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Radon in water standard samples for intercomparison experiments

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ABSTRACT

Radon (Rn-222) is the heaviest noble radioactive gas which is created during the alpha decay of radium (Ra-226), the natural element present in the environment, mainly in rock and soil. Due to radon solubility, it may be found in drinking water, thus being an additional hazardous factor for the population. That is the major reason why it is important to be able to determine its concentration in water with reasonable accuracy and well known uncertainty.

The aim of this paper is to thoroughly investigate the procedure of preparation of radon-in-water standards, which can be used in intercomparison measurements. The idea of the method is pumping radon gas of precisely determined activity through the well known water volume. The work focuses in particular on studying the influence of some factors on the procedure correctness, such as duration of relaxation time, the time of pumping radon from the source to chamber, the level of samples collection in water volume.

The setup allowed acquisition of 10 ml water samples, radon concentration values obtained in them ranged from approximately 0.5 Bq/l to 12 Bq/l, with uncertainties of about 17% to 4–5%, respectively.

The measured radon concentrations in water samples were generally lower than the calculated ones (systematic difference of about 13%), but the relation between these two quantities was linear in the whole range of measured values. It was also observed that there was no difference in results between the samples collected at different levels in the water volume. Furthermore, the optimal relaxation time was established. Therefore, the proposed method can be successfully used in radon-in-water intercomparison experiments.

1. Introduction

Radon (Rn-222) is the heaviest noble radioactive gas which decays through alpha particle. It is created during the radioactive alpha decay of radium (Ra-226), the element present in rock and soil, thus radon is one of the main sources of environmental natural radioactivity and the second leading cause of lung cancer (WHO, 2009). Radon, due to its solubility, is present in groundwaters, springs, and hence in drinking waters. This may be the additional hazardous factor for people which was reflected in European regulations (EU Directive 2013/51/EURATOM).

The measurements of radon concentration in different types of water have been carried out not only from radiation protection point of view (Przylibski et al., 2001; Zalewski et al., 2001; Bem et al., 2004; Kochowska et al., 2004; Todorovic et al., 2011, 2012) but they also are very useful for scientific investigations (Nevinsky et al., 2015; Csondor et al., 2016).

Radon concentration in water can be measured using different techniques, from which liquid scintillation counting seems to be the most common. All methods, however, need to be intercompared to ensure the quality and reliability of results. The idea of intercomparison experiment is to measure the water sample and then to compare the obtained result with the reference value of radon concentration in the sample. Sometimes this value is not known, e.g. if the sample comes from natural water springs (Przylibski et al., 2012). In that case the reference value is assumed to be an average value of all obtained results. In laboratory conditions there is a possibility to prepare water samples which can be considered as radon-in-water standard, free of radium and with a calculated radon concentration. There are some solutions for radon in water standard preparation. At the Czech Metrology Institute the standard has been prepared with the use of Ra-226 source in the form of thin foil mounted in the glass container which is filled with water. With a high emanation power radium decays to radon that is dissolved in water. (Havelka, 2009). The similar idea has been applied at some other institutions (Hutchinson et al., 1984; Dean and Kolkowski, 2004; Kitto et al., 2010). Another way of preparation of radon in water standard was presented by Karangelos et al. (2005). The radon gas emanated from a certified Ra-226 source was introduced to a big chamber. Then it was pumped from the chamber to a container with a known amount of water. All parts of the system were connected in a

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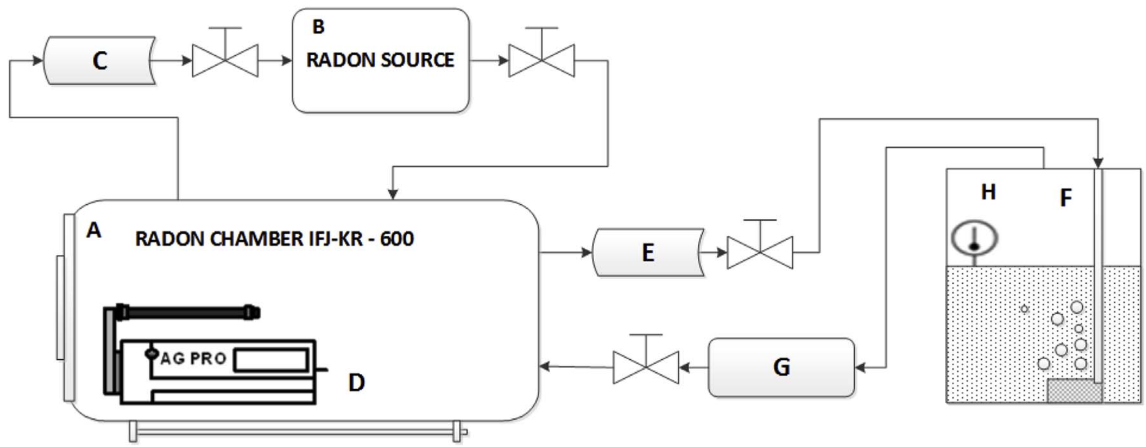

Fig. 1. Setup for preparation of water samples of known radon concentration.

closed circuit. An active monitor was used to register radon concentration in air in a circuit. The similar procedure for preparation of water samples with known radon concentration has been developed at the Laboratory of Radiometric Expertise, Institute of Nuclear Physics Polish Academy of Sciences, Kraków, Poland. Radon concentration in water is calculated theoretically and checked by measurements.

