Journal of Radiation and Nuclear Applications
An International Journal

http://dx.doi.org/10.18576/jrna/040104

Radon Standard Source in Different Countries with Different Principals

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Received: 23 Nov. 2018, Revised: 12 Dec. 2018, Accepted: 23 Dec. 2018.
Published online: 1 Jan. 2019.

Abstract: Radon is the most important source of natural radiation exposure. The inhalation of short-lived radon progeny in ambient atmosphere is responsible on half of internal exposure and can lead to lung cancer. Therefore, the measurement of radon and radon progeny concentrations is very important for dose evaluation. Due to these facts, measurements of the activity concentration of radon and its progenies are performed worldwide either at workplaces (e.g. mines) or in dwellings. So, it is necessary to have the calibration facilities in which the activity concentration of radon and its progenies can be measured under well-defined conditions. Improving of radon metrology also is necessary for the traceability of secondary measurements of radon or radon progeny concentration in air. Unfortunately, the possibility of checking equipment is not available permanently. Therefore, there is a necessity to develop metrology approaches for measuring of the concentration of radon and radon progeny using available equipment and simple procedures to interpret the data. Each measuring method requires a suitable calibration procedures and standards for accurate measurements.
Keywords: Radon, Radon standard, Radon Primary standard.

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 or 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 [1–5]. Nowadays, a large number of commercial radon and radon progeny monitors were developed and widely applied in environmental survey. For quality assurance on measurement, a large number of reference radon chambers were built for establishing radon and radon progeny standard in addition to for assessing the quality of those devices and instruments as well [6].

With the interest in the measurements of the ^{222}Rn in the environment, and within dwellings and working places increasing, the calibration of radon monitoring instruments is gaining an importance in addition to the health effects of radon and its progeny, high-quality radon measurement services are therefore needed to support national recommendations and legal requirements. A large number of ^{222}Rn measurement methods have been developed for different matrices which use a variety of sampling techniques and measurement instrumentation. Each method requires suitable calibration procedures and measurement standards. These methods include ^{222}Rn chambers, associated ^{226}Ra sources, for the generation and characterization of standard ^{222}Rn atmospheres, ^{222}Rn flux density standards, and others. These standards are defined in terms of different parameters, for example, the total ^{222}Rn activity, the ^{222}Rn activity concentration, Equilibrium Equivalent Concentration of Radon (EEC_{Rn}) or the ^{222}Rn exposure. The detection efficiency of equipment must be calibrated in order to quantitatively measure radiation. Equipment is often calibrated using reference/standard sources. However, the calibration of the equipment used to

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measure radon concentration in air is difficult because, unlike other radioisotopes, there are no standard sources for radon due to its short half-life and status as a noble gas. Based on these facts, improving radon metrology is then necessary for the traceability of secondary measurements of radon in air concentration. In spite of its important

contribution to irradiation, only few inter comparisons of either radon standards or radon in air concentration have been carried out, and furthermore these have revealed a spread in the results of about 5% to 40% [7].

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 [8], based on the detection of the alpha particles directly emitted by ^{222}Rn. This method was also adopted by Dersch et al [9, 10] and Spring et al. [7]. Other methods for standardization were based on the absolute measuring of ^{222}Rn activity by internal gas proportional counting [11], 4πγ NaI(Tl) measurement [7, 12] and liquid scintillation counting (LSC) [13, 14]. 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) [15–22]or using radon itself in an absolute calibration method [7].

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 are required [9, 10, 23–25]. 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 some of these methods was reported by De Felice [26]. 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.

Therefore, Existence of a primary and secondary ^{222}Rn radon standard is important, as it provides the opportunity to obtain reference sources designed to assure the traceability of radon measurements in various conditions. The international bodies, ICRP, WHO, IAEA, EC, only recommend maximum levels of radon concentrations in dwellings and work places, but require coverage of reports by measurement and confident data. Recently, the International Commission for Radiation Protection (ICRP) reconsiders by amplification with a factor of about two, the ICRP Publication 65 evaluation of its radiological risk.

The link between the primary and secondary standardization was necessary to be established, in order to obtain the recipients with radon gas standard, to be used for the calibration of the devices used by various laboratories.

