TRIPLE COINCIDENCE RADIOXENON DETECTOR

Justin I. McIntyre, Craig E. Aalseth, Theodore W. Bowyer, James C. Hayes, Tom R. Heimbigner, Scott Morris, and Paul L. Reeder

Pacific Northwest National Laboratory

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ABSTRACT

The Automated Radioxenon Sampler/Analyzer (ARSA) built by Pacific Northwest National Laboratory (PNNL) is one of the world's most sensitive systems for monitoring the four radioxenon isotopes ^{133}Xe, ^{133m}Xe, ^{131m}Xe, and ^{135}Xe. However, due to size, weight, and power specifications appropriate to meet treaty-monitoring requirements, the ARSA is unsuitable for rapid deployment using modest transportation means. To transition this technology to a portable unit that can be easily and rapidly deployed can be achieved by significant reductions in size, weight and power consumption if concentration were not required. As part of an exploratory effort to reduce both the size of the air sample and the gas processing requirements PNNL has developed an experimental nuclear detector to test and quantify the use of triple coincidence signatures (beta, conversion electron, x-ray) from two of the radioxenon isotopes (^{135}Xe and ^{133}Xe) as well as the more traditional beta-gamma coincidence signatures used by the ARSA system. The additional coincidence requirement allows for reduced passive shielding, and makes it possible for unambiguous detection of ^{133}Xe and ^{135}Xe in the presence of high ^{222}Rn backgrounds. This paper will discuss the experimental setup and the results obtained for a ^{133}Xe sample with and without ^{222}Rn as an interference signature.

OBJECTIVE

There are currently four automated platforms that have been developed to detect the four radioxenon isotopes, ^{133}Xe, ^{133m}Xe, ^{131m}Xe and ^{135}Xe. These systems primary mission is to monitor for clandestine underground nuclear explosions, and several publications discuss both the IMS and the radioxenon systems; CTBT/WGB/TL-11/5/Rev. 4 (1999); Auer et al. (2004); and L. E. DeGeer (1995). The Automated Radioxenon Sampler/Analyzer (ARSA) is one such system that is currently under commercial development, see Bowyer (2002). All of these systems sample large volumes of air and have low minimum-detectable-concentrations (MDC). These requirements have by necessity, led to large, heavy systems that are loud, consume large amounts of power and make them inappropriate for man-cartable equipment that can be easily and rapidly deployed, or deployed on moving platforms.

The current technology uses a "brute force" method to separate the ambient xenon from the air (ambient xenon accounts for less than 1 ppm of air). Current technology typically uses a series of chemical adsorbent materials (charcoal and mole sieve in the case of the ARSA) to collect and concentrate the xenon so that it can be counted in a small volume beta-gamma coincidence detector (see Reeder *et al* (2004) for a detailed description of the coincidence spectrometer). The collection method is very efficient at collecting ambient radon and with the large volumes of air collected the interference from the radon daughters would easily obscure all but the largest concentrations of radioxenon. To remove the radon, requires additional separation columns and traps and thus increases the size and power consumption of the system. Significant reductions in size, weight and power consumption can be achieved if preconcentration were not required and the removal of radon was no longer a necessity.

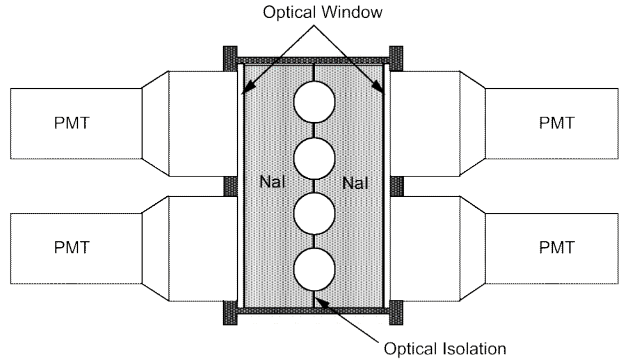

Figure 1. Diagram of the Nai (Tl) saddle detector for the ARSA system.

