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Gamma/neutron time-correlation for special nuclear material detection – Active stimulation of highly enriched uranium

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ABSTRACT

The time-correlated pulse-height technique can distinguish multiplying (special nuclear material) from non-multiplying sources. The technique relies upon the measurement of correlated photon–neutron pairs using organic liquid scintillation detectors. For such interactions, the distribution of measured neutron recoil energy versus the time-of-flight difference between correlated photons and neutrons are imprinted with the fission chain dynamics of the source. The theoretical time-of-arrival assuming the photons and neutrons are created in the same fission is calculated. Correlated pairs with longer time-of-arrival indicate delays caused by self-induced fission chains in a multiplying source. For the specific circumstances of simulated measurements of 25.4 kg of highly enriched uranium at 50 cm source to detector distance, correlated pairs from fission chains can arrive upwards of 40 ns later than correlated pairs with the same neutron energies from non-multiplying sources like ^{252}Cf at the same source detector distance. The use of detectors with ns scale time resolution and the use of pulse digitization allows for the distinction of these events.

This method has been used successfully in the past to measure a variety of plutonium-bearing samples. The particle transport code MCNPX-PoliMi has been used to simulate and validate these measurements as well. Due to the much lower signature emission rate of ^{235}U, this technique has not yet been used to measure the presence of highly enriched uranium. In this work we therefore explore the use of the time-correlated pulse-height technique with the introduction of an interrogating neutron source to stimulate fission. The applicability of ^{252}Cf, AmLi and a DD generator neutron sources is explored in a series of simulations. All three sources are viable options with their own pros and cons with the choice of appropriate source depending upon the intended application. The TCPH technique is envisioned as a viable measurement solution of special nuclear material in situations in which the presence of shielding material disqualifies the use of passive gamma spectroscopy or gamma spectroscopy reveals classified information on the special nuclear material's isotopic composition.

1. Introduction

1.1. Motivation

Many missions rely upon the detection and characterization of special nuclear material (SNM), including weapons grade plutonium (WGPu) and highly enriched uranium (HEU). Over time, the nature of these missions, each with unique detection requirements and intrinsic limitations, has changed repeatedly. During the Cold War, the large throughput of SNM at nuclear-weapons production facilities spurred the development of active interrogation techniques needed for criticality safety studies and monitoring at these

Abbreviations: BeRP, beryllium reflected plutonium; HDPE, high density polyethylene; HEU, highly enriched uranium; LINAC, linear accelerator; MOX, mixed oxide; NNSS, Nevada National Security Site; NORM, naturally occurring radioactive material; RPM, radiation portal monitor; SNM, special nuclear material; TCPH, time-correlated pulse height; TTAL, theoretical time of arrival line; WGPu, weapons-grade plutonium.

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facilities. These same techniques are now being applied in stockpile stewardship and arms control verification (Mihalczo et al., 2000). More recently, the interdiction of smuggled SNM in association with terrorism has gained heightened attention. Yet even within this narrower detection field, methods vary widely from active interrogation of cargo containers (Hall et al., 2007), to passive radiation portal monitors (RPM) at border crossings (Kouzes et al., 2008), to comprehensive mobile toolboxes for maritime law enforcement agencies (Cester et al., 2012).

Though certainly not an exhaustive list, closer study of these applications shows that every mission has unique constraints on the sources used (for active interrogation), the signatures analyzed, the effects of natural and induced background, the presence of shielding material and thus the ultimate design of the detection system (Runkle et al., 2012). For instance, measuring the prompt or delayed signature emitted by SNM actively interrogated with a pulsed source, such as a linear accelerator (LINAC), might produce excellent measurement results (Blackburn et al., 2007; Mullens et al., 2005), but the portability and affordability, among other considerations, could disqualify such a detection system for many SNM measurement missions.

Passive gamma spectroscopy coupled with a simple neutron detector, such as ^{3}He, is the foundation of most RPM systems. While WGPu's high neutron emission rate, primarily due to ^{240}Pu, as well as many strong gamma emission lines both offer prominent detection signatures, widespread agreement exists that passive detection of HEU presents a significant challenge (Kouzes et al., 2008; Cester et al., 2012). HEU's neutron emission rate is many orders of magnitude lower than that of WGPu, and HEU's low energy gamma lines (in particular the 186 keV line associated with ^{235}U) are easily shielded by any dense high-Z material. The attenuation coefficient representing the probability per unit distance of travel that a gamma photon undergoes any significant interaction in a material can be expressed independent of material density by the mass interaction coefficient, $\frac{\mu}{\rho}$. For lead, a typical gamma shielding material, the mass interaction coefficient for the 186 keV ^{235}U line is given by $\frac{\mu}{\rho} \simeq 1.2\ \frac{\text{cm}^2}{\text{g}}$. Given the natural density of lead of 11.34 $\frac{\text{g}}{\text{cm}^3}$, less than a quarter of 186 keV gamma lines would travel through a 1 mm sheet of lead without interacting (Shultis and Faw, 2000). Passive gamma spectroscopy relies upon measuring a statistically significant number of un-collided, full energy gamma photons. This means that only modest amounts of shielding are required to render passive gamma spectroscopy of HEU moot and that self-shielding limits detectable gamma emissions to a thin outer shell of SNM (Cester et al., 2012).

