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V. Ye. Khan, O. S. Lagunenko, V. O. Krasnov, O. O. Odintsov, O. K. Kalynovskyi, V. M. Bezmylov, V. O. Kashpur, P. V. Sabenin, O. A. Svirid, A. V. Tkach

Institute for Safety Problems of Nuclear Power Plants, NAS of Ukraine, 36a, Kirova st., Chornobyl, 07270, Ukraine

Radioactive Aerosols in Sub-Reactor Room 304/3 of the Chornobyl NPP Shelter Object within Conditions of Operation of the New Safe Confinement

Keywords:
New Safe Confinement,
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lava-like fuel-containing materials,
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volumetric activity,
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dispersity

Radionuclide composition, volumetric activities (VA) and dispersity of aerosols sampled in 2019–2023 in room 304/3 of the Shelter object, where lava-like fuel-containing materials (LFCM) penetrated after the accident, are presented. The availability of transuranium elements in radionuclide composition of aerosols in room 304/3 in ratios close to those in LFCM shows that the aerosol content continues growing as result of lava degradation. The carriers of radioactive products of accident are, as a rule, aerosols with the activity median aerodynamic diameter larger than 1 μm. It evidences their dispersion origin. Substantial enrichment of aerosols in room 304/3 by ^{137}Cs and ^{90}Sr, relatively to lava composition, indicate the presence of at least two sources influencing radionuclide composition of aerosols. It is confirmed by the values of parameters of disperse composition of aerosol.

Introduction

The lava-like fuel-containing materials (LFCM) incorporating a large amount of nuclear fuel and radioactive fission products were formed during the first days after the Chornobyl NPP accident in 1986. The main lava mass had produced in the sub-reactor room 305/2. From that place, after big horizontal LFCM flow was formed, they penetrated through a breach in room 304/3 wall and other rooms on the mark 9.000. In addition, after two vertical flows (big and small) produced, the lavas ingressed in the bottom rooms of ruined building of Unit 4 and generated LFCM accumulations in the rooms of system of steam-distribution corridor (mark 6.000) and rooms of system of bubbler pool (mark 3.000 and 0.000, accordingly) [1, 2].

Stiffened lavas represent nuclear, radiation and radioecological hazards. According to expert estimates, the LFCM contain from 60 to 110 tons of irradiated fuel, i. e. 32–58% of initial fuel loaded into the reactor [2]. Their behavior is insufficiently understood and has no substantiated forecast.

Although the accident occurred almost 38 years ago, data on the condition of LFCMs and, the more so, on their degradation are extremely scarce. The first studies concern March 1990 [3], when an alcoholic smear, a smear onto an ashless filter, and an aerosol sample were taken from the lava “tongue” at the south-western entrance of room 210/7. In 1995, aerosols were taken onto a filter in room 305/2, wherefrom the lava entered into room 210/7 and then into room 012/15 [4]. About 70% of particles were spherical with the diameter of 1–3 μm.

In 2008–2009, aerosol samples were taken in rooms 304/3 and 305/2, where there are large LFCM accumulations [5]. Then, the attention was focused on aerosols in room 304/3 [6]. As noted in [7], “in the period of 2009–2011, the radioactivity maximum in the distribution of α-active aerosols with respect to aerodynamic diameters (AD) shifted from the interval 2.0–10 μm to 0.6–2 μm." Starting from the autumn of 2011, studies in room 304/3 were continued using an air stream ejected from nozzles at a velocity of 41 m s^{-1} for raising the dust from the LFCM surface. The authors concluded that the "relative concentration of fine (AD smaller than 0.8 μm) particles bearing α-emitting radionuclides increased with increasing time of dust accumulation, and that of coarse particles decreased" [8]. Unfortunately, the content of main radionuclides (^{90}Sr, ^{137}Cs, ^{154}Eu, ^{155}Eu, Pu, $^{239+240}$Pu, ^{241}Am) in aerosols and LFCMs is not given in [5–9]. If isotope ratios in the aerosols were the same as in LFCMs, this fact would be indicative for LFCM degradation.

The researches of 2010–2014 in sub-reactor rooms of the Shelter object [3, 10, 11] showed that due to lava degradation, spontaneous transition of degradation products in aerosol state is observed. In addition, the number of radioactive aerosols, which are the product of LFCM degradation, depends on lava type.

