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APPLICATION OF A SCINTILLATION GAMMA-SPECTROMETER FOR DETERMINATION OF RADON CONTENT IN WATER

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Experimental studies have been carried out to determine the sensitivity of a mobile scintillation gamma-spectrometer to radon-222 in mineral water samples for the selected measurement geometry and the minimum measurable activity of radon-222 in such samples. The measurement results of radon content in mineral water samples obtained using such gamma-spectrometer are presented too.

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INTRODUCTION

Natural concentrations of uranium and, consequently, radium-226 in soils and rocks are sources of radon-222. Radon can also be present in water as a result of its transfer from soils and rocks. In many countries, drinking water comes from underground sources such as springs, wells, and artesian wells. As a rule, the concentration of radon in water that comes from such sources is higher than in water from surface sources, like water reservoirs, lakes, rivers. Groundwater, the main therapeutic factor of which is radon, is very popular all over the world [1].

Radon is a radioactive monoatomic inert gas, the heaviest of the noble gases: its density at 0°C is 9.81 kg/m^3, which is almost 7.6 times more than the density of air. The solubility of radon in water is 460 ml/l. All radon isotopes are radioactive and have short half-lives: the half-life of ^{222}Rn is 3.82 days, ^{220}Rn (thoron) is 55.6 s, and ^{219}Rn is 3.96 s. Due to very short half-lives of radon isotopes ^{219}Rn and ^{220}Rn, the mineral water supplied for the procedures in sanatoriums contains only the 222nd isotope of radon – ^{222}Rn [2].

Radon baths can be prepared both artificially from radium-226, which is available in the form of dissolved salts, or by using natural mineral radon water extracted from drilled wells. The radioactive radon gas is not detected by standard methods. If suspicions of radon presence are not unfounded, measurements can only be carried out using special equipment [3].

The purpose of the work was to determine the sensitivity of a scintillation gamma-spectrometer MKS-AT6101C manufactured by SPE "ATOMTEX", Minsk [4] to Rn-222 radionuclide for the selected measurement geometry, to assess the need for usage of shielding in order to reduce background radiation when measuring specific activity of this radionuclide in water samples, as well as to determine the lowest threshold of the minimum measurable activity. Determination of the sensitivity parameter will form the basis for activity calibration of this gamma-spectrometer for measuring radon-222 content in water.

1. MATERIALS AND METHODS

As a measuring container for water samples, a glass jar of 0.5 l volume with a metal lid, which is closed using a seaming machine, was used. The choice of optimal containers and methods of their sealing is described in [1]. Collected water samples with various radon-222 content were placed in these measuring containers. As a gamma-ray detector, a scintillation detector with a NaI(Tl)-crystal, having 63×63 mm in size, which is part of a MKS-AT6101C gamma-spectrometer, was used. The processing of gamma-spectra was carried out using applied software "ATAS" (SPE "ATOMTEX") [5]. In order to reduce the external background noise, lead shielding was additionally manufactured, and a special tripod was used to fix the detector in a vertical position. During measurements, lead shieldings with different thicknesses (50 and 15 mm) were used. Water samples were measured vertically with a metal lid facing down directly on the end surface of the detector.

Measurement of radon-222 in water samples was carried out using its daughter decay product – Bi-214, which is in equilibrium with the parent radionuclide, with the registration of its photopeak with gamma-ray energy of 609.3 keV and a quantum yield of 0.461 photons/decay.

The sensitivity S, (imp•kg)/(s•Bq) of the MKS-AT6101C gamma-spectrometer to water samples containing radon-222 was calculated according to [6] using the formula (1):

$$S = \frac{N - N_{ф}}{УА}, \qquad (1)$$

where N – the integral count rate in the energy range from 510 to 720 keV when measuring a water sample containing radon-222, counts/s; $N_ф$ – the integral count rate in the energy range from 510 to 720 keV when measuring a background water sample, counts/s; $УА$ – the specific activity of radon-222 in a water sample at the time of measurement, Bq/kg.

Calculation of the specific activity of radon-222 in a sample was carried out for a MKS-AT6101C gamma-spectrometer according to the formula:

$$УА\left(Rn^{222}\right) = \frac{n_{sample} \cdot \left(\frac{A}{n}\right)_{st} \cdot \left(\frac{Y_{Rn-222}}{Y_{Cs-137}}\right)}{m_{sample}} \cdot e^{\lambda t}, \qquad (2)$$

where n_{sample} – the count rate in the photopeak at energy of 609.3 keV for a given sample, counts/s; m_{sample} – the weight of a given sample, kg; $\left(\frac{A}{n}\right)_{st}$ – the coefficient obtained using a reference radioactive solution containing Cs-137 radionuclide; λ – the decay constant of radon-222, equal to 0.181 days-1; t – the sample aging time, days; $e^{\lambda t}$ – the correction coefficient due to the decay of radon-222; $\left(\frac{Y_{Rn-222}}{Y_{Cs-137}}\right)$ – the ratio of gamma-ray yields for energies 609.3 and 661.6 keV (46.1 and 85.1 %, respectively).

