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Title: ^{135}Xe Measurements with a Two-element CZT-based Radioxenon Detector for Nuclear Explosion Monitoring

^{135}Xe Measurements with a Two-element CZT-based Radioxenon Detector

Abstract

Measurement of elevated concentrations of xenon radioisotopes ($^{131\text{m}}\text{Xe}$, $^{133\text{m}}\text{Xe}$, ^{133}Xe and ^{135}Xe) in the atmosphere has been shown to be a very powerful method for verifying whether or not a detected explosion is nuclear in nature. These isotopes are among the few with enough mobility and with half-lives long enough to make their detection at long distances realistic. Existing radioxenon detection systems used by the Comprehensive Test Ban Treaty Organization (CTBTO) suffer from problems such as complexity, need for high maintenance and memory effect. To study the response of CdZnTe (CZT) detectors to xenon radioisotopes and investigate whether it is capable of mitigating the aforementioned issues with the current radioxenon detection systems, a prototype detector utilizing two coplanar CZT detectors was built and tested at Oregon State University. The detection system measures xenon radioisotopes through beta-gamma coincidence technique by detecting coincidence events between the two detectors. In this paper, we introduce the detector design and report our measurement results with radioactive lab sources and ^{135}Xe produced in the OSU TRIGA reactor. Minimum detectable concentration (MDC) for ^{135}Xe was calculated to be $1.47 \pm 0.05 \text{ mBq/m}^3$.

Keywords: Radioxenon Detection; Nuclear Explosion Monitoring; Beta-Gamma Coincidence; CZT; Coplanar Detector; Digital Pulse Processing

1. Introduction

Several noble gas radioisotopes are released as by-products of nuclear explosions. Many of them have short half-lives and will quickly decay to sub-detectable levels after the explosion. However, four radioxenon isotopes, ^{131m}Xe ($t_{1/2} = 11.93$ d), ^{133m}Xe ($t_{1/2} = 2.19$ d), ^{133}Xe ($t_{1/2} = 5.25$ d) and ^{135}Xe ($t_{1/2} = 9.14$ h), have relatively large fission yields and long half-lives which make their detection at long distances several days or even months after the release to the atmosphere realistic. Table 1 lists characteristic energies for the decay of these radioxenon isotopes [1].

The International Monitoring System (IMS) has been established to install and employ radioxenon detectors in various locations around the globe to measure atmospheric concentration of these radioxenon isotopes for any signs of nuclear weapon tests. These stations have been able to detect signs of nuclear explosions in the past. Following the Fukushima nuclear accident, ^{133}Xe was detected only four days after the incident at Pacific Northwest National Laboratory (PNNL) [2], 16 days after the accident in Xi'an, China [3] and one month after the accident at the Darwin IMS noble gas station in Australia [4]. ^{131m}Xe and ^{133}Xe were also detected 11 days after the accident in Beijing [5].

The announced October 2006 North Korean nuclear test was also confirmed by radioxenon detection 12 days after the test in Yellowknife, Canada [6]. It was the first real test showing the technical capabilities of the verification system established by the CTBTO [7].

The North Korean nuclear weapon tests in 2010 and 2013 were also confirmed by radioxenon monitoring in various IMS stations. In 2010 multiple detection of radioxenon isotopes at various IMS stations in Eastern Asia (two in Japan and one in Russia) also indicated that a possible low-yield nuclear test had been conducted by North Korea [8], [9]. Observation of rapid changes in the ratios of ^{133}Xe and ^{131m}Xe at measurement stations in Japan and Russia indicated that they belonged to the underground nuclear test conducted by North Korea, 7-8 weeks prior (on February 12, 2013) [10], [11].

Four different radioxenon detection systems are currently deployed at various IMS stations to monitor atmospheric concentrations of xenon radioisotopes. These detection systems work either based on beta-gamma coincidence technique (ARSA (USA), SAUNA (Sweden) and ARIX (Russia)) [12]–[14] or high-resolution gamma-ray spectroscopy (SPALAX (France)) [15].

Table 1 Half-lives and characteristic energies for the decay of ^{131m}Xe , ^{133m}Xe , ^{133}Xe , and ^{135}Xe [1].

