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Study of atmosphere parameters of the IVV-2M reactor hall

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ABSTRACT

The paper presents the results of a study of radioactive noble gases and from decay products in the atmosphere of the reactor hall of the research nuclear reactor IVV-2M. The distribution of short-lived ^{88}Rb and ^{138}Cs activity by sizes of aerosol particles was measured in the range of 0.5–1000 nm. It is shown that radioactive aerosols are characterized by three main modes with AMTD 2–3 nm, 7–15 nm and 400 nm. About 70% of aerosol activity is due to ^{88}Rb. The equilibrium factor between ^{88}Kr and ^{88}Rb is 0.2 ± 0.1. The total concentration of aerosols particles was measured using an aerosol diffusion spectrometer. The value of unattached fraction of radioactive aerosols in the atmosphere of reactor hall IVV-2M was f = 0.15–0.25 at the average total aerosol particles concentration from 20,000 cm^{-3} to 53,000 cm^{-3}.

1. Introduction

The detail information about the parameters of gas-air environment at the workplace is one of the key points in organizing principles of radiation safety at the atomic industry. Information about physical and chemical properties of radioactive gases and aerosols allows the correct use of personal protective equipment and reasonable decrease of the effective dose to personnel when performing various technological procedures during operation at power, and during the repair period [1].

Radioactive aerosols occur in the atmosphere due to both natural and anthropogenic factors. Natural radioactive aerosols appear as a result of an interaction of natural non-radioactive aerosols with the decay products of radon isotopes ^{220}Rn and ^{222}Rn as well as with ^{7}Be. Anthropogenic radioactive aerosols are the result of activities of nuclear power plants and plants for the processing of irradiated nuclear fuel as well as the nuclear weapon tests [2].

The formation of radioactive aerosols at nuclear power plants is caused by the operation of a nuclear reactor. During the operation of a nuclear reactor, a large amount of fuel fission and activation products are formed. Among them, radioactive noble gases (RNG) have a special place [3]. In terms of radiation exposure, RNG are not so dangerous, as their decay products [4].

The process of RNG decay with the formation of radioactive decay products is a unique physical process. When a noble gas atom decays in air, an isolated, usually positively charged, radioactive atom is formed. No other physical processes (evaporation, explosive spraying, plasma spraying, etc.) can result in the appearance of an isolated radioactive atom in air under normal conditions. This effect is well known in the decay of natural radioactive noble gases ^{222}Rn and ^{220}Rn. About 85–88% of free atoms formed during the decay of radon isotopes are positively charged [5]. Radioactive decay products of noble gases exist in the air as free atoms for a very short time (<1 s) [6,7]. The free atoms, interacting with trace gases and water vapor, form highly dispersed aerosol up to 15 nm in size, traditionally called the unattached fraction. The size for such radioactive particles is represented by the value of the activity median thermodynamic diameter (AMTD). During the decay of polonium isotopes, most of the recoil nuclei of ^{212}Pb and ^{214}Pb are also formed as free atoms with the subsequent formation of highly dispersed aerosols of the unattached fraction. The clusters of the unattached fraction can be neutral or positively charged. It is necessary to take into account that the process of neutralization of a cluster of an unattached fraction is possible. During the interaction of the unattached fraction with existing aerosols, radioactive aerosols of a larger diameter of about 100–400 nm (attached fraction) are formed.

The effect of the formation of isolated radioactive atoms during the decay of RNG is not typical only for the decay products of radon. Similar processes can also be observed during the decay of artificial

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RNGs, such as ^{88}Kr and ^{138}Xe. However, in practice, studies of the unattached fraction and the further formation of medium- and coarse-dispersed aerosols during the decay of these RNGs have not been carried out. The ratio of the activity concentration of the maternal radionuclides and their decay products is characterized by an equilibrium factor. A similar parameter has been well studied for the natural radioactive noble gas radon [4–7]. However, for such RNGs as ^{88}Kr and ^{138}Xe, the equilibrium factor under real conditions also has not been studied.

Another important parameter of the atmosphere is activity distribution by aerosol particles sizes. This information is important for a more accurate assessment of equivalent and effective doses during inhalation intake of radionuclides into the human body. Depending on the disperse composition of radioactive aerosols, the dose coefficients can differ by about an order of magnitude. In the presence of RNG with their decay products in the atmosphere, it is important to estimate the unattached fraction in relation to the total number of radioactive atoms in the atmosphere.

