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Comparison of phoswich and ARSA-type detectors for radioxenon measurements

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Abstract The monitoring of atmospheric radioxenon to ensure compliance with the Comprehensive Nuclear Test Ban Treaty (CTBT) has driven the development of improved detectors for measuring xenon, including the development of a phoswich detector. This detector uses only one PMT to detect β–γ coincidence, thus greatly reducing the bulk and electronics of the detector in comparison to the ARSA-type detector. In this experiment, ^{135}Xe was produced through neutron activation and a phoswich detector was used to attain spectra from the gas. These results were compared to similar results from an ARSA-type β–γ coincidence spectrum. The spectral characteristics and resolution were compared for the coincidence and beta spectra. Using these metrics, the overall performance of the phoswich detector for β–γ coincidence of radioxenon was evaluated.

Keywords Radioxenon · β–γ coincidence · ARSA · Phoswich

Introduction

The Comprehensive Nuclear Test Ban Treaty (CTBT) dictates an outright ban on nuclear explosives testing, for any reason. To ensure compliance with this treaty, a verification regime has been established, and as part of this regime, an international monitoring system (IMS) has been developed. The IMS consists of seismic, acoustic, infrasound, and radionuclide monitors worldwide. The radionuclide monitors collect particulates and xenon. Xenon isotopes are produced as fission products and from the decay of fission products in a nuclear explosion, and are difficult to contain due to the mobile and inert nature of the gas. As such, the presence of significant quantities of radioxenon gas can be an indication of a nuclear explosion. However, radioxenon gas can also be produced through other operations, like medical isotope production facilities or commercial reactor operations. To distinguish between these routine operations and a nuclear explosion, the xenon gas samples are analyzed using β–γ coincidence spectroscopy, which utilizes the unique signature of each isotope of xenon to determine the relative concentrations of each isotope, thus giving valuable information about the source of the gas. Because fission gases may travel a significant distance before reaching a detector, they are often present only in small quantities. Thus, the CTBT requires that the radioisotope detectors have a minimum detectable concentration of 1 mBq m^{-3} [1].

To meet this requirement, Pacific Northwest National Laboratory has developed an automated radioxenon sampler/analyzer (ARSA), that collects gas and measures the concentrations of the four prominent isotopes: ^{131m}Xe, ^{133g}Xe, ^{133m}Xe, and ^{135g}Xe. The ARSA detectors are β–γ coincidence detectors that utilize the betas and conversion electrons given off by the various radioxenon isotopes and the subsequent emission of the gamma rays to identify the isotope. The system uses two NaI(Tl) crystals that surround four beta cells. Each NaI crystal and beta cells is connected to a photomultiplier tube (PMT) that detects the light given

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off by the scintillator when struck by a gamma or beta particle, respectively. The signal is then processed using a series of electronics and logic controls to identify pulses from the NaI crystal and beta cells that occur in coincidence, based on the timing between the two events [2]. A 2-dimensional histogram plotting gamma energy versus beta energy and showing coincidence counts is produced, and can be used to uniquely identify each of the four radioxenon isotopes. An ARSA-type detector was constructed at The University of Texas at Austin and is used in this experiment.

While the ARSA-type detector demonstrates good efficiency and resolution, the large number of cells and associated electronics makes the system somewhat cumbersome, and the need to gain match all of the PMTs makes the system more difficult to deploy in an automated setting. In an effort to simplify the system, a phoswich detector has been developed. Like the ARSA-type detector, the phoswich detector uses beta–gamma coincidence spectroscopy to identify radioxenon isotopes; however, it does not use time-based coincidence from separate detectors. Instead, the phoswich detector relies on the use of pulse shape analysis (PSA) based on the fast and slow time characteristics of two different scintillator materials to identify beta, gamma and coincidence events. In the phoswich well design used in this experiment, the sample gas is injected into a cell surrounded by plastic scintillator material and a CsI crystal. A digital signal processor on-board the readout electronics performs the pulse shape analysis using the difference in rise times of the two materials and sums over characteristic intervals of the pulse to classify an interaction as occurring in one or both parts of the detector and to compute the fraction of energy deposited in each scintillator [4]. This information is then binned into a 2-dimensional energy spectrum.

