MONTE CARLO ANALYSIS OF AN ORGANIC GLASS BETA CELL FOR RADIOXENON DETECTION

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ABSTRACT – Detection of radioactive xenon in atmospheric samples is the only technology in the International Monitoring System that can unambiguously identify a detected event as nuclear. Currently deployed systems use beta-gamma coincidence detection to identify the various xenon isotopes in atmospheric gas samples. These systems use plastic scintillator to detect the beta particles and sodium iodide to detect the correlated X-rays and gamma rays. The plastic scintillator has hollow, cylindrical cell geometry with the atmospheric sample inside to create a near- 4π detection geometry. One limitation of plastic scintillator is the so-called "memory effect", whereby radioactive xenon diffuses into the plastic during the measurement; this effect raises the background for subsequent measurements. Stilbene organic scintillators have been previously explored to mitigate the memory effect but suffers from machining difficulty due to its brittle nature. Here, we explore an organic glass scintillator that was recently develop by Sandia National Laboratories as an alternative material for the organic beta detection cell. This material has improved brightness compared to plastic and stilbene and is very easy to machine. The glass can be easily melted in a laboratory and cast into various shapes using custom molds. An MCNP model of a radioxenon detection system based on organic glass and sodium iodide has been developed. This model will be used to conduct a simulation study to characterize the detection sensitivity compared to a traditional plastic beta cell.

INTRODUCTION

The Comprehensive Nuclear Test Ban Treaty prohibits nuclear weapons testing around the world; compliance with this treaty is enforced by the International Monitoring System (IMS), which consists of 321 globally distributed stations that constantly collect data to detect and identify explosions [1]. These stations consist of sensors based on four technologies: seismic, hydroacoustic, infrasound and radioxenon. Detection of radioactive xenon in atmospheric samples is the only technology in the IMS that can unambiguously identify a detected event as nuclear.

Currently deployed systems for radioxenon monitoring use beta-gamma coincidence detection to identify the various xenon isotopes in atmospheric gas samples [2]. These systems use plastic scintillator to detect the beta particles and sodium iodide to detect the correlated X-rays and gamma rays. The plastic scintillator has hollow, cylindrical cell geometry with the atmospheric sample inside to create a near-4-pi detection geometry. One limitation of plastic scintillators is the so-called "memory effect", whereby radioactive xenon diffuses into the plastic during the measurement; this effect raises the background for subsequent measurements, which decreases the minimum detectable activity in the system [3]. Stilbene organic scintillator has been previously explored to mitigate the memory effect, but it is difficult to machine into the hollow geometry needed for radioxenon detection [4].

Here, we explore an organic glass scintillator that was recently develop by Sandia National Laboratories as an alternative material for the organic beta detection cell. This material has higher brightness than plastic and stilbene and can be melt-cast into a variety of shapes and sizes [5 - 7].

Furthermore, the higher brightness results in improved resolution, which will reduce the spectral regions of interest used to identify the various radioisotopes from one another. This improved spectral discrimination will ultimately improve the sensitivity of beta-gamma coincidence detection compared to the plastic-based systems that are deployed in the field. In this work, we perform a Monte Carlo study to evaluate the impact of the organic glass energy resolution on beta-gamma coincidence spectra from the various radioxenon isotopes. A standard plastic-based system is simulated for comparison.

MATERIALS AND METHODS

The Monte Carlo code MCNPX-PoliMi [8] was used to model beta-gamma coincidence detection in the typical well detection geometry. The gamma-ray detector was modeled as a cylinder with a hollow "well" in the center where the organic beta cell was placed. The organic beta cell was modeled as organic glass with the same dimensions as a traditional beta cell; a plastic beta cell was also modeled for comparison. Figure 1 shows the simulation geometry.



Fig. 1. MCNPX-PoliMi model geometry

Table 1 lists the material composition of the plastic and organic glass modeled in this work. Because the materials have a similar Z_{eff} and density, it is expected that they will have a similar detection efficiency for the incident electrons from the radioxenon; however, recent work has shown that the energy resolution of organic glass is better than that of plastic scintillator [9]. The energy resolution of an organic glass pillar was measured to be 10.3% at 478 keV, which is significantly better than the typical plastic scintillator resolution of 13.7% at the same energy.

Material	Density	Composition	Zeff	Light output			
	(g/cm^3)	(atom %)		(photons/MeV)			
Plastic (EJ-200)	1.023	Н 52.43	3.38	~10,000			
		C 47.57					
Organic glass	1.096	H 45.59	3.82	~16,000			
		C 53.17					
		Si 1.24					

Table 1. Material composition of the plastic and organic glass scintillators.

The xenon decay sources were specified using the MCNPX-PoliMi Single Decay Option, which was recently modified for improved radioxenon simulations [10]. These improved source definitions were validated using experimental data with a variety of pure radioxenon samples. Table 2 lists the correlated emission products for the radioxenon isotopes of interest. In each simulation, the xenon was modeled as a volumetrically homogeneous source inside of the beta cell, as shown in Fig. 1.