Fig. 1,a,b show gamma-spectra for mineral radon water samples measured using a gamma-spectrometer based on a semiconductor Ge(Li)-detector DGDK-80B and a gamma-spectrometer MKS-AT6101C, respectively.

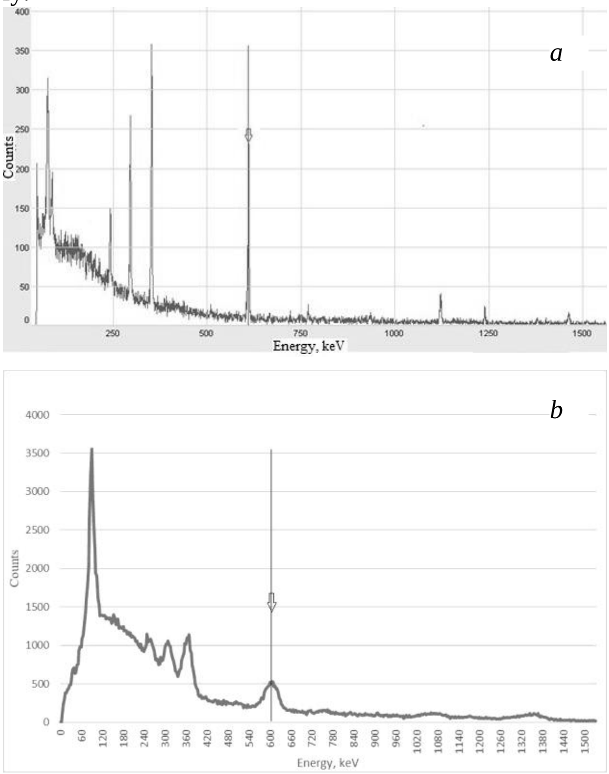

Fig. 1. Gamma-spectrum of a sample of mineral radon water measured using a gamma-spectrometer based on a semiconductor Ge(Li)-detector DGDK-80B. The concentration of Rn-222 in the sample is 1.7 kBq/kg, measurement time – 1200 s (red arrow points on the peak with the energy of 609.3 keV) (a); gamma-spectrum of a sample of mineral radon water measured using a MKS-AT6101C/gamma-spectrometer with NaI(Tl)-scintillation detector. The concentration of Rn-222 in the sample is 1.7 kBq/kg, the measurement time is 1200 s (red arrow points on the peak with the energy of 609.3 keV) (b)

The mean value of the sensitivity of a MKS-AT6101C gamma-spectrometer to water samples containing radon-222 for the selected geometry (0.5 l glass jar), determined by the formula (1), was ((0.0047± 10)%) counts·kg/s·Bq.

Minimum measurable activity (MMA) is the smallest activity of a radionuclide in a measured sample, which with the help of a given measuring installation, using given method of analysis of instrumental radiation energy spectrum can be determined for a defined time so that the uncertainty of type A due to the count rate in the photopeak for the line with the energy of the region of interest doesn't exceed a specified value. Depending on the conditions and parameters of the measurement, the MMA is essentially the lower dynamic level of the activity measurement range, for which the requirements for the permissible uncertainty of type A must be met.

The MMA value for a given radionuclide under constant background conditions with expanded measurement uncertainty (k = 2) was calculated according to [7] using the formula (3):

$$A_{\min}(t_0) = \frac{200\sqrt{N'_{\Sigma}}}{\xi(E_{\gamma i}) \cdot t_0 \cdot I_{\gamma i}}, \qquad (3)$$

where N'_{Σ} – the number of counts registred in the photopeak, counts/s; $\xi(E_{\gamma i})$ – the value of the efficiency of gamma-rays registration in the photopeak of the given energy $E_{\gamma i}$ for a given radionuclide and for the selected measurement geometry, Bq·s^{-1}; $I_{\gamma i}$ – the yield of gamma-rays ith energy $E_{\gamma i}$ for a given radionuclide, %; t_0 – the measurement time, s.

In order to determine the MMA value distilled water, samples in 0.5 l glass jar were measured using a MKS-AT6101C gamma-spectrometer during 600…7200 s in 3 measurement geometry options: 1st – with 50 mm thick lead shielding, 2nd – with 15 mm thick lead shielding, 3rd – without lead shielding. The measuring scheme for the water samples is shown in Fig. 2, and the MMA values are presented in Table 1.

