DIGITAL PULSE SHAPE ANALYSIS WITH PHOSWICH DETECTORS TO SIMPLIFY COINCIDENCE MEASUREMENTS OF RADIOACTIVE XENON

W. Hennig¹, H. Tan¹, W. K. Warburton¹, and J. I. McIntyre²

XIA LLC¹ and Pacific Northwest National Laboratory²

Sponsored by National Nuclear Security Administration Office of Nonproliferation Research and Engineering Office of Defense Nuclear Nonproliferation

Contract No. DE-FG02-04ER89121

ABSTRACT

The Comprehensive Nuclear-Test-Ban Treaty establishes a network of monitoring stations to detect radioactive xenon in the atmosphere from nuclear weapons testing. One such monitoring system is the Automated Radioxenon Sampler/Analyzer (ARSA) developed at Pacific Northwest National Laboratory, which uses a complex arrangement of separate beta and gamma detectors to detect beta-gamma coincidences from the xenon isotopes of interest. The coincidence measurement is very sensitive, but the large number of detectors and photomultiplier tubes requires careful calibration. Simplifying this coincidence measurement system while maintaining its performance is the objective of the research described here.

It has been suggested that beta-gamma coincidences could be detected with only a single photomultiplier tube and electronics channel by using a phoswich detector consisting of optically coupled beta and gamma detectors (Ely, 2003). In that work, rise time analysis of signals from a phoswich detector was explored as a method to determine if interactions occurred in either the beta or the gamma detector or in both simultaneously. However, this approach was not able to detect coincidences with the required sensitivity or to measure the beta and gamma energies with sufficient precision for radioxenon monitoring.

In this paper, we present a new algorithm to detect coincidences by pulse shape analysis of the signals from a BC-404/CsI(Tl) phoswich detector. Implemented on fast digital readout electronics, the algorithm achieves clear separation of beta only, gamma only and coincidence events, accurate measurement of both beta and gamma energies, and has an error rate for detecting coincidences of less than 0.1%. Monte Carlo simulations of radiation transport and light collection were performed to optimize design parameters for a replacement detector module for the ARSA system, obtaining an estimated coincidence detection efficiency of 82-92% and a background rejection rate better than 99%. The new phoswich/pulse shape analysis method is thus suitable to simplify the existing ARSA detector system to the level of a single detector per sample chamber while maintaining the required sensitivity and precision to detect radioxenon in the atmosphere.

OBJECTIVE

The Comprehensive Nuclear-Test-Ban Treaty establishes a network of monitoring stations to detect radioactive xenon in the atmosphere from nuclear weapons testing. One such monitoring system is the Automated Radioxenon Sampler/Analyzer (ARSA) developed at Pacific Northwest National Laboratory (Reeder, 1998). The ARSA system consists of a pair of large NaI(Tl) scintillator crystals holding four cylindrical fast plastic scintillator (BC-404) cells which are optically isolated from the NaI(Tl). The cells are filled with the xenon gas to be counted, which decays by emitting gamma rays or X-rays in coincidence with beta particles or conversion electrons. The plastic scintillator is meant to absorb all beta particles and conversion electrons, while the longer range gamma rays and X-rays will mainly be absorbed in the NaI(Tl) scintillator. Each BC-404 cell and each NaI(Tl) crystal is coupled to a pair of photomultiplier tubes (PMTs) and is read out by independent electronic channels. The sensitivity for detecting xenon isotopes is greatly increased by requiring coincidence between the signals from the PMTs coupled to the NaI(Tl) and the signals from the PMTs coupled to the BC-404.

While obtaining high coincidence detection efficiency and resolutions of about 25% for characteristic 80keV gamma rays, the current ARSA system design results in significant operational complexity. The principle of time based coincidence, while effective in suppressing the background, requires separate signals from the NaI(Tl) and the BC-404, i.e., separate PMTs and readout electronics. In particular, the 12 PMTs require careful gain matching and calibration, and as the PMT gains change with time, voltage and temperature, the system easily drifts out of calibration.

To improve the current ARSA system a new concept for coincidence measurements was explored previously (Ely, 2003) based on rise time analysis of signals from a phoswich detector. The phoswich detector consisted of a 0.04 inch thick $CaF_2(Eu)$ crystal (decay constant 940ns) optically coupled to a 2x2 inch NaI(Tl) crystal (decay constant 250ns) and was read out by a single PMT. The $CaF_2(Eu)$ is used as the beta detector to absorb beta particles and conversion electrons and the NaI(Tl) acts as the gamma ray detector to absorb gamma rays and X-rays. By integrating the signal from the PMT in a charge integrating preamplifier and acquiring pulse waveforms with a fast digital pulse processor, the scintillator in which radiation interacted could be determined by the signal rise times. While this method of pulse shape coincidence detection worked well to distinguish $CaF_2(Eu)$ only events and NaI(Tl) only events, coincident events in both scintillators were not easily identified by this algorithm and/or choice of scintillators and it was deemed challenging to separate the individual gamma and beta contributions with any precision.

