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Original Article

Large-volume and room-temperature gamma spectrometer for environmental radiation monitoring

Romain Coulon*, Jonathan Dumazert, Tola Tith, Emmanuel Rohée, Karim Boudergui

CEA, LIST, F-91191 Gif-sur-Yvette Cedex, France

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ABSTRACT

The use of a room-temperature gamma spectrometer is an issue in environmental radiation monitoring. To monitor radionuclides released around a nuclear power plant, suitable instruments giving fast and reliable information are required. High-pressure xenon (HPXe) chambers have range of resolution and efficiency equivalent to those of other medium resolution detectors such as those using NaI(Tl), CdZnTe, and LaBr₃:Ce. An HPXe chamber could be a cost-effective alternative, assuming temperature stability and reliability. The CEA LIST actively studied and developed HPXe-based technology applied for environmental monitoring. Xenon purification and conditioning was performed. The design of a 4-L HPXe detector was performed to minimize the detector capacitance and the required power supply. Simulations were done with the MCNPX2.7 particle transport code to estimate the intrinsic efficiency of the HPXe detector. A behavioral study dealing with ballistic deficits and electronic noise will be utilized to provide perspective for further analysis.

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1. Introduction

Gamma spectroscopy can be used to provide isotopic information about radioactivity. This identification then allows the diagnosis of a situation using information given by the nature of the radioactive materials. To discriminate between different isotopes with high efficiency and reliability, the energy resolution has to be as low as possible. The lowest possible resolution can be achieved by cryogenic detectors (high-purity germanium detectors), which are expensive and not suitable for use outdoors. A room temperature gamma spectrometer with quite good resolution has been developed during the last decades. CdZnTe semiconductors and LaBr₃:Ce scintillators provide an energy resolution of around 2–4% at 662 keV [1,2]. However, their cost increases dramatically with the volume and these devices need to be stabilized against external temperature variation.

Therefore, the high-pressure xenon (HPXe) technology is investigated for the CEA LIST for these problematic environmental conditions. The state of development is presented in this paper.

2. State of the art

HPXe chambers have been under development by many laboratories since the 1980s. The element xenon is a noble gas with a high charge number; xenon facilitates good gamma stopping power when used under pressure. At 50 bar, the density reaches 0.5 g/cm³ and gamma ray stopping power has a value between those of germanium and sodium iodine. Another interesting particularity is the process for charges carrier creation and, more specially, the Fano factor of this process (0.13) [3]. The intrinsic energy resolution is three times lower than those of room-temperature inorganic scintillators using NaI(Tl) or LaBr₃:Ce [4]. To address the industrial need for room temperature spectrometers, xenon is thus an important research path.

To build a spectroscopy-grade HPXe detector, some requirements have to be taken into account. The properties of the xenon gas for spectroscopy have been studied in detail by Dmitrenko et al. [5] and Bolotnikov and Ramsey [6].

3. General properties

Thanks to electronic band evolution of Xe₂^{*} dimers (decrease of excited states compared with ionizing states) according to gas density, the W-value decreases as the gas pressure increases [6–8]. The increase of the charge carrier number and the gamma ray

* Corresponding author.

E-mail address: romain.coulon@cea.fr (R. Coulon).<http://dx.doi.org/10.1016/j.net.2017.06.010>1738-5733/© 2017 Korean Nuclear Society, Published by Elsevier Korea LLC. This is an open access article under the CC BY-NC-ND license (<http://creativecommons.org/licenses/by-nc-nd/4.0/>).

stopping power are the main motives to apply very high pressure to the xenon chamber. However, a discrepancy of the energy resolution is observed when the density gets above 0.55 g/cm^3 [5]. The intrinsic energy resolution is approximately equal to 0.6% until a density value of 0.55 g/cm^3 is reached; the intrinsic energy resolution then increases to 5% at a density value of 1.6 g/cm^3 . Above 0.55 g/cm^3 the Fano factor increases (due to δ -ray recombination) and small liquid droplets appear and lead to electronegative clusters, which absorb electrons with kinetic energy below the ionizing energy level [6,9].

In practice, the pressure has to be between 0.5 g/cm^3 and 0.6 g/cm^3 . A device is exempt from French regulation rules if the pressure volume product does not exceed 200 bar L [10]. A 4-L chamber could then be built without the requirement of periodic control. The gamma ray stopping power obtained in a 4-L chamber makes possible the use of an HPXe chamber for environmental radiation monitoring.

Stability against temperature change is an issue for many industrial applications. Other room-temperature probes such as scintillators or CdZnTe suffer from temperature dependency, which affects the gain and the energy resolution. At relatively low pressure ($<0.6 \text{ g/cm}^3$), xenon shows perfect temperature stability in terms of density and charge carrier collection [6,9,11]. This particularity remains the main advantage of HPXe chambers in comparison with other room temperature spectrometers, which require temperature stabilization.

The use of an HPXe chamber could offer a gain in temperature stability and a cost reduction compared with other conventional systems. The first requirement to operate as a spectrometer is the achievement of a high purity level of xenon gas.