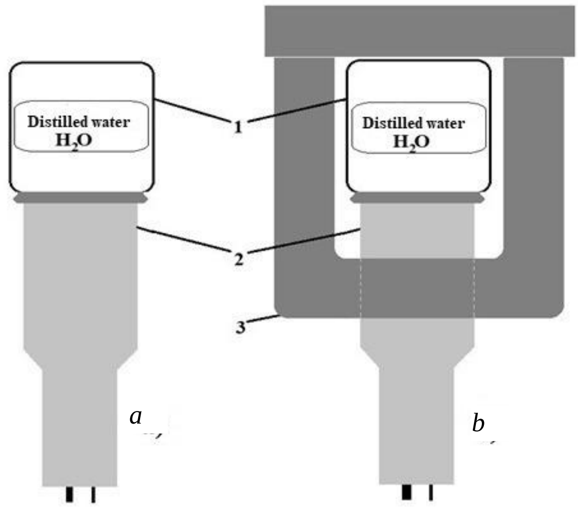

Fig. 2. Measuring scheme for water samples for the determination of the MMV value: a – without lead shielding and b – with lead shielding (50 and 15 mm of thick): 1 – distilled water in a glass jar with the volume of 0.5 l; 2 – detector of the scintillation gamma-spectrometer MKS-AT6101C; 3 – lead

From Table 1 one can draw a conclusion that the usage of lead shielding leads to the decrease of MMA values of radon-222 for the MKS-AT6101C gamma-spectrometer in water samples for the selected geometry (0.5 l glass jar) by 1.8…2.0 times when the thickness of the field shielding is equal to 50 mm and by 1.4…1.5 when it is equal to 15 mm.

Table 1

MMA values of radon-222 for the MKS-AT6101C gamma-spectrometer in water samples for the selected geometry (0.5 l glass jar)

Measurement geometry	MMA, Bq						
	Measurement time, s						
	600	900	1200	1800	2700	3600	7200
with lead shielding (50 mm)	290	190	140	90	60	50	25
with lead shielding (15 mm)	370	250	190	120	80	60	30
without lead shielding	530	350	260	180	120	90	50

2. RESULTS AND DISCUSSION

Radon water samples, when placed in every investigated container, were measured repeatedly at the specific intervals of time in the term of 19 days, and the specific activity of radon-222 at the time of measurement was determined. Logarithmic dependence of the measured activity to the time, elapsed since sampling, as well as linear approximation of this dependence is shown in Fig. 3.

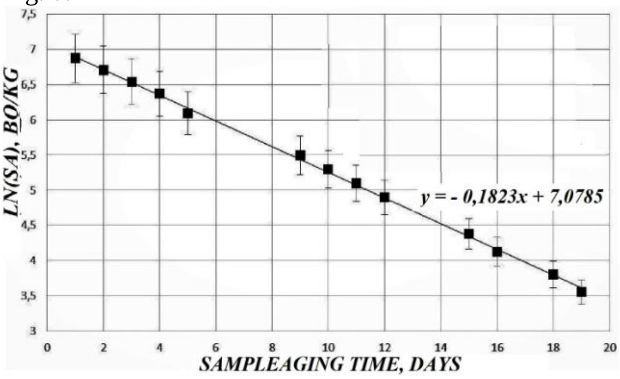

Fig. 3. Logarithmic dependence of measured specific activity of radon-222 of the sample on sample aging time, as well as its linear approximation

As it can be seen from Fig. 3, the data of the specific activity of radon-222 in the measured water sample is well approximated by a straight line, and the slope of this straight line (the first factor in the approximation equation) is close to the constant decay of radon-222 (0.181 day-1), which indicates the absence of radon leakage from a 0.5 l glass jar with a metal lid

In order to confirm the correctness of the results obtained for the determination of radon-222 concentration in mineral radon water samples using a MKS-AT6101C gamma-spectrometer interlaboratory comparisons were performed. Measurements of 2 samples of mineral radon water for the selected geometry (0.5 l glass jar) were carried out in 5 various laboratories of 4 organizations in Belarus.

The results of the measurements of the samples in the laboratories of the following organizations in Belarus are presented in Table 2:

1. Educational institution “International Sakharov Environmental Institute of Belarusian State University”, Laboratory of nuclear spectrometry of the Chair of Nuclear and Radiation Safety (Lab. № 1); type of measuring instrument (MI): a gamma-spectrometer based on a coaxial semiconductor detector GCD-100210 “BSI” and a digital multichannel analyzer (MCA) “HEXAGON”.
2. Scientific institution “JIPNR-Sosny”, Laboratory of experimental nuclear physics research and expert analyses of radioactive materials (Lab. № 2a); MI: a gamma-spectrometer based on a coaxial semiconductor Ge(Li)-detector DGDK-80B.
3. Scientific institution “JIPNR-Sosny”, Laboratory of experimental nuclear physics Research and expert analyses of radioactive materials (Lab. № 2b); MI: a scintillation gamma spectrometer MKS-AT6101C.
4. Scientific institution “JIPNR-Sosny”, Radiation safety department. (Lab. № 3); MI: a gamma spectrometer based on a coaxial semiconductor detector GEM-30185 “ORTEC” and MCA “Davidson” 2056-4k, № 27-P49LA.
5. Joint Belarusian-Russian CJSC “Isotope Technologies” (Lab. № 4); MI: a gamma spectrometer based on a coaxial semiconductor detector GMX40P4-76 “ORTEC”.
6. State institution “Republican center for hydrometeorology, control of radioactive contamination and environmental monitoring” (Lab № 5): MI: a gamma-spectrometer based on a coaxial semiconductor detector GEM-S8530 “ORTEC”.

Table 2

Measurement results of water samples containing radon-222 in various laboratories of Belarusian organizations

Sample number	Specific activity of radon-222, Bq/kg						
	Lab. № 1	Lab. № 2a	Lab. № 2b	Lab. № 3	Lab. № 4	Lab. № 5	Mean value
7	770±230	705±140	715±140	790±160	730±150	720±140	738
24	1040±305	870±170	840±170	890±180	870±170	850±170	893

As a result of the carried out research, experimental data was obtained, and the value of the sensitivity of a mobile scintillation gamma-spectrometer MKS-AT6101C to radon-222 in mineral water samples for the selected measurement geometry was determined. Values of the minimum measurable activity of radon-222 in such samples were determined depending on the measurement time. The correctness of the results of radon-222 concentration determination in mineral radon water samples obtained using this gamma spectrometer was confirmed during the interlaboratory comparison.

There are several sanatoriums in Belarus where either natural mineral radon water extracted from wells or artificially prepared radon water is used for therapeutic purposes. The laboratory of experimental nuclear physics research and expert analyses of radioactive materials of the scientific institution "JIPNR-Sosny" is accredited to carry out analyzes for the determination of radon-222 concentration, including in water, and since 2010, such analyzes have been regularly carried out using a gamma spectrometer based on a coaxial semiconductor detector [1, 2]. All collected water samples (there may be about hundreds of them) must be delivered to the scientific institution "JIPNR-Sosny" where they are measured at laboratory conditions.

The measurement technique for the determination of radon-222 concentration in water has been developed using a mobile gamma-spectrometer MKS-AT6101C. It makes it possible to determine the specific activity of radon-222 in water samples in the range of 50…10000 Bq/kg with expanded uncertainty ($k = 2$) not exceeding 20%.

The use of a mobile scintillation gamma spectrometer MKS-AT6101C for the determination of radon-222 concentration in water samples will allow to control the process of preparation of radon baths from radon waters extracted both from wells, and, especially, from radon waters obtained directly at sanatoriums' locations by using radon generators based on radium-226 salts preparation. It makes it possible to be more precise when determining radiation doses received by patients when taking radon baths, and consequently, it will improve the quality of their treatment.

Mobility of the MKS-AT6101C scintillation gamma-spectrometer makes it possible to measure radon content in drinking water in wells directly at settlements where they are located, without transporting samples to specialized laboratories if such spectrometers are equipped with portable shielding.

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ПРИМЕНЕНИЕ СЦИНТИЛЛЯЦИОННОГО ГАММА-СПЕКТРОМЕТРА ДЛЯ ОПРЕДЕЛЕНИЯ СОДЕРЖАНИЯ РАДОНА В ВОДЕ

Д.А. Хакимов, И.В. Жук, М.К. Киевец

Проведены экспериментальные исследования по определению чувствительности мобильного сцинтилляционного гамма-спектрометра к радону-222 в пробах минеральной воды для выбранной геометрии измерений и минимальной измеряемой активности радона-222 в таких пробах. Приведены результаты измерения содержания радона в пробах минеральной воды, полученные с помощью такого гамма-спектрометра.

ЗАСТОСУВАННЯ СЦИНТИЛЯЦІЙНОГО ГАММА-СПЕКТРОМЕТРА ДЛЯ ВИЗНАЧЕННЯ ЗМІСТУ РАДОНУ У ВОДІ

Д.А. Хакимов, І.В. Жук, М.К. Києвец

Проведено експериментальні дослідження з визначення чутливості мобільного сцинтиляційного гамма-спектрометра до радону-222 у пробах мінеральної води для обраної геометрії вимірювань і мінімальної вимірюваної активності радону-222 у таких пробах. Наведено результати вимірювання змісту радону в пробах мінеральної води, які отримані за допомогою такого гамма-спектрометра.