In this paper, we describe an improved method to detect coincidences with a phoswich detector and single channel of readout electronics. The method uses the signal directly from the phoswich detector, without a charge integrating preamplifier, and determines the scintillator(s) in which the interaction occurred by analyzing the signal over characteristic time periods, thus detecting coincidences. Applied to radioxenon monitoring, the method provides beta-gamma coincidence detection and energy measurement of both beta and gamma energies with a greatly simplified measurement setup.

RESEARCH ACCOMPLISHED

1. Development of Pulse Shape Analysis with Prototype Phoswich Detector

The pulse shape analysis algorithms were developed using a prototype phoswich detector consisting of a 1" diameter by 1" thick CsI(Tl) crystal optically coupled to a 1" diameter, 1-mm thick disk of the plastic scintillator BC-404 on the front end. The detector was illuminated with a variety of solid sources or Xe and Rn gas enclosed in small plastic bags. Using an XIA Pixie-4 digital spectrometer directly connected to the PMT coupled to the detector, we acquired waveforms of the detector signals and found the three basic types of events shown in Figure 1: a) slow rising and slow falling pulses corresponding to interactions only in the CsI, b) very fast pulses with high amplitude corresponding to interactions only in the BC-404, and c) combinations of the previous cases corresponding to coincident interactions in both scintillators. Limiting the signal bandwidth of the Pixie-4 analog front end reduced the amplitude of the fast BC-404 signals without affecting the slower, low amplitude CsI signals. A single channel could thus accommodate the highest beta particle energies expected from the radioxenon decays while still obtaining sufficient precision for low amplitude X-ray signals.



Figure 1: Pulse waveforms from the prototype phoswich detector (shown in insert). There are three types of events: CsI only pulses, BC-404 only pulses and combination pulses depositing energy in both parts of the phoswich detector.

Analyzing the acquired waveforms off-line, we calculated the signal rise time and various filter sums over different regions of the waveform for each acquired pulse. These properties allowed a clear distinction of the three event types described above and thus can be used to detect coincidences between the gamma and beta radiation from the samples. The properties can further be used to calculate the energies deposited in each part of the phoswich detector and display events in 2D energy scatter plots as shown in Figure 2. In the graph, CsI only events fall on the vertical axis (plastic energy is zero) and plastic only events fall on the horizontal axis (CsI energy is zero). Coincidence events form two horizontal bands, corresponding to 30 keV X-rays or 81 keV gamma rays in coincidence with betas of varying energy. Note that the 30 keV X-ray coincidence events are also well separated from the beta-only events.



Figure 2: Energy scatter plot for ¹³³Xe colored by event type. CsI only events fall on the vertical axis, plastic only events fall on the horizontal axis, and coincidence events form horizontal bands at characteristic X-ray or gamma ray energies (fixed photon energy deposited in the CsI and varying beta energy deposited in the BC-404).

By defining thresholds for the rise time and energy values, the three event types can be separated and binned into separate energy histograms as shown in Figure 3. The energy resolution at the 81keV peak is 16.9% for Eco, the energy deposited in the CsI in CsI only events, and 17.1% for Ecb, the energy deposited in CsI in coincidence events, which is significantly better than resolutions achieved with the current ARSA detector. The energy resolution is good enough to resolve even the 52 keV Iodine escape peak in both Eco and Ecb. In experiments with ⁶⁰Co, we obtained resolutions of 5.2% for Eco and 5.4% for Ecb for the 1.3 MeV peak. This is comparable to resolutions routinely achieved with a standard CsI detector and demonstrates that our pulse shape algorithms do not degrade energy resolution compared to standard pulse processing methods.



Figure 3: Energy histograms for ¹³³Xe formed by projecting the data of Figure 2 onto the two energy axes. The left graph shows the energy deposited in the CsI for CsI only events (Eco) and combination events (Ecb). The right graph shows the energy deposited in the plastic for plastic only events (Epo) and combination events (Epb).

The error rate of the algorithm was estimated from measurements with ²⁴¹Am which emits alpha particles and Xrays in coincidence. In the setup shown in Figure 1, the algorithm classified 30% of detected events as BC-404 only events and 3.5% as coincident events. When an Al shield was inserted between source and detector to stop all alpha particles, the fraction of BC-404 only events and coincident events dropped to 0.2% and 0.1%, respectively. Since there can be no true coincidences without the alpha particles, the measured 0.1% must be either random coincidences, scattering of a single X-ray from one scintilator to the other, or non-coincident events wrongly classified as coincident by the algorithm. As a worst case estimate, we can thus determine that the algorithm wrongly classifies events as coincidences at a rate of less than 0.1%.