Calibration with a primary standard is performed, as example, via the realization of the activity concentration C by means of reference atmosphere, i.e. with the aid of a radon activity standard A and a calibration volume V: C = A / V. The system under test is entered into a reference volume to which the radon gas standard is applied. The value of the activity concentration measured by it and is compared, at the reference moment, with the activity concentration which results from radon gas standard and the calibration volume. These data are used to calculate the calibration factor for the indication of the system under test.

Calibration with a secondary standard is performed via the realization of the activity concentration by means of a reference measuring system. The system under test is entered into a closed volume together with the reference measuring system. The values of the activity concentrations of the two systems are compared at a reference moment. The data are used to calculate the calibration factor for the indication of the system under test.

With such a calibration (primary or secondary), the system under test also becomes a secondary standard and can be used as national standard at a state institute or a radiation protection institute. The relative uncertainties of a calibration by means of a secondary standard lie in the range from 2.5% to 8% (k=2), depending on the radon activity concentration. The uncertainties for a calibration by means of a primary standard are usually smaller (up to 2% (k=2)), as a radon gas activity standard is used.

The uncertainty stated is the expanded uncertainty of measurement obtained by multiplying the standard uncertainty by the coverage factor k = 2. It has been determined in accordance with the "Guide to the Expression of Uncertainty in Measurement" [27] normally, the value of the measured lies with a probability of approx. 95% within the assigned interval of values. The achievable measurement uncertainty depends on the measurement conditions and on the system under test.

Standardization systems on ^{222}Rn measurements in the atmosphere were developed in several countries by different laboratories. For example, the Environmental Measurement Laboratory (EML) in the USA, the Australian Radiation Laboratory (ARL; present the Australian Radiation Protection and Nuclear Safety Agency), the Environmental Engineering Center of the Japan Atomic Energy Agency (JAEA Ningyo-toge), the National Radiological Protection Board (NRPB; present the Radiation Protection Division, a part of the Centre for

Radiation, Chemical and Environmental Hazards of the Health Protection Agency) in the UK and Physikalisch-Technische Bundesanstalt (PTB) in Germany are known well in all over the world as the national references on radon measurements in their countries [20].

Traceability chains for radon measurement reference systems in the national metrology institutes are realized in two ways. In some cases, laboratories measure the gamma emitting daughters (^{214}Pb and ^{214}Bi) of ^{222}Rn gas traceable to ^{226}Ra Standard Reference Materials (SRMs). Alternatively, measurements performed in some laboratories are traceable to radon primary standards in which radon itself is measured with an absolute method [7]. The traditional methods used for the standardization of radon sources for calibration are often obtained by bubbling air through a radium solution standard source. However, there are many problems associated with this method with respect to radiation safety and calibration accuracy because this method uses a radioactive solution [14]. A number of other radon sources are used for calibration. For a radon source placed in a container of finely powdered radium [14], the radon emanation rate in the container is assumed to be 100%. Other sources are used to determine the radioactivity of radon by measuring the radioactivity of the short half-life progeny nuclide after the radon in the container reaches radioactive equilibrium.

2 Examples of the Radon Standard

A radon standard source was realized using a method of evaluating the radioactivity of the radon from a solid radium source based on the difference in radioactivity of the radium and the short half-life progeny of the remaining radon in the radium source [15]. In addition, by using the obtained radon standard source, the calibration of radon measurement devices is greatly simplified and the measurement accuracy is improved.

Emanation rate of radon to a vapor phase from a solid phase of the radium source depend on the physical and chemical states of the source. The activity of the radon emanating from the source can be deduced by measuring the activities of both the radium and the radon progeny remaining inside the solid source. A radon standard source was realized by sealing in a container the radiation source used to evaluate the radioactivity of the radon in the above-mentioned manner.

The following equation expresses the relationship between the activity and the photon counting rate, N, of the radiation detector:

$$A = \frac{N}{\eta\varepsilon} \qquad (1)$$

where η is the photon yield and ε is the absolute efficiency of the detector at the induced gamma energy.

By measuring the activities of the radium and one of the short half-life progeny nuclides of the radon in the radiation source, the radioactivity of the radon emanated in the vapor phase can be obtained as

$$A_{Rn} = \frac{N_{Ra}}{\eta_{Ra}\varepsilon_{Ra}} - \frac{N_P}{\eta_P\varepsilon_P} \qquad (2)$$

where A_{Rn} is the emanated radon activity, the suffixes Ra and P indicate values for radium and the progeny nuclide of radon, respectively.

