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Development of a radioxenon measurement system and its application in monitoring Fukushima nuclear accident

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HIGHLIGHTS

- A system for detection of radioxenon isotopes has been developed.
- The system was mainly comprised of two HPGe detectors and a gas source cell.
- The efficiency of 81 keV gamma-ray for ^{133}Xe was calibrated to be 0.50.
- The MDA for ^{133}Xe was determined to be 10 mBq during 1 day measurement time.
- The system was successfully applied in ^{133}Xe monitoring at Xi'an of China.

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ABSTRACT

An HPGe gamma-ray spectrometry system for radioxenon isotopes detection has been developed. The system was comprised of two low-energy planar HPGe detectors that were horizontally and oppositely placed, and a gas source cell made of carbon fiber in both sides was used. The spectra obtained simultaneously by the two HPGe detectors were analyzed by summing spectrum method. After the radioactive concentration of ^{133}Xe was determined with three internal gas proportional counters, the efficiency of 81 keV gamma-ray for ^{133}Xe was calibrated to be 0.50 (1), and the Minimum Detectable Activity (MDA) for ^{133}Xe detection was determined to be 10 mBq during 1 day measurement time. Finally, the system was applied in atmospheric ^{133}Xe monitoring at Xi'an of China after the Fukushima nuclear accident.

1. Introduction

As xenon isotopes can easily vent from the cavity formed in underground nuclear explosion, the Comprehensive Nuclear-Test-Ban Treaty Organization (CTBTO) pays particular attention to the four radioxenon isotopes (Bowyer and Schlosser, 2002; Matthias and Timo, 2010; Perkins and Casey, 1996), namely, ^{131m}Xe, ^{133}Xe, ^{131m}Xe and ^{135}Xe. As shown in Table 1, these isotopes have relative large fission yields and half-lives long enough to offer realistic detection probabilities on continental or even global distances. Considering the respective radioactive half-lives of the four xenon radioisotopes and the activity levels in case of a nuclear event (Bowyer et al., 2002), it is very probable that the total sample activity will be controlled by that of ^{133}Xe, so ^{133}Xe will be primarily studied in our research work.

In monitoring atmospheric radioxenon isotopes, detection of collected xenon samples is typically based on either β–γ coincidence spectrometry using scintillators (Ringbom et al., 2003; Bowyer et al., 1996; Dubasov et al., 2005; Le Petit et al., 2013) or high resolution gamma-ray spectrometry using HPGe detectors (Fontaine et al., 2004; Stocki et al., 2004; Le Petit et al., 2006; Zhang et al., 2009). Fontaine et al. (2004) developed an atmospheric radioxenon isotopes monitoring system based on one HPGe detector, and the minimum detectable concentration (MDC) for ^{133}Xe was determined to be 0.15 mBq/m^3 relative to 86 m^3 air sampling volume and 1 day measurement time. Stocki et al. (2004) and Le Petit et al. (2006) improved detection sensitivity of the system for ^{131m}Xe and ^{131m}Xe by replacing the aluminum window of source cell with carbon fiber window, and the efficiency of 81 keV for ^{133}Xe was calibrated to be about 20%. In 2009, Zhang et al. (2009) developed an improved radioxenon gamma-spectrometry counting system based on one broad energy germanium detector (BEGe) and a counting cell with 0.7 mm carbon fiber window, and the efficiency of 81 keV was calibrated to be about 22%. In the papers mentioned above, radioxenon

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Table 1
Fission yields and main gamma-rays of radioxenon isotopes.

Radionuclides	Half-live	Fission yield (%)		Gamma-rays	
		Independent	Cumulative	Energy (keV)	Branching ratio (%)
^{131m}Xe	11.9 days	2.41×10^{-7}	4.51×10^{-2}	163.9	1.96
^{133m}Xe	2.19 days	4.23×10^{-3}	1.98×10^{-1}	233.2	10.3
^{133}Xe	5.25 days	1.46×10^{-3}	6.72×10^{0}	81.0	38.0
^{135}Xe	9.14 h	1.20×10^{-1}	6.60×10^{0}	249.8	90.0

Fig. 1. The photograph of the measurement system.

isotopes were detected only using one HPGe detector and the efficiencies of 81 keV were less than 22%. The full energy peak (FEP) efficiencies of HPGe detector are an important factor in decreasing the detection sensitivity limit, so, if the gamma and X-rays from radioxenon isotopes are simultaneously detected with two HPGe detectors in order to increase the FEP efficiency, detection sensitivity for radioxenon isotopes may be improved. In this work, an HPGe gamma-ray spectrometry system has been developed for radioxenon isotopes measurement, which were mainly made up of two low-energy planar HPGe detectors and gas source cell that was made of carbon fiber in both sides. In the following, we will describe the principle and performance of the system, and the application of the system in surveying ^{133}Xe from the Fukushima nuclear accident is also described.