4. Xenon purification and conditioning

To avoid charge recombination, electronegative impurities (mainly oxygen) have to be filtered out. A purity level below parts per billion has to be achieved. Xenon with a purity level below 10 ppm is the best purity grade of commercially available purity-grade xenon (research grade). Thus, a dedicated purification process has to be built to achieve this extra level of purity. The system under development at CEA LIST is shown in Fig. 1.

The process circuit is preliminarily set to an ultrahigh vacuum level of 10^{-11} bar and temperature of 175°C is applied to cause the overall internal skin to desorb. The research-grade xenon is injected and then transferred through a purifier to the container. Xenon

displacement is ensured by the vaporization/condensation cycles, which are controlled by cryo-cooling (LN_2). A gatekeeper filter (Entegris, Billerica, MA, USA) is installed in the CEA LIST device. This filter is based on a metal alloy using chemisorption to remove impurities. It operates at room temperature but requires precise flow rate control. The gas reaches a sub-parts per billion level after many purification cycles. Spark purification, because it is fast and effective, is currently the best purification technique [12]. Spark discharge produces ultraclean titanium dust that traps electronegative impurities and organics.

The purity monitor measures the electron lifetime in the gas. An electron cloud between the two plate electrodes, which are weakly polarized (recombination mode 10 V/cm), is created by external stimulation using a gamma source. The pulse length at the output of the slow charge preamplifier has to reach a value above 5 ms to validate the sub-parts per billion level of the xenon gas [13].

When the xenon gas has reached the sub-parts per billion purity level, the detector volume can be filled. The density has to be strictly controlled. The pressure and the temperature of the gas can be measured at the end of the process. Then, the density is evaluated using an isothermal abacus [13]. Instrumentation has to guarantee a relative accuracy below 1%. Another approach to density measurement consists of dielectric constant measurement, which is a function only of the density [12]. The purity control chamber (fast charge preamplifier) can be used for capacitance measurement after a calibrated pulse generation.

Saturation level of charge collection and optimal charge drift velocity are achieved using an adequate polarization voltage. This voltage is a function of the xenon density and temperature [9]. Complete saturation is achieved using a voltage equal to 7 kV/cm , regardless of the xenon density. In practice, and especially for large volume detectors, a voltage value of 2 kV/cm has been recommended by C. Levin [14] as the best compromise.

The drift velocity has to be maximized from a dynamic range point of view but also in terms of electronic noise filtering (signal-to-noise ratio). The addition of a polyatomic quencher has been studied by V. Dmitrenko [15]. Hydrogen (H_2) is the candidate that is most compatible with purification techniques. A hydrogen concentration addition of 0.2–0.3% increases the drift velocity by a factor of 6, without degradation of the spectroscopic performance [16,17]. However, the quencher is efficient at a bias voltage above 7 kV/cm and the HPXe chamber remains as a slow detector in comparison to scintillators or cadmium zinc telluride (electron mobility equal to $1 \text{ mm}/\mu\text{s}$ in pure xenon).

5. Chamber design for the spectrometer

Parallel-plate chambers with Frisch grids comprised the first design of HPXe technology. A 17-L chamber called Kseniya was built by the National Nuclear Research University (MEPhy) in the early 1980s for space radiation measurement [18,19]. It was installed on the MIR station and operated for 11 years, maintaining a stable energy resolution (3.5% at 662 keV) in severe conditions in terms of temperature gradient and cosmic irradiation flux [20]. In the 1990s, parallel plate chambers with Frisch grids were developed in the United States for earth applications [14]. The small volume of 0.16 L permitted the achievement of an energy resolution of 2.5% at 662 keV, but some limits were noted for industrial applications [21,22]. The useful volume of gas was only 30% due to the electric field inhomogeneity effect. As such, a large dead area produced an important Compton continuum in the gamma spectrum. Moreover, the Frisch grid structure induced additional acoustic noise. The cylindrical chambers allowed an optimization of the useful volume of gas (close to 100%), but vibration sensitivity remained a problem. To limit the acoustic noise, the Frisch grid has to be made with

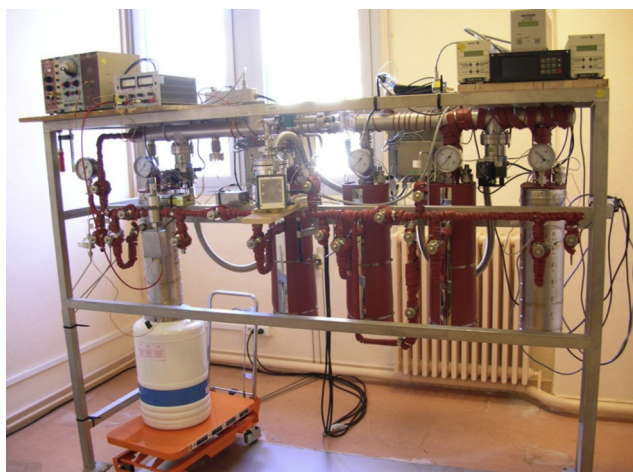


Fig. 1. Xenon purification apparatus of CEA LIST.

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