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SAMPLE PREPARATION FOR THE EFFECTIVE ACCUMULATION AND DETECTION OF THE BETA-ACTIVE Rn-222 DECAY PRODUCTS[†]

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The sample preparation method and the results of experimental measurements of the concentration of beta-radioactive aerosols (the decay products of Radon-222 in the air) are presented. The experimental equipment includes an electrostatic aerosol collector and a time spectrometer based on the PMT with a plastic scintillator. The accumulation of aerosols on the foil lasted for about 12 hours. The activity of accumulated aerosols was measured in the time interval of 0 to 300 minutes. The use of the time analyzer spectrometer, the proposed aerosol accumulator and the method of processing the accumulated spectrum makes it possible to increase the sensitivity of the radiometer in comparison with the collection method based on air filters. Applying the time-spectrum development procedure to the constituent components makes it possible to reliably establish the connection of aerosols registered in the room with β-active decay products of radon-222: Po-218, Pb-214, Bi-214.
Keywords: Radon-222; beta – activity, decay, electrostatic methods, sample preparation
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Radioactive aerosols are a mixture of dust and radioactive particles, which are α-, β- and γ emitters. Decay products of Rn-222, $T_{1/2}$ = 3.823 days; Rn-220, $T_{1/2}$ = 0.9267 min; Rn-219, $T_{1/2}$ = 3.96 s and their daughter products – Po-218, $T_{1/2}$ = 3.167 min; Pb-214, $T_{1/2}$ = 26.833 min; Bi-214, $T_{1/2}$ = 19.833 min are deposited on particles suspended in the air. Radon continuously enters the atmosphere from terrestrial rocks: Rn-222 - during the alpha decay of Ra-226 nuclei (uranium-radium family), and Rn-220 – during the alpha decay of Ra-224 nuclei [1]. The Rn-222 isotope gives approximately 50-55% of the effective radiation dose every Earth inhabitant receives annually. The Rn-220 isotope adds another ~ 5-10% to this [1].

No less dangerous are the decay products of radon - beta-radioactive isotopes of lead, bismuth, and polonium, since they form aerosols that when they enter the human lungs and cause micro burns.

The combination of the developed method of analysis and electrostatic aerosol accumulator and time analysis of the sample activity makes it possible to associate aerosols recorded in a room with β-active decay products of radon-222. It also increases the reliability of recording Rn-222 and significantly reduces the measurement time compared, for example, with a technique based on the use of CR-39 films [2].

The references have known methods for assessing the radioactivity of aerosols - radon decay products by measuring gamma and beta radiation [3-5]. In these works, the registration of radon decay products is carried out, as a rule, using spectrometric amplitude analysis. The use of multilayer filters in the registration of beta aerosols is not effective enough because, simultaneously with the accumulation, the outflow of accumulated particles occurs.

In work [6] was used a two-electrode filter, which provides for the capture of radon beta- products, is considered but does not allow obtained a high sensitivity. The paper [7] considered the approach of direct deposition of decay products on the spherical surface of the Si detector. The potential on the surface is -3 kV. The concentration of aerosols and the sample activity are not high; the data accumulation and processing speed are also low. In [8] work is represented a multilayer filter for collecting aerosols. The air in the room was blown through a filter, allowing radon decay products to accumulate. The method's efficiency is low due to the complexity of sample preparation for analysis and the low efficiency of the blowing way.

SAMPLE PREPARATION AND MEASUREMENT TECHNIQUE

Reliable registration of radon isotopes and decay products [2] for gas mixture is an unfinished problem. Mainly significant difficulties are determined the low adsorption of radon by the filters. For the quantity determined of radon isotopes, it is also necessary to have radioactive gas samples. Their creation is significantly complex, especially for the long-term isotope - radon-222. The decay products, being positively charged, can be subject to electrostatic precipitation. The alpha particle escapes produce the positive charge of the ionization of the atomic shell. It is also possible to have ionization by the kinetic energy of the recoiled atomic nucleus. To accumulate the radioactive sample was used an

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electrostatic method. On a sheet of aluminum foil (~ 0.02 mm) with an area of ~ 1 m^2, which was placed on a horizontally stretched insulating thread, a potential of mines 1500 V was applied. The time exposure was 12 hours. Due to the difference in the potentials of the dust particles in the air and the filter (foil), it was possible to deposit the aerosols efficiently. After the exposure, the foil sheet was reduced to a cylinder, Ø5x0.2cm. The additional reduction of the sample allows us to significantly increase the concentration of radioactive particles per unit area of the scintillation detector. Fig. 1 shows a circuit of a voltage converter used for an electrostatic filter. Fig. 2 shows a detector part of the measurement setup.

