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Radioactive Aerosols in Sub-Reactor Rooms of the Shelter Object within Conditions of the New Safe Confinement

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volumetric activity,
density of radionuclide deposition,
activity median aerodynamic diameter,
dispersity

Radionuclide composition, volumetric activities, radionuclide deposition densities and dispersity of aerosols sampled in 2019–2022 in rooms 012/7, 012/15, 210/7, and in 2017–2022 in room 304/3 of the Shelter object are presented, where lava-like fuel-containing materials (LFCM) penetrated after the accident. It was stated that activity median aerodynamic diameter of carriers of radioactive products of accident in rooms 012/7, 012/15 and 210/7 increased significantly over the past 8–10 years and reached 10 μm. It evidences their dispersion origin. The availability of transuranium elements in radionuclide composition of aerosols in all rooms in the ratios that are close to similar ones in the LFCM shows that the aerosols arose as a result of lava degradation. Substantial enrichment of aerosols in rooms 304/3 by ^{137}Cs as well as ^{137}Cs and ^{90}Sr – in rooms 012/7, 012/15, and 210/7 relatively to lava compositions indicate the presence of two sources more influencing radionuclide composition of aerosols. It is confirmed by the values of parameters of disperse composition of aerosols.

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 lava produced several horizontal and vertical flows and penetrated, in particular, into the lower rooms of destroyed building of power Unit 4 (fig. 1). The "large vertical flow" reached sub-reactor room (mark 9.0 m), room 210/7 of Steam Distribution Corridor (6.0 m), and rooms 012/15 and 012/7 of Pressure Suppression Pool (3.0 and 0.0 m, accordingly) [1].

Stiffened lavas represent nuclear, radiation and radioecological hazards. According to expert estimates, the LFCM contain from 60 to 110 tons of irradiated fire, i.e. 32–58 % of initial fuel loaded into the reactor [2]. This behavior is not sufficiently studied and cannot lead to a good forecast. The evidence of LFCM degradation, which was proven in work [3], is radioactive aerosol and radionuclide depositions in the rooms, where lava accumulations were localized.

The researches of 2010–2014 in sub-reactor rooms of the Shelter object [3–5] showed that due to lava degradation, spontaneous transition of degradation products in aerosol state is observed. In addition, the amount of radioactive aerosol, which is a product of LFCM degradation, depends on the lava type.

On November 29, 2016, the Arch of the 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

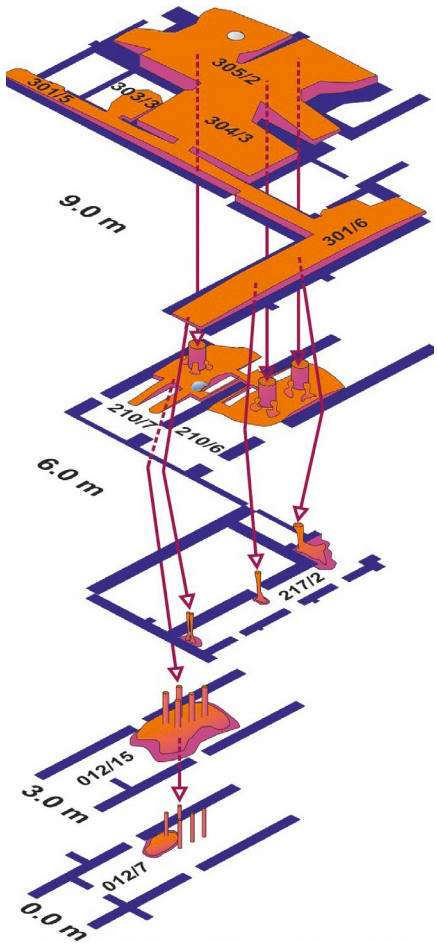

Fig. 1. LFCM accumulations and lava-flow direction

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 [6–8] demonstrated that spontaneous transition of LFCM degradation products in aerosol state has decreased. Because of the above, the values of volumetric activities of radionuclides turned out to be lower as compared to the results of 2010–2014 investigations.

In 2019–2022, the work to research the behavior of radioactive aerosols in sub-reactor rooms of the Shelter object was continued. The goal of work is to monitor the dynamics of radioaerosol situation in the rooms, where LFCM are localized, within the conditions of commissioning and operation of "New Safe Confinement – Shelter object" complex.

Data on rooms and LFCM

In accordance with the Technological Regulations of the Shelter object, rooms 012/7, 012/15, 210/7, and 304/5 are considered as "unattended" rooms. No works were performed during investigation period in these and adjacent rooms.