The presence of naturally occurring radioactive material (NORM) complicates any measurement system based upon particle counting, such as RPM systems. Cargo containing NORM and nuclear medicine patients must not trigger frequent false alarms by the gamma detection system. Although the natural neutron background is significantly lower than the natural gamma background, the natural neutron background can fluctuate significantly with location, time of day, the "ship effect" (increase of neutron background in presence of large quantity of high atomic number Z material leading to increased production of cosmic ray-induced spallation neutrons (Kouzes et al., 2008), among many other factors (Kouzes et al., 2008). Thus measurement systems are generally custom-designed for a specific mission and the myriad considerations and limitations that the mission entails and imposes.

1.2. New measurement technique and past work

The University of Michigan in collaboration with Sandia National Laboratories has developed a new measurement technique for distinguishing multiplying from non-multiplying sources and measuring subcritical multiplication. The spontaneous and induced fission reactions of certain isotopes in SNM emit temporally correlated photons and neutrons. Specifically, this measurement technique strides to detect the spontaneous fission of ^{240}Pu and the induced fission of ^{235}U and ^{239}Pu. Organic liquid scintillation detectors' fast timing and pulse shape discrimination capability provide the necessary detection and distinction of emitted photons and neutrons (Kaplan et al., 2013; Enqvist et al., 2013). These detector properties are crucial, because the time-correlated pulse-height (TCPH) technique relies upon the measurement of correlated photon–neutron (p–n) pairs emitted from fissions.

A measurement system consisting of four 7.62 cm diameter by 7.62 cm length cylindrical EJ-309 organic liquid scintillation detectors was utilized in this work and has a time resolution of approximately 1 ns (ELJEN Technology, xxxx). Pulses are acquired and digitized with a CAEN DT5720 portable self-contained digitizer that has a 2 V dynamic range, 250 MHz sampling rate and transfers data to a PC via USB at rates up to a maximum of 35 MB/s (CAEN, xxxx). Compared to systems using gas filled proportional counters, like ^{3}He, and shift register logic for measurements of multiplying sources, such as an active well coincidence counter, organic liquid scintillation detectors, in general, and this TCPH measurement system exhibit negligible dead time effects. Digitized pulses from these detectors are typically 20 ns in width. A p–n pair is classified as photon detection followed by the detection of a neutron within a 100-ns window. Thus one limiting factor for the system is concerned with high flux measurements in which more than ten million counts per second are recorded. Digitized pulses would occur close enough to each other in time within that 100 ns window that pulses might not be able to be easily deconvolved. The 35 MB/s maximum transfer rate of the USB digitizer similarly sets restrictions on high flux measurements. In practice, this can be overcome by introducing shielding material to the setup or increasing the source detector distance. In both cases the data acquisition rate is lowered at the expense of longer measurement times to acquire the same amount of TCPH data. Nothing but cost would preclude using a digitizer model capable of higher data transfer rates. Raising the threshold for minimum acceptable pulse heights presents an additional tool for dealing with high count rate measurement scenarios. The threshold setting is visually discernible in all TCPH plots as the white horizontal strip between zero pulse height and the chosen threshold setting. However, for sources measured passively in the past and those simulated in this paper, count rates do not approach these theoretical limits.

Returning to the p–n pairs, the current system requires these two interactions to occur in separate detectors. However, previously mentioned limitations are not due to limitations in the TCPH method but they were instead implemented to simplify the subsequent data analysis. Only two parameters are required for each p–n pair. These are the measured energy deposition caused by the neutron interaction (in Mega-electron Volts electron equivalent energy (MeVee)) and the time difference between the photon and neutron detection (ns). In practice, the time and energy of any interaction above threshold is recorded and an offline post-processing script sorts out the relevant information for correlated p–n pairs.

A theoretical time of arrival line (TTAL) is superimposed on a surface plot of these two measured values. As shown in Fig. 1, the abscissa reflects the time-of-arrival difference between the photon and neutron, while the ordinate depicts the measured light output caused by the neutron energy deposition in the detector. The color scale depicts the rate of p–n pairs in counts per second. The superimposed TTAL represents the theoretical maximum time delay between a photon and neutron emitted from the same fission and travelling over a fixed distance (source-to-detector front face). Because the detector material possesses non-infinite stopping power, photons and fast neutrons can interact anywhere within the detector volume. However, the time spread caused by this