2. Measurement system

We used two same HPGe detectors of planar type (referred as HPGe1# and HPGe2#, correspondingly) from Eurisys with a FWHM of 0.68 keV at 122 keV of ^{57}Co. The detectors have a crystal height of 19.5 mm and diameter of 70 mm, and the thickness of Ge dead layer and carbon window for detection are 0.5 μm and 1.1 mm, respectively. The two HPGe detectors were horizontally and oppositely placed, and shielded with 1.5 mm copper and 10 cm ordinary lead. The photograph of system is presented in Fig. 1. Spectra were recorded and processed by the two Normad-Plus data acquisition and GammaVisionV5.2 software.

A gas source cell with carbon fiber detection windows and stainless steel body was designed. Inner radius and outer radius of the stainless steel body were 50 mm and 70 mm, respectively, and the height was 10 mm. The thickness of carbon fiber detection windows was 0.6 mm, which minimized the adsorption of X-rays and gamma-rays of gas sample effectively and improved the system detectable sensitivity. The photograph of the gas source cell is presented in Fig. 2. The results of performance test showed that the cell have good air tightness and can bear in the pressure from 10 Pa to 200 kPa. As seen in Fig. 3, the gas source cell was placed between both HPGe detectors during the measurement duration.

The two spectra, which were recorded simultaneously by two HPGe detectors, were summed into one spectrum using spectrum summing function of GammaVisionV5.2 software. Energy and resolution of the system were calibrated from 20 keV to 500 keV with selected gamma- and X-ray lines from three radioactive point sources (^{133}Ba, ^{57}Co and ^{241}Am). As a result, the FWHM at 81 keV for ^{133}Xe was calibrated to be 0.67 keV.

Many background gamma spectra of the system were measured for a period of a few days. Fig. 4 presented a gamma spectrum of background as an example. The majority of the peaks in the background spectra can be identified as being from the Ra and Th decay chains and Pb X-rays were mainly from radon daughters ^{214}Pb and ^{212}Pb (Debertin et al., 1988). Because level-activity lead was not use in lead shield, the 47 keV gamma-ray from ^{210}Pb in lead shield was obviously observed. By analyzing the background spectra with the total summation method (ORTEC, 2003), n_b (the counts rate of the region of interest (ROI) for 81 keV) were determined to be 1.3×10^{-2} counts per second (cps). ROI was defined as $\pm 1.25 \times$ FWHM of the peak centroid of 81 keV for ^{133}Xe. And also, the integral background of measurement system was determined to be 3.1 cps.

3. Efficiency calibration and detection sensitivity

3.1. FEP efficiency calibration

The radioactive concentration of ^{133}Xe was obtained with the method (Makepeace et al., 1994) by three internal gas proportional counters. Here we provide a summary of the measurement procedure.

Enriched U_3O_8 (90% ^{235}U) sealed in a quartz capsule with a volume of 1 cm^3 was irradiated for 1 h at the core of Xi'an pulsed reactor where the thermal neutron flux was approximately 10^{13} n cm^{-2} s^{-1} and fission product gases were produced. After 7 days of cooling, when the krypton and other xenon isotopes decay away quickly and do not interfere with the ^{133}Xe of interest, the quartz tube was crushed inside the source preparation equipment presented in Fig. 5, and then the ^{133}Xe sample was imported into the mixing chamber and mixed with a counting gas (P10:90% Ar_2 + 10% CH_4) with a 100 kPa pressure. Finally, the homogenized gases were charged simultaneously into the three internal gas proportional counters (L counter, M counter and S counter in Fig. 5) and a gas source cell presented in Fig. 5.

The three copper gas counters with volumes of 267.1, 178.1, and 89.2 cm^3, have an identical end structure to eliminate the effects of non-uniform electrical fields at the end. Each counter was equipped with a high-voltage supply, preamplifier, amplifier, discriminator and data-acquisition unit. After the ^{133}Xe gas sample was filled into the three counters, the measurements were carried out at certain high voltages on the corresponding plateaus.