to detect shielded nuclear material, is based on utilizing delayed neutrons and photons between accelerator pulses. While most active interrogation systems have focused on delayed neutron and gamma-ray signatures, there is an increasing need to bring faster detection and acquisition capabilities to field inspection applications. This push for decreased interrogation times, increased sensitivity, and mitigation of false positives requires that detection systems take advantage of all available information. Collaborative research between Idaho National Lab (INL), Idaho State University's Idaho Accelerator Center (IAC), Los Alamos National Laboratory (LANL), and Oak Ridge National Laboratory (ORNL), has focused on exploiting actively-induced, prompt radiation signatures from nuclear material within a pulsed photonuclear environment. To date, these prompt emissions have not been effectively exploited due to difficulties in detection and signal processing inherent in the prompt regime as well as an overall

poor understanding of the magnitude and yields of these emissions. Exploitation of prompt radiation (defined as during an accelerator pulse/photofission event and/or immediately after (< 1 μs)) has the potential to dramatically reduce interrogation times since neutron yields are more than two orders of magnitude greater than delayed emissions. Recent preliminary experiments conducted at the IAC suggest that it is indeed possible to extract prompt neutron information within a pulsed photon environment. Successful exploitation of prompt emissions is critical for the development of an improved robust, high-throughput, low target dose inspection system for detection of shielded nuclear materials.

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Key words: Active-interrogation, photonuclear, prompt emissions, nonproliferation

INTRODUCTION

For more than a decade, Idaho National Laboratory (INL), in collaboration with the Idaho Accelerator Center (IAC) and Los Alamos National Laboratory (LANL), has been developing high-energy photon interrogation systems. Through funding from the Department of Energy's Office of Nonproliferation and National Security (NA22) and, most recently, the Department of Homeland Security's (DHS) Domestic Nuclear Detection Office (DNDO), this research and development has focused on the detection of nuclear materials, especially highly enriched uranium (HEU), within various shielding configurations such as cargo containers [1-5]. The current prototype nuclear material detection system based on the Pulsed Photonuclear Assessment (PPA) technique is illustrated in Figure 1. The primary components of this system are a specially designed 2-12 MeV Varitron accelerator and an array of custom-built and commercially-available Photonuclear Neutron Detectors (PND) [3] as well as co-located, unshielded Geiger-Müller (GM) gamma-ray detectors.

The current PPA detection system utilizes delayed emission from photonuclear reactions in nuclear materials. Interrogation is accomplished by stimulating materials with a bremsstrahlung photon spectrum produced by a 2-12 MeV Varitron accelerator that is nominally operated at an endpoint energy of 10 MeV. Photonuclear interactions within elements such as ^{235}U result in delayed neutron and photon emissions. These emissions are collected by the arrayed detectors and processed to determine the presence of nuclear materials. Delayed neutrons, however, constitutes only a very small fraction of the emissions produced during photonuclear reactions. The magnitude of prompt neutrons produced during the interrogating pulse of photons or shortly thereafter, can be orders of magnitude greater than the delayed emissions. There is strong evidence that these actively-produced prompt radiations can have distinguishable characteristics

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to facilitate nuclear material detection and identification. This approach offers the potential to dramatically reduce interrogation times since, increase inspection throughputs, and limit the dose to cargo. This paper highlights the joint research being conducted by the INL, ORNL, IAC and LANL in support of this effort.

UTILIZATION OF PROMPT RADIATION

Active non-intrusive interrogation technologies, such as the PPA system, apply an external source of radiation to generate characteristic emissions or absorptions from various materials of interest within an inspected object. While many interrogating sources have been proposed, the focus for most technologies has been charge-less radiations (specifically gamma-rays, x-rays, and neutrons) due to the high penetration capability into and out of shielded configurations. The PPA interrogation system is currently based on a bremsstrahlung photon source having an endpoint energy of 10 MeV. Pulses are normally 1-3 μs in length with a repetition rate of 60-125 Hz. Data suggests, however, that the efficacy of the photonuclear technique could be improved by increasing the endpoint energies in excess of 20 MeV.[6]

