State of Art of Radon and Thoron Volume Activity Measurements

Volodymyr Skliarov
Department of Information and Measuring Technologies
Kharkiv National University of Radio Electronics
Kharkiv, Ukraine,
Head of Department
National Scientific Centre "Institute of Metrology"
Kharkiv, Ukraine
vladimir.skliarov@gmail.com

Oleksandr Degtiarov
Department of Information and Measuring Technologies
Kharkiv National University of Radio Electronics
Kharkiv, Ukraine
oleksandr.degtiarov@nure.ua

Oleg Zaporozhets
Department of Information and Measuring Technologies
Kharkiv National University of Radio Electronics
Kharkiv, Ukraine
oleg.zaporozhets@nure.ua

***Abstract*— A control, monitoring and reduction of the dose load from ionizing radiation to the population is a national task, which is determined and coordinated by a number of Laws and Resolutions of the Cabinet of Ministers of Ukraine.**

Based on the accumulated experience in the creation and operation of the state primary standard for the unit of volumetric activity ^{222}Rn, a methodology for creating the state primary standard for the unit of volumetric activity ^{222}Rn (radon) and ^{220}Rn (thoron) is proposed.

The natural background radiation consists of cosmic radiation and radiation from natural radionuclides (^{232}Th, ^{226}Ra, ^{40}K) common in the Earth's crust, soil, air, water and other objects of the external environment. Some part of the territory of Ukraine is located on the Ukrainian crystal shield with significant reserves of ^{226}Ra and ^{232}Th and, as a result, ^{222}Rn (radon) and ^{220}Rn (thoron) emanations from the soil surface into the environment, housing and industrial premises.

The purpose of this work is to develop the structure and concept of constructing the state primary standard of the unit of volumetric activity ^{222}Rn and ^{220}Rn based on the accumulated experience in the creation and operation of the state primary standard of the unit of volumetric activity ^{222}Rn.

Keywords*— *state primary standard, radon chamber, thoron chamber, volumetric activity, radon generator, thoron generator.

I. INTRODUCTION

The greatest contribution to the irradiation of the body, primarily of its bronchopulmonary system, is made by the radioactive gas ^{222}Rn and its decay products, due to which certain groups of professionals and the population can receive extremely high radiation doses exceeding the maximum permissible levels.

Radiation safety standards of Ukraine regulate the average annual Equivalent Equilibrium Volumetric Activity (EEVA) of radon isotopes in the air of buildings:

– in the premises of buildings and structures under construction and reconstructed for operation with the constant presence of people, the level of action for the average annual EEVA of radon-222 in the air is 50 Bq/m^{-3}, the average annual EEVA of radon-220 is 3 Bq/m^{-3};

– the level of actions for the average annual EEVA of radon-222 in the breathing zone in the air of premises operated with the constant presence of people is 100 Bq/m^{-3}; and for EEVA radon-220 - 6 Bq/m^{-3};

– if the specified action levels are exceeded, countermeasures for children's, sanatorium and health resorts and health-care institutions, as well as public premises are mandatory: for residential premises - only with the consent of the owner of the home. In this case, the latter should be provided with complete information on radiation doses and health risks;

– if the average annual total EEVA of radon-222 and radon-220, after carrying out anti radon activities, cannot be reduced below the level of 400 Bq/m^{-3} (the level of actions of unconditionally justified intervention), then the decision on further actions belongs to the relevant state bodies, the order of which is regulated by a separate document.

Thus, one of the units of such physical quantities in the field of ionizing radiation and nuclear constants is the volumetric activity of ^{222}Rn and ^{220}Rn. The existing fleet of working measuring equipment contains equipment for determining both the volumetric activity of the initial radionuclide and its daughter decay products (DDP).

II. MEASUREMENTS STANDARDS AND CHAMBERS IN THE FIELD OF RADON AND THORON MEASUREMENTS

The state primary standard of the unit of volumetric activity ^{222}Rn was created in 1997 and improved in 2011 at the National Scientific Centre “Institute of Metrology” (Kharkiv, Ukraine).