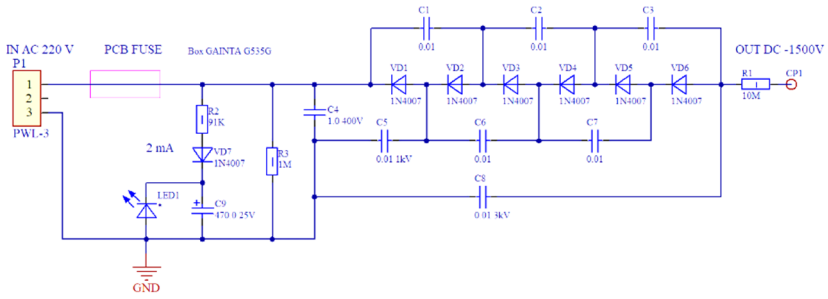

Figure 1. Power supply for an electrostatic filter

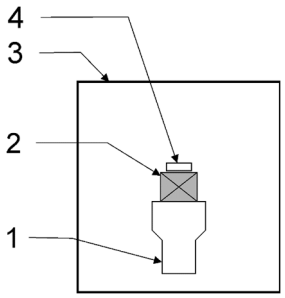

Figure 2. Detector system. 1 – PMT R1307 Hamamatsu, 2 – UPS-923A plastic detector, 3 – lead background shield, 4 – probe sample

RESULTS AND MEASUREMENTS

In this work, it was assumed that the activity of particles in the indoor air aerosol collected on the filter is mainly determined by the decay products of the isotope of radon: Rn-222 ($T_{1/2}$=3.82 days). The contribution of other isotopes – Rn-220 ($T_{1/2}$ = 55.6 s), Rn-219 ($T_{1/2}$ = 3.96 s) was neglected due to the short half-life compared to Rn-222 and lower prevalence. It was assumed that possible isotopes that were in the room and got on the radiometer filter and then disintegrated during the measurement process are isotopes Rn-222, Po-218, Pb-214 and Bi-214:

$$^{222}\text{Rn} \rightarrow {}^{218}\text{Po} \rightarrow {}^{214}\text{Pb} \rightarrow {}^{214}\text{Bi} \rightarrow \ldots;\ {}^{218}\text{Po} \rightarrow {}^{214}\text{Pb} \rightarrow {}^{214}\text{Bi} \rightarrow \ldots;\ {}^{214}\text{Pb} \rightarrow {}^{214}\text{Bi} \rightarrow \ldots;\ {}^{214}\text{Bi} \rightarrow \ldots. \quad (1)$$

The decay of isotopes accumulated in the filter is described by the following system of equations:

$$\frac{dN_1}{dt} = -\lambda_1 N_1$$ – decay of the accumulated products on the filter ^{222}Rn (N_1)

$$\frac{dN_2}{dt} = \lambda_1 N_1 - \lambda_2 N_2$$ – production ^{218}Po (N_2) from decay of ^{222}Rn (N_1) and decay of the accumulated products on the filter (N_2)

$$\frac{dN_3}{dt} = \lambda_2 N_2 - \lambda_3 N_3$$ – production of the ^{214}Pb (N_3) from decays of ^{218}Po (N_2), ^{222}Rn (N_1) and decay of the accumulated products on the filter ^{214}Pb (N3)

$$\frac{dN_4}{dt} = \lambda_3 N_3 - \lambda_4 N_4$$ – production ^{214}Bi (N_4) from decays of the ^{214}Pb (N_3), ^{218}Po (N_2), ^{222}Rn (N_1) and decay of the accumulated products on the filter ^{214}Bi (N_4) (2)

where N_1, N_2, N_3, N_4 – quantity of the radioactive nuclei ^{222}Rn, ^{218}Po, ^{214}Pb, ^{214}Bi respectively.

The activity of the source A can be related to the number of radioactive nuclei N and the detector efficiency ε as follows: (-dN/dt) = A; (-dN/dt) · ε = λ·N·ε = A·ε. In the calculation was assumed that ε = 1.