On November 29, 2016, the Arch of New Safe Confinement (NSC) was installed in its design position over the Shelter object. The NSC creation resulted in the changes of temperature-moisture mode inside the object, and in gradual drying of water accumulations and drop in air humidity in its rooms. It contributes in dust resuspension, including from LFCM surface, and in generation of dust by bottom sediment surfaces produced as result of drying of radioactively contaminated water accumulations.

The researches in sub-reactor rooms of the Shelter object conducted in 2017–2018 [12–14] demonstrated that spontaneous transition of LFCM degradation products in aerosol state has decreased. Because of the above, the values of volumetric activities (VA) of radionuclides turned out to be lower as compared to the results of 2010–2014 investigations.

In 2019–2023, the work to research the behavior of radioactive aerosols in sub-reactor rooms of the Shelter object was continued [15]. The goal of work is to monitor the dynamics of radioaerosol situation in the rooms, where LFCM are localized, within the conditions of operation of "New Safe Confinement — Shelter object" complex. That is the subject of this paper. It covers the studies performed in room 304/3.

Data on room 304/3 and LFCM

In accordance with the Technological Regulations of operation of the "New Safe Confinement — Shelter object" complex, room 304/3 is classified as "unattended" one. No works were performed in the research period in this room.

Room 304/3 for monitoring and measuring devices is located on mark 9.300 between the axes 45^{+300}–47_{-300} and rows $Ж^{+400}$ — $И_{-900}$. Because of high exposure dose rate (EDR) values and collapse of structures, rooms 305/2 and 304/3 remain insufficiently investigated. The main data on LFCM amount and location are based on the results of drilling works, measurements of neutron and thermal fluxes, and on examination with video cameras [16–19]. According to the calculations [16], there is about 6 t of uranium in room 304/3. The black LFCM filled all the square of room (Fig. 1). Upper LFCM boundary, which filled the room, reaches the mark 10.300. One should consider that they produced a single accumulation. The surface of LFCM is heterogeneous, coated with cracks and consists of individual pieces. Visually, lava structure is coarse-posed, there are a lot of cavities in solidified lava, some of them have the volume of 1–2 dm^3. LFCM volume makes: 50–70 m^3, fuel mass- 4–8 t uranium. The square of horizontal projection of LFCM surface makes 63 m^2. Estimated

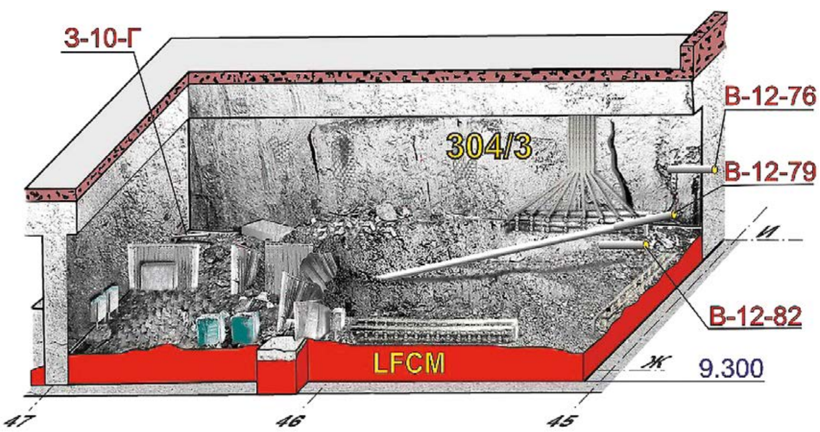

Fig. 1. Reconstruction of room 304/3 conditions

area of open surface of LFCM accumulation in room 304/3, in line with work [20] data, makes 312±38 m^2.

Tools and techniques to monitor radioactive aerosols

The aerosols were sampled by H810 SAIC blower at about 100 L min^{-1} rate. The 20 cm^2 stacks of Petrianov filters, consisting of AFA RSP-20 and AFA RMP-20 layers, were used. Two AFA filters ensured complete trapping of aerosols. Since there was no access to room 304/3/2, the samples were taken via the borehole B-12–76 drilled in room 304/3, by way of connecting the sampler to casing pipe with 10 cm internal diameter and 2 m length (Fig. 2).