2. Monte Carlo Simulations of Phoswich Well Detector for Radioxenon Monitoring

While the prototype detector was sufficient to develop the algorithms for detecting coincidences and measuring individual energies with good precision in a single channel of electronics, it has very low coincidence detection efficiency since at least half of the beta radiation and the gamma radiation will be emitted away from the detector. Using Monte Carlo simulations of light collection and radiation transport, we therefore studied a phoswich well detector design in which a BC-404 cell holding the radioxenon will be enclosed in a 3" cylinder of CsI, see Figure 4.

2.1. Light Collection Simulations

The unusual geometry of the phoswich well detector will affect the uniformity of light collection in the detector. We therefore carried out Monte Carlo simulations of the light collection for the geometry of the proposed detector, the test detector, and other geometries using Monte Carlo code DETECT2000. Figure 5 shows the distribution of light collection efficiency in the 3" prototype detector with a reflectivity of 0.95 or 0.99 for the diffusive reflector on the outer surface of CsI. We found that placing a structure inside the CsI crystal somewhat degrades the uniformity of

the light collection efficiency, especially if the coating on the outside of the CsI crystal has a low reflection coefficient. From the volume-weighted probability distribution of the light collection efficiency, we estimate that the energy resolution of mono-energetic gamma rays, due <u>only</u> to the non-uniformity of the collection efficiency in the crystal, increases from 2.4% without a cell to 2.9% with a spherical cell and 3.8% with a cylindrical cell (for the conservative estimate of the reflectivity, i.e. 0.95). Optimizing uniformity thus requires minimizing of the number of interfaces, paying careful attention to their shape, and maximizing the reflection coefficient of the reflector on the outside of the CsI crystal. However, compared to a typical measured resolution of ~7% at 662 keV (resulting from photostatistics, crystal non-uniformities and energy non-linearities), the effects of the embedded cell are small. Adding these numbers in quadrature, we anticipate the phoswich detector energy resolution to not worsen significantly at the lower energies from the Xe isotopes due to effects of light collection arising from either the inclusion of the BC-404 cell or its specific geometry, provided that the reflectivity is kept high and the cell is not unreasonably shaped.



Figure 4: Geometry the phoswich well detector. The outer 3" × 3" cylinder is the CsI(Tl) crystal. Embedded inside is the 1" × 1" BC-404 counting cell whose wall thickness may be up to 5 mm. The counting cell contains the xenon gas which is fed into the cell through the slender tube. Left: sketch of the detector size; middle: side view of the detector; right: top view of the detector.



Figure 5: Geometric distribution of light collection efficiency in the prototype detector. The left figures are distributions of light collection efficiency in the prototype detector with two typical outer reflectors (reflection coefficient = 0.95 and 0.99, respectively). The right figure is the volume-weighted probability distribution of light collection efficiency.



2.2. Radiation Transport Simulations

Major Xenon Decay Energies

Figure 6: Interaction probability of major radioxenon decay energies in each component of the phoswich well detector for radiation emitted from the center of the counting cell. The thickness of the BC-404 cell varied from 5mm to 1mm.

Monte Carlo simulations of the radiation transport in the phoswich detector were performed to determine optimum values of design parameters and to estimate the coincidence detection efficiency. Using Monte Carlo code PENELOPE, several characteristic energies of gamma rays, X-rays, beta particles and conversion electrons from radioxenon were simulated for the well detector and also for the prototype detector to compare the simulation to the experiments. The simulations showed that most beta particles or conversion electrons will be absorbed in the BC-404 and most X-rays or gamma rays will be absorbed in the CsI, as intended (see Figure 6). A small fraction of high energy gamma rays will escape from the detector, and a small fraction of low energy beta particles will be absorbed in the xenon gas, i.e., they will not be detected. The thickness of the BC-404 cell has to be adjusted to compromise between preventing betas from reaching the CsI (thicker wall) and reducing the chance of photons interacting with the plastic (thin wall). A wall thickness of 2-3 mm is a good compromise.