The aim of this study is determination of the equilibrium factor between anthropogenic radioactive noble gases and their progeny and proportion of the unattached fraction activity in comparison with the total activity concentration of radioactive atoms in the atmosphere in the IVV-2M reactor hall atmosphere.

2. Object of study

The object of this investigation is the activity concentration of RNG, activity concentration and distribution activity by sizes of aerosol particles and total concentration and size distribution of non-radioactive aerosols in the atmosphere of the reactor hall of the Institute of Nuclear Materials (INM) in Zarechniy, Russia.

In INM research water-water reactor IVV-2M for various materials research and the radioisotope production is operating [8]. As a result of more than 55 years of operation of the reactor, a certain amount of ^{235}U fuel got into the first cooling circuit of the reactor core. It results in the RNG production under the fission reaction in first circuit water. As a result, anthropogenic RNG (xenon and krypton) can diffuse into the atmosphere of the reactor hall. The RNG atoms can turn into atoms of radioactive metals (cesium and rubidium) due to radioactive decay. Free metal atoms in the atmosphere can form unattached fraction – solid aerosols with very small (1–2 nm) particle diameters [9]. Later such aerosols can interact with non-radioactive aerosols forming radioactive aerosols with diameter 200–500 nm. The scheme of the formation of free atoms of the decay products of ^{88}Kr and ^{138}Xe, unattached fraction of these radionuclides and attachment to aerosol particles is shown in Fig. 1.

Radioactive aerosol particles in the studied atmosphere are mainly represented by ^{88}Rb and ^{138}Cs atoms, which form clusters by joining water molecules. Such unique situation allows to investigate the equilibrium factor between anthropogenic RNG and their decay products, which has not been studied yet. It is an important parameter of radioactive substances in atmosphere for radiation safety assessment of a nuclear reactor. Since the studied atmosphere is a rather dynamic, aerosol particles in the form of daughter products of the decay of technogenic RNG have a high probability of deposition on structural elements. Thus, the equilibrium in the atmosphere between the parent and daughter atom can be disturbed.

3. Instruments and methods

The study of the equilibrium factor was carried out with the simultaneous measurement of RNG and radioactive aerosols concentrations and the total concentration of aerosol particles with the use of approach, described in Ref. [10].

Measurements of the RNG activity concentration were carried out by creating an excess pressure in a special Marinelli vessel, developed in the Institute of Industrial Ecology UB RAS (Fig. 2) and subsequent measurements on a coaxial gamma spectrometer with a HPGe detector. The 3-L Marinelli metal vessel made of stainless steel 1.5 mm thick and a caprolon liner 5 mm thick [3] allows to create excess pressure up to 15 atm.

Radioactive aerosols with RNG decay products were collected by the sampling system using a 20-cascade diffusion battery, also developed in the Institute of Industrial Ecology UB RAS (Fig. 3), in combination with aerosol filters. The battery consists of aluminum blocks equipped with wire screens separated by rubber rings to ensure laminar flow between the elements. Generally, four blocks with five screens in each were used [11–13]. The sampling elements are made of brass meshes with a wire thickness of 65 μm and a cell opening width of 100 μm. After pumping air with rate 39 l/min (air velocity 32.5 cm/s), the cells of the diffusion battery were reassembled and measured by a beta radiometer. After beta radiometry, the analytical filters were analyzed using a gamma spectrometer to measure radionuclide composition of aerosols in the atmosphere.

Since the atmosphere of the reactor of IVV-2M is presented by aerosol particles of short-lived radionuclides, the activity concentration was corrected for radioactive decay during the sampling process:

$$A_V^{aerosol} = \frac{I \bullet \lambda}{\varepsilon \bullet \upsilon \bullet (1 - exp(-\lambda \bullet t_1))}, \quad (1)$$

where $A_V^{aerosol}$ – activity concentration of radioactive aerosols on the trapping element, Bq/m^3;

λ – constant half-life of a mixture of radioactive aerosols in the reactor hall, min^{-1};
υ – air flow through diffusion battery – 0,039 m^3/min;
ε – beta particle detection efficiency for a specific type of capture element;
I – count rate from the trap element corrected for decay during measurement, sec^{-1};
t_1 – diffusion battery sampling time, min

Activity concentration of RNG collected with metal vessel and compressor was calculated using eq. (2).

$$A_V^{RNG} = \frac{A_\gamma}{\eta \bullet V}, \quad (2)$$

where A_V^{RNG} – activity concentration of RNG in reactor hall atmosphere, Bq/m^3;

A_γ – activity of each RNG radionuclide, measured with HPGe detector and calculated taking into account decay half-life, Bq;
η – pressure in metal Marinelli vessel, atm;
V – volume of metal vessel, m^3

The coefficient f characterizes the proportion of aerosol radioactive particles of the unattached fraction to their total number captured by the trapping elements of the diffusion battery.