The sampling of aerosol particles and their in-size classification was made using 5 cascade impactor IBF-5K. This device makes particle gradation on five ranges of aerodynamic diameter (AD): <0.5 μm; 0.5–1.2 μm; 1.2–3.7 μm; 3.7–8.5 μm; 8.5–17.0 μm. As the fifth stage (<0.5 μm), finely dispersed filter was used, which allowed more completely catching submicron-sized aerosols. The sampling was held during 1–2 weeks. Temperature and relative humidity were measured using Elitech RC4HA/C register and TH mini hygrothermometer with remote sensor of temperature-humidity.

Sample beta-activity was measured by the device MKS-01R in 4–5 days, when daughter products of radon and thoron completely decayed. As a result, total activity was identified of long-existing beta-emitting nuclides — Chornobyl accident products (Σβ) in taken samples, which include ^{90}Sr + ^{90}Y and ^{137}Cs isotopes.

The next measurements of radioactive substances were carried out at Canberra company gamma-spectrometer consisting of semiconductor detector GL2020R made of ultrapure germanium and 500-mkm thick beryllium window, and 8192-channel amplitude pulse analyzer. The measuring range covers the energies from 10 to 1,400 keV. The detector has 0.57 and 1.2 keV resolution for 122 keV gamma-quanta energies (gamma-line of ^{57}Co) and 661.6 keV (gamma-line of ^{137}Cs), accordingly.

In aerosol after radiochemical separation, ^{90}Sr activity was determined by beta-radiometry, and the activities of Pu, $^{239+240}$Pu and ^{241}Am — by alpha-spectrometry measurements.

Concentration of aerosols carrying the accident product

Over 2019–2023 period, 36 aerosol samples were taken in room 304/3. Relative moisture of air in the room during sampling period was varying from 40 to 99 %, temperature — from 3 to 20 °C (Fig. 3). The lowest values of relative moisture were registered in winter period, and the highest values — in period from June to October.

One should note, the results of visual survey of room 304/3 realized in 90s of the last century have demonstrated that the room is dry, when walking in it, the dust is rising [21, 22].

As Fig. 4 shows, in 2019–2023 volumetric activity (VA) of Σβ in the room was varying within the range of 0.05–1.3 Bq/m^3, and ^{241}Am VA was varying within the range of $2.0 \cdot 10^{-4}$–$6.3 \cdot 10^{-3}$ Bq/m^3. At the same time, trend line demonstrates ^{241}Am VA drop in room air in contrast to Σβ VA.

In most cases, increased aerosol VA was observed under relatively low air moisture in the room, and decreased VA coincided with times with high moisture value.

In line with radiometry and gamma-spectrometry measurements of samples, ^{137}Cs/Σβ ratios in aerosols during 2019–2023 was fluctuating from 0.17 to 0.90 under mean value 0.64. From obtained mean value of ^{137}Cs/Σβ follows that ^{137}Cs input in Σβ made 64 %. Correlation dependence of ratio over time was not established.The aerosol samples were analyzed by radiochemical method. Be-