Nuclide	^{131m}Xe	^{133m}Xe	^{133}Xe	^{135}Xe
Half-life	11.93 d	2.19 d	5.25 d	9.14 h
Gamma-rays (keV)	163.9	233.2	81.0	250.0
Gamma-ray abundance (%)	1.96	10.3	37.0	90.0
X-ray, K-shell (keV)	30.	30.	31.	31.
X-ray abundance (%)	54.1	56.3	48.9	5.2
Beta, Max. Energy (keV)	-	-	346.	905.
Beta abundance (%)	-	-	99.	97.
CE, K-shell (keV)	129.	199.	45.	214.
CE abundance (%)	60.7	63.1	54.1	5.7

ARSA, SAUNA and ARIX detection systems employ plastic and inorganic scintillators. In these systems, the plastic scintillator is used for detecting beta and conversion electrons. The plastic scintillator is surrounded by a NaI (TI) scintillator, which detects gamma and x- rays. While beta-gamma coincidence detection improves the sensitivity of these detection systems compared to the high resolution gamma spectroscopy method, these systems suffer from problems such as complexity, bulkiness, and memory effect due to the use of plastic scintillators. Systems using high-resolution HPGe detectors, namely the SPALAX system, require regular maintenance due to the cryogenic cooling system, and are therefore not well suited to unattended applications.

To address these issues, several detection systems have been developed during the past decade [1], [16]–[23]. These systems were able to address some of the aforementioned issues while still meeting the IMS requirements. To further address these issues a new radionuclide detection system was designed and developed at OSU based on the well-known room temperature semiconductor material CdZnTe (CZT).

CZT detectors are capable of superior energy resolution compared with other scintillator-based beta-gamma coincidence detectors currently in operation. Another advantage of CZT detectors is their capability to be operated at room temperature, making the system lower maintenance compared to radionuclide detection systems relying on the maintenance-intensive, cryogenically-cooled HPGe detectors.

The new CZT-based radionuclide detection system is also expected to significantly reduce the memory effect [24] which is currently observed with radionuclide detectors that use plastic scintillators. The high-Z nature of the CZT material is anticipated to minimize radionuclide diffusion into the detector elements compared to the low-Z nature of plastic scintillator materials. The reduction in memory effect for the CZT-based detection system should improve the Minimum Detectable Concentration (MDC) of this device.

The prototype system introduced in this work uses only two CZT detectors. This prototype is only meant to characterize the response of the system to coincident electron and photon emissions from radionuclides, as opposed to a fully-populated system that would more completely cover the 4π solid angle of sample emissions from all radionuclides of interest. This paper mainly covers the system's design, its characterization and measurement results for ^{135}Xe . The MDC of this radionuclide was also estimated and will be discussed in more detail. Other radionuclide's measurements can be found elsewhere [25].

2. Detector Geometry

2.1. Two-Element Coplanar CZT detector (TECZT)

The prototype TECZT system uses two coplanar CZT crystals from Redlen technologies [26] each with dimensions of $10\times 10\times 10\text{ mm}^3$. The use of CZT for spectroscopy necessitates the implementation of a single charge carrier anode configuration to overcome loss of charge carriers and degradation of energy resolution due to the low mobility-lifetime of holes. Among the different single carrier implementations, the coplanar grid technique [27] was chosen for our CZT detectors over a pixelated anode design because the coplanar design requires fewer signal channels and no imaging is necessary for measuring radionuclides.

In coplanar detectors, the two separate anode grids form collecting and non-collecting electrodes. By applying a slightly different bias voltage between these two grids, a strong electric field is generated near the anode surface. This electric field only affects the electrons when they are in close proximity to the anode. The difference in voltage between the collecting and non-collecting grids pulls electrons toward the collecting grid and pushes them away from the non-collecting grid. The coplanar anode design also almost completely eliminates the signal contribution from the slow-moving, more frequently trapped holes. The anode side of one of the coplanar CZT crystals used in this work is shown in Fig. 1B). The anode strips and pitch sizes are both $263\text{ }\mu\text{m}$.

Fig. 1. A) Illustration of the TECZT radioxenon detector design; B) Photograph of the anode side of a CZT crystal from Redlen Technologies used in the TECZT radioxenon detector showing the coplanar anode pattern; C) assembled and sealed TECZT radioxenon detector showing the two CZT crystals installed on either side of the gas cell.