If a high-energy photon beam is utilized in an inspection system to interrogate cargo, the emitted radiation will be in the form of both prompt and delayed neutrons and photons if nuclear material present. The fission process, whether induced by photons or neutrons, immediately liberates energetic gammas and neutrons within $<10^{-14}$ seconds after interaction with the stimulating source radiation. Prompt emissions also occur between interrogating pulses from fissions induced by delayed neutrons slowing down to thermal energies. These emissions are produced with a spectrum of energies but in discrete numbers: typically, 0-6 neutrons, 0-20 photons [7]. On average, however, roughly prompt 2.5 neutrons and 7 prompt photons are emitted from the thermal neutron fission of ^{235}U. Yields from photofissions of materials such as

^{235}U and ^{239}Pu are of similar magnitudes. Delayed neutrons and photons will also be emitted from the neutron rich daughters and unstable fission products. Delayed neutrons are emitted on timescales of milliseconds to several seconds after the fission process but in much less abundance than the prompts. On average, only 0.0158 delayed neutrons are emitted from a thermal ^{235}U fission; yet at higher interrogation energies, the ratio of prompt-to-delayed neutrons can be even greater. Figure 2 illustrates the ratio of prompt to delayed neutron emissions for (γ,f) reactions at various gamma energies in selected materials of interest.

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There is a wide range of prompt emissions that could possibly be exploited to detect nuclear material including:

- Prompt gamma/x-rays: These include x-ray fluorescence (XRF, ~ 10-100 keV), Compton scattering (~ 100-500 keV), positron annihilation (511 keV), and nuclear resonance fluorescence (NRF, ~ 100s of keV), fission gamma-rays (<10 MeV).
- Prompt Neutrons: These are neutrons that are produced directly from the "flash" event, such as (γ, n) or (γ, f) absorption reactions and are material dependent. Because of the slow speed of neutrons (relative to photons) they can be observed between accelerator beam pulses and spectroscopically analyzed by time-of-flight (TOF) and/or pulse height techniques.

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Through the exploitation of both promptly emitted gamma rays and prompt neutron emissions, which are orders-of-magnitude greater than delayed neutron emission rates (originating long after the prompt effects have subsided), it is possible that the current PPA system could be significantly enhanced so that increased inspection throughput rates, improved detection sensitivities, and decreased delivered doses during an inspection can be realized. In addition, there is nothing within the utilization of prompt emissions which would preclude the

use of other types of active inspection technologies (i.e., x-ray radiography and delayed radiation measurements). The benefits of using these prompt emissions are clearly evident; however, the utilization of prompt emissions is not without significant challenges.

CHALLENGES RELATED TO UTILIZING PROMPT EMISSIONS

As noted earlier, the challenges of prompt radiation detection primarily arise from the interactions with the interrogating radiation. These include induced prompt background effects from non-nuclear materials such as (γ,n) reactions in lead, high count-rate effects, detector "recovery" effects associated with the intense interrogating radiation pulse, and the difficulties of "fast" spectroscopy associated with these prompt emissions. The primary difficulty in successfully performing prompt radiation measurements is that the fraction of emitted radiation is usually small compared to the magnitude of the interrogating photon beam. During the early development of the PPA system, INL attempted to detect and utilize gamma-ray emissions occurring within 1 μs of the flash. This early study included the response characterization of several very fast scintillators in the pulsed environment. Initial results indicated that for gamma-ray counting, a BC-418 (with a gated Hamamatsu photomultiplier tube) was a leading candidate for this type of application. However, because of the magnitude of the interrogating flash, which can be in excess of 10^{12} photons per pulse, successful utilization of the plastics to detect prompt emissions was not realized. This was primarily due to photomultiplier tube (PMT) saturation and pulse pile-up effects. As a result of these tests, focus was given to utilization of the delayed emissions for nuclear material identification. Fortunately, since this initial study, there have been a number of technological advances which have enabled the renewed investigation into prompt emission utilization.

