NOVEL BETA-GAMMA COINCIDENCE MEASUREMENTS USING PHOSWICH DETECTORS

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ABSTRACT

The Pacific Northwest National Laboratory developed an Automated Radio-xenon Sampler/Analyzer (ARSA) for the Comprehensive Nuclear-Test-Ban Treaty (CTBT) to measure four radio-xenon isotopes, ^{131m}Xe, ^{133m}Xe, ^{133}Xe, and ^{135}Xe, originally using a beta-gamma coincidence counting detector. Betas and conversion electrons are detected in a cylindrical plastic scintillation cell and gammas and x-rays are detected in a surrounding NaI(Tl) scintillation detector. This paper reports investigations into a novel method to measure beta-gamma coincidences using a phoswich detector with state-of-the-art pulse shape discrimination techniques. A thin CaF_2(Eu) and thick NaI(Tl) crystal phoswich detector, with the use of pulse shape discrimination, is able to separate gamma and beta responses in a single detector unit. A single detector allows for simplification of the hardware and calibration needed for the counting system. In addition to identifying radio-xenon, the phoswich detector has the potential for detection of other radioactive noble gas species, such as radon, via daughter gamma and direct alpha emission, allowing more information to be gleaned from a single sample. The experimental configuration and results of a phoswich beta-gamma measurement will be presented.

OBJECTIVE

Of all the fission products produced in an underground nuclear explosion, only noble gases with relatively low-interaction cross-sections are likely to escape. Noble gases like xenon are, therefore, the isotopes used to detect clandestine nuclear explosions, and are part of the Comprehensive Nuclear-Test-Ban Treaty (CTBT) detection task assigned to the International Monitoring System (IMS). The IMS has been mandated to establish a worldwide network of detector systems capable of detecting the four radioxenons: ^{131m}Xe, ^{133g}Xe, ^{133m}Xe, and ^{135g}Xe. The proposed detection systems need to be sensitive to very low concentrations of radioxenon and be automated for remote operation. These four radioxenon isotopes each have unique, though overlapping, signatures that can be exploited using beta-gamma coincidence techniques. Table 1 lists the half-life, fission yield, prominent gamma rays, x-rays, beta endpoint energy, and dominant conversion electrons (CE) for ^{131m}Xe, ^{133g}Xe, ^{133m}Xe, and ^{135}Xe (Browne and Firestone (1986), ENDF/B-VI). Pacific Northwest National Lab (PNNL) has developed an Automated Radioxenon Sampler/Analyzer (ARSA) to detect these four radioxenon fission products. The ARSA is designed to be an automated system that separates and concentrates the ambient xenon (0.087 ppm in air) from the air, and uses a beta-gamma coincidence counting system to determine the concentrations of these four radioxenon isotopes as described in detail in Reeder *et al* (1998) and Reeder and Bowyer (1998). The detection system of the ARSA consists of four separate cylindrical gas cells used on a rotational basis to provide continuous monitoring. The walls of the gas cells are made from 1.2-mm thick plastic scintillator (Bicron BC-404) and are 5-cm-long hollow cylinders with a diameter of 1.25 cm (see Figure 1) so that in addition to containing the gas, the cells are also beta detectors. The scintillating light produced in the cell walls is collected at each end of the cell through two small photomultiplier tubes (PMT). The gamma detector consists of two optically separated 5-in. by 8 in. NaI(Tl) crystals that surround the four beta cells (see Figure 2). Each NaI(Tl) crystal is viewed by two 3" sideways looking PMTs. The output signals are added in coincident circuits to reduce PMT dark noise and to select out the beta-gamma coincidence signals. For the ARSA configuration, the gas responses in each cell are viewed by at least four PMTs requiring numerous electronic circuits and configuration time to gain-match and calibrate each PMT.

In order to reduce the required amount of hardware and maintenance overhead, we have investigated an alternate system of viewing the beta-gamma coincident signals. This novel system uses a phoswich detector to investigate differences between beta, gamma, and beta-gamma coincident pulses, and to separate the beta and gamma contribution from a single beta-gamma coincidence pulse.

Table 1. Half-lives, fission yields, principal radiation, and abundances of xenon fission products.