The main goal of this work was to study the influence of some factors on the correctness of preparation of water samples with the calculated radon concentration. These factors are: relaxation time (time when water stays undisturbed after pumping radon gas before taking a sample for measurement), the level of water sampling and the time of pumping radon gas through the system. The optimized procedure for preparation of water samples that can be used in intercomparison measurements or as secondary standards for routine checking of the measurement set-up has been proposed.

2. Materials and methods

The idea of the method and the setup used is presented in Fig. 1.

A particular amount of radon gas is collected in the certified radon source B (Pylon RN-1025). This amount depends on the required radon concentration in water sample. In the first step radon gas is transferred from the source B to the radon chamber IFJ-KR-600 (A) in order to obtain a known radon concentration inside the chamber. The chamber is filled with radon using peristaltic pump MasterFlex (C). The AlphaGUARD PQ2000 PRO radon monitor (D) is placed in the chamber to measure radon concentration inside it. The method of radon in air measurement using AlphaGUARD monitor is accredited by Polish Centre of Accreditation (certificate No. AB 788 issued in 2007, valid to 2019). Polish Centre of Accreditation is a member of the International Laboratory Accreditation Cooperation (ILAC Mutual Recognition Arrangement). During many audits the traceability of the equipment was confirmed. The next step is to pump radon gas through the entire system using the gas-tight piston pump (E). During that radon is dissolved in the known volume of water in the vessel F. It is important to use distilled water in which radon content is almost zero. The vessel F is made of PMMA, tightly sealed, so that radon cannot escape from it. The air inlet is located at the bottom of the vessel. The outlet is above the water level. For security purpose the vessel is filled with water to about three-quarters of its height, so that water does not enter the chamber through the tubes. To be absolutely sure that no water droplets get into the chamber, the safety vessel (G) between the main vessel and the chamber has been mounted. A thermometer (H) is placed inside the vessel F. The measurement of water temperature is necessary to accurately determine Ostwald coefficient. The air flow direction is shown in Fig. 1. The most important technical details of the setup (A-H) are given below:

Radon Calibration Chamber IFJ-KR-600 **(A)**	- length: 2.20 m; diameter: 0,60 m
	- steel sheet: 3.0 mm thick
	- volume: 608.0 ± 2.4 dm^3
	- control board with valves Swagelock type and Tygon tube
	- control of humidity, temperature, inside fan
Radon source RN-1025 (Pylon) **(B)**	- active material: Radium-226
	- form: dry powder
	- half-life: 1600 ± 7 y
	- source activity: 52.3 kBq (± 4%)
	- gas phase: Rn-222
	- Rn-222 production: 6580 Bq/min
	- max. flow rate: 10 dm^3/min
Pump MasterFlex®, model 7591-07 **(C)**	- speed regulation: ± 3%
	- max speed: 650 rpm
	- flow rate: 1 dm^3/min ÷ 10 dm^3/min
Alpha GUARD PQ2000 PRO **(D)**	- type of radon detector: ionization chamber (HV = 750 VDC)
	- operation mode: 3D alpha spectroscopy and current mode
	- total/active detector volume: 0.62 dm^3/0.56 dm^3
	- measurement range: 2 ÷ 2000000 Bq/m^3
	- detector filling mechanisms: passive diffusion (10/60 min cycle)
	- flow mode (1/10 min cycle)
	- calibration error: 3% (plus uncertainty of used reference)
	- sensors: air temperature: 15 °C ÷ + 60 °C; air pressure: 800 mbar ÷ 1050 mbar
	air humidity: 0% ÷ 99%
	- system linearity error: < 3% within total range
	- type of Rn-progeny filter: glass fibre (retention factor > 99.9%)
Piston Pump **(E)**	- speed regulation using voltage unit
	- flow rate: 9 dm^3/min
	- gas-tight
Vessel **(F)**	- length: 2.20 m; diameter: 0,60 m
	- volume: 5.6 dm^3
	- material: PMMA, tightly sealed
Security vessel **(G)**	- volume: ca. 0.12 dm^3
Thermometer **(H)**	- range 0 ÷ 50 °C; accuracy: 0.5 °C