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Improvements of low-level radioxenon detection sensitivity by a state-of-the art coincidence setup

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HIGHLIGHTS

- A new electron/photon spectrometer for radioxenon measurements has been developed.
- High resolution coincidence measurement of a ^{131m}Xe standard is presented.
- A detection efficiency of 22 ± 4 counts/s/kBq for ^{131m}Xe is obtained.
- MDA of 0.9 mBq for ^{131m}Xe is obtained from a 3.9 day background measurement.
- A complex environmental measurement is presented.

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ABSTRACT

The ability to quantify isotopic ratios of 135, 133 m, 133 and 131 m radioxenon is essential for the verification of the Comprehensive Nuclear-Test Ban Treaty (CTBT). In order to improve detection limits, CEA has developed a new on-site setup using photon/electron coincidence (Le Petit et al., 2013. J. Radioanal. Nucl. Chem., DOI : 10.1007/s 10697-013-2525-8.). Alternatively, the electron detection cell equipped with large silicon chips (PIPS) can be used with HPGe detector for laboratory analysis purpose. This setup allows the measurement of β/γ coincidences for the detection of ^{133}Xe and ^{135}Xe; and K-shell Conversion Electrons (K-CE)/X-ray coincidences for the detection of ^{131m}Xe, ^{133m}Xe and ^{133}Xe as well. Good energy resolution of 11 keV at 130 keV and low energy threshold of 29 keV for the electron detection were obtained. This provides direct discrimination between K-CE from ^{133}Xe, ^{133m}Xe and ^{131m}Xe. Estimation of Minimum Detectable Activity (MDA) for ^{131m}Xe is in the order of 1 mBq over a 4 day measurement. An analysis of an environmental radioxenon sample using this method is shown.

1. Introduction

An International Monitoring System (IMS) has been installed for the verification regime of the Comprehensive Nuclear Test Ban Treaty Organization (CTBTO). The IMS is composed of three waveforms technologies (seismic, infrasound, hydro-acoustic) and two radionuclide technologies (atmospheric aerosols and noble gas) station types spread around the globe for the detection (at 95% certainty) and the identification of any kind of nuclear test (atmospheric, underwater, surface or underground tests) of 1 kT or more within 10 days.

Low energy confined nuclear explosions are very challenging to detect. Thanks to their good diffusion characteristics, radioactive noble gases are difficult to contain. Among the noble gases emitted from a nuclear event, four radioisotopes of xenon (131 m, 133 m, 133 and 135) have nuclear decay data (cf. Table 1) and fission yields compatible with the detection and the characterization within a few days of a nuclear explosion (Saey, 2007). They are therefore potential key indicators of a nuclear test and the IMS noble gas stations are designed for their detection. Four systems were initially developed: the ARIX, the ARSA, the SAUNA and the SPALAX (Prelovskii et al., 2007; Reeder et al., 1998; Ringbom et al., 2003; Fontaine et al., 2004). The first three setups are implementing β–γ coincidence measurements using plastic scintillators coupled to a NaI(Tl) crystal whereas the SPALAX is performing a direct HPGe γ/X-ray measurement.

As can be seen in Table 1, unlike ^{133}Xe and ^{135}Xe, the γ emission yields for the metastable radioxenon (^{131m}Xe and ^{133m}Xe) are quite low and they are not distinguishable via their X-ray emission. This is a major drawback of the direct γ measurement (implemented by the SPALAX systems). On the other hand, the β/γ detection setups (the other IMS systems) have a typical energy resolution of 30%, which is far too high to allow unambiguous distinction between the K-shell Conversion Electron peaks and the β background (from

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Table 1
Main decay mode of the four relevant radioxenon isotopes, with associated emission probabilities in bracket (Chisté et al., 2013).

Isotopes	^{131m}Xe	^{133m}Xe	^{133}Xe	^{135}Xe
Half-life (days)	11.93	2.19	5.24	0.38
Major γ-ray energy (keV)	163.9 (2.0%)	233.2 (10.3%)	81.0 (37.0%)	249.8 (90.0%)
Kα X-ray energy (keV)	29.7 (43.9%) (Xe)	29.7 (45.7%) (Xe)	30.9 (38.5%) (Cs)	30.9 (4.1%) (Cs)
Kβ X-ray energy (keV)	33.7 (8.3%) (Xe)	33.7 (8.6%) (Xe)	35.1 (7.3%) (Cs)	35.1 (1.0%) (Cs)
β Endpoint energy (keV)			346.4 (99.1%)	905.0 (96.0%)
K shell conversion electron energy (keV)	129.4 (61.0%)	198.7 (63.5%)	45.0 (52.9%)	214.0 (5.7%)

^{133}Xe and ^{135}Xe) in the xenon/cesium K X-ray region (31 ± 3 keV). In order to improve the radioxenon detection, a new setup was developed by CEA and the Canberra Semiconductors company. It consists in a well-type NaI(Tl) detector and a gas cell equipped with two large area (1200 mm^2) Passivated Implanted Planar Silicon detectors (PIPS). First results and a complete detailed description of this setup are reported in (Le Petit et al., 2013). For laboratory purposes, the gas cell equipped with PIPS (hereafter called PIPSbox) can also be coupled to an HPGe detector. This setup allows complete distinction between ground state (^{135}Xe and ^{133}Xe) and metastable (^{131m}Xe and ^{133m}Xe) xenon thanks to the high resolution of the HPGe, and complete discrimination between ^{131m}Xe and ^{133m}Xe thanks to the high energy resolution of the PIPS detectors. Additionally, the photon energy resolution allows clear separation between radon progenies and radioxenon peaks. This is particularly useful for the detection of low-level of radioxenon in an environmental sample containing trace amounts of radon and its progenies.