2.2. Detector Assembly

The coplanar CZT detectors were installed face-to-face on opposite sides of the detector gas cell (Fig. 1C). The gas cell was 3-D printed at OSU using polylactic acid (PLA), a commonly used desktop 3D printing material [28]. The opening around crystals was sealed using a high vacuum sealant (Torr Seal) to avoid leakage of radioxenon [29].

The CZT crystals were mounted in close proximity to the PCBs in order to overcome noise issues associated with long leads between the anodes and the preamplifiers. The TECZT detector assembly and the preamplifier and subtraction circuit PCBs were mounted inside an EMI shielded enclosure, shown in Fig. 2A), to reduce noise.

Fig. 2. The full TECZT radioxenon detection system and electronics, showing A) the radioxenon sample syringe, gas injection tube, TECZT detector assembly, preamplifiers, and subtraction circuits, and B) the two-channel digital pulse processor.

3. Measurements and Methods

3.1. Characterization with Radioactive Lab Sources

General and coincidence characterization of the TECZT detector was performed using laboratory sources. A FPGA-based, custom two-channel digital pulse processor (DPP), designed and built at OSU, was used to process pulses from the TEZCT detector (Fig. 2B). Detector pulses were sampled using a 200 MHz, 12-bit resolution

ADC on each channel. Digital trapezoidal shaping was performed in MATLAB. The gain matching method [30] was used to compensate for electron trapping inside the CZT crystals. Different subtraction gains and different electrode biasing voltages were set for the detector and the energy resolutions of different radioactive laboratory sources were measured in order to find the optimal settings for the system. The best cathode and grids bias voltages were found to be -1000V and +80V, respectively.

In the TECZT system, coincidence detection was performed in real time in the FPGA. Coincidence events are captured and transferred to the PC when the two detectors are triggered within Coincidence Time Window (CTW), which is adjustable from 0 to 1.275 μ s with 5 nsec steps (ADC's sampling period). Coincidence Detection Unit continuously monitors trigger signals from each channel and issues a one-shot signal (coincidence ready) when both channels have been triggered within the CTW. A simplified diagram of the FPGA design is shown in Fig. 3.

Since the arrival time of pulses from a coplanar CZT detector is a function of the interaction depth, an experiment was conducted to investigate the time range of the coincidence events. A ^{60}Co source was placed on the top of the gas cell (Fig. 4A) and the rate of the coincidence events from the simultaneously-emitted 1.17 and 1.33 MeV gamma rays was recorded versus the coincidence time window set in the FPGA. ^{60}Co emits two cascading gamma-rays (1.17 and 1.33 MeV) and thus is well suited for this experiment.

Fig.3. Diagram of the coincidence firmware in the FPGA.

Fig. 4B shows the results of these measurements. As shown in this figure, the coincidence count rate reaches a flat region at about 0.75 μ s. This is close to the maximum electron drift time in this CZT crystal with detector biasing of 1000 V ($\sim 1 \mu$ s) [31]. For the rest of the measurements in this work, the CTW was set to 0.75 μ s.

Fig.4. (A) Geometry of the measurements used to find the optimum Coincidence Time Window (B) Coincidence count rate versus Coincidence Time Window measured using ^{60}Co .

A trapezoidal filter with rise time of $0.75\ \mu\text{s}$ and flat top of $1.0\ \mu\text{s}$ was used to process both single-event pulses and double-event pulses. Double-event pulses were included as valid events since a photon could undergo a Compton scatter and then a photoelectric event in the same CZT detector. The trapezoidal filter peaking and flat top times were chosen to generate a filter response such that a Compton-photoelectric event pair would be included as a valid pulse by the triggering and pileup rejection firmware modules. Fig.5 shows a double-event pulse captured by the DPP. The response of a trapezoidal filter, with a peaking time of $0.5\ \mu\text{s}$ and a flat top of $1.0\ \mu\text{s}$, is also shown in Fig. 5.

After finding the optimum operating conditions for the TECZT detector, radioactive lab sources were used to characterize the energy resolution of the system. Fig. 6 and 7 show energy spectra of ^{137}Cs and ^{60}Co collected from the two CZT detectors.

