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Low-level activity measurement of $^{131}Xe^{m}$, $^{133}Xe^{m}$, ^{135}Xe and ^{133}Xe in atmospheric air samples using high-resolution dual X–γ spectrometry

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Abstract

High-resolution dual X and γ spectrometry has been optimized to improve the sensitivity in analyzing radioxenon from environmental samples, especially for the metastable isotopes ($^{131}Xe^{m}$ and $^{133}Xe^{m}$). The full-energy peak (FEP) efficiency curve in the 20–700 keV range has been established with a global uncertainty better than ±3% using a Monte Carlo simulation. The minimum detectable activity concentrations (MDC) achievable at the laboratory for $^{131}Xe^{m}$, $^{133}Xe^{m}$, ^{133}Xe and ^{135}Xe are presented and the influence of ^{133}Xe activity on the metastable radioxenon MDCs is evaluated.

Keywords: Comprehensive nuclear test ban treaty; Radioxenon monitoring; X-ray spectrometry; γ-ray spectrometry; Monte Carlo

1. Introduction

As part of the International Monitoring System (IMS) for the Comprehensive Nuclear Test Ban Treaty (CTBT), radioactive isotopes of xenon will be continuously monitored by a worldwide network of 40 radioxenon stations. The French Atomic Energy Commission (CEA) has designed and developed a system complying with the technical specifications of the CTBT organization. This system, named Spalax (for Système de Prélèvement d'air Automatique en Ligne avec l'Analyse des radioXénons), is fully automated. It extracts, purifies and concentrates gaseous xenon from atmospheric air and measures four radioactive isotopes of xenon ($^{131}Xe^{m}$, ^{133}Xe, $^{133}Xe^{m}$ and ^{135}Xe) by high-resolution γ-ray spectrometry (Fontaine et al., 2004). In order to support the station network, an international network of 16 laboratories (CTBTO Preparatory Commission (CTBTO), 2002) will chiefly guarantee the quality of the data and, if necessary, analyze samples coming from stations with, in principle, a higher level of sensitivity.

The presence of the radioactive isotopes of xenon in the atmosphere may be characteristic of nuclear explosion, reactor operation, fuel reprocessing, medical isotope production or other human activities. The ^{133}Xe background in the northern atmosphere is several mBq/m^3 with strong temporal variations. In order to discriminate between nuclear explosions and civilian sources, it is necessary to measure simultaneously and with the best accuracy the four isotopes. Indeed, this will allow the determination of the relevant isotopic ratios such as $^{133}Xe^{m}/^{131}Xe^{m}$ and $^{135}Xe/^{133}Xe$.

The goal of this paper is to present the metrological aspects associated with a radioxenon measurement and to evaluate the lowest detection limits likely to be obtained in laboratory.

2. Detector selection to analyze a xenon air sample

The four xenon radioisotopes have distinct decay properties, which can be used for their identification and quantification. $^{131}Xe^{m}$ (half-life: $T_{1/2} = 11.8$ d) and $^{133}Xe^{m}$ ($T_{1/2} = 2.2$ d) are isomeric states that decay to their ground states by highly converted γ-rays of 163.9 and 233.3 keV, respectively. The internal conversion is mostly K-shell, so the conversion electrons are in coincidence with Xe K X-rays (29.46, 29.78, 33.56, and 33.62 keV for both $^{133}Xe^{m}$ and $^{131}Xe^{m}$). ^{133}Xe ($T_{1/2} = 5.2$ d) and ^{135}Xe ($T_{1/2} = 0.38$ d) are β-emitters with 99 and 96% proceeding to the first

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excited states of ^{133}Cs and ^{135}Cs, from where they de-excite by 81 and 249.8 keV γ-rays, respectively. The low energy of the 81 keV transition results in substantial K internal conversion accompanied by Cs K X-rays (30.62, 30.97, 34.92, and 34.99 keV for both ^{133}Xe and ^{135}Xe). Two main approaches can be used to identify and quantify the xenon radioisotopes: the first is the high-resolution X–γ ray spectrometry, and the second a β-coincidence technique. Even though the former might be less sensitive than the latter, we chose high-resolution X–γ ray spectrometry, because it is easier to perform in the point of view of the analysis interpretation.