The coefficient f was calculated by eq. (3).

$$f = \frac{A_{unat.\,fraction}}{A_{total}}, \quad (3)$$

where $A_{unat.\,fraction}$ – total activity of radioactive aerosol particles represented by the unattached fraction (aerosol particles that

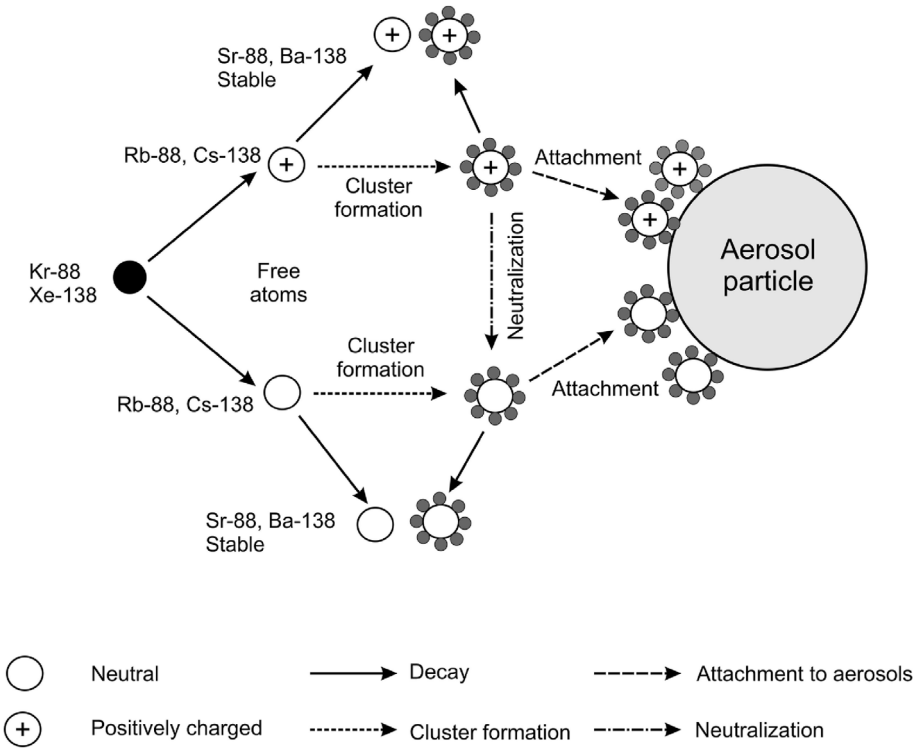

Fig. 1. The formation of free atoms of noble gases decay products, an unattached fraction, neutralization and attachment to aerosol particles.

Fig. 2. Metal Marinelli vessel for the measurements of RNG activity concentration.

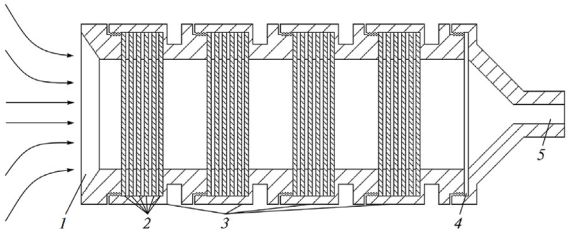

Fig. 3. Schematic diagram of the diffusion battery: (1) inlet, (2) trapping elements, (3) diffusion battery cascades, (4) last filter, and (5) outlet [11–13].

captured only by the mesh screens of the diffusion battery), Bq;

A_{total} – total activity of all radioactive aerosol particles captured by all elements of the diffusion battery, Bq.

Equilibrium factor was calculated by eq. (4).

$$EF = \frac{A_V^{daughter}}{A_V^{parent}}, \quad (4)$$

where $A_V^{daughter}$ – activity concentration of RNG decay products, Bq/m^3;

A_V^{parent} – activity concentration of parent atoms of RNG, Bq/m^3.