The compact and simplified nature of the phoswich detector increases the ease with which it can be deployed in the field, making it an excellent candidate for use as part of as IMS set-up. In order to use the phoswich detector in this capacity, however, the detector must be tested to ensure that its performance is comparable to that of the ARSA-type detector. The goal of this paper is to compare the performance of the ARSA-type and phoswich detector, with special attention given to the appearance and resolution of the spectra. Spectra of ^{135}Xe were taken with both detectors and spectral characteristics of the beta and coincidence spectra were compared. Finally, conclusions were drawn about the performance of the phoswich detector.

Experimental set-up

The radioxenon gas used in this experiment was produced at The University of Texas' nuclear engineering teaching laboratory (NETL) by activating stable enriched xenon gas in the 1.0-MW TRIGA reactor. To produce ^{135}Xe, ^{134}Xe was irradiated in the reactor and underwent a neutron capture reaction. The stable gas is enriched to 99.61% ^{134}Xe. The sample used in the ARSA-type detector was irradiated in beam port 2 (BP2), which is tangential to the core. The gas samples used for the phoswich detector were produced using in-core irradiation, irradiated in the lead-lined 3L facility, which lies within the 7L facility. Irradiation times and powers were adjusted to account for differences in fluxes to produce similar activities. The fluxes, irradiation times, and irradiation powers are listed for both irradiations in Table 1. Figure 1 shows a schematic of the reactor core and labels BP2 and the 7L facility. The samples irradiated in BP2 were irradiated in a 7-cm long hollowed aluminum vessel that was machined at NETL. This vessel holds approximately 1.75 cc of gas. The aluminum tube was connected to a Teflon Swagelok valve that was used in the gas transfer set-up. Due to the small size of the 3L, the gas sample produced for the phoswich run could not be irradiated in the aluminum vessel, but was instead irradiated in a small Teflon Swagelok valve with an end cap (valve part # PFA-43S4, end cap part #PFA-420-P). This allowed for a

Table 1 Irradiation time, power and fluxes

Detector	Isotope	Power (kW)	Time	Flux (n/cm^2/s)
ARSA	^{134}Xe	950	8 h	1×10^8
Phoswich	^{134}Xe	100	15 min	1×10^{11}

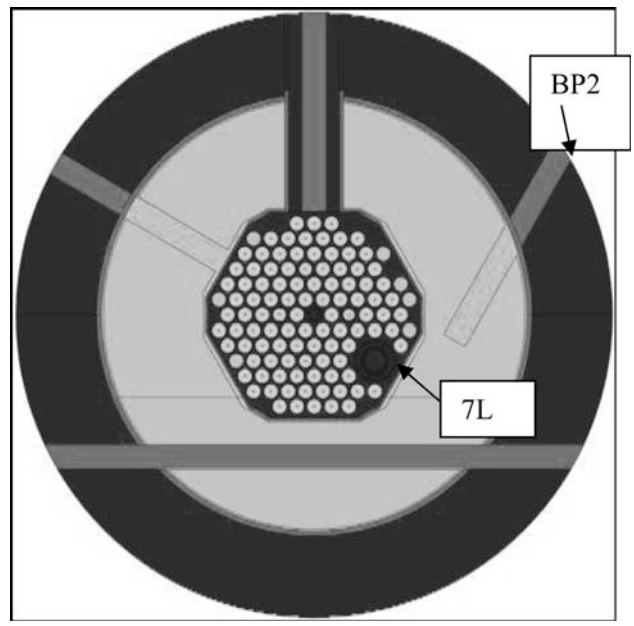

Fig. 1 Diagram of NETL reactor core showing 7L and BP2 locations

volume of approximately 0.5 cc of gas to be irradiated, producing ample activity to be measured.

Gas is transferred to the vessels for irradiation using the gas transfer set-up designed and built at NETL [3]. The set-up features thin Swagelok piping arranged in a cross shape with four connections. One side is connected to a pressure regulator, which then connects to the cylinders with stable xenon gas. A second side connects to a rubber hose that is attached to a vacuum line. The third and fourth sides remain open for the attachment of a detector and the gas sample vessel. To transfer gas, the sample vessel is attached, and a vacuum is pulled on the whole system, until the regulator indicates that negative pressure has been achieved. The vacuum is then shut off and the valve to this line closed, and the gas cylinder is opened, allowing gas to transfer to the sample chamber. Similarly, once the sample has been irradiated and gas must be transferred to the detector, the detector and gas sample vessel are both attached to the gas transfer apparatus. A vacuum is then pulled on the entire system, including the detector to ensure that as much residual gas as possible is removed from the detector. The vacuum valve is then closed and the detector and sample valves are opened, permitting gas to flow from the sample chamber to the detector. Because this transfer is of a volumetric nature, every effort was made to minimize the volume of the transfer apparatus; that is, short piping with a small diameter was used to allow for maximal transfer of sample to the detector.