Room 012/7 is located on the lowest height mark (0.0 m) of destroyed reactor building (Unit B). It is a part of pressure suppression pool system (PSP-1) of the Chornobyl NPP Unit 4. Its size is 6×72 m, and the height is about 3 m. From the east and west, adjacent rooms 012/6 and 012/8 are located, which are similar in their configuration and building materials. Between them, there are passages and technological apertures, through which the access to different points and air exchange are possible. Close attention is drawn to room 012/7 after the 1986 accident is due to the fact that this room appeared to be the lowest point reached by "large vertical flow" of lava; it covered a distance of approximately 10 m along the vertical from lava formation area – room 305/2 [9, 10]. The lower rooms of the Shelter object are the sites of origination of air flows carrying radioactive aerosols to free atmosphere through the technological apertures and untight sites at the upper levels of the Shelter object and through the Bypass system and Ventilation Stack. The lava accumulation in room 012/7 has elliptical shape in the plan with the longitudinal axis length of ~3 m and latitudinal axis length of ~2 m [10]. The largest height of this accumulation in the dome is ~0.8 m. The lower part of accumulation (~0.3 m) is covered with concrete that came here in 1986 in the course of erection of cascade wall of the Shelter object. In the experiments on dust suppression and LFCM localization in March 2000, southern part of accumulation was coated with a layer of EKOR-25MM organosilicon compound 1–3-cm thick [9]. Therefore, fully open horizontal projection of LFCMs is about 2.5 m^2. The lifetime of coating is expected to be about 100 years. According to expert estimates, this accumulation, called as PSP-1"heap", contains 1.0 ± 0.5 t of uranium [9].

Room 012/15 is located at height mark 3.0 m and is a part of pressure suppression pool system (PSP-2). The lava flew here along the steam-discharging pipes from room 210/7. The LFCM accumulation called as PSP-2 "heap" [9] is located between the axes 47^{+1000} – 48_{-1000} and rows I_{-3000} – J. Its northern edge in the course of the Shelter object erection was covered with the concrete and formed around 0.7-m layer over the floor. The surface of accumulation is covered by around 0.1-m thick pumice-like layer. The LFCMs are also present in five, 0.28 m in diameter, steam-discharging pipes, through which the lava penetrated into room 012/15.

According to [10], the volume of PSP-2 "heap" is $23 \pm 6\ m^3$. Having brown ceramic density of $2.14 \pm 0.34\ t\ m^{-3}$ and uranium content of 9–13 %, the total uranium amount in the "heap" is close to 3 t.

Room 210/7 and adjacent rooms 210/5, 210/6, 210/8, and 208/12 are the parts of Steam Distribution Corridor (SDC) system. The SDC area is about 630 m^2. The floor is located on mark 6.0 m, and the ceiling, on 8.0-m mark. Until 2018, there was a water accumulation in the southern part of SDC room, which washed out the soles of southern slopes of the lava. Room 208/12, in which sampling equipment was installed, is always dry. The LFCM came into rooms 210/6 and 210/7 through third and fourth steam-discharging valves from southeastern and southwestern parts of room 305/2. Through the other valves, "fresh" concrete flew to these room in the course of the Shelter object erection. The total LFCM volume in SDC is about 58 m^3 [10]. The uranium amount is 12 ± 6 t.

Rooms 305/2 and 304/3 played the key role in forming and spilling lava flows (see fig. 1). The reactor base on which the reactor cavity rests is located in room 305/2. Before the accident, the upper height mark of both rooms corresponded to +9 m, and the lower mark, to +12 m. After reactor explosion, due to catastrophic destruction, Central Hall, rooms 504/2, 305/2, 304/3, and rooms 303/3, 301/5 and 301/6, became a common space for air exchange and aerosol transfer.

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 [9–12]. According to the calculations [9], there are about 80 t of uranium in room 305/2 and about 6 t of uranium in room 304/3.

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.

The filters were placed as close as possible to LFCM accumulations. For this, filtering funnel was installed in room 012/7 directly on the sampler located at 1-m distance from southeastern edge of LFCM heap. In room 012/15, filtering funnel was connected to the sampler using polypropylene pipe with 2-cm internal diameter and 2-m length. In addition, the funnel with filters were located at 5-m distance from northern end of LFCM accumulation. In room 210/7, filtering funnel was connected to the sampler using the same pipe, which was introduced through the door opening into the area of southern part of LFCM accumulation. 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.

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.

The monitoring of radionuclide deposition density was made in rooms 012/7 and 210/7. To define the density of radionuclide deposition on the underlying surface, plastic cuvette was used, at whose bottom the filter paper was laid.

Near the sampling places in rooms 012/7, 012/15, and 210/7, temperature and relative humidity were measured using Elitech RC4HA/C register and TH mini hygrothermometer with remote sensor of temperature-humidity. In room 304/3, temperature and humidity were measured in the borehole B-12-76.

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 with 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 and radionuclide deposition samples, after radiochemical separation, ^{90}Sr activity was determined by beta-radiometry, and the activities of ^{238}Pu, $^{239+240}Pu$ and ^{241}Am – by alpha-spectrometry measurements.

Concentration of aerosols carrying the accident products

Room 012/7. Over 2019–2022 period, 35 aerosol samples and 10 samples of radionuclide deposition on underlying surface, were taken in room. The results of 2017–2018 survey period are presented in work [6].