4. Results and discussions

The atmosphere of the reactor hall is presented by such RNG as ^{41}Ar, ^{85m}Kr, ^{87}Kr, ^{88}Kr, ^{133}Xe and ^{135}Xe. Reliable measurement of the ^{138}Xe activity concentration was not possible due to its short half-life. The main radioactive aerosols present in the atmosphere of the reactor hall are ^{88}Rb and ^{138}Cs – decay products of ^{88}Kr and ^{138}Xe respectively. The decay products of other RNG are either stable isotopes or radionuclides with very long half-lives or low-energy beta emitters.

The fraction of ^{88}Rb in the total activity concentration of radioactive aerosols in the IVV-2M reactor hall is about 0.7. The ratio of the average activity concentrations of ^{88}Rb and ^{88}Kr demonstrated the equilibrium factor value – 0.2 ± 0.1.

Figs. 4 and 5 demonstrate the measurements results during three cycles of normal operation of nuclear reactor performed simultaneously with DAS 2702-M and diffusion battery [14]. It is evident that the distribution activity by aerosol particles sizes has three primary modes: with AMTD 2–3, 7–15 and 400 nm. The modes with AMTD less than 15 nm are small radioactive highly dispersed aerosols (2–3 nm) and nucleation nuclei (7–15 nm) of aerosols. Such fine particles are formed as a result of the radioactive decay of RNG caping from the space above the IVV-2M reactor. The mode AMTD 400 nm corresponds to aerosols of condensation nature. These aerosols connected with particles of dust and water present in the atmosphere of the reactor hall. The aerosol particles

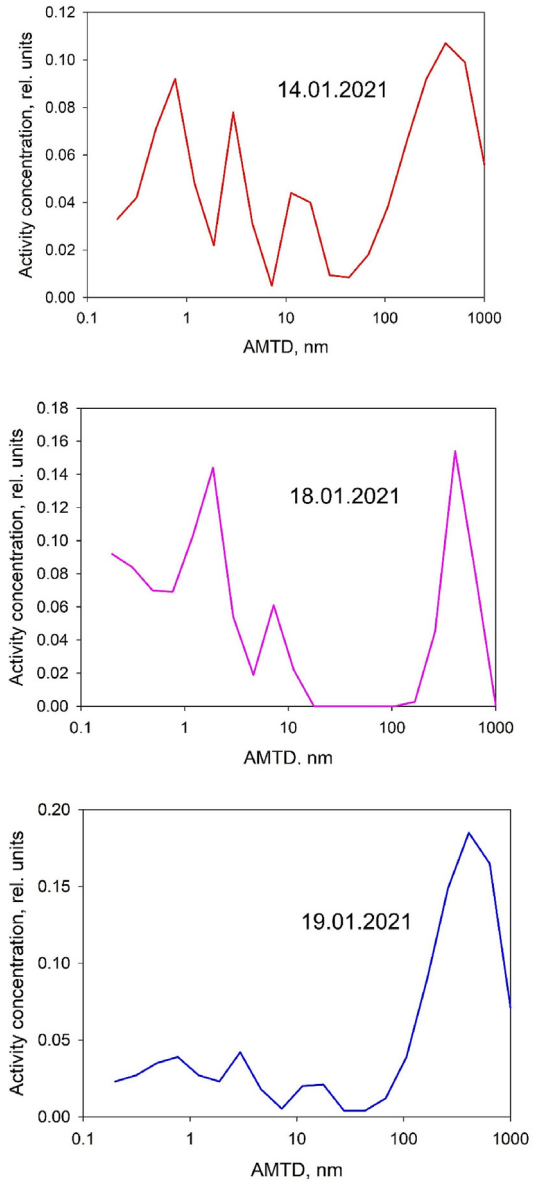

Fig. 4. Measurements of activity distribution on particle sizes in reactor hall atmosphere of IVV-2M reactor.

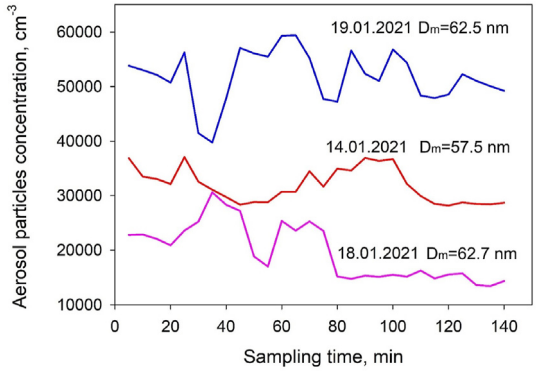

Fig. 5. Time dependent measurements of the concentration of non-radioactive aerosol particles in reactor hall atmosphere of IVV-2M reactor.