Nuclide	^{131m}Xe	^{133m}Xe	^{133g}Xe	^{135g}Xe
Half-life	11.93 d	2.19 d	5.25 d	9.14 h
Fission yield				
Independent (%)	2.41×10^{-7}	4.23×10^{-3}	1.46×10^{-3}	1.20×10^{-1}
Cumulative (%)	4.51×10^{-2}	1.98×10^{-1}	6.72×10^{0}	6.60×10^{0}
Gamma-rays				
Energy (keV)	163.9	233.2	81.0	249.8
Abundance (%)	1.96	10.3	37.0	90.0
X-rays (K-shell)				
Energy (keV)	30.	30.	31.	31.
Abundance (%)	54.1	56.3	48.9	5.2
Beta Spectrum				
Max. Energy (keV)			346.	905.
Abundance (%)			99.	97.
CE (K-shell)				
Energy (keV)	129.	199.	45.	214.
Abundance (%)	60.7	63.1	54.1	5.7

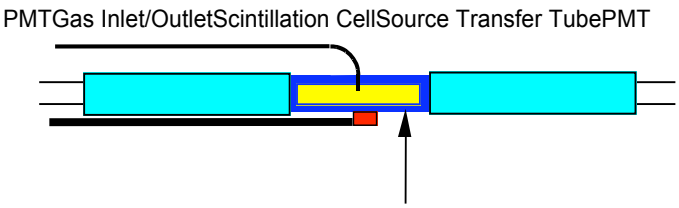

Figure 1. Schematic of the ARSA gas cell illustrating the dual PMT readout.

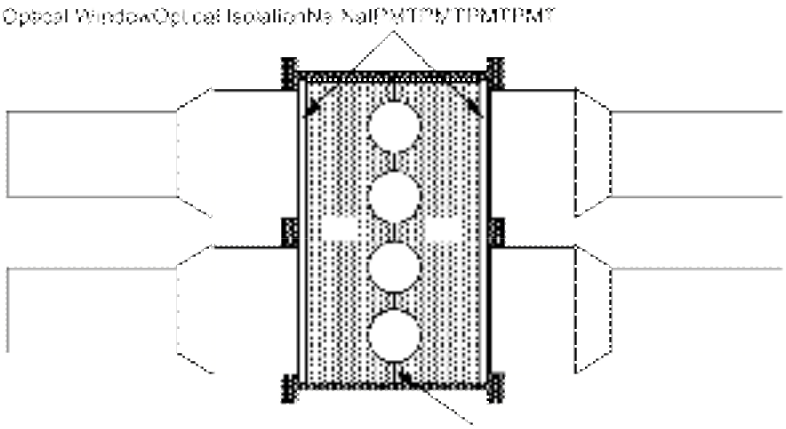

Figure 2. Schematic of the NaI(Tl) crystal of the ARSA detector.

RESEARCH ACCOMPLISHED

Phoswich Detector and Experimental Setup

The phoswich detector arose from the idea of combining two different phosphorous materials together in a sandwich that was viewed by a single light detection system. The term phosphorous sandwich was shortened to phoswich and is presently used to describe any dual scintillating material system viewed by a single light sensitive device. The two detector materials respond differently to incoming radiation and these different responses can be separated electronically. In practice, a material with a fast decay-time is used together with a slow decay-time material, both of which are read out by a single light sensitive device, such as a PMT. A pulse produced in the slow decay-time material will have a distinctively different shape than a pulse of the same amplitude produced in the fast decay-time material. The separation of the two responses is carried out in a post-processing phase. Typically integrating the pulses for a specific amount of time, and comparing the integrated pulse heights with the total pulse heights accomplish this separation. With the advent of fast digital processing, more sophisticated approaches to the separation process can be used. In addition, it may be possible to separate the gamma and beta contributions of a single beta-gamma coincident pulse. If this is possible, a single detector could be used to select individual beta-gamma pulses and perform isotopic identification based on the separated gamma and beta energies.

A phoswich detector for this investigation was custom built by Saint-Gobain Crystals and Detectors from a 2x2 inch NaI(Tl) cylindrical crystal with a 0.04-inch-thick window of calcium fluoride (CaF_2(Eu)) on one end. The NaI(Tl) crystal was separated from the CaF_2(Eu) crystal by a quartz optical window of 0.25-inch thickness as a result of the hydroscopic nature of the NaI(Tl). Pulses from a Na(Tl) crystal have typical rise-times of 250 ns (250ns decay-time of the NaI(Tl) material), while the response of the CaF_2(Eu) is somewhat slower, with typical rise-times of 940 ns. On the other end of the NaI(Tl) crystal, an ETI 9266 PMT was attached with a standard tube base mount. The crystals and PMT were packaged in an aluminum housing, which was threaded on the viewing end for the attachment of a gas cell. The gas cell was a short hollow cylinder fabricated at PNNL out of aluminum with inside dimensions of approximately 2-inch diameter by 1-inch depth. A gas-tight fitting was added on the end to allow filling of the cell. The PMT base used was an ORTEC 276 single-unit PMT base with preamp. The preamp power, PMT high voltage, and signals were connected to modules in a NIM crate. The pulses were collected and

processed by an XIA DGF4C digital pulse processor. Finally, the individual digitized pulses were sent to a computer for display and storage.