The effective emanation coefficient χ of a source, the fraction of radon gas transferred to the reference volume or equipment, is calculated using equation 3. In general, for emanation source the emanation coefficient is fulfilled by the equation $0 \leq \chi \leq 1$.

$$\chi_{Rn-222} = \frac{A_{Rn}}{A_{Ra}} = 1 - \frac{A_P}{A_{Ra}} \qquad (3)$$

The detection efficiency of the detector and the gamma-ray yield per disintegration of both the radium and the radon progeny nuclide are needed in order to determine the quantity of radon emitted in vapor phase using this equation, and the accuracy of these values greatly affects the result.

The German radon-measuring group at PTB maintains a research and calibration facility for the development of new accurate techniques for measuring the activity concentration of radon and its respective short-lived progeny (Figure 1) [17].

The traceability of ^{222}Rn reference atmospheres is achievable using radon gas activity standards and standard volumes, and is an established method. The activity concentration of ^{222}Rn is realized via a radon gas activity standard (the gas activity stated at a given time for the sealed vessel and a defined volume). Activity, volume (differential volume: reference volume minus air displaced by the radon monitor under test) and time measurement have to be traceable) [17]. Two typical procedures for the calibration of active radon monitors are used at PTB:

1) a primary method based on a reference activity concentration realized by a primary radon gas activity standard and a calibration volume, both values are traceable to national standards, and
2) a secondary method based on a calibration via a reference monitor enclosed in the same atmosphere as the system under test.

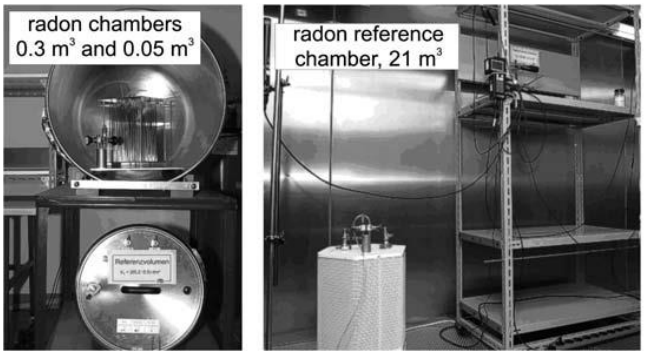

Fig. 1: Chambers available at PTB: chambers with volumes of 0.3 and 0.05 m3, radon reference chamber with 21 m^3.

The radon calibration facilities at PTB have provided the bases for the performance of reliable and exact measurements of radon and radon progenies in Germany. Since the establishment of the radon reference chamber in 1990 and its use in this field, PTB has succeeded in becoming one of the first addresses in the world for radon metrology. Its clients are national metrology institutes, radiation protection institutes and a number of companies.

The traceability of the activity concentration (C) is given by the activity of radium A (Ra-226) in the emanation source, the emanation coefficient χ of the emanation source and the reference volume V. For practical reasons, in calibrations a factor φ=1.0 of ten is induced to assign an uncertainty to the homogeneity of the activity in the volume [16].

$$C = \frac{A \times \chi}{V} \text{ or } \frac{A \times \chi}{V} \times \phi \qquad (4)$$

In the same way, the Radionuclide Metrology Laboratory from **IFIN-HH, Romania** presents the ^{222}Rn standard system using a Pylon solid ^{226}Ra source, and a glass circuit for circulation and recovery of ^{222}Rn in glass ampoules, at the 77K temperature. The radon can be recovered both in ampoules with liquid scintillator (LS), for absolute standardization of ^{222}Rn by liquid scintillation counting (LSC) [14], and in evacuated ampoules as gas, simultaneously or by the transfer of radon from gas into LS ampoules, in order to establish the traceability chain. The absolute standardization consists in the registration of the double coincidence counting rate in LS in equilibrium conditions.