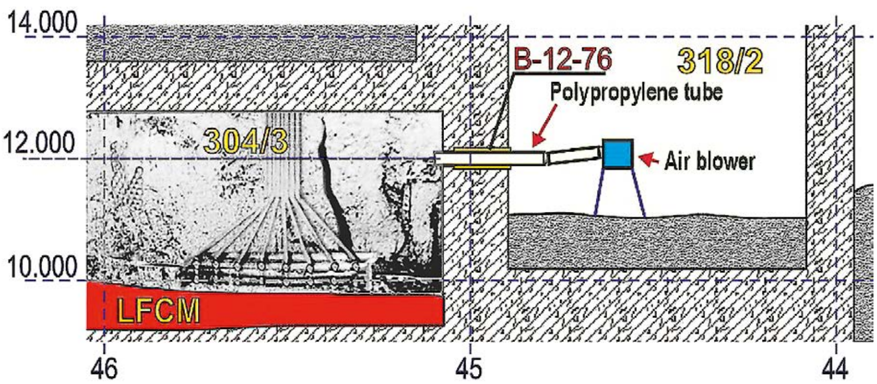

Fig. 2. Sampling of radioactive aerosols in room 304/3

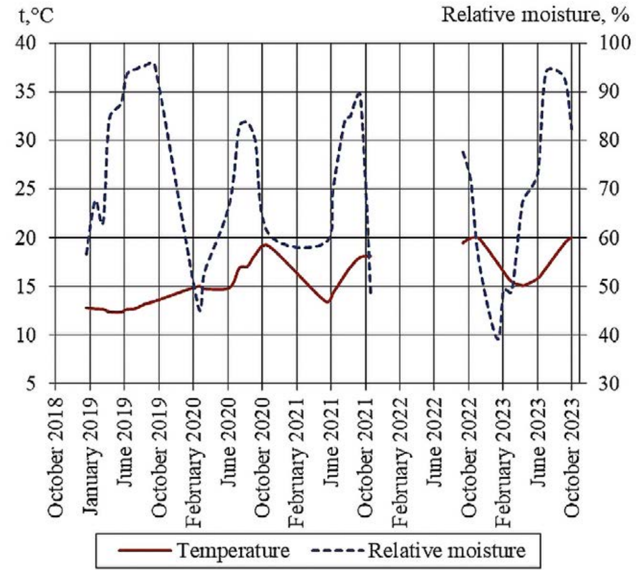

Fig. 3. Temperature and relative moisture in room 304/3 when sampling aerosols in 2019–2023

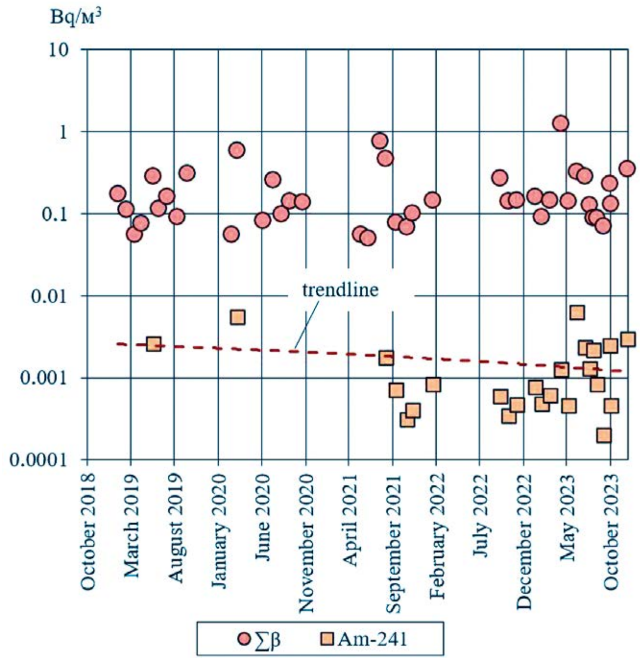

Fig. 4. VA of aerosols carring Σβ and 241Am in room 304/3 in 2019–2023

low the analysis results are shown of most active on Σβ aerosol samples taken on April 11, 2023.

Content of radionuclides in room 304/3 aerosols, Bq/m^3

^{90}Sr	0.19
^{137}Cs	0.88
Pu	$2.0 \cdot 10^{-4}$
$^{239+240}$Pu	$4.1 \cdot 10^{-4}$
^{241}Am	$1.3 \cdot 10^{-3}$
^{244}Cm	$1.9 \cdot 10^{-5}$
Σβ	1.3

In Table 1, mean values of nuclide activity ratios in aerosol samples taken during 2019–2023, and ratios in LFCM localized in room 304/3, are given. The data on radionuclide composition in LFCM within room 304/3 were obtained from database of Nuclear and Radiation Safety Division of the Institute for Safety Problems of Nuclear Power Plants of the National Academy of Sciences of Ukraine.

The data presented in Table 1 testify that in 2019–2023 period, the ratio of radionuclides in aerosols and LFCM of room 304/3 differed substantially due to aerosol enrichment by ^{137}Cs and ^{90}Sr relatively to ^{241}Am. At the same time, over time the aerosol enrichment by cesium and strontium is growing. The value of ratios of ^{241}Am/$^{239+240}$Pu activities in aerosols and in LFCM have close values. The discrepancies are associated with both measurement uncertainty and heterogeneity of LFCM composition [1]. The ratio of plutonium isotopes in aerosols remained identical to the values of analogous ratios in the LFCM. Thus, due to LFCM surface erosion, generation of radioactive dust occurs, which penetrates the air in room 304/3.

Dispersity of radioactive aerosols

Underway March — November 2023, to identify disperse composition of aerosols, 10 aerosol samples were taken with using five-cascade impactor IBF-5K. Assuming a priori the logarithmic normal distribution of aerosol particle size, we calculated the activity median aerodynamic diameter (AMAD) and standard geometric deviation (σ) (Table 2).