Initial testing of the detector system involved known calibration sources to ensure the normal response of the detector and to optimize the parameters of the XIA unit for resolution. The gamma radiation will, for the most part, transverse the CaF_2(Eu) window and deposit energy in the NaI(Tl) crystal that will produce a fast rising pulse in the electronic circuitry. Low-energy photons may, however, deposit some energy in the CaF_2(Eu), resulting in a slow rising pulse. The CaF_2(Eu) window is thick enough that any beta radiation should be completely deposited, forming a slow rising signal in the PMT. An example of the two distinct pulses is shown in Figure 3.

A ^{137}Cs source was used initially to view the detector response to photons, as well as to optimize the bias voltage of the PMT and the parameters of the XIA module. We observed that the resolution of the 662 keV peak from the ^{137}Cs source was maximized at a PMT bias voltage of 1,000V. Several parameters were optimized on the XIA module to ensure maximal resolution of the 662 keV peak of the ^{137}Cs source. The peaking time was adjusted to 4 μs, while the gap time was set to 2 μs. The lower-level discriminator threshold level was set to 8 (XIA units) to reduce background noise. After optimization, an energy spectrum of the ^{137}Cs source was produced (see Figure 4) that exhibits 7% resolution for the 662 keV peak.

To check the detector's response to beta particles, a ^{99}Tc source was placed directly in front of the detector with the gas cell removed. The detector responds to beta particles in a normal fashion, although the energy spectrum also contains the gamma signature from background radiation (see Figure 5). Although not performed in this investigation, the calibration sources can be used to convert the channels of energy into a common unit such as electron volts (eV).

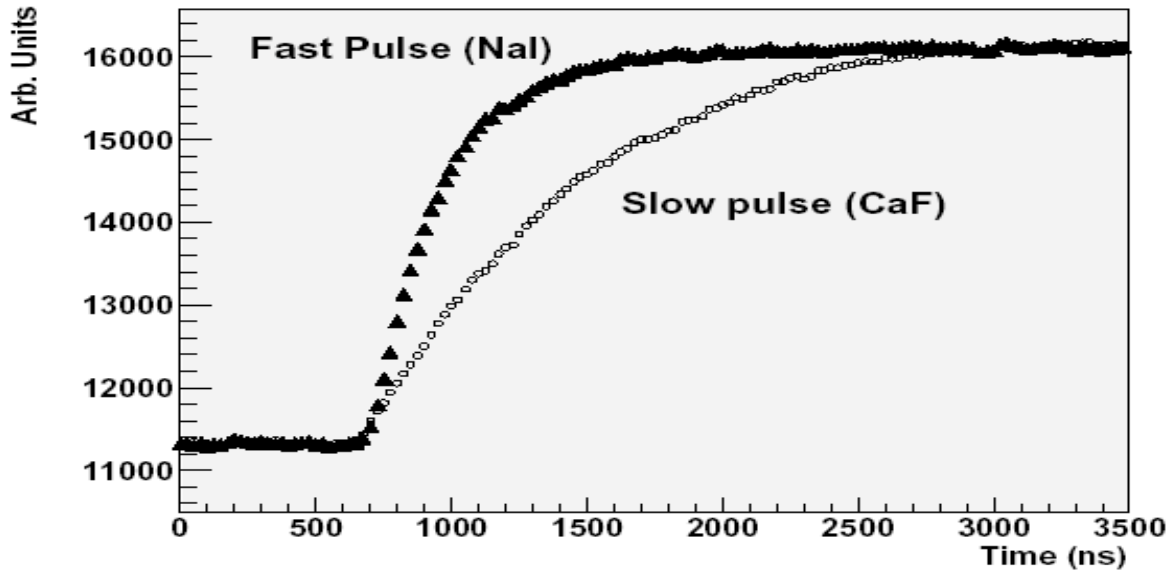

Figure 3. Example of the fast and slow rising pulses from the phoswich detector. The plot is obtained from the digitized XIA pulses.