The change over time in the total activity of the isotopes accumulated on the filter could be described by the system of the following equations:

– decay of the accumulated products on the filter ^{222}Rn ($N_1(0)$):

$$N_1(t) = N_1(0)e^{-\lambda_1 t}, \quad (3)$$

– production ^{218}Po from the decay ^{222}Rn, and decay of the accumulated products on the filter ^{218}Po ($N_2(0)$):

$$N_2(t) = \frac{\lambda_1 N_1(0)}{(\lambda_2 - \lambda_1)}\left[e^{-\lambda_1 t} - e^{-\lambda_2 t}\right] + N_2(0)e^{-\lambda_2 t}, \quad (4)$$

– production ^{214}Pb from the decay ^{218}Po, ^{222}Rn, and decay of the accumulated products on the filter ^{214}Pb ($N_3(0)$):

$$N_3(t) = \frac{\lambda_2\lambda_1 N_1(0)}{(\lambda_2 - \lambda_1)}\left[\frac{e^{-\lambda_1 t} - e^{-\lambda_3 t}}{(\lambda_3 - \lambda_1)} - \frac{e^{-\lambda_2 t} - e^{-\lambda_3 t}}{(\lambda_3 - \lambda_2)}\right] + \frac{\lambda_2 N_2(0)}{(\lambda_3 - \lambda_2)}\left(e^{-\lambda_2 t} - e^{-\lambda_3 t}\right) + N_3(0)e^{-\lambda_3 t}, \quad (5)$$

– production ^{214}Bi from the decay ^{214}Pb, ^{218}Po, ^{222}Rn, and also decay of the accumulated products on the filter ^{214}Bi ($N_4(0)$):

$$N_4(t) = \frac{\lambda_3\lambda_2\lambda_1 N_1(0)}{(\lambda_2-\lambda_1)}\left[\frac{1}{(\lambda_3-\lambda_1)}\left(\frac{e^{-\lambda_1 t}-e^{-\lambda_4 t}}{(\lambda_4-\lambda_1)}-\frac{e^{-\lambda_3 t}-e^{-\lambda_4 t}}{(\lambda_4-\lambda_3)}\right)-\frac{1}{(\lambda_3-\lambda_2)}\left(\frac{e^{-\lambda_2 t}-e^{-\lambda_4 t}}{(\lambda_4-\lambda_2)}-\frac{e^{-\lambda_3 t}-e^{-\lambda_4 t}}{(\lambda_4-\lambda_3)}\right)\right]+$$
$$+\frac{\lambda_3\lambda_2 N_2(0)}{(\lambda_3-\lambda_2)}\left[\frac{e^{-\lambda_2 t}-e^{-\lambda_4 t}}{(\lambda_4-\lambda_2)}-\frac{e^{-\lambda_3 t}-e^{-\lambda_4 t}}{(\lambda_4-\lambda_3)}\right]+\frac{\lambda_3 N_3(0)}{(\lambda_4-\lambda_3)}\left(e^{-\lambda_3 t}-e^{-\lambda_4 t}\right)+\quad N_4(0)e^{-\lambda_4 t}. \tag{6}$$

Figure 3. shows the results of the analysis of the activity of the sample (analysis time 300 min.), accumulated on the filter for a time interval of 12 hours using the system of equations (3-6).

The total activity of aerosols was measured in a closed room on the 1st floor of the building. The time of collecting aerosol particles by the electrostatic method was 12 hours. After installing the filter on the scintillation counter (detector - plastic), the change in the total beta activity of the filter was recorded duration 300 minutes, and the time of the accumulation of one channel was 1 minute. The time spectrometer was used from [8].

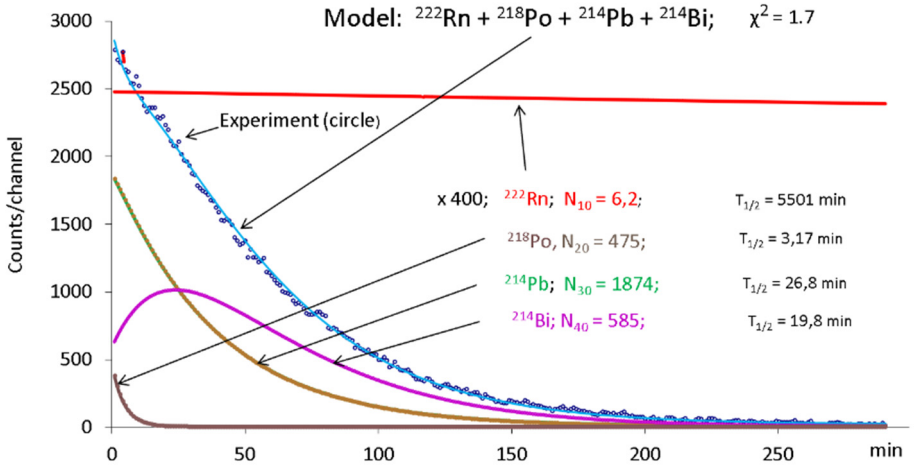

Figure 3. Results of processing the counts (counts/channel) of the decay products of the measured sample. The event pulse recorded by the detector is plotted along the vertical axis. N_{i0} – calculated initial values (activities) for beta-components