In room 304/3, the particles with AMAD of 2.8–11 μm only were most frequently ^{137}Cs carriers, one AMAD sample was smaller than 1 μm. The AMAD value of ^{241}Am-bearing particles was varying within the range 2.0–10.2 μm. Such a distribution testifies that the main mechanism of their origination had dispersion character [23]. That is why ^{241}Am-bearing particles, based on presented results of radiochemical research, should be considered as products of LFCM surface degradation [3].

The value of ^{137}Cs/^{241}Am ratios has demonstrated that at all impactor cascades, ^{137}Cs was present in excess amount relatively to its content in the lava. Taking into account the values $\sigma > 3$, one can assume that the aerosols in room 304/3 in 2023 were produced from different sources of radioactive aerosols.

Discussion

The above materials demonstrate that in room 304/3 volumetric activities, radionuclide composition

Table 1. Radionuclide ratios in aerosols and LFCMs in room 304/3

Object, year	$^{137}Cs/^{241}Am$	$^{90}Sr/^{241}Am$	$^{241}Am/^{239+240}Pu$	$^{239+240}Pu/Pu$	$^{137}Cs/\Sigma\beta$
aerosol, 2019	68	21	2.0	2.5	0.63
LFCM, 2019	7.8	22.0	1.6	2.3	0.21
aerosol, 2020	54	26	1.7	2.9	0.56
LFCM, 2020	7.7	21.7	1.6	2.4	0.21
aerosol, 2021	130	40	2.0	2.3	0.54
LFCM, 2021	7.6	21.4	1.6	2.4	0.21
aerosol, 2022	270	38	1.9	2.5	0.80
LFCM, 2022	7.5	21.1	1.6	2.4	0.21
aerosol, 2023	212	94	2.5	2.5	0.71
LFCM, 2023	7.4	20.8	1.6	2.4	0.21

and aerosol dispersity have changed significantly over the past five years.

The Table 1 data show that owing to continuing erosion of LFCM surface in room 304/3, radioactive dust is generated and penetrates in the room air. However, LFCM capability in generating aerosol particles is, evidently, reducing and entails aerosol VA drop in the room. As Fig. 4 shows, that ^{241}Am VA in room dropped at 2 times, given that its content in LFCM grows as result of β^--decay of ^{241}Pu isotope. The same picture was observed also in other controllable rooms containing LFCM (rooms 012/7, 012/15, 210/7) [15].

Mean annual contribution of ^{137}Cs in $\Sigma\beta$ three times exceeded the value of analogous contribution in the LFCM. Since during lava origination and its distribution along the Shelter object rooms, the LFCM were depleted by radiocesium owing to its evaporation, the sources of additional ^{137}Cs are ^{137}Cs-bearing aerosols from the other sources. These particles are, apparently, generated as result of degradation of surfaces, on which "condensation" cesium was earlier sorbed. The enrichment of aerosols by ^{90}Sr relatively to LFCM composition in 2020–2023 period (see Table 1) was, apparently, entailed by dust generation on bottom sediment surfaces, which produced as result of complete drying in 2019 of radioactively contaminated water accumulations in rooms, which are connected to room 304/3.

It is noteworthy that the mean annual $\Sigma\beta$ VA values in room 304/3 air are lower, than in other controllable rooms containing LFCM. For instance, in 2022 mean annual $\Sigma\beta$ VA values in room 304/3 air made 0.17 Bq/m^3, in room 012/7 air — 1.3 Bq/m^3, in room 012/15 air — 0.41 Bq/m^3, and in room 210/7 air — 0.53 Bq/ m^3 [15]. Moreover, in room 304/3, the square of LFCM surface, from which radioactive aerosols are generated, is much more larger, than in other rooms. On top of that, the LFCM in room 304/3 contain much more fuel amount, than the lavas of rooms 012/7, 012/15. Here it is necessary to address to morphology of Chornobyl lavas. Room 304/3 is filled by black LFCM, and in rooms 012/7, 012/15 and 210/7, the accumulation of brown LFCM is located.