In the period from 2004 to 2006, the standard participated in international comparisons on the topic COOMET.RI (II)-S1, ^{222}Rn (169 /UA/98) “^{222}Rn VOLUME ACTIVITY COMPARISON”. As a result of comparisons, the metrological characteristics were confirmed, which allowed Ukraine to confirm line CMC, which is necessary for the mutual recognition of certificates issued by national metrological institutes [1, 2]. In world practice, the basis for creating a radon or thoron atmosphere is chamber-rooms with different volumes. A liquid, solid-state or gaseous standard (standard) ^{222}Rn or ^{220}Rn is used as a source (generator) of radon or thoron.

The national standard for the unit of volumetric activity ^{222}Rn was created by specialists from the Physikalisch-Technische Bundesanstalt (PTB), Braunschweig, Germany in 1991.

Compare to other national standards of the unit of volumetric activity ^{222}Rn, it reproduces the standard radon

978-1-7281-9719-7/20/$31.00 ©20XX IEEE

atmosphere and the programmed radon atmosphere with the ability to set thermodynamic characteristics (temperature, pressure, humidity, aerosol concentration). The internal structure of a radon chamber with an internal volume of 21.035 m^3 is shown in fig. 1.

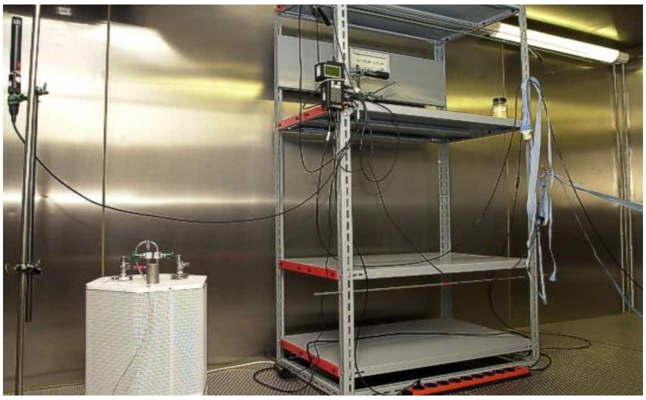

Fig. 1. Radon chamber of the National standard of Germany of the unit of volumetric activity ^{222}Rn.

The radon atmosphere in the radon chamber can be set warm, cold, dry, humid and standard for the possibility of researching measuring instruments (MI) created in Germany of volumetric activity and equivalent equilibrium volumetric activity ^{222}Rn [3].

The climatic system of the radon chamber operates in two modes with the temperature of the radon atmosphere from -20 °C to 10 °C and with the temperature of the radon atmosphere stable at any point in the range from 10 °C to 40 °C.

To reduce humidity, the condenser lowers the temperature. Thus, the humidity in the radon chamber can vary from 95 % to 5 %.

In addition to changing the thermodynamic characteristics of the radon atmosphere, the radon chamber allows experiments with aerosols of various sizes and concentrations [4]. The aerosol source is “Carnauba” wax. This material contains carbon-hydrogen molecules. “Carnauba” wax allows the production of aerosol particles of various sizes from 10 nm to 1 µm. “Carnauba” wax aerosols have unique medical and chemical properties in terms of shape, size; density and acidity of the environment. From a theoretical and experimental point of view, they are the ideal choice for working in a radon chamber. The spherical shape of wax aerosols is advantageous for adsorption by charged ions. The radon chamber was designed and created to provide stable environmental parameters during the calibration of ^{222}Rn measuring instruments and its DDP, which is achieved by an atmosphere control system, an effective aerosol generator and an aerosol cleaning air system. Environmental parameters are determined and measured within an uncertainty range of 0.5 % to 5.0 %. This provides ideal conditions for the calibration of both passive and active ^{222}Rn measuring devices with forced circulation of the investigated atmosphere. In addition, it makes it possible to change the equilibrium factor F between ^{222}Rn and its DDP.

When reproducing a unit of volumetric activity ^{222}Rn, a ^{222}Rn generator and a pulsed ionization chamber are used.

As part of the national standard of Germany, three pulse ionization chambers with different structures of the internal electrode are used. Also, as in the national standard of the United States, the international standards for the mass 226Ra [5, 6] were included in the national standard of Germany. A distinctive feature is the use of the absolute method for determining the activity of ^{222}Rn in the known "solid angle" (Picolo method). Gaseous radon enters the "solid angle" and, spreading over the volume, condenses. The alpha particles emitted by the radon atmosphere are measured with an alpha spectrometer, the detection efficiency depends only on the geometry of the volume and the detector efficiency is determined by the ratio of the "solid angle" through which the detector counts alpha particles.