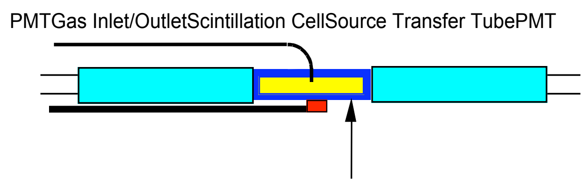

Figure 2. Schematic representation of plastic scintillation beta cell with photo-multiplier tubes, gas transfer line and calibration source transfer tube.

The ARSA nuclear detector (see figures 1 and 2) uses a beta-gamma (β, γ) coincidence to detect ^{133}Xe and ^{135}Xe. The two metastable xenon isotopes, ^{131m}Xe and ^{133m}Xe, emit only a conversion electrons (CE) and x-ray (for more detail see McIntyre (2000)). The x-rays and gammas are detected in the two NaI(Tl) saddle detectors (figure 1) and plastic scintillator beta cells occupy the four holes in the detector. The beta and (CE) are detected in the 6 cc plastic scintillation cell shown in figure 2 along with the two photomultiplier tubes (PMT), gas transfer tube and the calibration source transfer tube. The geometry provides nearly complete coverage for the decay process making the ARSA detector very efficient with greater than 60% efficiency for gammas and x-rays and greater than 80% efficiency for beta and CE.

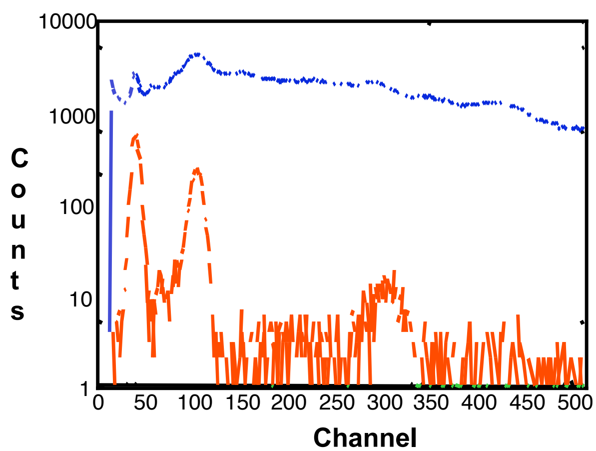

Figure 3. A beta-gated gamma-ray spectrum showing the significant reduction in the background when a beta-gamma coincidence is used.

The coincidence nature of the physics decay process allows for a dramatic reduction in the ambient background caused by cosmic rays and terrestrial gammas and x-rays. Figure 3 shows the difference in the between the gamma singles spectrum, the top blue curve and the beta-gated gamma spectrum, the lower red curve. The beta-gated spectrum clearly shows the

30-keV x-ray and 80-keV gamma-ray of ^{133}Xe as well as the 250-keV gamma-ray of ^{135}Xe. The reduction in the dominant Compton background in the ungated gamma spectrum is dropped by three orders of magnitude when a coincidence is required.

Figure 4 shows the beta-gated, x-ray/gamma response of ^{133}Xe (green circles) and ^{214}Pb plus ^{214}Bi (blue triangles). The ^{214}Pb and ^{214}Bi are daughter products of ^{222}Rn and are a significant source of interference. Note that ^{135}Xe is not present in this sample but it would appear at 250-keV and also has a significant interference problem caused by ^{214}Pb.

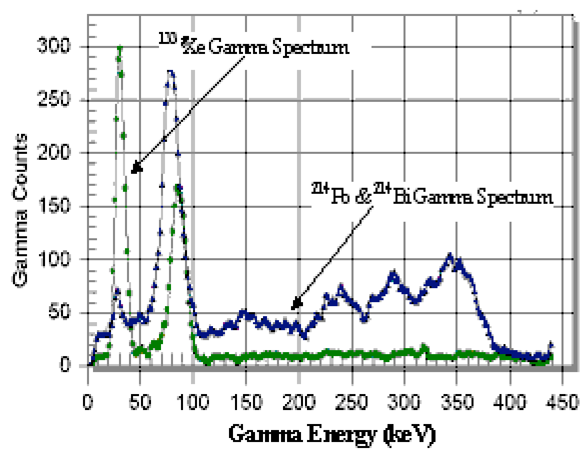

Figure 4. A beta-gated spectrum of a ^{133}Xe sample (green circles) and a beta-gated radon daughter spectrum, ^{214}Pb, (blue triangles).

