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Prototype of a primary calibration system for measurement of radon activity concentration

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HIGHLIGHTS

- A prototype of a calibration system for radon concentration is presented.
- The system uses a solid ^{226}Ra source and an HPGe detector.
- Calibration is provided within a systematic error of 4% and a random error $\sim 2\%$.

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ABSTRACT

To calibrate measurement devices for monitoring the activity concentration of ^{222}Rn in air, a prototype of a calibration facility is tested using a solid ^{226}Ra source and a high-purity germanium (HPGe) detector. An emanation box was mounted on the detector for online gamma measurements. Inside this box, a $32.8 \text{ kBq} \pm 3\%$ ^{226}Ra standard source was placed. An AlphaGUARD control radon monitor was connected to the emanation box with a pumping air system in an open flow mode as a reference monitor. The emanation coefficient of the source was controlled online by comparing the gamma activity of ^{214}Bi ($E_\gamma=609.3 \text{ keV}$), progeny of ^{226}Ra , to that of the calibration source. A standard ^{137}Cs source, installed within the emanation box, was used as a reference for gamma spectroscopy using the HPGe detector, with a total systematic error of 4% and a random error less than 2%. The ratio between gamma measurements and AlphaGUARD was 0.94 ± 0.4 ; which is within the 9% uncertainties of AlphaGUARD calibration.

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1. Introduction

The determination of the radon activity concentration in air requires the use of detectors calibrated with specific standard and reference materials. The use of primary and secondary radon (^{222}Rn) standards is important because it provides the opportunity to obtain reference sources or systems designed to assure the traceability of radon measurements in various conditions.

A radon primary standard source requires an absolute measurement of radon gas activity (in a glass flask or a bottle) or activity concentration in a reference atmosphere (a chamber or a box). The first absolute measurement method for ^{222}Rn was reported by Picolo (1996), based on the detection of the alpha particles directly emitted by ^{222}Rn . This method was also adopted by Dersch (1998, 2004) and Spring et al. (2006). Other methods for

standardization were based on the absolute measuring of ^{222}Rn activity by internal gas proportional counting (Busch et al., 2002), $4\pi\gamma$ NaI(Tl) measurement (Spring et al., 2006; Nedjadi et al., 2007) and liquid scintillation counting (LSC) (Cassette et al., 2006; Sahagia et al., 2010). Standardization systems for ^{222}Rn measurements in the atmosphere have been developed, either by measuring the gamma emitting decay products of ^{222}Rn gas traceable to a ^{226}Ra standard reference materials (SRM) (Sakamoto et al., 2005; López-Coto et al., 2007; Röttger and Honig, 2011; Heidary et al., 2011; Kim et al., 2012; Lee et al., 2013; Röttger et al., 2014) or using radon itself in an absolute calibration method (Spring et al., 2006).

The use of calibrated gaseous radon sources is suitable for most radon monitors to measure the activity of radon in air. However, the half-life of radon is not sufficiently long to permit high quality calibration in all cases, and secondary calibration systems (Falk et al., 1994; De Felice and Myteberi, 1996; Picolo et al., 2000; Dersch, 1998, 2004; Kotrappa, et al., 2005) are required as

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secondary standard. The procedure for secondary measurement has to be as simple as possible and the time to measure the activity of radon is to be short. A critical analysis of all these methods was reported by De Felice (2007). All of these activities in radon standardization could only be accomplished by the realization of a complex system for the generation, circulation, and recovery of the primary standardization and transfer of a radon primary source to secondary standards sources or systems.

The purpose of this work is to introduce a simplified, yet accurate, system for calibrating radon measurement devices. A radon standard system was constructed using a solid radium source with sufficient radon emanation rate of about 44%. The activity of emanated radon gas is determined by measuring the difference between the absolute activity of the solid ^{226}Ra source and the activity of a short-lived progeny of the same source determined using gamma-ray spectroscopy. This difference in activity indicates the amount of released radon activity into the system. A high purity germanium (HPGe) detector is used to measure the absolute activity of ^{226}Ra with online controlling of radon emanation (by measuring the ^{214}Bi activity, progeny of ^{226}Ra) to produce the released absolute activity of ^{222}Rn . This prototype of a calibration system was tested using AlphaGUARD (PQ2000Pro) monitor as secondary standard of radon activity concentration. Using the developed radon standard system, the calibration of radon measurement devices is greatly simplified and the measurement accuracy is improved.

2. Experimental setup and procedure

The concept of our standard of radon concentration measurement is to precisely measure the activity of a ^{226}Ra solid source and online control ^{222}Rn emanation by continuously measuring the ^{214}Bi activity of the ^{226}Ra source during the entire calibration period using γ -ray spectrometry (with an HPGe detector).