Fig. 5. A double-event pulse and the response of a trapezoidal filter with peaking and flat times of 0.5 μ s and 1.0 μ s, respectively.

The best energy resolutions obtained for 662 keV were 2.3% and 2.7% FWHM for CZT 1 and CZT 2, respectively.

In order to get the best energy resolutions, detectors must be irradiated from the cathode side of each CZT crystal inside of the gas cell. Since such a setup was not possible with the assembled gas cell, the sources were placed at the top of the gas cell, as shown in Fig. 4. Additionally, while CZT 1 shows better overall performance compared to CZT 2, the channel connected to CZT 2 shows better electronic performance (11.1 keV FWHM pulser resolution for CZT 2 vs. 11.6 keV FWHM for CZT 1).

It is worth mentioning that 31 keV peak from ^{137}Cs is not visible in energy spectra due to creating pulses whose amplitudes were below the noise level of the prototype readout electronics. To enhance the signal-to-noise ratio and making our detection system capable of detecting 31 keV from ^{133}Xe , $^{133\text{m}}\text{Xe}$ and $^{131\text{m}}\text{Xe}$, the hardware and firmware of our system were modified to allow detecting of 31 keV. For firmware modification, the fast trigger filter was replaced with a longer filter in FPGA device to have a better discrimination between the low-energy-triggered events and noise-triggered events.

Fig.6. Energy spectrum of ^{137}Cs from CZT 1 and CZT 2.

Fig.7. Energy spectrum of ^{60}Co from CZT 1 and CZT 2.

3.2 Radioxenon Measurements

3.2.1. ^{135}Xe

To produce ^{135}Xe stable and highly enriched (>99%) ^{134}Xe was first drawn into a polypropylene syringe using a simple hand-pumping system. The polypropylene syringe was chosen because of its low activity after neutron irradiation. The syringe was then placed in the Thermal Column of the OSU TRIGA reactor with a flux of $\sim 7 \times 10^{10} \text{ n/cm}^2 \cdot \text{s}$ for seven hours. Then the activated xenon gas was injected into the detector gas cell following a cooling time of about 17 hours.

Before starting data collection, the FPGA firmware was set to coincidence mode, in which only coincidence events can trigger the TECZT system. A 2D coincidence energy spectrum from ^{135}Xe collected by the TECZT detector is depicted in Fig. 8.

As can be seen, beta-gamma coincidence events from ^{135}Xe are dominated by two crossing lines at 250 keV extending from zero to the maximum energy of the beta distribution (905 keV) in one detector and a constant 250 keV energy (gamma-ray photopeak) in the other detector. A diagonal line in this 2D energy spectrum indicates full energy depositions from Compton events between the two detectors. Events on this line have summation energy equal to the energy of gamma ray emitted from ^{135}Xe (250 keV). This is an indication of the proper functioning of the coincidence mode in the TECZT detector when ^{135}Xe is injected into the gas cell. Fig. 9 shows the 2D energy spectrum when the FPGA's firmware was set to the free running mode. In this mode, regardless of arrival times, all the events were collected. Two spectral differences between the coincidence and free-running modes can be identified by comparing Fig. 8 and Fig. 9. First, in Fig. 9, the 250 keV lines extend to energies beyond the maximum energy of the beta particles (905 keV), which is to be expected since in free running mode the two particles can be absorbed in a single detector. Second, the diagonal energy line in Fig. 8 generated by Compton scatter events between the two detectors is not present in the free running spectrum in Fig. 9 since the probability of capturing coincidence events in the free running mode is very low.

Fig.8. 2D coincidence energy histogram from ^{135}Xe collected by TECZT and DPP2.

Fig.9. 2D energy histogram from ^{135}Xe collected by TECZT and DPP2 in free running mode (non-coincidence).

Fig. 10 shows the energy spectrum of ^{135}Xe in each CZT detector when the system was set to the coincidence mode. In both spectra, the 250 keV photopeak is superimposed on the beta distribution which extends up to the maximum beta energy of 905 keV. The energy resolution of the 250 keV photopeak was measured to be 4.4% and 4.7% FWHM for CZT 1 and CZT 2, respectively.