In addition to (β, γ) and (CE, x-ray) coincidences ^{133}Xe and ^{135}Xe have a triple coincidence (β, CE, x-ray) that can be exploited. Requiring three detectors to register something simultaneously would further reduce the background and reduce the interference from the dominant 80-keV ^{214}Pb and ^{214}Bi peak. This triple coincidence detection can be achieved by using a highly segmented detector. Such a detector could achieve Compton scattering background rates approaching zero and be able to dramatically suppress the radon interference. For long enough count times ^{133}Xe and ^{135}Xe could be detected without any sample purification or preconcentration. A radioxenon measurement device could be designed that could be carried to the field with similar sensitivity as the current state-of-the-art.

RESEARCH ACCOMPLISHED

The goal of this project was to build and test a new radiation detector for radioxenon that could eliminate the need for chemical separations altogether by measuring all of particles emitted from ^{133}Xe and ^{135}Xe simultaneously. This is a major step in the evolution of radioxenon detection techniques and represents the only conceivable approach to drastically changing this type of monitoring. It will allow real-time radioxenon detection from several platforms that are currently too restrictive for existing equipment because of weight, size and power requirements.

The major research of this project was to develop a segmented detector for CE and betas and an appropriate detector for x-rays. Figures 5 shows the test detector that was built. It consists of a 60 cc gas cell made up of two paddles of 1.0 mm thick scintillating plastic to detect the beta particles in one paddle and the CE in the other. A thin aluminized Mylar sheet has been stretched between the two paddles to prevent the scattering of scintillation light between the two paddles. Each paddle is viewed by four PMT's and crude position sensitivity is possible with very little post data acquisition processing. The scintillator planes are 2 cm apart to create a 60 cc gas cell (beta-cell). To prevent the scintillation light from one plane of the scintillator from being seen by the other the two planes are separated by a thin sheet of aluminized Mylar. The two scintillator paddles are 2 cm apart and a gas tight inlet tube is located at the top of the assembly for the introduction of gas samples. Two 5"x2" NaI detectors can be placed against each of the scintillator paddles to provide gamma and x-ray detection. These detectors have good energy resolution and high stopping power for the low energy gamma/x-rays from the

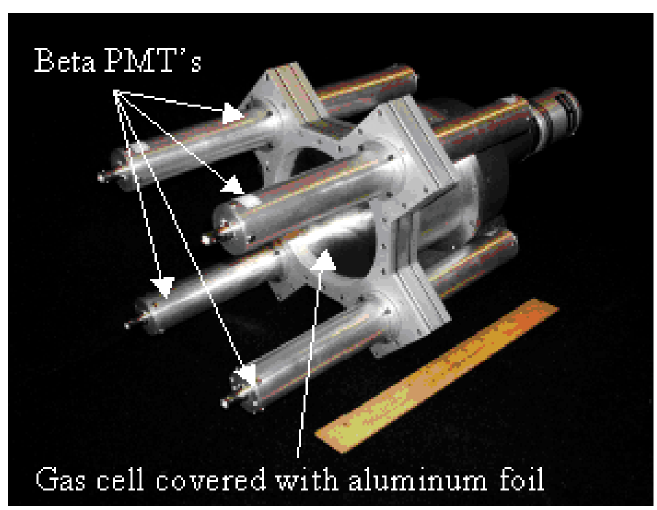

Figure 5. Pictures of the SSXD test detector (note 12" ruler for scale).

radioxenon isotopes. The detectors are also easy to calibrate and integrate into data acquisition systems. The detector assembly was energy calibrated with several sealed gamma-ray line sources and ^{133}Xe and radon gas sources. Modeling of the detector clearly indicated that the ambient background from a triple coincidence would be low and most of the events would be cosmic rays and have very high-energy deposition in the plastic and NaI.

