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Bias in the measurement of radon gas using ionization chambers: Application to SIR

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HIGHLIGHTS

- Flame-sealed glass ampoules are used for SIR gas international comparisons.
- Ampoules of ^{222}Rn are measured in ionization chamber and gamma-ray spectrometer.
- Ampoules parameters are studied: volume, position of sealing point and thickness.
- Discrepancy on glass thickness is observed.
- Ampoules discrepancy can bias the comparisons.

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ABSTRACT

Two main non-destructive techniques can be used to measure standard ^{222}Rn gas ampoules: well-type ionization chambers and gamma-ray spectrometry, the former being used in the Système International de Référence (SIR) for international comparison purposes. The reliability of these techniques requires that the variability of the flame-sealed gas glass ampoules used have a negligible influence on the detector response. This variability is studied in this work by considering three parameters: the volume of the ampoule, the position of the sealing point and the thickness of the glass. Results showed that variability of the gas ampoules induced measurement bias larger than the uncertainty of the standard sources.

1. Introduction

A system for primary activity measurements of ^{222}Rn, based on the defined solid angle (DSA) alpha counting of a cryogenic solid source, developed at the Laboratoire National Henri Becquerel (LNHB) (Picolo, 1996) has recently been upgraded (Sabot et al., 2016). Using a DSA alpha counter to measure ^{222}Rn standards is a primary method used not only by LNHB, but also by IRA-METAS (Spring et al., 2006), KRISS (Kim et al., 2012) and PTB (Dersch, 2004). This system allows the characterization of a radon gas standard in either a precisely manufactured metal container or in a flame-sealed glass ampoule. Unlike glass ampoules, using a metal container with a valve allows repetitive transfers and measurements. Therefore, metal containers are generally preferred at LNHB.

Preparation of radon gas standards in BIPM flame-sealed glass ampoules (Fig. 1, which will be referred to as “gas ampoule” in the following) is necessary for international comparisons with the Système International de Référence (SIR) of the Bureau International des Poids et Mesures (BIPM). This system is based on the detection of photonic emissions (e.g., from x rays, gamma emission and electron bremsstrahlung). For these measurements, which are the cornerstone of international traceability for standardizations of γ-ray emitting radionuclides, the ampoule is placed inside a well-type re-entrant ionization chamber (IC), producing a current which is compared to that obtained for a stable radium source (Ratel, 2007). The computation of the ratio between these two currents enables the evaluation of a so-called “equivalent activity,” characterizing the radionuclide under study. The use of these gas ampoules for radioactive gas standardization raises three important issues, which could affect the IC response and therefore the final results:

(1) The influence of the volume of the gas ampoule,
(2) The influence of the position of the sealing point, as ICs are known to be very sensitive to the geometry of the source,

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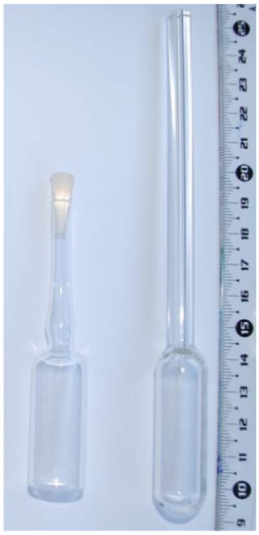

Fig. 1. BIPM ampoule used for liquid (on the left) and for gas (on the right).

(3) The influence of the thickness of the gas ampoule.

To address these issues, three different ICs (from two different laboratories: LNE-LNHB and BIPM), with different wall materials and thicknesses, were used. The first point was studied by comparing the IC response with radon ampoules of different volumes. To address the second point, and to evaluate the possible variation of the IC response, standards of radon gas in gas ampoules were prepared with different positions of the sealing point (Fig. 2) and measured in the three chambers. The third issue was studied by preparing radon sources in gas ampoules of various bottom thicknesses. It is worth noting that BIPM graciously received these gas ampoules from a national laboratory, which has expertise in ampoule manufacturing and that no particular specification on ampoule uniformity was provided.

2. Measurements

The study was conducted using a random selection of nineteen ampoules from a batch of BIPM gas ampoules available at LNHB.