The characteristics of the national standard of Germany are presented in table. 1.

The expanded uncertainty of this National Standard for the unit of volumetric activity ^{222}Rn is 2.5 % with a confidence level p = 0.95.

The reference installation for measuring the volumetric activity of radon (Republic of Belarus, Minsk, BelGIM) [7] consists of two chambers with a volume of 0.142 m^3 and 3.087 m^3. Both cameras are equipped with windows for visual observation, connectors for power, equipment, sensors and sampling systems. The reference installation is completed with six exemplary radon generators of various activity, which are certified on the basis of an exemplary ^{226}Ra solution. The created installation provides calibration and verification of working instruments of measuring equipment in the range from 15 to 50 % with a confidence level of 0.95. The range of reproduction of a unit of radon volumetric activity is from 20 $Bq \times m^{-3}$ to 60 $kBq \times m^{-3}$ with an expanded uncertainty of no more than 10 % at a confidence level p = 0.95.

The complex of impulse chambers of the Environmental Measurement Laboratory, New York [8, 9] is a stainless steel room with a volume of 19.2 m^3. The volumetric activity of ^{222}Rn is created in the range from 50 $Bq \times m^{-3}$ to 3700 $Bq \times m^{-3}$, the temperature is set and maintained in the range from 0 °C to 45 °C and the humidity is from 20 % to 90 %.

Pulsed ionization chambers certified by the National Institute of Standards and Technology (NIST, USA) using a ^{226}Ra reference solution are used as a reference device.

The US Bureau of Mines Radon test Chamber, Colorado [10] is a cylinder 2.13 m long and 1.52 m in diameter, respectively, has a volume of 3.9 m^3. The design of the chamber allows the selection of the generated volumetric activity. The volumetric activity of ^{222}Rn is created in the range from 37 $Bq \times m^{-3}$ to 37000 $Bq \times m^{-3}$. The concentration of aerosols is possible in the range from 5000 cm^{-3} to 200000 cm^{-3}.

The radon chamber of the U.K. National Radiological Protection Board [11] allows creating and maintaining a volumetric activity of ^{222}Rn up to 2000 $Bq \times m^{-3}$ in a volume of 43 m^3. The auxiliary equipment makes it possible to simulate a radon atmosphere with aerosol particles in the range from 100 nm to 300 nm with a concentration of up to 3×10^4 cm^{-3}. The chamber ensures uniformity of the volumetric activity of ^{222}Rn throughout the entire volume with the ability to control the temperature and humidity of the created radon atmosphere.

TABLE I. CHARACTERISTICS OF THE GERMAN NATIONAL STANDARD

Characteristic	Value
Temperature range, °C	from -20 to + 45
Relative humidity, %	from 5 to 95
Aerosol diameter, μm	from 30 to 300
Aerosol concentration, m^{-3}	from 10^{6} to 10^{13}
Volume activity ^{222}Rn, Bq×m^{-3}	from 10 to 100000
Volumetric activity of DDP ^{222}Rn, Bq×m^{-3}	from 10 to 100000
Equilibrium coefficient, F	from 0,1 to 1

The radon chamber of the Australian Radiation Laboratory (Victoria) [12] is a chamber with an internal volume of 7.2 m^{3} and the ability to create a volumetric activity of ^{222}Rn up to 60,000 Bq×m^{-3}. Auxiliary equipment provides control and maintenance of humidity in the range from 20 % to 50 %.

The calibration chamber of the Institute for Nuclear Protection (BACCARA, Saclay, France) [13] has a cylindrical shape and an internal volume of 1 m^{3}. Volumetric activity is provided in the range from 40 Bq×m^{-3} to 40 kBq×m^{-3}.