The research has demonstrated that at microlevel the LFCM represents heterogenous solid solution, whose "dilutant" is glass-like silicate matrix with large amount of diverse inclusions, among which, uranium oxides, uranium-zirconium-oxygen phase $U_xZr_yO_2$, technogenic uranium were detected, which contain zircon ("chernobylite") and metal globules [1]. The ratios of activity and mass of matrix and micro inclusions of black and brown LFCM indicates that these LFCM were formed under various conditions. The stability of glass-like structure in relation to the impact of both external and internal factors [24] depends on thermodynamic phase equilibrium (degree of structure homogenization), which directly depends on the conditions of their formation [25, 26]. Obviously, black LFCM have undergone more long or more intensive "annealing", than brown ones, which has resulted in a higher degree of homogenization of its constituent components. In this case, black LFCM are more stable thermodynamic system and, thus, they have less capability to aerosol generation.

The AMAD of carriers of radioactive products of accident in room 304/3 reaches the values of 10–11 μm. It testifies the fact that the origination of aerosol-bearings particles of accident products occurs, mainly, due to dispersion processes [23]. At the same time, taking into ac-

Table 2. Disperse composition of radioactive ^{137}Cs and ^{241}Am-bearing aerosols in room 304/3

Sampling period	Nuclide	Volumetric activity within AD ranges, Bq/m^3					Total volumetric activity, Bq/m^3	AMAD, μm	σ
		17÷8.5 μm	8.5÷3.7 μm	3.7÷1.2 μm	1.2÷0.5 μm	<0.5 μm			
07.03–15.03	^{137}Cs	$1.2 \cdot 10^{-2}$	$5.8 \cdot 10^{-3}$	$4.9 \cdot 10^{-3}$	$1.7 \cdot 10^{-3}$3	$1.3 \cdot 10^{-3}$	$2.3 \cdot 10^{-2}$	11.0	2.6
15.03–22.03	^{137}Cs	$5.3 \cdot 10^{-3}$	$2.6 \cdot 10^{-3}$	$3.9 \cdot 10^{-3}$	$4.8 \cdot 10^{-3}$	$2.2 \cdot 10^{-3}$	$1.9 \cdot 10^{-2}$	10.2	3.8
22.03–30.03	^{137}Cs	$2.7 \cdot 10^{-3}$	$2.6 \cdot 10^{-3}$	$9.2 \cdot 10^{-3}$	$2.3 \cdot 10^{-3}$	$3.8 \cdot 10^{-3}$	$4.3 \cdot 10^{-2}$	3.3	3.4
04.07–19.07	^{137}Cs	$1.0 \cdot 10^{-2}$	$1.8 \cdot 10^{-2}$	$3.2 \cdot 10^{-2}$	$1.4 \cdot 10^{-2}$	$7.5 \cdot 10^{-3}$	$8.2 \cdot 10^{-2}$	6.0	3.0
	^{241}Am	$1.4 \cdot 10^{-4}$	$3.1 \cdot 10^{-4}$	$4.7 \cdot 10^{-4}$	$2.1 \cdot 10^{-4}$	$1.2 \cdot 10^{-4}$	$1.3 \cdot 10^{-3}$	6.1	3.0
19.07–02.08	^{137}Cs	$1.2 \cdot 10^{-2}$	$9.4 \cdot 10^{-3}$	$4.7 \cdot 10^{-2}$	$2.3 \cdot 10^{-2}$	$1.9 \cdot 10^{-2}$	$1.1 \cdot 10^{-2}$	3.7	3.2
	^{241}Am	$2.1 \cdot 10^{-4}$	$1.7 \cdot 10^{-4}$	$5.8 \cdot 10^{-4}$	$9.4 \cdot 10^{-4}$	$3.2 \cdot 10^{-4}$	$2.2 \cdot 10^{-3}$	4.7	3.1
02.08–16.08	^{137}Cs	$8.3 \cdot 10^{-3}$	$5.1 \cdot 10^{-3}$	$1.4 \cdot 10^{-2}$	$2.5 \cdot 10^{-2}$	$5.4 \cdot 10^{-3}$	$5.8 \cdot 10^{-2}$	7.6	3.2
	^{241}Am	$1.4 \cdot 10^{-4}$	$5.9 \cdot 10^{-5}$	$1.4 \cdot 10^{-4}$	$4.0 \cdot 10^{-4}$	$8.9 \cdot 10^{-5}$	$8.4 \cdot 10^{-4}$	8.5	3.3
19.09–28.09	^{137}Cs	$2.8 \cdot 10^{-2}$	$2.2 \cdot 10^{-2}$	$2.2 \cdot 10^{-2}$	$3.8 \cdot 10^{-2}$	$2.5 \cdot 10^{-2}$	0.14	9.2	4.0
	^{241}Am	$5.6 \cdot 10^{-4}$	$3.9 \cdot 10^{-4}$	$3.1 \cdot 10^{-4}$	$5.4 \cdot 10^{-4}$	$6.7 \cdot 10^{-4}$	$2.5 \cdot 10^{-3}$	9.8	4.6
11.10–24.10	^{137}Cs	$9.4 \cdot 10^{-3}$	$8.9 \cdot 10^{-3}$	$1.1 \cdot 10^{-2}$	$2.6 \cdot 10^{-3}$	$1.6 \cdot 10^{-3}$	$3.3 \cdot 10^{-2}$	8.9	2.8
24.10–07.11	^{137}Cs	$3.3 \cdot 10^{-3}$	$1.4 \cdot 10^{-3}$	$4.5 \cdot 10^{-2}$	0.54	0.13	0.71	0.9	1.8
19.11–29.11	^{137}Cs	$1.4 \cdot 10^{-2}$	$1.0 \cdot 10^{-2}$	$4.1 \cdot 10^{-2}$	$9.7 \cdot 10^{-2}$	$8.2 \cdot 10^{-2}$	0.24	2.8	3.1
	^{241}Am	$7.3 \cdot 10^{-5}$	$6.8 \cdot 10^{-5}$	$5.5 \cdot 10^{-4}$	$1.3 \cdot 10^{-3}$	$1.0 \cdot 10^{-3}$	$3.0 \cdot 10^{-3}$	2.0	2.7