The ARSA-type detector system used in this experiment contains two NaI crystals and four beta cells, like the ARSA system. The beta cells are inserted into holes in the NaI assembly. Each beta cell is connected to two PMTs, as is each gamma cell, for a total of 12 PMTs. The entire apparatus is cased in lead housing to reduce background from cosmic radiation. The data processing was done by a nuclear instrumentation methods (NIM) component and the computer-detector interface was controlled by computer automated measurement and control (CAMAC) components. Gain matching was performed for the beta and gamma cells and the energy was calibrated using standard sources [3].

The phoswich detector used was developed by XIA LLC. It is a well detector with a plastic BC-404 capsule that holds the xenon gas sample, embedded in a 3-inch CsI(Tl) crystal. A filling tube extends through the crystal to the sample chamber [4]. The entire detector is surrounded by a 1/8″ thick copper housing. The BC-404 cell itself is an elongated sphere featuring two hemispheres (outer radius 11.5 mm) connected by a 5-mm-long cylinder (outer radius 11.5 mm). The wall thickness is 2 mm and the cell has an inside volume of 5.009 cm^3 [5]. Beta particles are mainly absorbed in the BC-404, while gamma particles are mainly absorbed in the surrounding CsI crystal. A single PMT is attached below the CsI and is connected to an XIA DGF Pixie-4 pulse processor [6]. Pulse shape analysis is performed by the Pixie-4's on board DSP and results are read out with an embedded CompactPCI/PXI computer. Data analysis was done in an Igor Pro 6.01 interface designed by XIA for use with the detector. The specific model used in this experiment is the XIA PW6 with an R1307 PMT. According to work conducted by XIA, both the CsI crystal and the PMT in this particular detector have a small amount of ^{40}K contamination, obtained during the manufacturing process. Chemical analysis revealed the concentration in the crystal to be around 130 ppm, and activity calculations confirmed this value. The result of said contamination is a 1,460 keV gamma peak, and in some cases a Compton background on the vertical axis of the 2-dimensional spectrum [7]. Evidence of this contamination did appear in the data gathered at low count rates.

Results

While the stable xenon gas that underwent irradiation was highly enriched in ^{134}Xe (99.61%), it still contains small quantities of the other xenon isotopes, most notably ^{132}Xe, which is present in 0.291% abundance. Thus these other isotopes are irradiated along with the ^{134}Xe, making the sample slightly impure. This impurity is augmented by the short-lived nature of ^{135}Xe, as it is the shortest lived of the radioisotopes with a half life of 9.14 h. Consequently, the activity of ^{135}Xe only dominates the sample for approximately 4.5 days, at which point ^{133}Xe decay begins to dominate the sample activity. The activity of various isotopes of xenon over time is shown in Fig. 2. Due to the transient nature of ^{135}Xe, data used in this experiment to characterize the detectors was taken from the first day of data collection to ensure that adequate ^{135}Xe was present.