Table 2 shows a comparison of the energy resolution for different energies from the radioactive lab sources and ^{135}Xe . The TECZT detector shows better energy resolution for all of the energies listed compared to the other available beta-gamma coincidence-based radioxenon detection systems, a direct result of using CZT detectors instead of scintillators. In addition, the background count rate for both total and coincidence events is significantly lower compared to the other detection systems, which should result in comparable or better MDC compared to other designs.

To increase the solid angle of the present system to almost 4π and therefore improve the detection efficiency of the system, the gas cell will be modified to accommodate six CZT crystals in a cubic formation. This modification is predicted to improve the geometric efficiency of the system by a factor of 15 to be about 83%.

Fig.10. Energy histogram, in coincidence mode, for each CZT detector from ^{135}Xe .

Table 2 Comparison of energy resolutions of TECZT detection system with current radioxenon detection systems work based on beta-gamma coincidence for different energies.

	Gamma Energy (keV)	TECZT (this work)	WASPD [23]	Phoswich [1]	SAUNA [13]	ARSA [31]	BGW [21]
Energy Resolution (FWHM, %)	88 (^{109}Cd)	11.2	31.5	25	14	NA	14
	122 (^{57}Co)	6	NA	24	NA	22	13
	662 (^{137}Cs)	2.3	13.6	8.9	7.3	12	8.7
	250 (^{135}Xe)	4.4	19.3	13	NA	9.6	NA
Background Rate (counts/s)	Total (all events)	0.2	1.26	3.29	7.5-12	30	5.5
	Coincidence Events	0.0036	0.02	0.06	0.03	0.1	0.025

4. Minimum Detectable Concentration

Radioxenon detection systems must be sensitive enough to detect trace amount of xenon gases to be eligible candidates for integration into the IMS. International Monitoring System requires that the Minimum Detectable Concentration (MDC) of all radioxenon detection systems must be less than or equal to 1 mBq/m³ for ¹³³Xe [32].

In this work, MDC of ¹³⁵Xe has been calculated. In general, the MDC can be calculated using the following equation:

$$MDC_{mBq/m^3 air} = 2.71 + 4.65 \sigma_0 \epsilon_\beta \epsilon_\gamma \beta_{BR} \gamma_{BR} \lambda^{21} \exp(-\lambda T_c) (1 - \exp(-\lambda T_p)) (1 - \exp(-\lambda T_A)) T_C * 1000 V_{air} \quad (1)$$

Where:

σ_0 : [Background counts + ($\sigma_{Interferences}$)²]^{1/2}

ϵ_β : β Efficiency

ϵ_γ : γ Efficiency

β_{BR} : β Branching Ratio

γ_{BR} : γ Branching Ratio

λ : Decay Constant (s⁻¹)

T_c : Xenon collection time (s)

T_p : Processing Time of Gas (s)

T_A : Acquisition Time of Counts (s)

V_{air} : Sampled Air Volume (m³)

The first term in Eq. (1) takes into account the traditional sensitivity expression using the standard deviation of the measured background and interference counts (σ_0) with the beta and gamma detector efficiencies and branching ratios respectively (ϵ_β , ϵ_γ , β_{BR} , γ_{BR}). The second term accounts for the decay of the xenon isotope during the collection time T_c , the processing time T_p , and the nuclear acquisition time T_A using the decay constant for the specific isotope λ . Finally, the third term accounts for the volume of air measured V_{Air} and the conversion to milliBecquerels.

Several factors can affect the MDC of radioxenon detection systems such as the background counts and memory effect from the previous sample measurements. Background itself is affected by both radioxenon itself for the 30 keV region and ²²²Rn daughters (²¹⁴Pb and ²¹⁴Bi) for the 81 keV and 250 keV regions. Therefore, to accurately calculate the MDC for a given radixenon isotope in a specific detector a careful accounting of the various backgrounds and interference terms is necessary.