count the values σ > 3, one can assume that the radionuclide composition of aerosols is, apparently, formed of more than one source, which generates aerosol particles. Unfortunately, now we do not know how LFCM degradation products get into the air. Without mechanical action and wind loads (these are the conditions of LFCM occurrence in the Shelter object), the mechanism of transfer of degraded solids into aerosols is unknown. Presumably, the aerosol generation from LFCM surface is influenced by electric charges. They undoubtedly arise upon radioactive decay.

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В. Є. Хан, О. С. Лагуненко, В. О. Краснов, О. О. Одінцов, О. К. Калиновський, В. М. Безмилов, В. О. Кашпур, П. В. Сабенін, О. А. Свирид, А. В. Ткач

Інститут проблем безпеки АЕС НАН України, вул. Кірова, 36а, Чорнобиль, 07270, Україна

Радіоактивний аерозоль у підреакторному приміщенні 304/3 об'єкта "Укриття" в умовах експлуатації нового безпечного конфайнмента

Представлено радіонуклідний склад, об'ємну активність радіонуклідів та дисперсність аерозолю, відібраного у 2019–2023 рр. у приміщенні 304/3 об'єкта "Укриття", куди внаслідок аварії протекли лавоподібні паливовмісні матеріали (ЛПВМ). Встановлено, що медіанний за активністю аеродинамічний діаметр (АМАД) носіїв радіоактивних продуктів аварії в приміщенні у 2023 р. досягає 10–11 μm. Це свідчить про їхнє диспергаційне походження. Наявність трансуранових елементів у радіонуклідному складі аерозолю у співвідношеннях, які близькі до аналогічних співвідношень ЛПВМ, показує, що аерозольні частки продовжують виникати внаслідок руйнування лави. Істотне збагачення у приміщенні 304/3 аерозолю ^{137}Cs і ^{90}Sr, що належать до складу лав, вказує на наявність ще двох джерел, що впливають на радіонуклідний склад аерозолю. Це підтверджується значеннями параметрів дисперсного складу аерозолю. За останні п'ять років об'ємна активність ^{241}Am в повітрі приміщення знизилася у 2 рази. З цього можна зробити висновок, що знижується здатність до генерації аерозолю скупчення ЛПВМ у приміщенні. При цьому зростає вплив на значення радіонуклідних співвідношень інших додаткових джерел аерозолю. За той же період спостережень сумарна активність довгоживучих бета-випромінюючих нуклідів-продуктів Чорнобильської аварії у приміщенні залишалася на одному рівні.

Ключові слова: новий безпечний конфайнмент, об'єкт "Укриття", лавоподібні паливовмісні матеріали, аерозоль, об'ємна активність, медіанний за активністю аеродинамічний діаметр, дисперсність.

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