Concept for a primary Romanian radon standard

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Abstract
The paper presents the concept of a complex system, aimed to assure the traceability of ^{222}Rn measurements, from the absolute (primary) standardization to the preparation and delivery of secondary standards, gas vials. The system will contain a solid ^{226}Ra source, a gas radon circuit, connections with a liquid scintillator vial and with glass vials. The absolute standardization of the ^{222}Rn, in equilibrium with all the short half life daughters, will be performed by the method of the Liquid Scintillation Counting (LSC). The system, and method, will allow our laboratory to take part in future international ^{222}Rn comparisons. The transfer of activity unit from the primary to the secondary standardization will be performed by the preparation of vials with ^{222}Rn gas, comparative measurements by LSC and a GeHP gamma-ray spectrometry system, or a well type NaI(Tl) crystal, and their link. The secondary standards will be used for the calibration of measurement instruments, for assurance of controlled radon atmosphere in "radon chambers", and for the validation of some calculation models for various detectors efficiency. The range of activities for secondary standards is in

agreement with the national measurement necessities.

***KEYWORDS**: ^{222}Rn, absolute standardization, secondary standards*

1. Introduction

Existence of a primary ^{222}Rn radon standard is important, as it provides the opportunity to obtain reference sources designed to assure traceability of radon measurements in various conditions. Several types of requirements, to be accomplished according to the national and international legislation and the aim of measurement (routine measurement, assurance of imposed legal levels, calibration, method validation), are the main constraints in designing the system.

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 [1-3]. Romania established recommended reference levels of radon concentration: 400 Bq m^{-3} and 200 Bq m^{-3} for existent, respectively new dwellings, and 400 Bq m^{-3} for work places. A special category is the occupational exposure to radon The resulting interval of measurement necessities is 75-3000 Bq m^{-3} and the corresponding interval of secondary standards is 0.75 - 30 kBq, accordingly. Referring the technical conditions, the users require stable mixtures of standard radon and carrier gas, excluding water vapors or solid impurities; the vials are required to assure the lack of any adsorption of radon on the walls.

All these requirements can only be accomplished by the realization of a complex system for generation, circulation and recovery, primary standardization and transmission of radon activity unit to secondary standards, such as presented in the paper.

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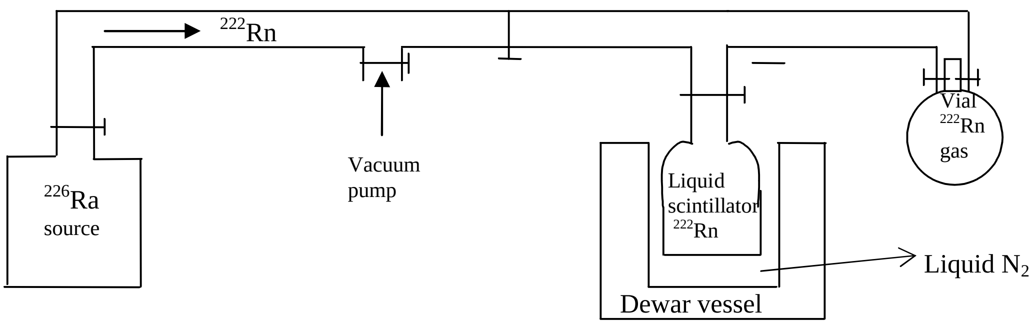
2. Presentation of the system concept and absolute standardization

2.1 The radon system

The system for generation, recovery and standardization is conceptually similar to that presented in paper [5], and contains: a solid ^{226}Ra source for controlled ^{222}Rn generation, in our case a 250 kBq unit; 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 reference volume vials and in secondary standards. The link between the liquid scintillator and gas vials is direct and allows the transfer between gas and scintillator; another possibility is to transfer radon from vials received in laboratory within comparative measurements. The whole system is placed in a ventilated box, with controlled circulation of radioactive gases, situated in the building of the Radioisotope Department.

Figure 1



2.2 Absolute standardization method

The absolute standardization is intended to be performed by the Liquid Scintillator Counting (LSC), by measuring the ^{222}Rn decay chain components in equilibrium.