The radon chamber of the Elliot Laboratory (CANMET, EMR, Ontario, Canada) [14] is a pressurized room with an internal volume of 30 m^{3} and a transition sluice with a volume of 3 m^{3}. The range of measurement and maintenance of temperature is from 10 °C to 35 °C, relative humidity is controlled in the range from 5 % to 99 %. Thermodynamic parameters are measured and maintained using a microcontroller air conditioning system. In addition to providing the volumetric activity of ^{222}Rn, the radon chamber allows the creation of the volumetric activity of ^{220}Rn. 6 radium standards with activities of hundreds to thousands of kBq are used as ^{222}Rn generators. Six thorium standards with activities of hundreds to thousands of kBq are used as Thoron generators.

The calibration radon chamber of the Paul Scherrer Institute, Switzerland [15] represents a sealed volume of 12.6 m^{3} with a 1.6 m^{3} transition sluice. A liquid radium source with the ability to create a radon atmosphere is used as a ^{222}Rn generator up to 15 kBq×m^{-3}. The microprocessor-based air conditioning system measures and maintains the following thermodynamic parameters: temperature from 5 °C to 40 °C, humidity from 20 % to 95 %.

The radon chamber of the Radiological Protection Institute of Ireland, Dublin [16, 17] is a complex of three modules: a chamber, a gateway, and a working compartment. The total volume of 8 m^{3} is provided with a system for monitoring thermodynamic parameters and emergency ventilation. The radon chamber is equipped with an observation window, sensors for monitoring the radon atmosphere, and rubber gloves for working inside the working compartment. The microprocessor system provides information exchange between the equipment in the working chamber and external devices. As a ^{222}Rn generator, a solid-state (dry, powdery) radium source with a volume of 5 liters, representing a powder of ^{226}Ra salt with a 100 % yield of ^{222}Rn, was used.

One of the last known ones created to date is the Thoron-Folgeprodukt-Kammer (PTB, Germany) daughter decay product chamber [18]. Thoron daughter decay product chamber (TKF) is The "Vötsch" sealed chamber with a volume of 6 m^{3} with the ability to measure and control temperature in the range from 10 °C to 70 °C and humidity from 10 % to 95 % (fig. 2).

Ten lead cubes with ^{228}Th sources are evenly distributed over the TFK volume (fig. 3). The internal structure and arrangement of the apparatus in the chamber of the daughter products of thoron decay is shown in fig. 4. Each source can be controlled separately, independently of the others. Each source extends and retracts by means of a threaded mechanism. Positioning is controlled by limit switches. At the same time, the generator control system ensures the homogeneity (uniformity) of the thoron atmosphere with a range of reproducible activities from 400 Bq×m^{-3} to 8 kBq×m^{-3}. With the possibility of increasing the activity up to 10 kBq×m^{-3} (^{220}Rn and ^{222}Rn) of a single applied activity.

Fig. 2. Chamber of Thoron decay daughter products (PTB, Germany).

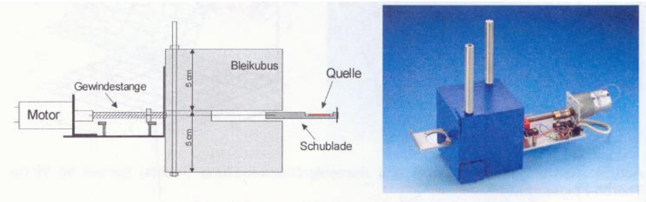

Fig. 3. Generator of ^{228}Th.

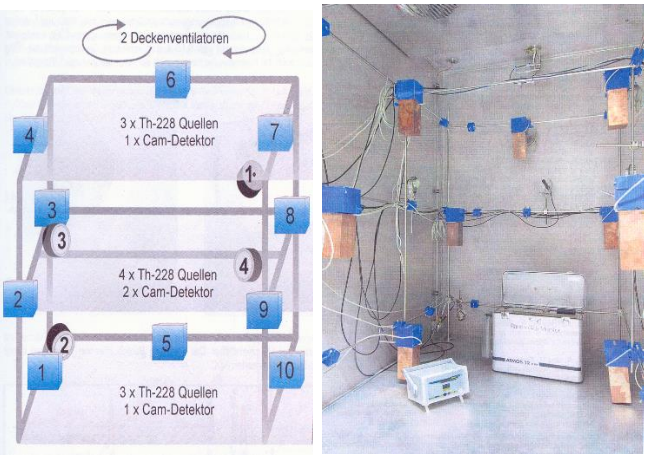

Fig. 4. Internal structure of the chamber of daughter decay products of thoron.