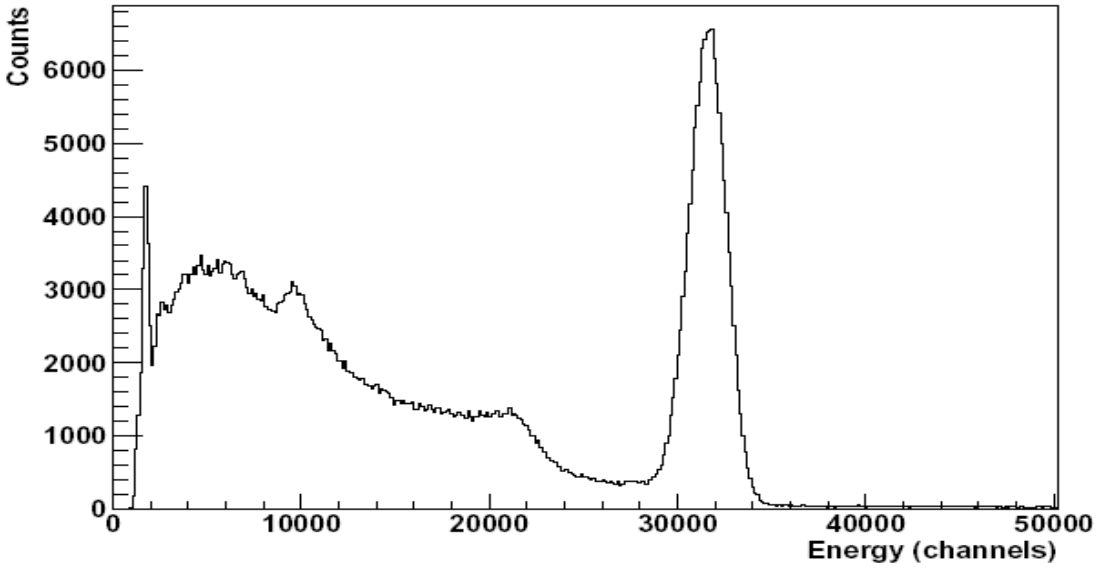

Figure 4. Energy spectrum of the phoswich detector when exposed to a ^{137}Cs calibration source.

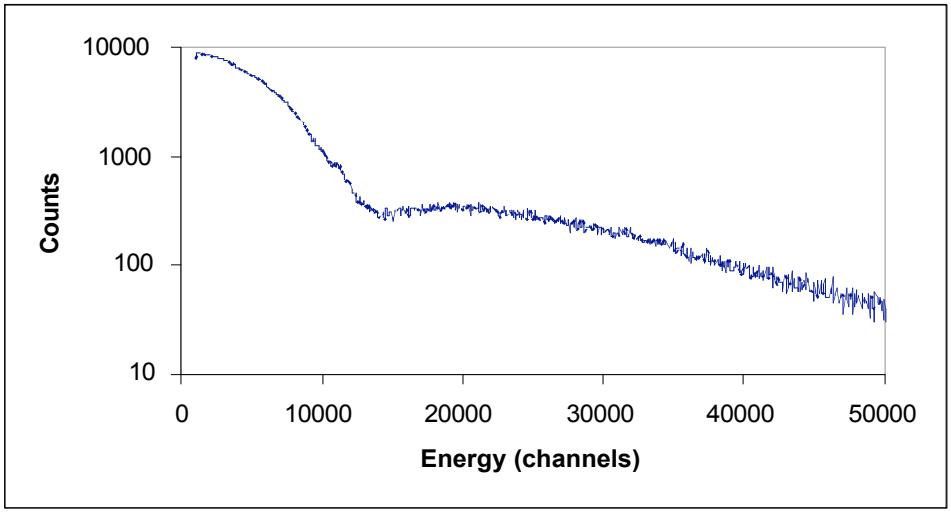

Figure 5. Energy spectrum of the phoswich detector from a ^{99}Tc calibration source. Both the beta response and gamma signature from background radiation are collected.