Table 1. Probabilities (in %) of simultaneous interactions of characteristic gamma rays and beta particles
from ¹³³ Xe in the phoswich well detector. Only events where betas interact with the BC-404 only and
gammas interact with the CsI only (bold) are coincidence events with good energy measurement.

	beta not interacting	beta interacting with BC-404 only	beta interacting with CsI only	beta interacting with both BC-404 and CsI
gamma not interacting	0.0	1.5	0.0	0.0
gamma interacting with BC-404 only	0.0	0.3	0.0	0.0
gamma interacting with CsI only	0.0	92.6	0.0	0.5
gamma interacting with both BC-404 and CsI	0.0	5.1	0.0	0.0

The interaction probabilities of the characteristic energies were then used to estimate the coincidence detection efficiency. For example, 81 keV gamma rays and 346 keV beta particles may be taken to represent the coincidence radiation from ¹³³Xe. Each has a certain probability to interact a) not at all, b) with BC-404 only, c) with CsI only, or d) with both BC-404 and CsI. The products of the probabilities for the altogether 16 combinations are shown in Table 1. Only events where betas interact with the BC-404 only and gammas interact with the CsI only are coincidence events with good energy measurement, shown in bold in Table 1. For ¹³³Xe, the estimated coincidence detection efficiency of the well detector is thus 92.6%, for other xenon isotopes of interest it is about 82%.

Table 2. Probability of external background gamma rays to interact with the phoswich well detector,
obtained from simulations. Only events directly aimed at the center of the detector were simulated;
estimated to be 1/27th of the overall background. The numbers shown have thus to be divided by 27
to obtain the overall background coincidence rate.

Gamma Energy (MeV)	not interacting	interacting with BC-404 only	interacting with CsI only	interacting with both BC-404 and CsI	interacting with both, < 250keV deposited in CsI
0.583	8.66%	0.22	88.95	2.17	0.86
1.461	24.20	0.49	73.05	2.26	0.82
2.614	30.18	0.48	66.87	2.47	0.68
5	32.35	0.14	64.33	3.18	0.22
10	29.08	0.05	65.55	5.32	0.07
20	23.22	0.01	66.70	10.07	0.01

To estimate the rejection ratio of the well detector, interactions of background radiation were also simulated. Table 2 shows the interaction probabilities for gamma rays with representative energies, coming from a single external location and directed towards the center of the detector. Of the simulated gamma rays, a majority interacts with CsI only and will thus be rejected by the pulse shape analysis. About 2-10% of simulated events interact with both scintillators and will be categorized as coincidences, but only 0.9-0.01% will deposit less than 250 keV in the CsI, the maximum gamma-ray energy from the xenon decay chain, i.e. most of the background coincidences can be rejected by a simple energy cut. In addition, the radiation will in practice be coming from a multitude of locations around the detector and most of the radiation will not be directed towards the BC-404 cell, only interacting with the plastic if scattered towards the center. Therefore, as a very crude estimate, we assume that the simulation represents only the fraction of the background radiation equal to the volume fraction of the BC-404 cell in the detector, and that the rest will not interact will the BC-404 cell at all. The simulated events thus represent only 1/27th of the background radiation, and the not simulated events will be rejected by the pulse shape analysis. This means that overall the background rejection ratio for the phoswich well detector is greater than 99.9% in the energy range of interest.

CONCLUSION

In summary, we developed an algorithm to detect beta-gamma coincidences in the signals from a BC-404/CsI(Tl) phoswich detector, using a single channel of readout electronics. The algorithm achieves clear separation of beta only, gamma only and coincidence events, accurate measurement of both beta and gamma energies, and has an error rate for detecting coincidences of less than 0.1%. Monte Carlo simulations of radiation transport and light collection were performed to optimize design parameters for a replacement detector module for the ARSA system, obtaining an estimated coincidence detection efficiency of 82-92% and a background rejection rate better than 99%. The new phoswich/pulse shape analysis method is thus suitable to simplify the existing ARSA detector system to the level of a single detector per sample chamber while maintaining the required sensitivity and precision to detect radioactive xenon in the atmosphere.

ACKNOWLEDGEMENTS

We would like to thank the PNNL ARSA development team headed by Theodore Bowyer for valuable information and insight on radioxenon collection, as well as Michael Momayezi from Bridgeport Instruments for his support in this research.

REFERENCES

Reeder, P.L., T.W. Bowyer, and R.W. Perkins, (1998) Journal of Radioanalytical and Nuclear Chemistry, Vol. 235, No. 1-2 (1998) 89-94

Ely, J. H., C.E. Aalseth, J. C. Hayes, T. R. Heimbigner, J. I. McIntyre, H. S. Miley, M. E. Panisko, and M. Ripplinger, (2003), Novel Beta-Gamma Coincidence Measurements using phoswich detectors, in *Proceedings of the 25th Seismic Research Review – Nuclear Explosion Monitoring: Building the Knowledge Base*, LA-UR-03-6029, Vol. 2, pp. 533-541.