Thus, the establishment of additional sources to increase the activity of the created atmosphere is ensured. In addition to calibration and safety, the production of a reference atmosphere must ensure its homogeneity. In the study of the radon atmosphere, this problem is not, due to the long half-life of ^{222}Rn.

The provision and elimination of the temperature gradient is also not a concern for ^{222}Rn and its daughter products.

However, ^{220}Rn has a short half-life of 55.6 s. Therefore, the source control system must provide constant replenishment and circulation of the thoron atmosphere. The simplest ventilation system, in the future, will be able to ensure circulation and uniformity. This can be proved by continuous monitoring by means of an air monitoring system using 4 detectors (fig. 5), by means of local measurements of ^{216}Po.

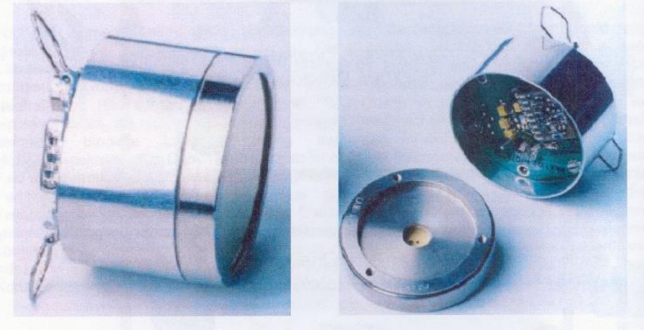

Fig. 5. Detector ^{216}Po.

Here, ^{216}Po, due to a half-life of 0.15 s, is constantly in equilibrium with ^{220}Rn, which in turn is proof of the homogeneity of the ^{220}Rn atmosphere. Signals are fed from 4 detectors to one ADC unit, where they are processed. The alpha spectrometer determines the values and generates a signal for constructing a spectrum with 0.3 MeV resolution, this allows you to select ^{216}Po (6.778 MeV, 0.16 s). The Atmos 12 DPX radon monitor, modernized for measuring 220Rn, and the universal ^{222}Rn and ^{220}Rn meter - RTM 1688-2, were used as recording devices.

One of the absolute methods for determining the ^{220}Rn concentration and ^{220}Rn DDP is the “double filter” method [19]. The method is insensitive to the presence of any concentration of ^{222}Rn. Turbulence, humidity and air temperature do not affect the measurement. This method was primarily developed for measuring ^{220}Rn and ^{212}Pb concentrations in work places in underground environments with high (^{222}Rn) concentration, but this methodology can be used for reference measurement of ^{220}Rn concentration in calibrators.

It is known that ^{222}Rn and its daughter products of its decay make a significant contribution to the dose load on the population. ^{220}Rn does not pose a significant problem due to its short half-life, but its DDPs make a significant contribution with long half-lives. The measurement of the ^{222}Rn concentration can be replaced by the measurement of its DPR using the equilibrium factor, which is usually used to estimate the ^{222}Rn dose. However, this principle cannot be used to determine the ^{220}Rn concentration. This is due to the fact that the equilibrium between ^{220}Rn and its DDP is influenced by many factors, such as the concentration of aerosols in the air and air flows, as well as the influence of the short-lived ^{220}Rn and the relative long-lived ^{212}Pb. To estimate the dose from ^{220}Rn and its DDP, it is necessary to measure both ^{220}Rn and ^{212}Pb.

The “double filter method” is based on the collection of DDP ^{220}Rn. The unit consists of a branch pipe, an inlet filter and an outlet filter (fig. 6). Air is drawn in through the inlet filter at a constant rate. Inside the nozzle, ^{220}Rn disintegrates and its DDP settles on the inner wall of the nozzle as well as on the outlet filter. A thin-walled plastic tube is used inside the nozzle, which can be compressed to a standard geometric shape (fig. 7) for subsequent measurement on a gamma spectrometer.

This method is implemented in an Italian toron calibration facility developed at ENEA (BAS-ION Instituto di Radioprotezione) (fig. 8).

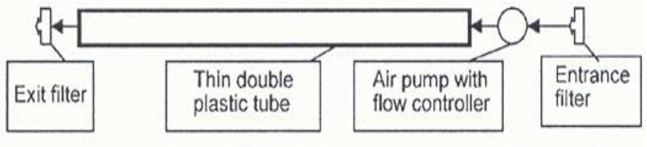

Fig. 6. Schematic representation of the "double filter" method.