DIGITAL ACQUISITION AND PROCESSING

State-of-the-art digital acquisition

Historically, the bulk of nuclear measurement techniques have relied on analog collection and processing techniques, which can be susceptible to high, count rate effects and pulse pile-up errors. In an analog system, once a signal is detected and processed, the information in the signal is forever lost and no possibility exists for post processing. Digital processing and fast digital analysis systems, initially developed for high energy physics studies, have progressed to the point that nanosecond (and in some cases sub-nanosecond) timing can be employed to acquire and process detector signals. Current state-of-the-art digitizers are capable of resolving input signals with 10-16 bits of accuracy at timescales down to about 0.1ns. Unlike fast digital oscilloscopes, these digitizers can collect and transfer data to a host PC at rates of up to several hundred megabytes (MB) per second. Digitizer models such as the Acqiris DC282 offer synchronous four channel sampling at up to 2 GS/s per channel, or interleaved dual- or single-channel sampling at up to 4 and 8 GS/s respectively. Programmable front-end electronics are used to provide a complete set of input voltage ranges, from 50 mV to 5 V full scale. With a bandwidth of 2 GHz, amplifier response can be optimized so that fidelity is ensured even with high count rate and high-frequency measurements. Through the utilization of this state-of-the-art digitizer combined with unique data collection and processing techniques, efforts are underway to utilize a number of novel detector types and collection systems to exploit information within the prompt regime.

Silicon Carbide (SiC) Detectors

Silicon Carbide (SiC) detectors are a relatively new class of radiation detectors. Developed by Westinghouse for in-core radiation measurements, they are composed doped layers of SiC

sandwiched between two metal contacts. Depending on the applied voltage, depletion regions of 10-100 μm can be induced in the detectors with band gaps on the order of 3.25 eV. Incident radiation generates several electron-hole pairs that are pulled apart under an applied electrical field, generating a current signal that is then amplified and recorded. Advantage of SiC detectors include: ultra-fast (sub-nanosecond risetimes) timing, capable of processing high count rates, can operate during and within nanoseconds of the end of an intense neutron pulse, and photon insensitivity [8].

Lanthanum Bromide (LaBr3) Scintillation Detector

Recent advances in scintillator material have resulted in the development of $LaBr_3$ detectors. These detectors offer improved energy resolution when operating at room temperature, fast emission and excellent temperature and linearity characteristics. Typical energy resolution at 662 keV is 3% as compared to sodium iodide detectors at 7%. The improved resolution is due to a photoelectron yield which is 160% greater than that achieved with sodium iodide. Another advantage of $LaBr_3$ is the nearly flat photo emission over a 70 degree C temperature range (~1% change in light output). The improvements in resolution allow more accurate peak discrimination in ranges where isotopes often have many overlapping peaks. This leads to better isotope detection classification. Experiments to utilize the spectroscopic abilities of the fast $LaBr_3$ crystals in the PPA environment are imminent.

Nuclear Materials Identification System (NMIS)

Developed by ORNL, NMIS is a 10 channel, 1 GHz sampling and processing system that measures the time distribution of coincidences between all pairs and specified triplets of input channels, and also the number of detector pulses that occur within a time window following a initial pulse. It is basically a 1 GHz shift register that samples the leading edge of input pulses to

time detector pulses and calculates the time distribution of pulses in a channel with respect to pulses in the same and other channels. The time dependent coincidences measure a fissile object's response to active interrogation (the interrogating radiation source is one input), or a naturally radioactive object's response to its own radiation. The value of the time dependent response is that it is a function of the object's neutron multiplication and leakage. The system's nanosecond timing precision makes it suitable for measurements of fissile metal (unmoderated) objects. For moderated objects the system can capture hundreds of microseconds of continuous data with nanosecond resolution. The dead time in any one of the input channels is 8 nanoseconds. The system can process and stay live for input rates up to 10^7 counts per second (total across all channels). The processor board is deeply buffered to handle the occasional large fission chain fluctuation. Acquisition stops cleanly on a data block boundary if the buffer becomes nearly full and continues automatically when the buffer is nearly empty again; a "time gap" message is inserted into the data stream so that the processor does not attempt time correlation of pulses across the gap.

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EXPERIMENTAL RESULTS

Although this research is at a preliminary stage, there have been some tantalizing results which indicate that these novel detectors, when utilized with a fast digital acquisition and processing system, can not only survive in the prompt regime, but can extract and process information regarding the presence of various materials of interest. In measurements conducted at the Inspection Technology Research and Development Laboratory (ITRDL) facility at IAC, a SiC detector was operated in an unshielded configuration in close proximity to the photon converter in order to look at prompt (γ,n) radiations coming from the lead collimator of the Varitron accelerator. Events were recorded only during the photon flash which is nominally