Two circuits, a glass and a metallic one, are to be used in the system. The glass circuit serves for the generation, circulation, recovery of radon and for elaboration of the measurement method. It can be used in parallel with a metallic circuit, containing all the necessary gauges for the control of pressure and temperature. The glass and metallic circuit are presented in Figure 2. The whole description of absolute standardization by liquid scintillation counting presented in details in [13,14]. The absolute standardization is done by using the Liquid Scintillator Counting (LSC), by measuring the short half-life ^{222}Rn decay chain components in equilibrium. In addition to relatively, by the gamma–ray spectrometry using a high resolution HPGe system and by the measurement in a CENTRONIC IG12/20A well type ionization chamber, with all types of recipients [13, 14]. With a high efficiency HPGe detector, the measurements were made on the two main full absorption peaks (photopeaks): 352 keV (Pb-214) and 609 keV (Bi-214).

Fig.2: A glass and metallic circuits in Radionuclide Metrology Laboratory presents the 222Rn Standard System.

The system for generation, recovery and standardization of ^{222}Rn contains: a solid ^{226}Ra source for controlled ^{222}Rn generation; a gas circuit connected to a vacuum and cryogenic system; a system for quantitative transfer of radon in frozen liquid scintillator; quantitative recovery of radon gas in secondary standards. The link between the liquid scintillator and gas recipients is direct and allows the transfer between gas and scintillator; another possibility is to transfer the radon from vials received in laboratory inside the circuit. Figure 3 presents the concept of the radon standard.

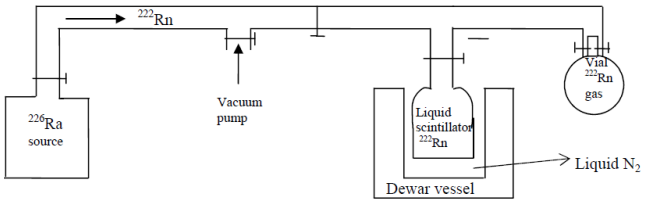

Fig. 3: The scheme of the radon standard system in Radionuclide Metrology Laboratory presents the 222Rn Standard System.

A comprehensive analysis of Romanian group papers [13, 14] in the Radionuclide Metrology Laboratory from IFIN-HH, Romania gives us these points as conclusion:

- The primary radon standard was conceived as a complex structure, involving equipment and methods designed to assure the continuous traceability chain: obtaining of ^{222}Rn from a solid ^{226}Ra source; absolute standardization; transfer of the ^{222}Rn activity unit from the primary to secondary standard systems; preparation of recipients with standard gas radon, to be delivered to the final users.
- Two variants of systems were conceived and set up:
- Initial, glass variant (ii) Final, metal variant, with the radon circulation fully controlled.
- The absolute standardization of ^{222}Rn, in equilibrium with its short half-life daughters was done by the liquid scintillation method.
- The relative measurements and calibrations were done with a HPGe spectrometric system and an ionization chamber.
- The combination of the absolute standardization by LSC with secondary systems measurements: HPGe gamma-ray spectrometer and CENTRONIC IG12/20A ionization chamber were used for two types of operations:
- Determination of the activity of radon gas recipients with a satisfactory uncertainty.
- Calibration of the two secondary systems, in terms of counting efficiency and respectively calibration factor, with satisfactory uncertainties.
- A complete traceability chain, allowing the transfer of the activity unit from the primary to the working standards was experimentally established.
- Studies on the liquid scintillator and geometry efficiency were performed, in order to demonstrate the correctness of the work hypotheses.

The Korea Research Institute of Standards and Science (**KRISS**) have developed a primary system for the activity standardization of gaseous radon-222 [21]. The KRISS radon primary measurement system is based on the defined solid angle counting method which has been introduced for radon measurements by LPRI [8] for the first time. The combined uncertainty measured by the radon primary measurement system at KRISS is known to be 0.14% [21] and several types of containers including glass ampoules are used to transfer the gaseous radon standards. After transferring the measured radon gas into containers, a measurement for the residual radon can be made again to confirm a complete transfer.

A glass ampoule source of gaseous ^{222}Rn standardized by the radon primary measurement system based on the defined solid angle counting method was used to calibrate the reference ionization chamber for gamma-emitting radionuclides at KRISS [20]. A radon standard source was prepared by transferring the gaseous radon into a flame-sealed glass ampoule (20mL) after measuring it by the KRISS primary measurement system [20, 21].