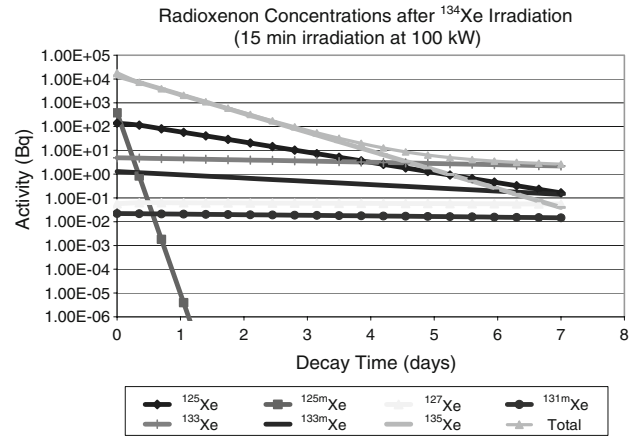

Fig. 2 Activity contributions from radioxenon isotopes

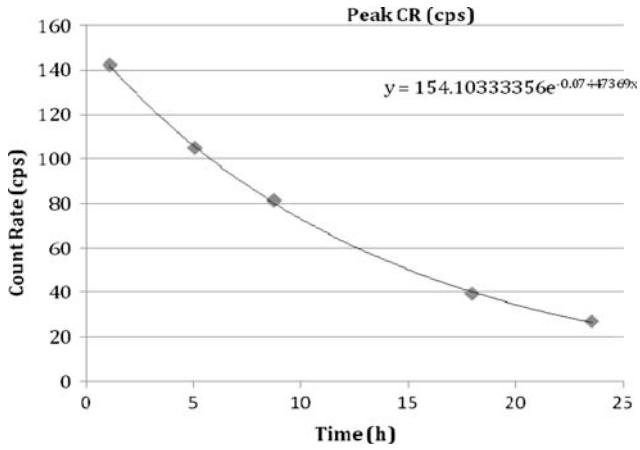

Fig. 3 Count rate of ^{135}Xe versus time

To verify that the isotope produced and detected was, in fact, ^{135}Xe, a half-life measurement was made using the raw phoswich data. The count rate in the peak region was calculated at five points over the course of a day and plotted versus time. The resulting graph is presented in Fig. 3. An exponential curve fits yields the equation shown on the graph, with a decay constant $\lambda = 7.45 \times 10^{-2}$ h^{-1}. This equates to a 9.31 h half-life, which is within 2% of than the known half-life of ^{135}Xe, 9.14 h. Thus the count rate data collected by the phoswich detector confirms the presence of ^{135}Xe with a high degree of accuracy.

The ^{135}Xe spectra from both the ARSA-type and phoswich detectors are shown below in Fig. 4. Both spectra feature the characteristic ^{135}Xe peak at 250 keV, and also show peaks at 81 and 30 keV. The 81 keV peak is a gamma peak from ^{133}Xe, while the 30 keV peak is an X-ray. The two spectra are comparable; however, notable differences do exist. The 250 keV peak on the phoswich spectrum appears wider than the comparable peak on the ARSA spectrum. The full width at half maximum (FWHM) of the 250 keV peak on the phoswich detector is 30 keV, resulting in a resolution of 12%. For the ARSA detector, the FWHM of the 250 keV peak is about 24 keV, making the resolution 9.6%. Thus the ARSA detector provides slightly better resolution. The 81 keV gamma peak is also far narrower and clearer on the ARSA-type detector than the phoswich detector. Another prominent feature of the two spectra is the difference in beta distribution. The beta counts on the phoswich detector appear to be clustered at lower energies, and the tailing off of the beta distribution as the energy reaches a maximum (910 keV) is less dramatic, as well.

The differing beta distributions between the two plots is even more evident in the surface plots, shown in Fig. 5. The