composed of 10^{12} gamma-rays per 3-μs pulse from the Varitron accelerator operating at 125 Hz with an endpoint energy of 10 MeV. The SiC detector was placed along the beam axis at a distance of one meter from the photon converter. The SiC was biased at -900V and was processed by using a VT120 fast preamplifier leading directly to an Acqiris digitizer, which was sampling at a 2Gsample/sec rate. The digitizer triggered on each accelerator pulse and recorded pulses over slightly greater than the 3 μsec burst. Typical traces are shown in Figure 3. In the top trace, large RF transients (separated by 3μsec) are visible at the beginning and near the end of the trace. These transients were used to define the starting and end of the accelerator pulse. The bottom two plots show expanded time-scale views. The large pulses (>10mV (y-axis)) have a rise time of about 2ns and a FWHM of 4-5 ns. This shape closely corresponds to pulse shapes from neutron pulses taken with a ^{252}Cf source. A preliminary estimate of the time-of-flight characteristics of these events was made. Figure 4 shows the distribution of times to detection following a total of 1000 accelerator pulses. The counts start appearing at times of ≈60-120ns which is consistent with the 1m transit time of ≈0.5-1 MeV neutrons. This 0.5-1 MeV average neutron energy is typical for a 10 MeV-endpoint energy bremsstrahlung beam interacting with lead in the collimator. The counts at smaller times are likely from noise although higher energy neutrons or photons can not be ruled out.

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In a related experiment, tests were conducted again utilizing the Variton accelerator, although in this test, various materials were located within the INL Wood Calibration Pallet and prompt signatures were monitored with a fast EJ-200 (BC-408 equivalent) plastic scintillator. The pallet was located 2.5 m from the photon converter and was positioned so that the central void was located along the photon beam axis. The scintillator was located at a 45^{o} angle to the beam axis and was positioned 2.85 m behind the edge of the pallet. Three separate tests were conducted

with the pallet using (1) no material (2) bismuth (3) depleted uranium (DU). Detected signals from the scintillator were processed only if they fell within the 3 μs of the accelerator pulse. Figure 5 illustrates both the raw and integrated counts for 1000 pulses of the accelerator. This detector is sensitive to both neutrons and photons although the light output for neutrons is much less than for photons of equal energy. In the pulse height spectra it appears that the scintillator is successful at detecting the prompt fission neutrons from the DU as well as the (γ,n) neutrons from the bismuth. For the bias voltage used on the PMT in the experiment, these neutron signals will all be under 100 mV. At large pulse heights, the signal is due primarily to high energy photons. The absence of any high-Z material in the pallet would allow for a large fraction of the beam to penetrate through and scatter into the scintillator. By simply integrating the counts, the signals from the DU (primarily because of the prompt neutrons) can be differentiated from other non-nuclear materials and from cases where material is located within the pallet. It should be remembered that it both the SiC case and in the test with the scintillator, these results were obtained over an acquisition period of only 3 ms.

CONCLUSIONS

The utilization of prompt radiation signatures in the PPA environment have the potential to greatly enhance the detection and identification of shielded nuclear material. By drawing on state-of-the-art technology such as novel detectors and fast digital acquisition systems, the joint team of INL, IAC, ORNL and LANL will continue to develop the techniques and processes to exploit the abundant information available during and shortly after a pulse from an interrogating photon beam. Preliminary measurements and simulations conducted at the respective facilities have shown great potential for utilizing prompt radiation emissions for the detection of nuclear

material. The use of prompt signatures would allow for increased throughput, decreased inspection times and lowered delivered dose to the interrogated cargo.

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Figure 1. The transportable PPA interrogation system including the Varitron accelerator along with custom neutron and gamma detectors.

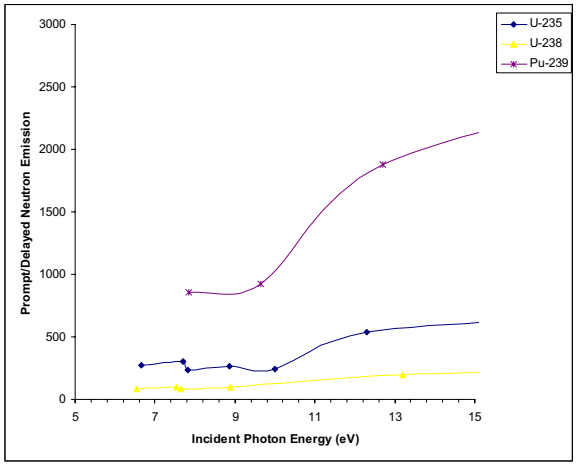

Figure 2. Ratio of prompt to delayed neutrons at various interrogating photon energies.