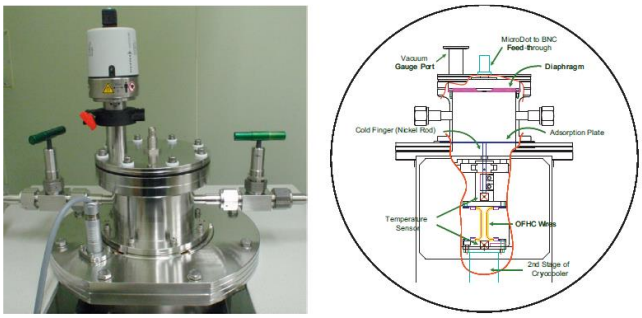

Fig. 4: KRISS radon chamber and its schematic diagram.

The efficiency, as the calibration result, of the ionization chamber for radon and its progeny was in affair agreement with that calculated by using the energy-dependent efficiency of the ionization chamber and in a good agreement with that measured by using an aqueous ^{226}Ra standard solution source from NIST. Authors in [20] concluded that the reference ionization chamber may be a good secondary standard for disseminating the gaseous radon standard traceable to the radon primary measurement system.

The traceability on radon measurements at the Environmental Engineering Center of the Japan Atomic Energy Agency (JAEA) Ningyo-toge was discussed in [28]. The primary standard is a radium solution provided by the NBS, and the secondary standard is the method with gas-filled ionization chambers calibrated with the solution. It was experimentally confirmed that the method has high accuracy for the radon measurement. Through the intercomparison experiments among the international reference institutes, it was also confirmed that the reliability

and consistency of measurement techniques at the JAEA Ningyo-toge has been retained since 1984. It shows that the calibration and measurement techniques associated with the traceability system constructed were maintained well at the JAEA Ningyo-toge. Furthermore, it was illustrated in this paper how to manage the traceability with radon reference chamber and the method standardized at the JAEA Ningyotoge.

Three years ago, the first prototype of radon EEC_{Rn} and radon activity concentration is presented in Russia at Ural federal university [29–32]. A high purity germanium (HPGe) detector is presented as a reference measuring device to produce more precise and less complicated calibration system of radon concentration and EEC_{Rn} measuring equipment.

In this prototype of ^{222}Rn primary standard, the small emanation box was mounted on the high purity germanium (HPGe) detector for online gamma measurements. Inside this box, a solid ^{226}Ra standard source was placed. HPGe detector was used to measure the absolute activity of ^{226}Ra with online measuring of radon emanation coefficient by measuring of the ^{214}Bi activity. This prototype of a calibration system was tested in opened and closed scheme with different ^{226}Ra sources using Alpha GUARD monitor [30, 31], shown in Figure . The range of standardized radon activity concentration in dependence on ^{226}Ra source activity and calibration scheme was from 300 to 30000 Bq/m^3. In the case of EEC_{Rn} measurements, it was demonstrated that more precise and less complicated calibration of EEC_{Rn} measuring equipment can by conducted by the use of HPGe as a reference measuring device [29]. the developed radon concentration standard systems, the calibration of radon and EEC_{Rn} measurement devices are considerably simplified and the measurement accuracy is improved with level of uncertainty not more than 4 %.

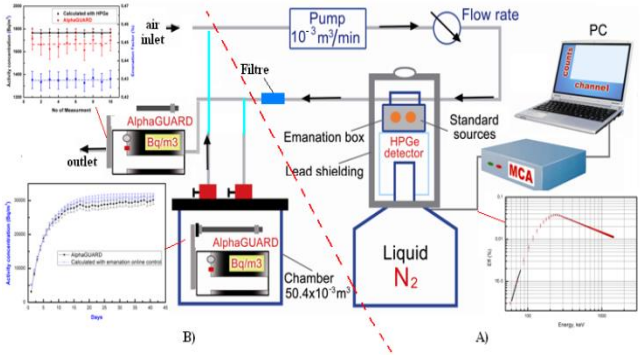

Fig. 5: Experimental set up of radon activity concentration calibration system.

3 Conclusions

Obviously, there are many methods to calibrate radon instruments depend on the nature gaseous state of radon and the its small lifetime. These methods are based mainly on the measurement technique and the availability for calibration. In the end, the continuity of the calibration of the radon measuring instruments should be continuing. Therefore, each country should have or should create a radon standard for calibration.

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