^{222}Rn decay chain:

$$ {}^{222}_{86}Rn \xrightarrow[3.8235\ d]{\alpha} {}^{218}_{84}Po\ (\text{Ra A}) \xrightarrow[3.05\ \text{min}]{\alpha} {}^{214}_{82}Pb\ (\text{Ra B}) \xrightarrow[26.8\ \text{min}]{\beta^-} {}^{214}_{83}Bi\ (\text{Ra C}) \xrightarrow[19.9\ \text{min}]{\beta-} $$

$$ {}^{214}_{84}Po\ (\text{Ra C'}) \xrightarrow[165\ \mu s]{\alpha} {}^{210}_{82}Pb\ (\text{Ra D}) $$

The complete equilibrium is established after a time $T_{equilibrium}$ = 4.667 h. The decay chain contains three alpha, ^{222}Rn, ^{218}Po, ^{214}Po and two beta –gamma high beta energy, ^{214}Pb and ^{214}Bi, emitters. The main equations allowing the calculation of each vial activity was established in the paper [5] The basic relation, between the counting rate, R, s^{-1}, and individual activities of ^{222}Rn and daughters A_i, s^{-1}, is :

$$ R = \varepsilon_{Rn222} s_{Rn222} A_{Rn222} + \varepsilon_{Po218} s_{Po218} A_{Po218} + \varepsilon_{Pb214} s_{Pb214} A_{Pb214} + \varepsilon_{Bi214} s_{Bi214} A_{Bi214} + \varepsilon_{Po214} s_{Po214} A_{Po214} e^{-\lambda \tau_{eff}} \quad (1) $$

where ε are detection efficiencies, practically as high as 100% in LSC, S are emission intensities ; $\lambda = 4200\ s^{-1}$ is the decay constant of ^{214}Po ; τ_{eff} is the effective dead time of the LSC.

In equilibrium condition, relation (1) becomes:

$$R = A_{Rn\,222} \cdot (4.0151 + 1.0091 \cdot e^{-\lambda \tau_{eff}}) \qquad (2)$$

Relation (2) allows for the direct determination of ^{222}Rn activity from R, if τ_{eff} is well determined. It depends both on the LSC imposed dead time and the source counting rate. The details of this correction are given in paper [5]. Sources with activities less than 5000 Bq are prepared, by the quantitative transfer of the gas radon in frozen liquid scintillator, at 77K. The standardization of vials containing radon will be performed by the use of the liquid scintillation system existent in the laboratory, based on the principle of triple to double coincidence ratio (LSC-TDCR) [6]. The measurement is performed 5 hours after the sample preparation, in equilibrium conditions. The evaluation of uncertainties will summarize the type A-statistics and type B-components, which are due to background counting, detection efficiency, radon recovery in the scintillator volume.

2.2 Obtaining of secondary standards

For obtaining of secondary standards and transfer of the radon activity unit from the primary to the secondary standards, the same system, such as presented in Figure 1 is used. The proposed sequence of operations is: one accumulates the desired radon activity in the source, which is then released in the system and completely recovered as gas in a reference glass vial, with the volume precisely determined, V_1. The vial is then sealed and left for 5 h, in order to get the equilibrium. The vial is measured, in a fixed geometry, with the use of a gamma spectrometry system, GeHP (GeLi), or a well type NaI(Tl) detector; counting rates corresponding to the established energetic intervals are registered. Then, the vial is again attached to the radon system and the gas is recovered in the LS vial. The LS vial is absolutely standardized, after a 5 h interval. The following calculation relations are used

$$N_{\gamma} = \varepsilon \cdot A \qquad (3)$$

A is the ^{222}Rn activity of the gas vial***, ε*** is gamma measurement system efficiency, both unknown quantities.

A is then precisely determined by absolute standardization in LSC. From relation (3), one determines the detection efficiency for the reference vials, and the transfer of activity unit is carried for the secondary standard.

After the determination of the efficiency for various types of reference vials (recipients), their production is possible, as well as their relative standardization, by gamma-ray spectrometry method.

3. Conclusions

- The main requirements and the overall conception of a primary radon standardization system is presented

- The system functions allow for the coverage of the entire traceability chain in ^{222}Rn measurement, from the international level, to the end users.

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REFERENCES

[1 INTERNATIONAL COMMISSION ON RADIOLOGICAL PROTECTION: Protection against Radon-222 at Home and at Work. ICRP Publication 65. Pergamon Press (1993).

[2] INTERNATIONAL ATOMIC ENERGY AGENCY Safety Series 33 (2003)

[3]WORLD HEALTH ORGANIZATION: WHO Reggional Office for Europe: Indoor Air Quality: A risk-based approach to health criteria for radon indoors (1996)

[4] EUROPEAN COMMISSION: Commission Recommendation of 21 February 1990 on the protection of the public against indoor exposure to radon. European Communities. 90/143/Euratom (1990)

[5] CASSETTE, P., SAHAGIA, M., GRIGORESCU, E.L., LEPY, M.C., PICOLO, J.L. Standardization of ^{222}Rn by LSC and comparison with α- and γ-spectrometry. Appl. Radiat. Isot. 64(2006) 1465-1470

[6] RAZDOLESCU, A.C., BRODA, R., CASSETTE, P., SIMPSON, B.R.S., WYNGAARDT, W.M., The IFIN-HH triple coincidence liquid scintillation counter, Applied Radiation Isotopes 64(2006) 1510-1514.