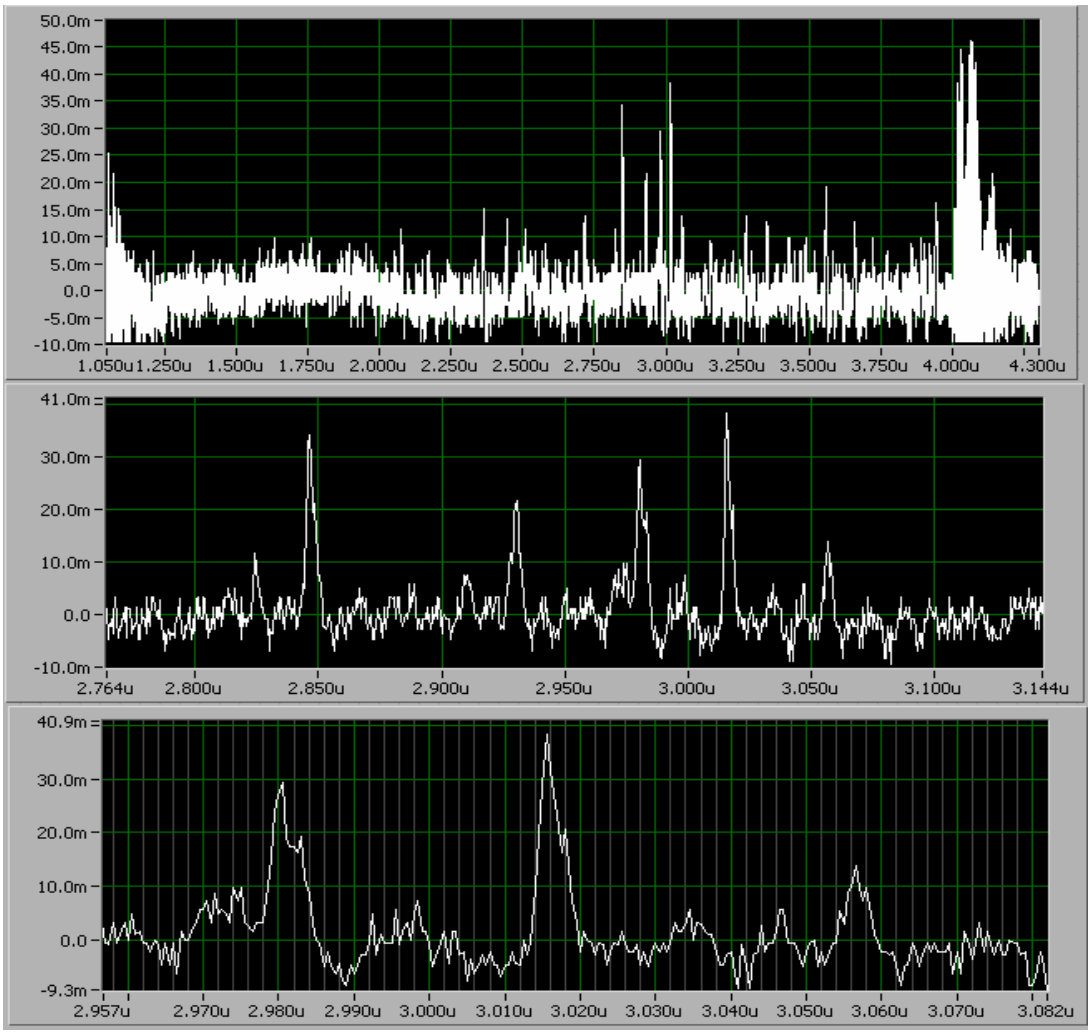

Figure 3. Signals acquired during the flash of the Variton accelerator using a SiC detector.