Each PMT had to be gain matched so that nonlinear response was minimized. With each PMT for a single paddle giving the same response it was possible to sum the PMT signals together to give a total signal that was proportional to the total energy deposition in each paddle. Figure 6 show a histogram of the response of paddle 1 vs. paddle 2. The counts in the histogram require signals from both paddles and indicated either a (β, CE) coincidence or a high energy cosmic ray going through both paddles. The triangular low-energy region was the region where (β, CE) coincidence from the ^{133}Xe were most likely to occur and the triangular high-energy region was where cosmic rays were most likely to occur and could be rejected from further analysis.

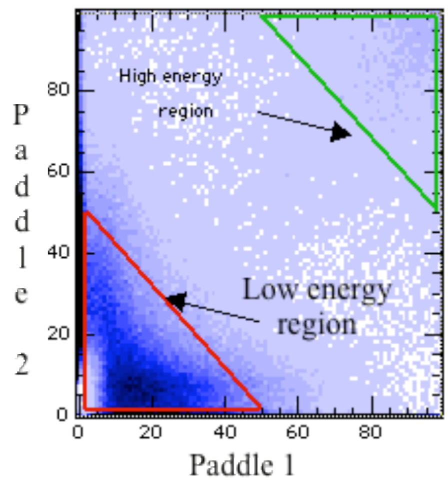

Figure 6. This 2-dimensional histogram is the summed response of all four PMT's on beta-cell paddle 1 vs. the summed response of all four PMT's on paddle 2.

An additional analysis parameter was calculated to give a crude position of the events in a given paddle. This parameter used the response from all four PMT's of a single paddle and is plotted in figure 7. The green indicates no counts and the white indicates high counts. The highlighted lines indicate saturation of the amplifier-ADC channel for one of the PMT's. For future detectors the ability to accurately determine event position would allow an additional method to reduce the interference of radon. Radon gas particles after the first alpha decay will bind strongly to the detector walls and hence emit subsequent alphas and betas from decays in the same location.

Additional studies indicated that the detector would be sensitive to the levels of stable xenon that were injected into the gas cell because xenon is a high-density gas. The detector has the greatest efficiency when there are small volumes of Xe gas. The other constituents of ambient air, O_2, N_2, etc. play a much smaller role in γ/x-ray attenuation and β/CE energy loss than xenon. Modeling also indicated that the solid angle would be sufficient for the test detector (~20%), but it is anticipated that later models for an early proto-type would attempt to achieve 50% or better. The current ARSA beta cell has a solid angle of ~80%.

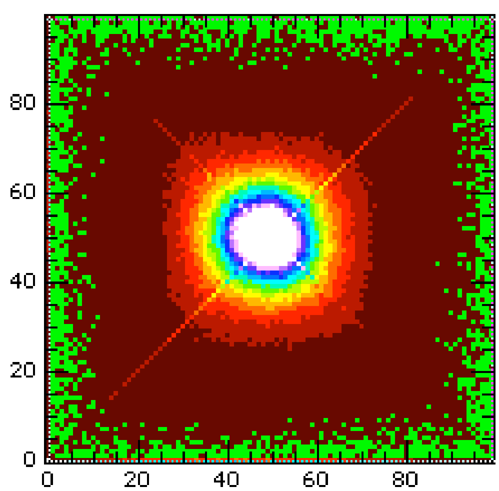

Figure 7. A 2-dimensional histogram that gives a crude position plot of events for one of the beta-cell scintillating paddles.