The analysis of the decay curve and determination of the initial activities of the components is performed by approximating the experimental data of the theoretical curve, which is the sum of the calculated activities on the aluminum foil ($N_1(t)$, ^{222}Rn) + ($N_2(t)$, ^{218}Po) + ($N_3(t)$, ^{214}Pb) + ($N_4(t)$, ^{214}Bi) duration 300 minutes.

Fit quality criterion is:

$$\chi^2 = \frac{1}{n-5}\sum_1^n\left(\frac{N_{exp}-N_{th}}{\delta i(N_{exp})}\right)^2, \tag{7}$$

where n – the number of experimental points form a decay curve; N_{exp} – registered counts (activities); $\delta_i(N_{exp})$ – absolute error of a single measurement, $\delta_i = \sqrt{N_{i,exp}}$.

During the fitting, the initial activities of N_1, N_2, N_3 and N_4 was varied. As a result, the following values of the initial activities of beta-active components on the filter were obtained: $A_{10} = N_1(0)$ (^{222}Rn) = 6.2; $A_{20} = N_2(0)$ (^{218}Po) = 475; $A_{30} = N_3(0)$ (^{214}Pb) =1874; $A_{40} = N_4(0)$ (^{214}Bi) = 585. The relative error of approximation, estimated based on the root-mean-square error (RMS) of the spectrum by expression (8) was 4.5%, $\chi^2 = 1.7$.

$$\delta_{\bar{x}} = \frac{1}{(\sum_1^n N_{i,exp})/n}\sqrt{\frac{1}{n-1}\sum_1^n\left(N_{i,exp}-N_{i,th}\right)^2}, \tag{8}$$

RESULTS AND DISCUSSION

The results of the measurements and the analysis of the decay curves indicate the presence in the filter (Al-foil) of the alpha-active isotope of radon Rn-222, beta-active products of decay of radon - Pb-214, Po -218 and Bi-214. The ratio of initial activities of Pb-214 / Bi-214 for the studied room was 3.2:1.

The presence of Rn-222 alpha emitters in the filter leads to a noticeable improvement in the agreement between the model and experimental curves. An exception to the analysis of the Rn-222 isotope worsens the quality of the fit, with the value of $\chi^2 = 6.5$, RMS = 6.7%. Thus, the analysis results (χ^2 values, activity values of the components) indicate that the activity of aerosols accumulated in the filter is formed mainly by beta-active products of the decay of the radon isotope Rn-222.

CONCLUSIONS

The results of measurements and processing confirm a significant increase in the sensitivity of the method due to the proposed electrostatic method of the sample preparation and an increase in the reliability of measurements due to the use of time analysis of the decay curve and a scintillation counter.

Application of the proposed technique allows to reduce a sample accumulation time of ~ 12 hours and a processing time of ~ 5 hours to obtain a statistical error of fitting ~ 4.5%.

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ПІДГОТУВАННЯ ПРОБ ДЛЯ ЕФЕКТИВНОГО НАКОПИЧЕННЯ ТА РЕЄСТРАЦІЇ БЕТА-АКТИВНИХ ПРОДУКТІВ РОЗПАДУ Rn-222

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Представлена методика підготування проб та результати вимірювань концентрації бета-радіоактивних аерозолів - продуктів розпаду Радону-222 у повітрі. До складу експериментального тракту входять електростатичний збирач аерозолів та сцинтиляційний часовий спектрометр на базі ФЕП з пластиковим сцинтилятором. Накопичення аерозолів на фользі тривало близько 12 годин. Вимірювання активності накопичених аерозолів проводилося в інтервалі 0-300 хвилин. Використання часового аналізатора, запропонованого накопичувача аерозолів і методу обробки накопиченого спектра дає змогу суттєво підвищити чутливість радіометра у порівнянні з методикою збору на основі повітряних фільтрів. Застосування процедури розвинення часового спектра на складові компоненти дає змогу надійно встановлювати зв'язок зареєстрованих в приміщенні аерозолів з β-активними продуктами розпаду радону-222: Po-218, Pb-214, Bi-214.

Ключові слова: Радон-222; бета-активність, розпад, електростатичний метод, підготування проб