Xenon Analysis and Discussion

After the initial check of the detector and optimization for energy resolution, the gas cell was re-attached to the phoswich detector and xenon gas injected. Data were collected from the phoswich detector for xenon gas over a period of approximately 4 hours and yielding approximately 2.5 million pulses for analysis. The XIA module to a binary file recorded the digitized pulses. A program developed for a similar analysis was used to read and manipulate the data using the framework of the data analysis package ROOT as described in Brun and Rademakers (1997).

The data were first sorted according to the amount of time the pulses took to develop from 10% to 90% of the full pulse height, and also with respect to energy. The results are plotted in the 2-D histogram of Figure 6. The fast and slow rising pulses can be separated into two distinct distributions. An energy spectrum of pulses with rise-times less than 1,000 ns is compared to longer pulses in Figure 7, illustrating different energy distributions for

slow and fast pulses. It appears that this detector system can differentiate between pulses formed in the NaI(Tl) from pulses originating in the CaF_2(Eu) window.

As illustrated in Figure 6, there are also a significant number of pulses with rise-times in-between the two distinct distributions. In particular, there are several clusters of these pulses with rise-times in the middle region that appear as ridges. These ridges are most prominent near the fast rise-time distribution and decreasing in number toward the slow rise-time distribution. In addition, the pulses in these ridges increase in energy as the rise-time increases. This behavior is similar to what would be expected from beta-gamma coincident pulses in a phoswich detector. The gamma in a beta-gamma coincidence has a single energy, while the beta has a range of energies. Therefore, one would expect a peak in the fast rise-time distribution, with a ridge of longer rise-times and increasing energy as the variable-energy beta pulse is combined with the gamma pulse in the PMT of the phoswich detector. Also, since the beta distribution is peaked at low energy, the pulses in this 2-D plot should decrease in number as the rise-time increases. The pulses in the middle rise-time region forming the ridges of Figure 6 are promising candidates for beta-gamma coincident pulses. In order to illustrate these candidate pulses in another way, the energy distributions for the fast rise-time pulses (less than 800 ns) were compared to pulses with slightly longer rise-times (800-1200 ns) and displayed in Figure 8. In the fast rise-time energy spectrum there are three dominant peaks that are mimicked in the medium rise-time energy distribution. In addition, the peaks of the medium rise-time distribution are all at higher energy, which corresponds with the expectation of a beta response added to the gamma pulse.

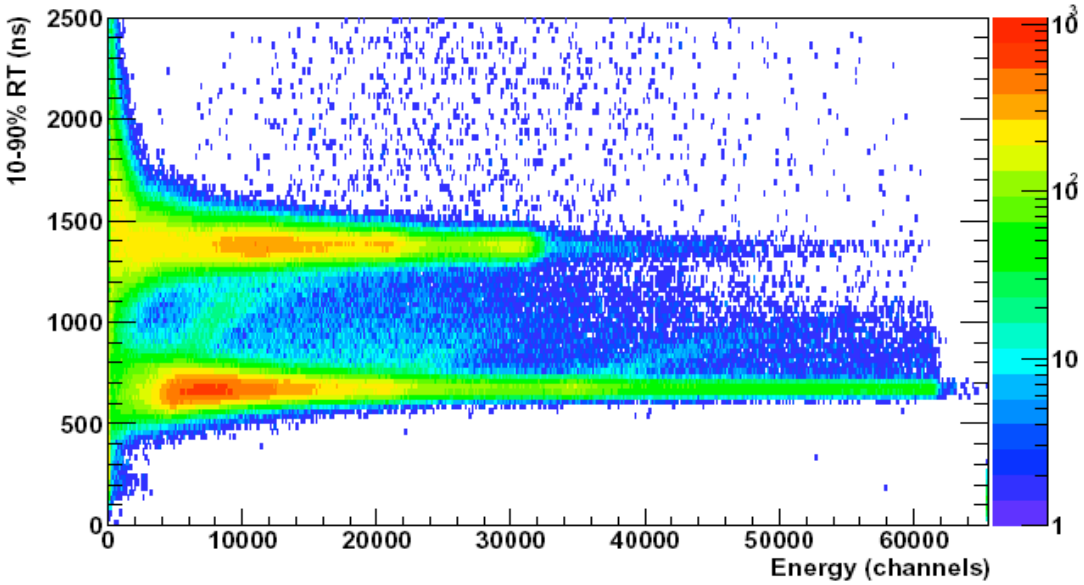

Figure 6. Rise-time (10-90%) versus energy for the xenon pulses in the phoswich detector.