After the gain matching of the PMT's was completed data was taken using a high- level ^{133}Xe spike (~100 Bq). The ^{133}Xe spike was injected into the cell and the double coincidence response was measured (see figure 8, red curve). The 30-keV x-ray and 80-keV gamma-ray lines are both present and the ratio between the two peaks is well within the range of the current ARSA beta-gamma detector. This indicates that the efficiency for both detectors is comparable for the low energy gamma and x-ray. Only the low energy gamma-ray region (0-150 keV) is shown, as this is the region of most interest. The detector electronics were than modified to accept only triple coincidence events (both scintillator paddles and one of the NaI detectors) and the results are presented in figure 8 as well (green curve). The 30-keV peak is clearly present while the 80-keV is suppressed. It is not clear why there were any events in the 80-keV peak as these events are not triple coincidence events and it is felt that some sort of cross talk in

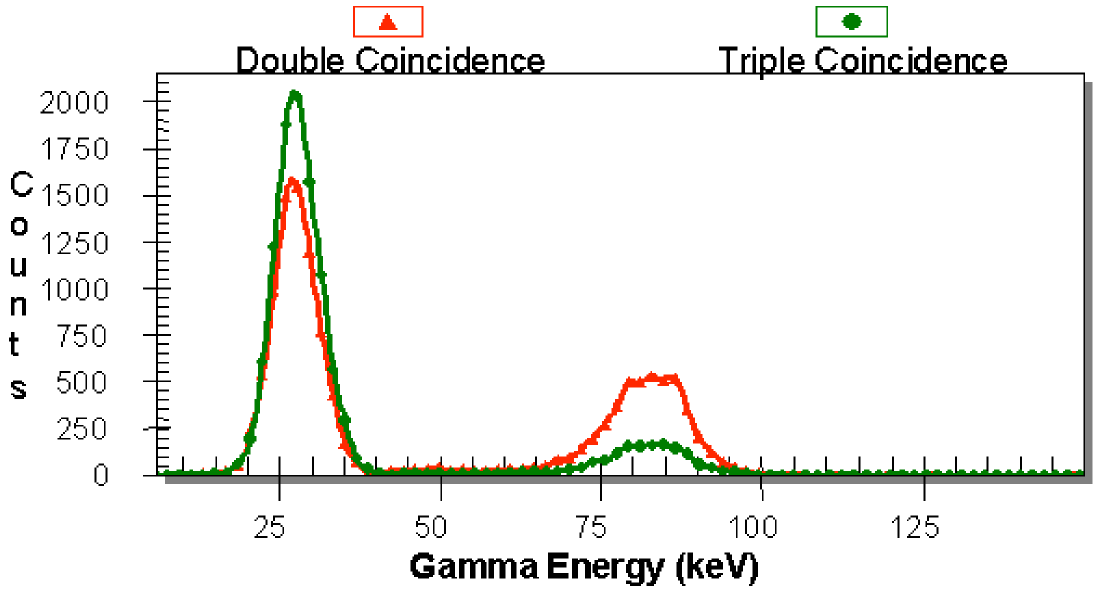

Figure 8. Histogram showing beta gated gamma spectrum with beta only gating (red triangles) and beta plus CE gating (green circles).

the electronics must be occurring. In an ideal detector there would be no counts in the 80-keV region and further optimization of the detector geometry and the readout electronics is necessary to achieve this result.

The next test was to add a spike of comparable strength of radon and measure the double and triple coincidence events. The results of this test are shown in figure 9. As in figure 8 the green curve represents the double coincidence gated gamma-ray spectrum and the red curve is the triple coincidence gated gamma spectrum. A significant reduction in the Compton scattered background is apparent in the triple coincidence data. A cut on high energy beta plus CE shows that the radon interference is reduced dramatically and it will now become possible to measure much lower levels of radioxenon concentrations in the presence of high radon backgrounds. As in figure 8 the green curve is a double coincidence and it is clear that the radon daughter ^{214}Pb x-ray at 80-keV is a significant interference term for the 80-keV gamma ray of ^{133}Xe. For lower concentrations of ^{133}Xe even the 30-keV line will be completely obscured by the radon interference. A significant reduction of the Compton scattering interference is seen and much lower (2-3 orders of magnitude) concentrations of ^{133}Xe can now be measured with a high radon background.

Using the solid angle, efficiency and the ambient background that the detector registered an MDC of ~7 Bq/m^3 of air was determined for the current test detector. This result is very high in comparison to the state of the art by approximately 4 orders of magnitude and is driven by the small quantity of gas that is sampled (1 liter in this case). Crude concentration techniques would dramatically decrease this MDC. The MDC falls linearly with an increase in sampled gas. Such concentration methods would increase the size and power consumption of a unit with a trade off in much lower MDC's (2-3 orders of magnitude).