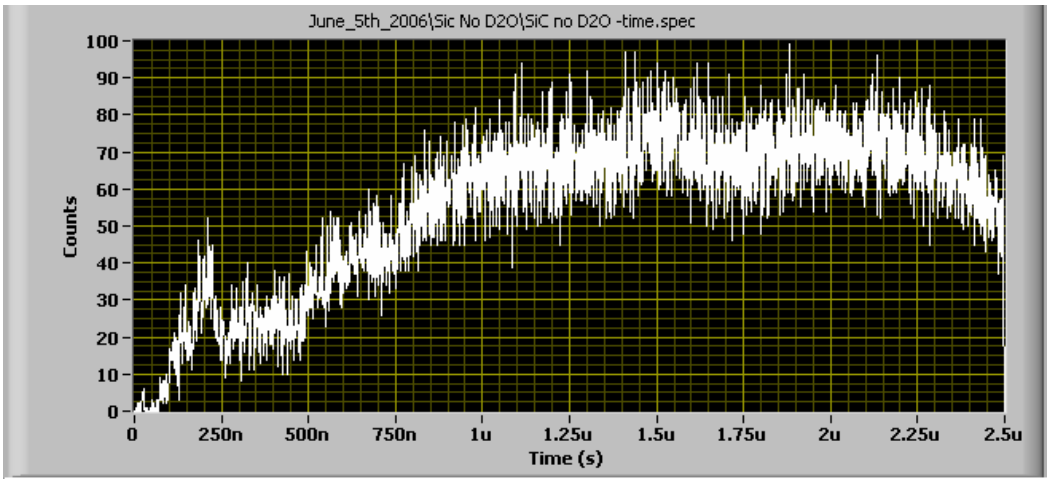

Figure 4. Time-of-flight measurements of prompt (γ,n) neutrons acquired with a SiC detector.

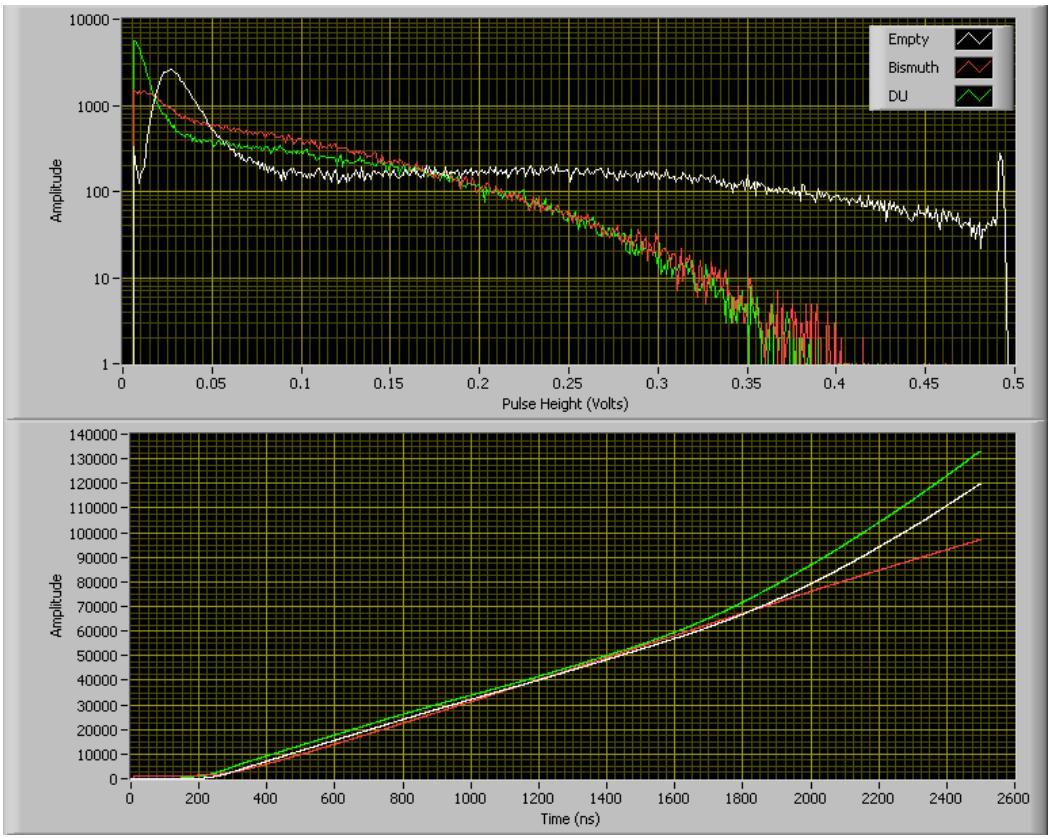

Figure 5. Pulse height spectra and total integrated counts from a plastic scintillator tested with various shielded materials in the wood INL Calibration Pallet.