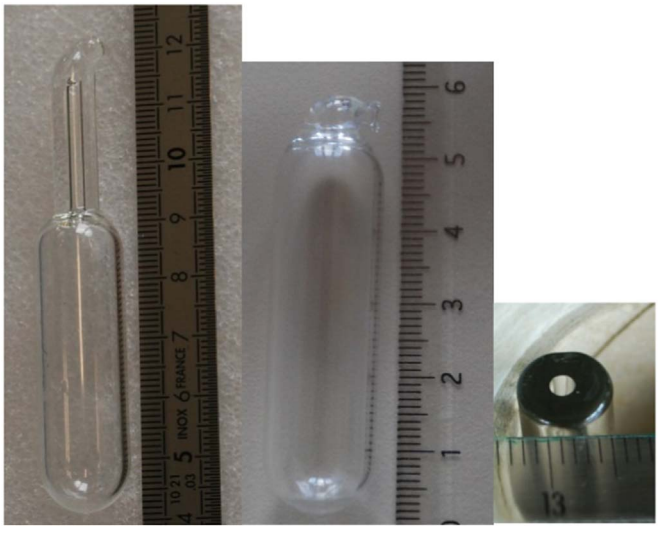

Fig. 2. BIPM gas ampoule sealed with long neck (on the left) and with a short neck (in the middle), and a view of the tube unsealed on the right.

2.1. Preamble

All the ampoules studied were normalized by the activity given by the DSA alpha-particle counter (after destruction of the ampoule and cryogenic pumping to the DSA). The uncertainties of measurements with the DSA are given in Table 1.

As already mentioned, three different ICs were used in two different laboratories: two chambers (named 2 A and 6D) at LNHB, and the SIR chamber number 389 (Ratel, 2007) at BIPM. The characteristics of the chambers are given in Table 2.

Measurements are decay-corrected and expressed at a common reference date. A dedicated plastic holder for reproducible positioning in the well of the IC is always used.

2.2. Influence of the ampoule volume and sealing point

In measuring radon gas, the ampoule volume must be considered because the gamma emission arises mostly from solid progenies which are attached to the surface of the ampoule (which is a function of the volume). The volumes of the ampoules were precisely measured using weighing of degassed distillated water. First, the empty ampoules were weighed. Then they were filled with water and weighed again, taking into account the position of the future sealing point. The relation between volume and weighing enabled the deduction of the effective volume of the ampoules. The uncertainties are given in Table 3. Since the relative uncertainty is lower than $8.5\ 10^{-5}$, the value of 10^{-4} used in our calculations is a conservative one.

Three gas ampoules were filled with nominally 5.5 mL radon gas and flame-sealed. Their volumes and characteristics are given in Table 4. One ampoule was sealed with a short “neck”, whereas another was sealed with a long one. The third ampoule was chosen because it had a visibly thicker glass bottom (see Section 2.3).

The responses of the three ICs, as a function of the internal ampoule volume, are presented in Fig. 3 (results normalized to 1), and the uncertainties are given in Table 5. It can be observed that the responses of the three chambers display the same trend in response for the three ampoules. Note that the intention is not to compare the responses given by the three chambers for a single ampoule, because this response depends on the characteristics of the chamber, but rather the trend of the response curve for each chamber.

The volume difference between ampoules 1 and 2 is 1.10 (1) %. Nevertheless, in each curve, the responses to ampoules 1 and 2 are consistent, whereas ampoule 1 is sealed with a long neck and ampoule 2 with a short one. This shows that the internal volume and the position of the sealing point do not seem to be the parameters with the largest influence on the measurements.

In the 3 curves of Fig. 3, the result for ampoule 3 is not consistent with the results for ampoules 1 and 2, although its volume is close to that of ampoule 2 (the difference is 0.55 (1) %). Furthermore, ampoules 2 and 3 are sealed with a short neck. The only difference between ampoule 3 and the other two is the thickness of the ampoule bottom.

2.3. Influence of the thickness of the ampoule bottom

The thickness of the glass ampoule bottom was measured by x-ray absorption: a very thin metal rod with an electroplated ^{241}Am source was introduced inside six ampoules, and the photon flux through the bottom was measured using a collimated High Purity Germanium (HPGe) well detector with a nominal crystal size of 52.3 mm diameter $\times$ 57.3 mm height and a 0.5 mm thick Be window, having a 20 mm deep well of 10 mm diameter. The measurement of the attenuation of three characteristic photon emissions of ^{241}Am [21.16 keV (E_1), 26.34 keV (E_2) and 59.54 keV (E_3)] was used to quantify the glass thickness using the Beer-Lambert exponential law. Measuring the photon emission both without (I_0) and with (I) a glass ampoule between the source and the detector, the thickness (x) of the material is deduced