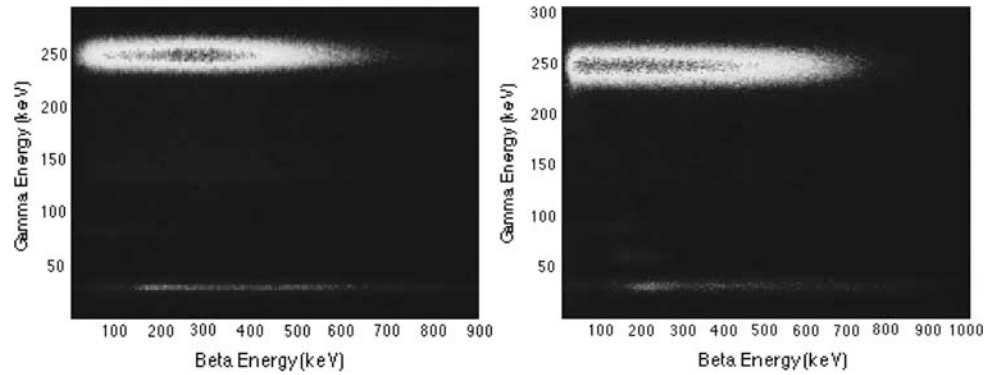

Fig. 4 ^{135}Xe spectra from the ARSA-type and phoswich detectors

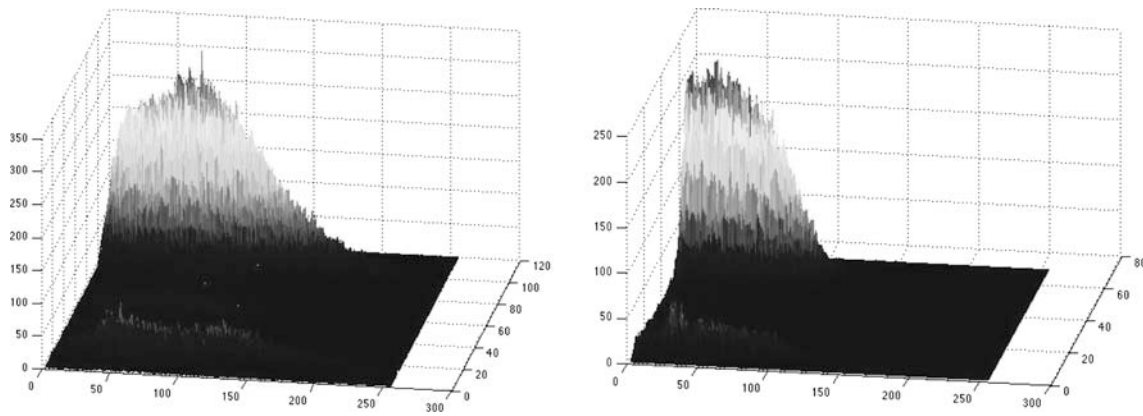

Fig. 5 3D surface plots of ^{135}Xe from ARSA-type and phoswich detectors

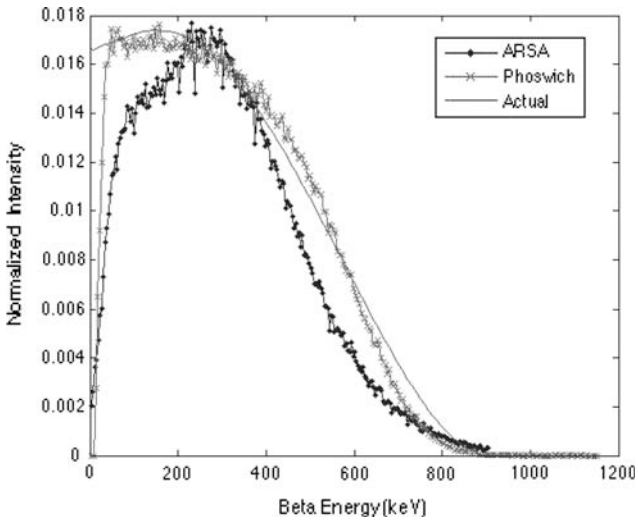

Fig. 6 Theoretical, ARSA-type and phoswich distributions

vertical axis is a measure of counts, while the x and y axis show beta and gamma channel, respectively. From these graphs it is clear that the beta distribution is wider and falls off more slowly for the ARSA-type detector for both the 250 keV and 30 keV peaks, while the distribution for the phoswich detector rises and falls more sharply. The actual beta distribution for the 250 keV region of interest is shown in Fig. 6. Also plotted in Fig. 6 are the beta curves for the ARSA-type and phoswich detectors. The shape of the phoswich curve is very close to the true beta distribution. The peak for the ARSA spectrum is around 300 keV, which is higher than for the true distribution and the phoswich spectrum. This difference in peak energy is due to the difference in thicknesses of the BC 404 scintillating material. The phoswich detector has a thicker beta cell, allowing all radioxenon beta particles to be stopped in the cell. In the ARSA detector, the plastic is not thick enough to completely stop all high energy beta particles; instead, the particles partially deposit their energy in the cell resulting in a maximum energy deposit of around 300 keV for a perpendicularly incident beta. This leads to an artifact peak at 300 keV that appears in the ARSA beta distribution.

Conclusions

The data collected in this experiment indicate that spectra collected with the phoswich detector are comparable to those collected by the ARSA-type detector. While the resolution was slightly lower, the peaks did appear where expected and the count rate data did result in an accurate calculation of the half life of ^{135}Xe. A surprising difference between the detectors was the shape of the beta distributions. Since both detectors utilize the sample BC-404 material for beta detection, this difference may be the result of the differing thicknesses. The phoswich detector also offers the significant advantage of ease of use and transportability. Because the well design features only one PMT and does not depend on time coincidence, it eliminates the need for gain matching and greatly reduces the physical size of the detector, making it easier to deploy in the field. The set-up for this detector is also far simpler than the set-up for the ARSA-type detector, though it still requires some degree of familiarity with the detector to optimize settings. Overall, the phoswich detector proves to be a promising new detection technology that could be employed as part of the IMS for radionuclide monitoring.

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