Additional improvements can be made in the overall detector design and potentially the readout electronics. For instance using a segmented phoswich style detector as outlined in Ely (2003). Phoswich detectors are a set of two or more scintillating detectors that give a different response depending on the particle detected. This would have the effect of having both the gamma and beta detectors viewed by half the total number of PMT's that would normally be used, reduce the overall detector size, and provide superior beta and CE separation.

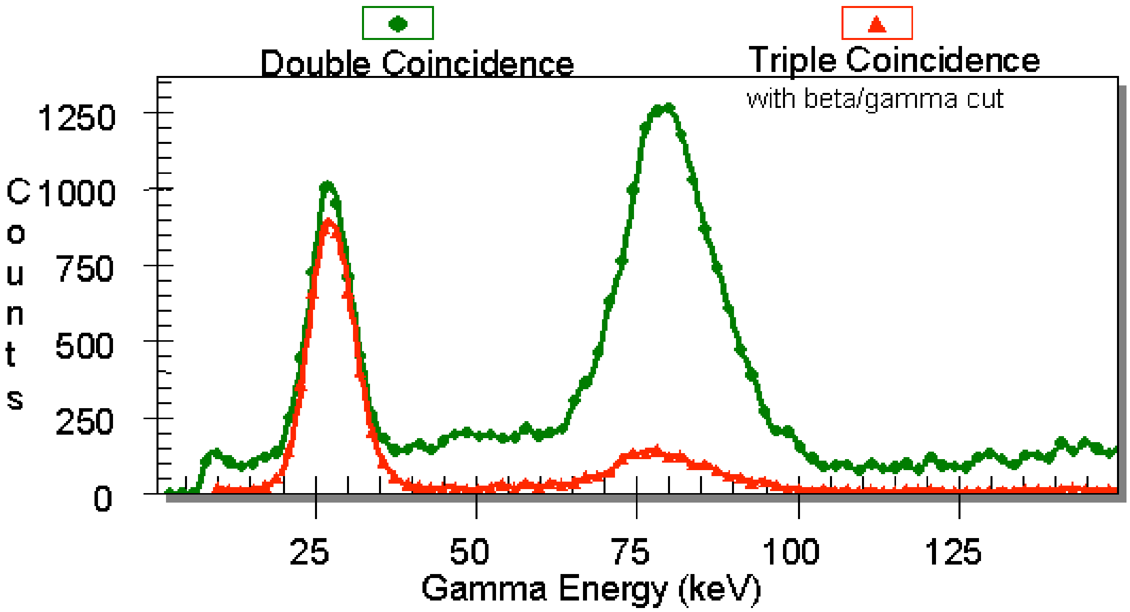

Figure 9. This histogram shows the beta-gated gamma spectrum of a mixed ^{133}Xe and radon spike with equal source strengths of both isotopes.

CONCLUSIONS

From data taken from the test detector it is clear that is possible to detect ^{133}Xe with little to no radon separation and little to no lead shielding. This significant detection enhancement opens the possibility of developing portable systems that can be automated for remote site deployment. Additional R&D will be required to optimize and shrink the current test detector but the fundamental nuclear signatures are present and measurable.

It is important to note that with a properly designed detector, data acquisition system gamma-ray singles, beta singles, (β, γ) coincidences, and (β, CE, x-ray) triple coincidence signals can be detected simultaneously. This increases the ability of the system to detect other radioxenon isotopes (^{131m}Xe and ^{133m}Xe).

Further research to find methods and processes to reduce the current chemical separation systems in size and power consumption would further increase the sensitivity of remotely deployed systems. In this scenario most of the noble gases would be collected to some degree and all would be counted in the nuclear detection system. It is envisioned that such as system would be deployed remotely and not be a hand carried device. Modification to the current test detector to provide a more compact and portable detector would be useful to measure high level radioxenon levels such as above ground nuclear explosions and reactor or criticality accidents.

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