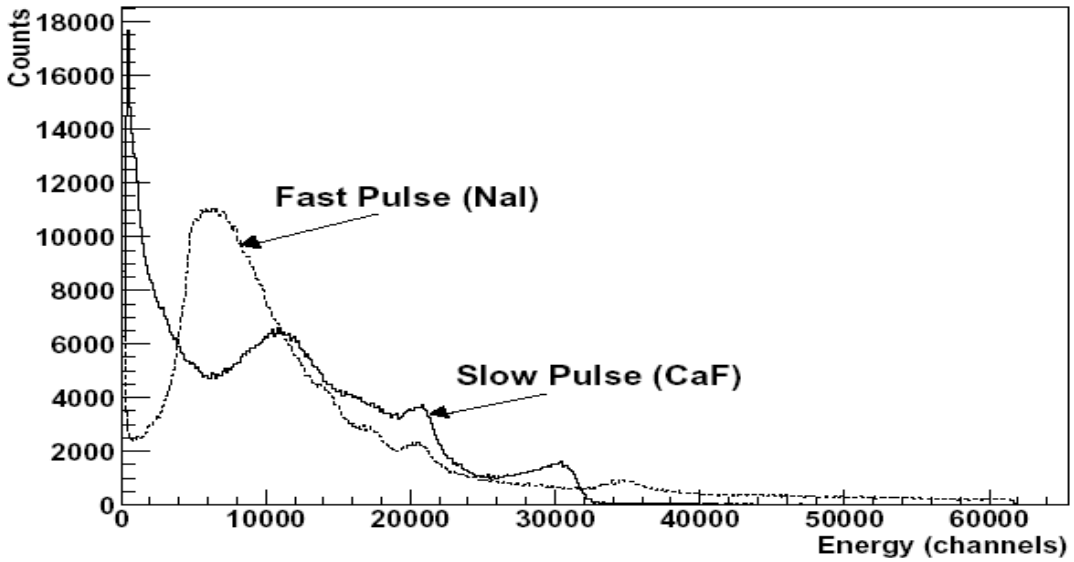

Figure 7. Energy spectra for the fast and slow rise-time pulses of xenon produced in the phoswich detector.

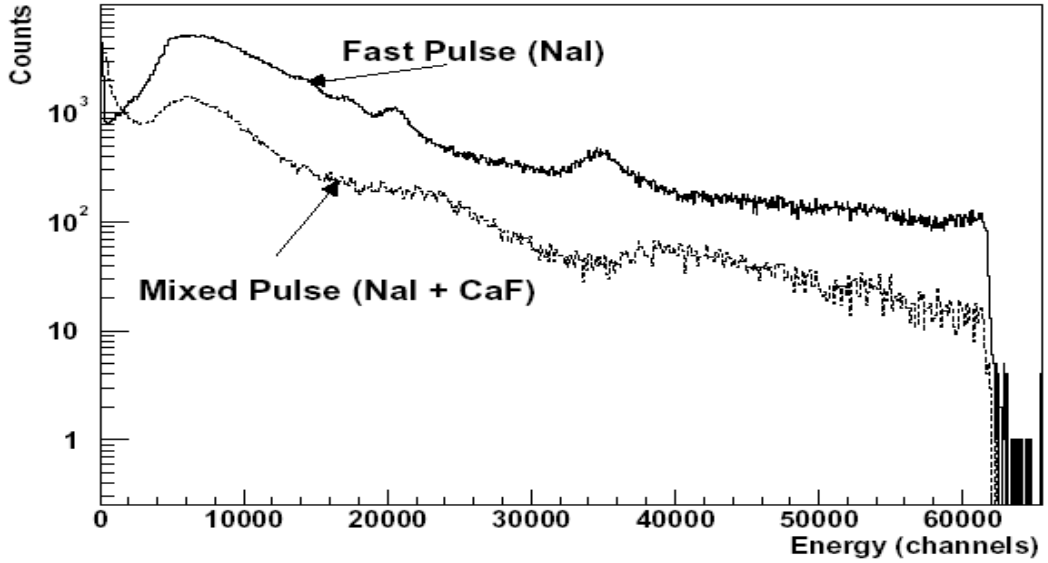

Figure 8. Energy distribution of xenon pulses with fast and medium rise-times from the phoswich detector system.