An AlphaGUARD (PQ2000Pro) radon monitor was used in this work as a reference device. A 10-min FLOW mode is standard for our version of the AlphaGUARD PQ2000Pro equipment. Some years ago, our device was calibrated using a NIST calibration source; Source code (SRM 4973-16), a liquid in heat sealed polyethylene capsule with a ^{226}Ra activity of 487.2 ± 6.2 Bq with a ^{222}Rn emanation factor of 0.877 ± 0.14 . This source was calibrated by pulse ionization chambers with the NIST primary radon measurement system (Collé et al., 1990). Also our AlphaGUARD was compared with another AlphaGUARD, which was calibrated by a primary standard source, at the All-Russian Scientific Research

Institute of Physic Technical Measurements (Mendeleev, near Moscow). There was a good correlation between the two devices with a total error (systematic and random) of calibration of $\sim 10\%$. Our own calibration using the NIST source resulted in a total calibration error (including source activity and emanation coefficient errors, random errors during measurements, etc.) of approximately 8–9%.

The experimental arrangement for the calibration system is shown in Fig. 1. A small emanation box ($2.1 \times 10^{-4} \text{ m}^3$) was installed on the HPGe detector. Inside this box, a $32.8 \text{ kBq} \pm 3\%$ ^{226}Ra standard source was mounted. The activity of ^{226}Ra was directly measured using the 186 keV line, and the results were compared with the standard value reported in the original certification document of the standard ^{226}Ra source ($33 \text{ kBq} \pm 5\%$), called the passport data. Both values were practically identical (with a level of uncertainty of 3%). The emanation box was connected to a radon monitor AlphaGUARD and pumping system ($10^{-3} \text{ m}^3/\text{min}$ with a flow rate controller) in an open flow mode. Fresh air was drawn through the flow rate controller and pumped to the emanation box, reaching the AlphaGUARD monitor through an air filter. With a good quality pump and good quality flow controller used in the current work, the flow rates did not change by more than $\sim 1\%$ during the experiment.

Radon emanating from the solid radium source was transferred by the flow to the AlphaGUARD monitor. The radon concentration was measured directly by the AlphaGUARD monitor in the flow open mode every 10 min. At the same time, the gamma spectrum of the ^{226}Ra solid source was measured continuously during time intervals of 20 min. The emanation factor of the ^{226}Ra emanating source was controlled online by measuring the ^{214}Bi ($E_\gamma = 609.3 \text{ keV}$) activity in the ^{226}Ra source and comparing the measured value with the activity of ^{226}Ra in the source. The radon emanation factor χ was calculated using

$$\chi_{222\text{Rn}} = \frac{A(222\text{Rn})}{A(226\text{Ra})} = 1 - \frac{A(214\text{Bi})}{A(226\text{Ra})} \quad (1)$$

where A is the activity. In general, for the emanation source, the emanation factor should be in the range $0 \leq \chi \leq 1$.

Because the radon gas was removed very quickly from the emanation box by the air flow, there was a very small effect of the wall-attached radon decay products in the volume of the emanation box. To determine the wall-attached radon decay products, the emanation source was removed from the emanation box and the empty emanation box was measured using a γ -spectrometer. Within the limits of the measurement error, there was a very small additional background (less than 0.2% of the measured activity) in

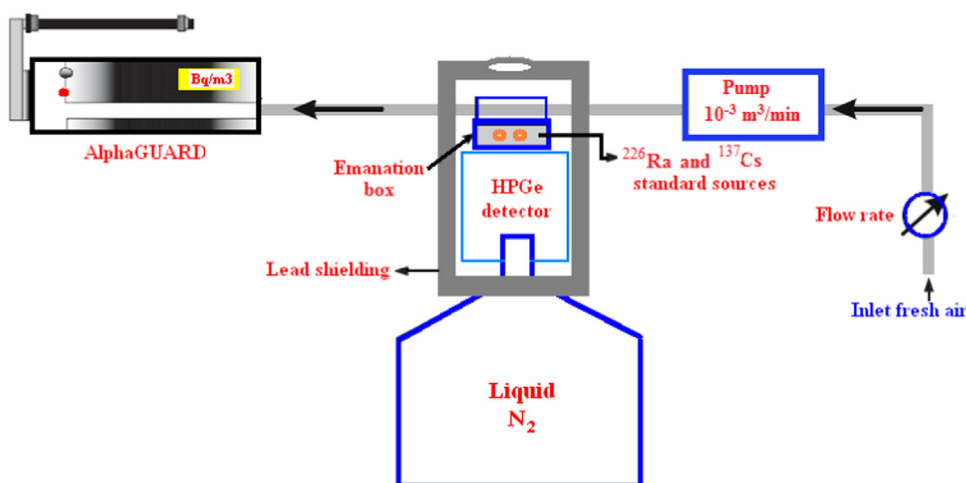


Fig. 1. Schematic of the experimental arrangement.

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