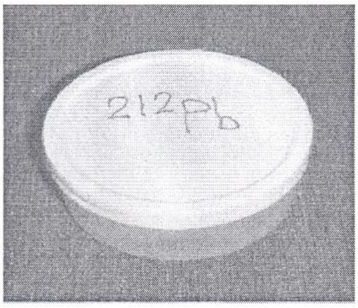

Fig. 7. Standard vial for gamma measurements.

Fig. 8. Aluminum cassette implementing the "double filter" method.

III. RESULTS AND DISCUSSION

Thus, the analysis performed allows us to conclude that the existing radon or thoron chambers consist of a complex of systems and modules. Based on the review of sources [3, 7–18], it is possible to determine the main systems and modules of the structural diagram:

1. Chamber-room for creating an air mixture "radon atmosphere", "thoron atmosphere" with a sealed volume from 0.1 m^3 to 78 m^3. The size and equipment of the cell rooms varies depending on the purpose of use. Chambers with a volume of up to 3 m3 are relatively small and are used to calibrate and transfer a unit of volumetric activity to measuring instruments that measure only ^{222}Rn or ^{220}Rn without taking into account DDP. Chambers-rooms oriented to work with the equipment measuring the DDP have a larger volume and, as a rule, have a system of aerosol generators with a system for monitoring the concentration of aerosols [4].

2. Generators ^{222}Rn or generators ^{220}Rn exist in three types: liquid, solid and gaseous.

Liquid generators consist of an acidic solution of ^{226}Ra or ^{228}Th salts to produce ^{222}Rn or ^{220}Rn, respectively. The salt solution is placed in a glass vessel, in which equilibrium is reached between ^{222}Rn, ^{220}Rn and their DDP during ten half-lives. Then, the accumulated ^{222}Rn or ^{220}Rn is pumped into the chamber-room.

Solid-state sources consist of ^{226}Ra or ^{228}Th dry salts. The salt can be mixed with the carrier and placed on the backing of the container where ^{222}Rn or ^{220}Rn accumulates.

Vessels with condensed ^{222}Rn are used as gaseous radon generators [6].

3. A system for measuring and monitoring thermodynamic parameters (temperature, humidity) of the air environment is necessary to simulate the natural operating conditions of devices, as well as to take into account the influence of environmental parameters on the formation of DPR.

4. The system for ensuring the uniformity of the volumetric activity of ^{222}Rn or ^{220}Rn in the entire volume consists of a complex of fans located in the volume of the chamber-room or a distributed system of ^{220}Rn generators.

5. The control system of the ^{222}Rn or ^{220}Rn generators provides opening/closing of the generators during work.

6. The system for determining the volumetric activity of ^{222}Rn or ^{220}Rn consists of transfer standards and/or α-, β-spectrometers that determine the presence of either ^{222}Rn/ ^{220}Rn or their DPR.

7. Control system of the external environment.

8. Control system.

IV. SUMMARY

Having got acquainted in detail with the execution of the state standard of the unit of volumetric activity 222Rn, it can be stated with confidence that such conditions can be realized on its basis, i.e. By improving the existing one, reproduction, storage and transmission of not only the volumetric activity unit ^{222}Rn, but also the reproduction, storage and transmission of the volumetric activity unit ^{220}Rn can be provided.

To do this, you only need to perform the following modifications on it:

- equip the reference measuring chamber (RMC) with a system of electromechanical opening / closing of the cover for placement inside the RMC equipment;
- to place temperature and pressure sensors inside RMC;
- to equip RMC with mounts to accommodate equipment;
- to equip the internal volume of the chamber with a system of video cameras for reading readings from equipment during research;
- to replace obsolete valves on the vacuum system and on the ^{222}Rn generators;
- to develop an electronic system and an algorithm for programmed control of the state primary standard of the unit of volumetric activity ^{220}Rn;
- to create the thoron chamber as a separate complex of systems and modules for reproducing, storing and transferring the unit of volumetric activity ^{220}Rn into the state primary standard of the unit of volumetric activity ^{220}Rn and evaluate uncertainty of radon and thoron volumetric activity according to [20].