The pulses with medium rise-times are candidates for beta-gamma coincident pulses, but must be distinguished from either a single gamma or beta pulse. In addition, for the intended use of the phoswich, the beta-gamma coincident pulses must be separated into constituent pieces. This aspect of separating out the gamma and beta contributions of an individual pulse is quite different from the normal phoswich use where single particle pulses are sorted by rise-times. Generally, we can describe a pulse produced by a single particle as a single exponential with the rise-time characterized by a single parameter. For a pulse arising from two particles in a phoswich detector, the shape should be described by two exponentials and two rise-times. The difference in the rise-times should result in a pulse with an initial fast-rising shape, followed by a slow-rising tail. This difference would be distinct from a fast pulse with both a fast initial piece and tail or a slow pulse with a slow initial piece and tail. To investigate this expectation for beta-gamma coincident

pulses, the initial rise-time of the pulse (10-50%) is plotted against the final rise-time of the pulse (50-90%), for pulses with energies between 4,000–14,000 channels (see Figure 9). The energy constraint is used to ensure pulses have similar total pulse height. Two peaks, corresponding to the fast and slow pulses, are shown in Figure 9 with a relatively smooth connecting distribution. No other peak, which would correspond to anticipated fast initial pieces and slow tails, is observed. The data of Figure 9 indicate this technique does not identify beta-gamma coincident pulses. A further investigation to develop an appropriate method of identifying and selecting beta-gamma coincident pulses is required.

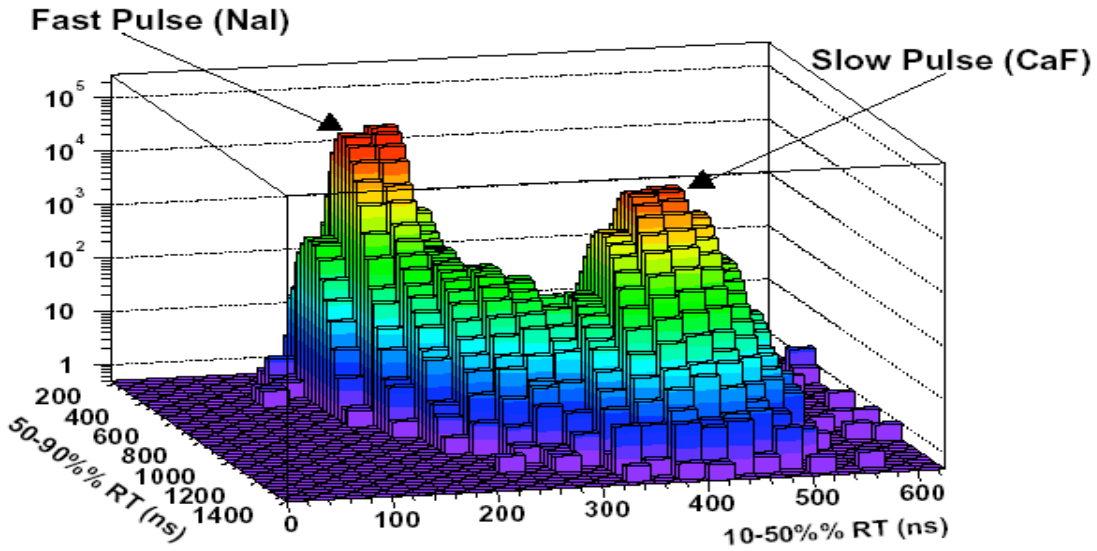

Figure 9. Distribution of the initial (10-50%) versus final (50-90%) rise-time of the xenon pulses in the energy range of 4000 – 14000 channels.

CONCLUSIONS AND RECOMMENDATIONS

A NaI(Tl) and CaF_2(Eu) phoswich detector system was built and responds to both gamma and beta radiation sources. It is possible to effectively discriminate between pulses created by either a gamma or beta particle with this system. It appears, however, that the discrimination of pulses resulting from by both a gamma and beta response in a beta-gamma coincidence event is not straightforward. There are indications that beta-gamma coincident events can be identified, and more investigation is warranted to develop an algorithm with better discriminating power than was used in this preliminary investigation. For example, a technique to fit each individual digitized pulse with an appropriate two-parameter function may be useful. In addition, different detector materials may provide a better differentiation than the choice of NaI(Tl) and CaF_2(Eu) and should be investigated. Finally, although the beta-gamma coincident events may ultimately be separable from single gamma or beta events, it may be challenging to separate the individual gamma and beta contributions of a single pulse with any precision.

ACKNOWLEDGEMENTS

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