In the laboratory, we opted for a broad-energy germanium (BEGe) detector (active area of 3800 mm^2 with a 1 mm-thick aluminium entrance window) manufactured by the Canberra company. This detector has the advantage of covering the 15 keV–2 MeV energy range with a relatively good efficiency over the entire energy range. Furthermore, its resolution at low energy (0.54 keV at 30 keV and 0.72 keV at 122 keV) is equivalent to the one of a conventional planar high-purity germanium (HPGe) detector, whereas its resolution at high energy is comparable with that of an HPGe coaxial detector. This detector is therefore well suited to xenon radioisotopes measurement both for the X-ray multiplets zone (in the 29–35 keV energy range) and for the γ-rays (with energies below 300 keV). The passive shielding is made up of two layers: the outer one consists of 13 cm-thick low-activity lead, and the inner one, which is close to the crystal, consists of 2 cm-thick ultra-pure lead ($\leqslant 10^{-3}$ Bq/kg).

3. Full-energy peak efficiency calibration curve in the energy range 20–700 keV

Once the detector has been chosen, it is necessary to establish the full-energy peak (FEP) efficiency calibration with the best possible accuracy, especially at low energies. There are several methods of achieving this:

- Use the xenon isotopes themselves as standards for the FEP efficiency calibration. This method has the advantage of using the same sample geometry. But it presents the drawback of requiring a substantial level of activity, especially for the ^{133}Xe^{m} and ^{135}Xe isotopes given their short half-lives.
- Use certified X–γ ray radioactive sources such as a low-density resin labelled by a multigamma standard in sample geometry or point sources. In this case, a method of transferring the efficiency curve corresponding to the sample geometry must be applied. This practice has the disadvantage of leading to significant uncertainties in the determination of the FEP efficiency at low energies owing to the relatively high uncertainty associated to X-ray emission intensities.
- Use a method that overcomes the problem of X- and γ-ray emission intensities. Monte Carlo type probabilistic codes are particularly well adapted to this problem. Nevertheless, they do require the full knowledge of the features of the detector and more particularly of the volume of the germanium crystal that is truly sensitive to the radiation. The manufacturers are generally unable to provide this information. Therefore, the user must evaluate the germanium dead layer thickness to model the detector. However, when low-energy (<100 keV) photons are concerned, analytical models have been developed to determine the germanium FEP efficiency with a good level of accuracy (Hansen et al., 1973; Cohen, 1980); this opportunity has been used to calculate the germanium dead layer thickness as explained hereafter.

3.1. Empirical method for BEGe dead layer thickness determination

The analytical model presented here is based on the work of Hansen et al. (1973). It is only suitable for low-energy photons (<60 keV). Some assumptions have to be made for the FEP efficiency calculation: the dead zone layer of the germanium crystal is supposed to be constant over the entire diameter, and the efficiency of the total charge collection is considered to be complete in the sensitive volume. Furthermore, events lost from the full-energy peaks due to the escape of germanium X-rays are ignored for the energy range under study (20–60 keV).

The FEP efficiency curve at energy E (ε_E) of the detector ($E<60$ keV) is given by the following equation:

$$\varepsilon_E = \frac{\Omega_{\text{eff}}}{4\pi} \times (1 - \exp(-\mu_{\text{Ge}}(E)D)) \frac{\tau_{\text{Ge}}(E)}{\mu_{\text{Ge}}(E)} \prod_i \exp(-\mu_i L_i), \tag{1}$$

where μ_i and L_i are, respectively, the total linear attenuation coefficient and the mean path for the ith absorber of thickness x_i, placed between the source and the sensitive part of the detector (in the present case, the end cap material and the germanium dead layer). τ_{Ge} and μ_{Ge} are, respectively, the germanium photoelectric and the total linear attenuation coefficients (Berger and Hubbel, 1999).D is the detector-sensitive thickness.

The effective solid angle Ω_{eff} is given by

$$\frac{\Omega_{\text{eff}}}{4\pi} = \frac{1}{2}\left(1 - \frac{d + Z(E)}{\sqrt{[(d + Z(E))^2 + r_{\text{d}}^2]}}\right), \tag{2}$$

where d is the distance between the source and the front face of the crystal, r_{d} is the detector radius, and $Z(E)$ is the mean interaction depth for photons of energy (Hansen, 1973)

$$Z(E) = \frac{1 - \exp(-\mu_{\text{Ge}}D) \times (1 + \mu_{\text{Ge}}D)}{\mu_{\text{Ge}}[1 - \exp(-\mu_{\text{Ge}}D)]}. \tag{3}$$

All sample measurements are performed on top of the detector and, therefore, at a high solid angle. In this case, the average path crossed by a photon in a layer is larger