Acknowledgement

The authors express his sincere gratitude to colleagues from Scientific Centre of Quantum measurements of National Scientific Centre Institute of Metrology for discussions and advice when structuring and writing this paper.

REFERENCES

[1] Resolution of the Cabinet of Ministers of Ukraine of December 28, 2016 №1041. About the statement of the State program of development of the reference base for 2018-2022.

[2] V. Skliarov et al. 2009 Metrologia 46 http://kcdb.bipm.org/ 10.1088/0026-1394/46/1A/06006.

[3] A. Honig, A. Paul, S. Röttger, U. Keyser “Environmental control of the German radon reference chamber,” Nuclear Instruments and Methods in Physics Research, 1998, vol. 416, pp. 252–530.

[4] A. Paul, U. Keyser “A reference aerosol for a radon reference chamber,” Nuclear Instruments and Methods in Physics Research, 1996, vol. 368, pp. 819–824.

[5] R. Collé, J.M.R. Hutchinson, M.P. Unterweger “The NIST Primary Radon-222 Measurement System,” J. Res. Natl Inst. Stand. Technol, 1990, vol. 95, pp. 155–165.

[6] R. Dersch, U. Schotzig “Production and measurement of Rn-222 standard,” Appl. Radiat. Isot., 1998, vol. 49, No 9, pp. 1171–1174.

[7] S.A. Soroka at al. “Principles of construction of reference radon chambers,” ANRI, 2006, No 1 (44), pp. 34–37.

[8] J.M. Fisenne, A.C. George “A calibration and quality assurance program for environmental radon measurement,” J. Res. Natl. Inst. Stand. Technol, 1995, pp. 127–134.

[9] J.M. Fisenne, A.C. George “Radon measurement intercomparisons,” Health Physics, 1983, vol. 45(2), pp. 553–556.

[10] R.F. Droullard, T.H. Davis, E.E. Smith, R.F. Hoiub “Radiation hazard test facilities at the Denver Research Centre,” Information Circular 8965, US Department of the interior, bureau of mines, Denver, CO. PB-84-190248 (1984).

[11] K.D. Cliff “U.K. National Radiological Protection Board Radon Calibration Procedures,” J. Res. Natl. Inst. Stand. Technol, 1990, vol. 95, pp. 135–138.

[12] S.B. Solomon, W. Gan, J.B. Peggie “OECD/NEA Radon intercalibration and intercomparison programme,” Pacific region intercomparison programme Commonwealth Department of Health, Australian Radiation Laboratory, Yallambie, Victoria, ARL/TR068, ISSN 0157-1400 (April 1985).

[13] J.L. Picolo, D. Pressyanov, P. Blanchis, M. Barbier “A radon-222 traceability chain from primary standard to field detectors”, Applied Radiation and Isotopes, No 52(2000), pp. 427–434.

[14] J. Biggy “The National Radon/Thoron Test Facility,” Proceeding of seminar on Radon Risks, Ottawa, Canada, 29 Oct. 1992, pp. 61–83.

[15] D. Azimi-Garakani “A comparison of different radon chamber,” Nuclear Instruments and Methods in Physics Research, Section B, vol. 71, Issue 1, pp. 99–102.

[16] “Measurement of radon and radon daughters in air”, NCRP report No 97.

[17] IEC 61577-1 “Radiation protection instrumentation – Radon and radon decay product measuring instruments. Part 1General requirements,” IEC, 2000.

[18] A. Röttger, A. Honig and D. Arnold “The German thoron progeny chamber – Concept and application,” Applied Radiation and Isotopes, Available online 26 January 2009.

[19] R. Falk, G. Akerblom, L. Nyblom “A method for the determination of thoron and thoron progeny concentration at workplace and thoron concentration in calibration chambers,” Swedish Radiation Protection Authority (SSI), Radiation Protection Dosimetry, 2008, pp. 1–5.

[20] Zakharov I.P., Vodotyka S.V., Shevchenko E.N Methods, models, and budgets for estimation of measurement uncertainty during calibration // Measurement Techniques, 2011, Volume: 54 Issue: 4, pp. 387-399, DOI: 10.1007/s11018-011-9737-5.