In general σ_0 in the MDC equation can be calculated using [33]:

$$\sigma_0 = \sqrt{BckCnt_{total}^2 + \sigma_{BckCnt}^2 + InterferenceCnt + \sigma_{InterferenceCnt}^2 + MemoryCnt + \sigma_{MemoryCnt}^2} \quad (2)$$

In this work, $BckCnt_{total}$ in the coincidence region of interest (ROI) was calculated using the method described in [34]. Using this method $BckCnt_{total}$ can be defined as the product of four terms:

$$BckCnt_{total} = [S_{Bck(ROI)}] CTWTATB [N_{cp}] \quad (3)$$

1- $S_{Bck(ROI)}$: Summation of the background count rates in CZT1 and CZT2 for each ROI

2- CTW: The Coincidence Time Window = 1 μ s for TECZT

3- T_A : Acquisition Time of Counts = 24 hrs

3- T_B : The Background Measurement Time = 48 hrs for TECZT

4- N_{cp} : Number of Coincidence Permutations = 2 for TECZT

To find the associated background counts in CZT1 and CZT2, a 48-hour background measurement was performed in coincidence mode. The ROI for the ^{135}Xe were defined based on its coincidence decay emissions (250 keV gammas in coincidence with 905 keV betas) and using 2 times the FWHM of the energy resolution for that ROI.

To estimate MDC, the following assumptions and conditions were considered:

(1) For simplicity, background counts due to interferences, such as those from radon daughters and their associated variance, were ignored ($InterferenceCnt + \sigma InterferenceCnt = 0$);

(2) Since we anticipate that our detection system doesn't show a significant memory effect, counts related to memory effect and its associated variance were also ignored ($MemoryCnt + \sigma MemoryCnt = 0$);

(3) Typical values from the ARSA system [35] for xenon collection time ($T_C = 8$ h), processing time ($T_P = 5.45$ h), and acquisition time ($T_A = 24$ h) were used;

(4) The products of the branching ratios ($\beta_{BR} \cdot \gamma_{BR}$) were taken from literature (%90 for the coincidence decay emissions of 250 keV gammas with 905 keV betas from ^{135}Xe) [36];

(5) The typical air volume V_{air} (20 m^3) is given by the Xe collection system which is a distinctive value in monitoring stations and independent of the detector [34]. However, this parameter was adjusted by a factor equivalent to the ratio of active gas volume in the TECZT and ARSA ($1 \text{ cm}^3/6.14 \text{ cm}^3$) to account for differences in active gas volume in the two detectors. Therefore, a value of 3.26 m^3 was used for V_{air} in the MDC formula;

(6) The beta-gamma coincidence detection efficiency ($\epsilon_{\beta, \epsilon_{\gamma}} = 0.032$) of the TECZT for ^{135}Xe ROI was estimated by performing a Monte Carlo analysis using the MCNP6 code.

Based on the aforementioned assumptions, MDC of ^{135}Xe for TECZT was calculated to be $1.47 \pm 0.05 \text{ mBq/m}^3$. One way to improve the MDC of the TECZT system is to enhance its geometric detection efficiency by increasing the number of CZT crystals to six. Our calculations show that increasing the number of CZT crystals to six will improve detection efficiency of the system by a factor of 15. Therefore, MDC of ^{135}Xe is estimated to be 0.09 mBq/m^3 for the 6-CZT version.

Another improvement in the MDCs of the CZT-based systems can be achieved by using a larger gas cell volume. In future work, $20 \times 20 \text{ mm}^2$ CZT crystals will be used in order to accommodate an 8 cm^3 gas cell volume. The increase in gas cell volume (V_{air}) while maintaining a high geometric efficiency will provide a factor of 8 improvements in MDC compared to the current 1 cm^3 gas cell volume.

5. Conclusion

In this work, a two-element detection system was designed and built to study the response of CdZnTe detectors to xenon radioisotopes through beta-gamma coincidence counting. A custom algorithm was developed to capture beta-gamma coincidence events in a FPGA device in real-time. The coincidence detection system was tested using lab sources and ^{135}Xe produced in OSU TRIGA reactor. Our measurement results show significant improvements in the energy resolution and background count rates. The energy resolution of 250 keV from ^{135}Xe was measured to be 4.4% FWHM, which shows substantial improvement compared to other beta-gamma coincidence detection systems. MDC of ^{135}Xe for this system was calculated to be $1.47 \pm 0.05 \text{ mBq/m}^3$. Based on the MDC results of the prototype two-element system, it was found that a 6-CZT design will have MDCs of less than 1 mBq/m^3 for all radioxenons. Further improvements to the general design can be made by using a larger gas cell volume and enhancing its geometric detection efficiency by increasing the number of CZT crystals to six.

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