The Effect of Radon Concentration at Liquid Nitrogen Temperature

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Abstract. Experimentally obtained high concentrations of radon in an isolated container of uranium-containing minerals using liquid nitrogen. A simple device is proposed, the essence of which is based on the transformation of gaseous radon into a liquid. It is known that liquid nitrogen has a boiling point (-195.75) ºC, and gaseous radon turns into liquid at (-62) ºC, and at (-70) ºC turns into a solid state. The low temperature of liquid nitrogen greatly exceeds the liquefaction and solidification temperature of radon. It follows that if you put a radioactive outgoing source, for example, pieces of uranium ore, into a heat-insulated container, such as a thermos or a Dewar vessel into which liquid nitrogen is poured, then radon will accumulate at the bottom of the container in a solid state. As liquid nitrogen evaporates, gaseous radon will concentrate inside the container. The technical result is to increase the efficiency of obtaining radon with emanation and the use of the device directly in stationary conditions, for example, in medical institutions. Replacing the solutions of radium salts with pieces of uranium ore from the Elkonskiy deposit from Yakutia significantly simplifies and reduces the cost of preparing radon fluids for medical procedures.

1. Introduction

The main part of the radiation population of the Earth receives from natural sources of radiation. The whole process of evolution took place under the influence of cosmic radiation and radioactive substances of the earth's crust [1].

Different types of ionizing radiation affect organisms in different ways. The nature of the impact largely depends on whether the radionuclide is inside the body (that is, the body is exposed to internal radiation) or it is located outside the body (external radiation) [2].

Radioactive gas radon has a negative effect on the human body, which can later lead to a number of diseases, in particular, to lung cancer. But also a certain amount of it can cause a positive effect on the human body, thanks to α-radiation, which occurs during the decay of radon. Therefore, it is used in medical practice for the preparation of radon baths, which are based on the use of radioactive water or air enriched with radon-222. The specificity of the therapeutic effect of radon waters is ionizing...
radiation, accompanied by the decay of radon and its daughter products [3]. This treatment is prescribed to patients if the resulting beneficial effect will significantly exceed the damage from the effects of α-radiation arising from the decay of radon atoms [4]. When different types of radon procedures are dispensed, the doses to patients are determined by the duration of the procedures, the characteristics of the radiation source and the conditions of the patients' exposure [5].

Radon waters (according to Russian balneological waters) include mineral waters, the radon content in which must be at least 185 Bq/l (MAC of radon for drinking water - 120 Bq/l). This value is rather conditional, since the required intensity of irradiation can be regulated by time parameters. So, in Poland the minimum treatment rate is 375 Bq/l, in France - 370 Bq/l, in the Czech Republic - 1192 Bq/l, in Germany - 6885 Bq/l, in Hungary - 3 Bq/l.

In Russia, the most famous sources of radon waters in the Urals are in the Sverdlovsk and Chelyabinsk regions, the Republic of Bashkiria, the Caucasus, Siberia and the Far East [6].

According to the Radiation Safety Standards NRB-99/2009: the critical way of exposing people to radon-222 contained in drinking water is the transfer of radon into the room air and the subsequent inhalation intake of radon daughter products into the body. The intervention level for $^{222}\text{Rn}$ in drinking water is 60 Bq/kg. For mineral and medicinal waters, special standards are established [7].

2. Experimental part

This paper presents a method for obtaining high radon concentrations using liquid nitrogen. Liquid nitrogen has a temperature of -195.75 °C, and gaseous radon turns into liquid at -62 °C and at -70 °C turns into a solid state. Hence, the temperature of liquid nitrogen significantly exceeds the liquefaction and solidification temperature of radon. Therefore, if you place a radioactive emanating source, for example, pieces of uranium ore in the form of pieces of metasomate with veinlets of brannerite, granite and marble fractions in a thermally insulated container into which liquid nitrogen is poured, then radon will accumulate at the bottom of the container in a solid state. As the liquid nitrogen evaporates, gaseous radon will be concentrated inside the container.