2. Detection technique

The detection of the four radioxenon isotopes is implemented in two different ways depending on the nuclide: one will quantify ^{131m}Xe or ^{133m}Xe by analyzing a xenon *K*α X-ray gated electron spectrum, whereas ^{133}Xe and ^{135}Xe will be quantified by analyzing β-gated photon spectra. The result from the β-gated photon spectrum could be compared to the analysis of a cesium *K*α X-ray gated electron spectrum. This is mainly relevant for ^{133}Xe which has an internal conversion coefficient of 1.43 (for a K-CE at 45 keV) compared to 0.06 for ^{135}Xe.

Activities can be calculated from the modified classical formula (Knoll, 2010):

$$A = \frac{C}{K T_{acq} P_{decay} \varepsilon_{photon} \varepsilon_{e-}} \quad (1)$$

where K is a correction factor (in our case, K is correcting the decay during counting), P_{decay} is the probability of the considered event, ε_{photon} and ε_{e-} are the photon and electron detection full-energy-peak (FEP) efficiency respectively, T_{acq} is the acquisition time and C are the net counts corresponding to the considered event.

The corresponding event probabilities can be calculated, as follows:

- For the ^{131m}Xe isotope, the probability of having a 129.4 keV K-shell conversion electron (K-CE) followed by a 29.7 keV *K*α X-ray is 43.9%, as we must have a 129 keV K-CE for each K X-ray measured.
- For ^{133m}Xe, the probability of having a 198.7 keV K-shell conversion electron (K-CE) followed by a 29.7 keV *K*α X-ray is 45.7%, for the same reason as for ^{131m}Xe.
- For ^{133}Xe, the probability of having an electron from its β spectrum ($E_{electron} \in [0; 346$ keV]) followed by an 81 keV photon is 37.0%, as all β particles from ^{133}Xe have an energy lower than 346 keV. And the probability of having a 45.0 keV K-shell conversion electron (K-CE) followed by a 30.9 keV *K*α X-ray is 38.5% (we cannot distinguish the two K-CE of ^{133}Xe: 45.0 keV and 43.6 keV (0.41%)).
- For the ^{135}Xe isotope, the probability of having an electron from its β spectrum ($E_{electron} \in [0; 905$ keV]) followed by a 249.8 keV photon is 90.1% (as all β particles from ^{135}Xe have an energy lower than 905 keV).

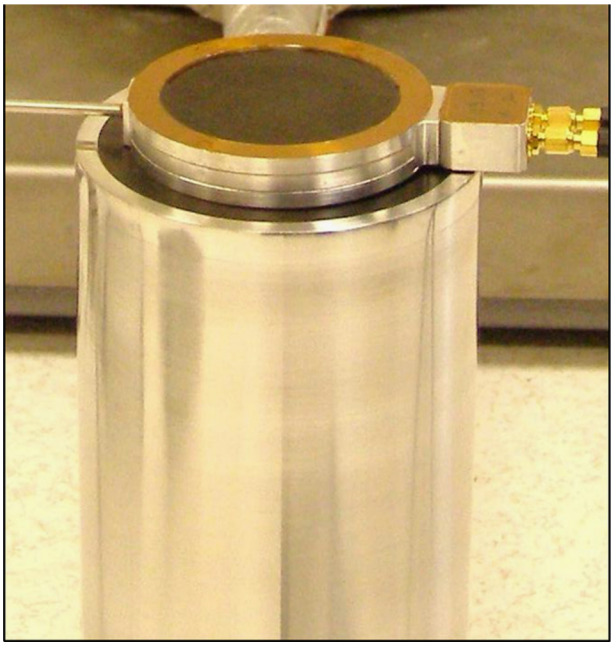

Fig. 1. Picture showing the PIPSBox (electron detection) on top of the HPGe detector (photon detection).

3. Experimental setup

The detection setup is composed of the PIPSBox presented in (Le Petit et al., 2013) and a BEGe 5030 HPGe detector from the Canberra Semiconductors company, a picture of the setup is shown in Fig. 1. The PIPSBox is an 11 cm^3 (active volume) gas cell (under a pressure lower than 900 mBar) equipped with two 500 μm thick and 1200 mm^2 large PIPS detectors dedicated to the electron measurement. The cell is made out of low background materials; it is fitted on both faces with thin carbon epoxy window (600 μm). 500 μm thick PIPS detectors allow full energy deposition of electrons up to 400 keV, while keeping a good electron resolution (11 keV full width at half maximum (FWHM) at 130 and 160 keV) as shown in Fig. 2. The BEGe 5030 is a High Purity Germanium detector (HPGe) with a 30 mm thick and 50 cm^2 area crystal. It has a typical FEP efficiency of 20% and an energy resolution lower than 650 eV at xenon and cesium K X-ray energies. The detectors are shielded by a first 5 cm layer of low