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	^{131m} Xe	^{133m} Xe	¹³³ Xe	¹³⁵ Xe
Half-life	11.84 d	2.2 d	5.25 d	9.14 h
Gamma energy (keV)			81.0	250
X-ray energy (keV)	29.5-34.6	29.5-34.6	30.6–36.0	30.6–36.0
Beta endpoint energy (keV)	—	—	346	905
Conversion electron energy (keV)	129	199		

Table 2. Coincident decay emissions from radioxenon isotopes of interest.

In this work, the energy dependence of the organic glass resolution was assumed to be the same as that of plastic scintillator. The data from the plastic scintillator were shifted to match the organic glass resolution at 478 keVee and then re-fit to obtain the Gaussian energy broadening parameters needed for MCNP, shown in Eq. (1)

$$FWHM = a + b\sqrt{E + cE^2},\tag{1}$$

where FWHM is the energy resolution in MeV, E is the energy deposited in MeV and a, b, and c are the fit parameters [11]. Figure 2 shows the resulting energy resolution distribution and Table 3 shows the fit parameters for each function.



Fig. 2. Energy resolution as a function of energy for plastic scintillator and organic glass.

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Material	а	b	С
Plastic (EJ-200)	-0.007544	0.08907	0.8383
Organic glass	-0.004337	0.07093	0.4077

Table 3. Energy resolution fit parameters for Eq. 1 for plastic and organic glass scintillators.

RESULTS

The MCNPX-PoliMi models were used to perform simulations of each individual radioxenon isotope (^{131m}Xe, ^{133m}Xe, ¹³³Xe, and ¹³⁵Xe) to obtain a beta-gamma coincidence spectrum. Figures 3 – 6 show the beta-gamma coincidence spectra for each of these four isotopes with the plastic scintillator and organic glass beta cell. The metastable isotopes (^{131m}Xe and ^{133m}Xe) decay by single-energy conversion electron emission in coincidence with an X-ray of approximately 30 keV. In the beta-gamma coincidence spectra for these isotopes (Figs. 3 and 4) decay mode appears as an island centered at the corresponding energies. Consequently, the improved resolution of the organic glass is evident in the spectra for the metastable isotopes (¹³³Xe and ¹³⁵Xe) decay by beta emission in coincidence with X-ray and/or gamma-ray emission. These decay modes appear as a distribution in the organic detector centered at the appropriate X-ray and gamma-ray energy, as shown in Figs. 5 and 6.



Fig. 3. Simulated ^{131m}Xe beta-gamma coincidence spectra for using plastic (a.) and organic glass (b.) beta cells; the color-bar scale is logarithmic counts.



Fig. 4. Simulated ^{133m}*Xe beta-gamma coincidence spectra for using plastic (a.) and organic glass (b.) beta cells; the color-bar scale is logarithmic counts.*



Fig. 5. Simulated ¹³³Xe beta-gamma coincidence spectra for using plastic (a.) and organic glass (b.) beta cells; the color-bar scale is logarithmic counts.



Fig. 6. Simulated ¹³⁵Xe beta-gamma coincidence spectra for using plastic (a.) and organic glass (b.) beta cells; the color-bar scale is logarithmic counts.

To investigate the impact of the organic glass resolution on a mixed radioxenon sample, a simulation was performed with all four radioxenon isotopes present (^{131m}Xe, ^{133m}Xe, ¹³³Xe, and ¹³⁵Xe). Figure 7 shows the beta-gamma coincidence spectrum for this mixed-source simulation. As expected, all features from the individual radioxenon decays are readily apparent in the mixed source distribution. In Fig. 7b, the conversion electron peaks from ^{131m}Xe (129 keV), and ^{133m}Xe (199 keV) are better isolated than in the plastic spectrum shown in Fig. 7a.

To better illustrate the impact of the improved energy resolution, the integrated spectra in the beta cell is plotted in Fig. 8. The improved resolution in the glass scintillator makes the conversion electron peaks easier to distinguish above the continuum from the beta decays than when using the plastic scintillator. The energy resolution of the organic glass is 17% better than plastic at 129 keV and 19% better than plastic at 199 keV, which will directly result smaller regions of interest in the spectroscopic analysis. The improved isotopic discrimination will ultimately improve the sensitivity of beta-gamma coincidence detection compared to the plastic-based systems that are deployed in the field.



Fig. 7. Simulated beta-gamma coincidence spectra for using plastic (a.) and organic glass (b.) beta cells with a mixed source of ^{131m}Xe, ^{133m}Xe, ¹³³Xe, and ¹³⁵Xe.



Fig. 8. Simulated energy spectra in the plastic and organic glass beta cells with a mixed source of ^{131m}Xe , ^{133m}Xe , ^{133m}Xe , and ^{135}Xe .

SUMMARY AND CONCLUSIONS

An MCNP model of a radioxenon detection system based on organic glass and sodium iodide scintillators has been developed and used to explore the sensitivity to the beta-gamma coincidence decay of the four primary isotopes of radioxenon used in atmospheric nuclear explosion monitoring. The improved resolution in the glass scintillator makes the conversion electron peaks from the metastable radioxenon isotopes easier to distinguish above the continuum from the beta decays from other radioxenon isotopes.

Future work will experimentally verify the energy resolution function of the organic glass. In addition, we will explicitly quantify the improvement to radioxenon minimum detectable activity for a range of isotopic mixtures.

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