concentration varies from $2.0 \cdot 10^4$ to $5.3 \cdot 10^4$ cm^{-3} and depends on technological process in the reactor hall. The registration of the unattached fraction of radioactive aerosols shows that the processes of formation of radioactive aerosols during the decay of radon isotopes and ^{88}Kr and ^{138}Xe isotopes have a similar behavior. This effect was previously observed only in a pilot experiment carried out at the Institute of Reactor Materials [15]. This paper confirms and complements the previously obtained materials. The fundamental difference between this paper and [15] is the simultaneous determination of the activity concentrations of RNG and aerosol particles in the atmosphere of the reactor hall.

The activity concentration of RNG changed insignificantly during the expose of the diffusion battery. The average value of the ^{88}Kr activity concentration was $(1.1 \pm 0.3) \cdot 10^4$ Bq/m^3. The total activity concentration of radioactive aerosols sampled by the diffusion battery ranged from $1.6 \cdot 10^3$ to $3.3 \cdot 10^3$ Bq/m^3.

The unattached fraction of radioactive aerosols f was estimated as part of the particles activity with AMTD less than 15 nm from the total activity of radionuclides captured on the diffusion batteries is in the range from 0.15 to 0.25. The average size of the aerosol particles in the reactor hall based on the results of DAS measurements was from 50 to 65 nm. Various technological processes in the reactor, which lead to an increase in the overall aerosol particle concentration, slightly reduce the unattached fraction. Such a decrease can be explained by an increase in the probability of the attachment of ^{88}Kr and ^{138}Cs clusters to condensation nuclei. The figures show a significant dependence of the part of the highly dispersed aerosols with AMTD 1–2 nm on the total concentration of aerosol particles. At a concentration of aerosol particles in the second half of the sample, which is the most significant for the interpretation of the results, ~50,000–55,000 cm^{-3}, the activity of the highly dispersed aerosols with AMTD 1–2 nm was no more than 0.05. At a concentration of aerosol particles of about 30,000 cm^{-3}, this value increased to 0.09, and at a particle concentration of only 15,000 cm^{-3}, it increased to ~0.14.

The dose coefficients for the isotopes ^{88}Kr and ^{138}Cs (type F) are fairly close. However, the values of dose coefficients differ significantly for radioactive aerosols with different values of AMTD. For example, for AMTD 3, 10, 30, and 300 nm, the dose coefficients ^{88}Kr and ^{138}Cs are $4.8 \cdot 10^{-11}$, $(4.0–4.1) \cdot 10^{-11}$, $(2.7–2.8) \cdot 10^{-11}$ and $(1.2–1.3) \cdot 10^{-11}$ Sv/Bq [16]. Such a noticeable difference in the dose coefficients for the unattached fraction and medium-sized aerosols demonstrates the importance of obtaining detailed information on the size distribution of radioactive aerosols in the reactor hall.

5. Conclusions

Investigation in the IVV-2M reactor hall atmosphere demonstrated the follow results:

1. The conducted studies made it possible to assess in detail the radiation situation in the reactor's hall during the operation of the IVV-2M reactor:
   - the average value of the ^{88}Kr activity concentration was $(1.1 \pm 0.3) \cdot 10^4$ Bq/m^3;
   - activity concentration of radioactive aerosols is in the range from 1.6 to 3.3 kBq/m^3;
   - nearly 70% of the activity of radioactive aerosols is due to ^{88}Rb.
2. The equilibrium factor between ^{88}Kr and ^{88}Rb is 0.2 ± 0.1. Taking into account the higher intensity of air exchange in the reactor hall compared to conventional rooms, this value is quite close to the values of the equilibrium factor between radon and its decay products [5–7].

3. The distribution activity by aerosol particles sizes has three primary modes with AMTD 2–3, 7–15 and 400 nm. The presence of these size modes demonstrates that the processes of formation of highly dispersed aerosols and nucleation modes, as well as attachment to condensation aerosols, occur for decay products of "light" RNGs similarly to the processes known for radon decay products [5–7,10–12].
4. The total aerosol concentration in reactor hall is in the range from $2.0 \cdot 10^4$ to $5.3 \cdot 10^4$ $см^{-3}$ and depends on the on technological process in the reactor hall. The unattached fraction of radioactive aerosols f (particles with AMTD less than 15 nm) from the total number of radionuclides captured on the diffusion batteries is in the range from 0.15 to 0.25.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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