Radon concentrations were measured with an Alpha Guard radon radiometer (manufactured in Germany). Alpha GUARD is a high-capacity, portable, battery-powered or networked radon monitor with a measurement range of 2 MBq/m$^3$. In standard operation, the gas to be measured is diffused through a large area glass fiber filter into the ionization chamber. That is, only gaseous radon-222 can pass through a glass fiber filter. In addition to the diffusion mode, Alpha GUARD devices also have an active mode of operation, in which gas is pumped into the ionization chamber using a pump [2] [8]. In standard operation, the gas to be measured is diffused through a large area glass fiber filter into the ionization chamber. That is, only gaseous radon-222 can pass through a glass fiber filter. In addition to the diffusion mode, Alpha GUARD devices also have an active mode of operation, in which gas is pumped into the ionization chamber using a pump [8] [9].

Equivalent equilibrium volume activity (EEVA) of radon for a nonequilibrium mixture of short-time affiliated products of decay in the air is called a volume activity of radon. It is in balance with affiliated products of decay which has the same value of latent energy, as well as the given nonequilibrium mixture. It is calculated by the following formula [10]:

$$EEV_{\text{Rn}} = VA_{\text{Rn}} \times F, \text{ Bq/m}^3$$

where $VA_{\text{Rn}}$ is a volume activity of radon, $F$ is a balance factor between radon and products of its decay which can accept values from 0 to 1. In the absence of experimental data about a mean value of this factor, it is accepted as $F = 0.5$ [11]. The given value has been used in this work.

The experiment with liquid nitrogen was carried out in several stages: measurements inside the thermos for several days, measurements inside the thermos at different levels, inside the Dewar vessel. Experiment No.1: To obtain high concentrations of radon in the upper part of the thermally insulated container (in our case, a thermos), in which liquid nitrogen is poured, pieces of uranium ore wrapped in gauze, pieces of brannerite veins, were put. Since it is impossible to measure radon in the liquid state, measurements are made after complete evaporation of liquid nitrogen, when a radon gas concentrate is obtained inside.
The highest concentrations of radon within four days changed in the following sequence: 73.22 ± 12.37 Bq / m$^3$; 496.57 ± 74.47 Bq / m$^3$; 959.06 ± 143.81 Bq / m$^3$; 462.8 ± 69.41 Bq / m$^3$.

It was established experimentally that when used as a container for a radon concentrator a 2-liter thermos, for two days after placing the ore in a vessel with liquid nitrogen, which is closed by a thermally insulated lid, the nitrogen evaporates completely. And the maximum radon concentration inside the thermos is 959.06 ± 143.81 Bq / m$^3$.

Experiment No.2: Also, for clarity, experiment No.2 was carried out in the same thermos at three different levels inside the thermos. Measurements began to be carried out after 3 days of exposure in a vessel of active ores weighing 403.49 grams, where liquid nitrogen was poured. The results are shown in table 1.

| Table 1. Experiment No.2 results. |
|-----------------------------------|
| 4 cm from the base of the thermos | 10 cm from the base of the thermos | 16 cm from the base of the thermos |
| $V_{ARn}$, Bq/m$^3$ | $EEV_{ARn}$, Bq/m$^3$ | $V_{ARn}$, Bq/m$^3$ | $EEV_{ARn}$, Bq/m$^3$ | $V_{ARn}$, Bq/m$^3$ | $EEV_{ARn}$, Bq/m$^3$ |
| First day of measurement | 858 ± 93.43 | 429 ± 46.71 | 698.76 ± 64.1 | 349.38 ± 32.05 |
| Second day of measurement | 591.6 ± 295.8 ± 27.68 | 64.1 | 450.43 ± 45.57 | 225.21 ± 22.78 |
| Third day of measurement | 1037.33 ± 767.2 ± 30.61 | 49.78 | 24.89 | 61.23 | 30.61 |

The maximum radon value is observed on the third measurement day, practically at the bottom of the thermos (4 cm from the base): 1037.33 ± 49.78. And on the same day, at another level inside a thermos (10 cm from the base), the radon activity is less by almost 250-300 Bq / m$^3$. For clarity, the results are shown in figure 1.

![Figure 1. Experiment No.2 results.](image)

Experiment No. 3: The experiment was also carried out with a Dewar vessel, inside which were placed active pieces of uranium ore weighing 500.19 grams. Measurements began to be carried out after 7 days of aging in a vessel where liquid nitrogen was poured. Table 2 shows the indications of radon activity over several days.
Table 2. Experiment No.3 results.

| Days of measurement               | Average radon activity readings |
|-----------------------------------|----------------------------------|
|                                   | VA\(_{\text{Rn}}\) Bq/m\(^3\) | EEVA\(_{\text{Rn}}\) Bq/m\(^3\) |
| First day of measurement          | 17.06 ± 4.82                    | 8.53 ± 2.41                      |
| Second day of measurement         | 691.2 ± 420.1                   | 345.6 ± 210.05                   |
| Fourth day of measurement         | 338.5 ± 51.63                   | 169.25 ± 25.81                   |

3. Calculation of effective dose of irradiation during radon procedures

During various types of radon procedures, patient dose rates are determined by the duration of the procedures, the characteristics of the radiation source and the patient's exposure conditions [5]. If, during airborne radon procedures, the patient breathes clean air, in which radon EEVA is close to its value in atmospheric air, then the effective dose of its exposure is almost entirely determined by the gamma radiation of the daughter products of radon decay in the cabin air, in which the patient takes an air radon bath. Estimation of the effective dose of the patient under these conditions is calculated by the formula:

\[ E = 1.65 \times T \times C_{\text{Rn}} \times 10^{-10}, \text{ mSv / procedure} \]  

(2)

where \( T \) - procedure duration, min; 
\( C_{\text{Rn}} \) – radon activity in the cabin air (Bq / m\(^3\)) under conditions of complete radioactive equilibrium with its decay products.

The effective dose of internal exposure of the patient due to inhalation intake of radon and its daughter decay products with inhaled air is determined by the value of EEVA of radon in the air of the respiratory zone \( C_{\text{Rn}}^{\text{air}} \) (Bq/m\(^3\)) and the duration of the procedure and is calculated by the formula:

\[ E = 1.625 \times C_{\text{Rn}}^{\text{air}} \times T \times 10^{-7}, \text{ mSv / procedure} \]  

(3)

where \( T \) - procedure duration, min; 
\( C_{\text{Rn}}^{\text{air}} \) – EEVA of radon in the air of the breathing zone (Bq / m\(^3\)).

If, when receiving airborne radon procedures, the patient breathes air from the room in which the radon baths are installed, then in calculating the effective doses to the patient under these conditions, the contribution of radon inhalation and its inhaled air flow should also be taken into account. This component of the effective dose of the patient is determined by the formula (2), and to estimate the total dose, the dose is added to it due to external exposure of the patient obtained by the formula (3).

In radiation hygiene, it is customary to calculate doses for the maximum possible values of the radon volume activity under these conditions [12].

Let us calculate, using formula (3), the value of the patient's effective dose due to inhalation intake of radon and its daughter decay products with inhaled air, taking the maximum VA of radon from experiment No.2 1037.33 ± 49.78 Bq / m\(^3\) (EEVA of which 518.6 Bq / m\(^3\)), we take the duration of the procedure for 15 minutes:

\[ E = 1.625 \times 518.6 \times 15 \times 10^{-7} = 12640.87 \times 10^{-7} = 1.26 \text{ mSv / procedure}. \]

At this value of the radon volumetric activity of 1037.33 ± 49.78 Bq / m\(^3\) when the patient receives the radon bath in the cabin, the effective dose to the patient under these conditions using formula (2) will be (we will take the procedure for 15 minutes):

\[ E = 1.65 \times 15 \times 1037.33 \times 10^{-10} = 25673.91 \times 10^{-10} = 0.0025 \text{ mSv / procedure}. \]

This means that what radiation dose the patient will receive (at the same radon concentration) depends on the procedure. For therapeutic purposes, use natural or artificially prepared (by physical means) radon water. In addition to general and local radon water baths, other types of radon procedures are used: “dry”, or air-radon, baths, gynecological irrigation and micro enemas, inhalations, drinking radon waters, etc. [13].
With radon procedures with a commonly used therapeutic dosage, the dose of radiation to critical organs of patients ranges from 1 to 5 mSv [14].

4. Conclusion

Thus, it was experimentally obtained that using liquid nitrogen and uranium ore, radon can be concentrated to high levels by overshoot radon activity levels, which can reach almost 1000 Bq / m³.

With additional experiments with different active ores, it is possible to obtain greater or lower concentrations of radon activity, which can, if necessary, be used to treat diseases with the help of radon baths. It has been proven that it is possible to create a cryogenic radon concentrator with a storage device in the form of a Dewar vessel, and to apply a concentrator for the treatment with radon.

For the practical use of concentrated radon, it is possible to technically make a bubbling device for concentrating and moving radon gaseous concentrate for medical use.

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