As fig. 2 shows, in 2019–2022 volumetric activity (VA) of ∑β was varying within the range of 0.15–7.2 Bq/m^3, and ^{241}Am VA was varying within the range of $3.9 \cdot 10^{-4}$ – $4.3 \cdot 10^{-2}$ Bq/m^3. At the same time, trend line demonstrates ^{241}Am VA drop in room air in contrast to ∑β VA.

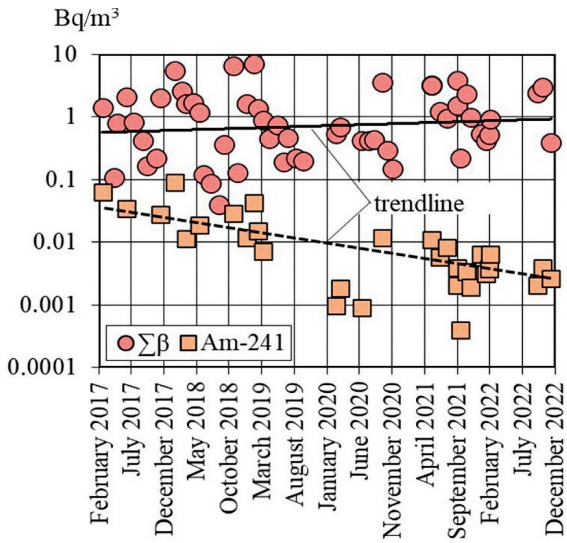

Fig. 2. VA of aerosols carrying ∑β and ^{241}Am in room 012/7 in 2017–2022

In line with radiometry and gamma-spectrometry measurements of samples, ^{137}Cs/∑β ratios in aerosols during 2019–2022 was fluctuating from 0.16 to 0.74 under mean value 0.4. In addition, correlation dependence of ratio over time was not established. From obtained mean value of ^{137}Cs/∑β follows that ^{137}Cs input in ∑β made 40 %.

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.

As fig. 3 shows, 2019–2022, ∑β deposition density was varying within the range of 23–1,500 Bq/(m^2 · day). Deposition density of ^{241}Am was varying within the range of 0.093–15 Bq/(m^2 · day). Trend lines indicate a drop in deposition density of ∑β and americium in this room.

During 2019–2022, ratios of ^{137}Cs/∑β in deposition density was changing from 0.1 to 0.37 under mean value 0.23. Correlation dependence of ratio over time was not established. From obtained mean value of ^{137}Cs/∑β it follows that ^{137}Cs input in ∑β radionuclide deposition made 23 %.

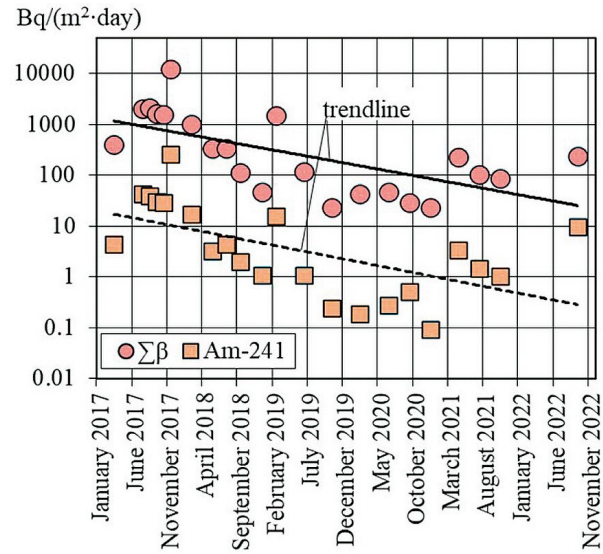

Fig. 3. Deposition density of ∑β and ^{241}Am in room 012/7 in 2017–2022

Aerosol and radionuclide deposition samples were analyzed by radiochemical method. The analysis results of most active in aerosol ∑β sampled on September 16, 2020, are shown below. The data shows that the ratio of ^{137}Cs/^{241}Am activities is equal to 62, at the same time, relative standard deviation from mean value of annual activities ratio made 43 %. The value of ^{137}Cs/∑β activity ratio made 0.21 under relative standard deviation from mean annual value of this ratio made 47 %.

Content of radionuclides in aerosol in room 012/7, Bq/m^3

^{90}Sr	1.4
^{137}Cs	0.74
^{154}Eu	$7.9 \cdot 10^{-4}$
^{238}Pu	$2.3 \cdot 10^{-3}$
$^{239+240}$Pu	$5.3 \cdot 10^{-3}$
^{241}Am	$1.2 \cdot 10^{-2}$
^{244}Cm	$2.2 \cdot 10^{-4}$
∑β	3.6

The analysis results samples are also shown with highest in 2020 ∑β deposition density obtained when exposing deposition collector from February 25 to July 11.

Deposition density of radionuclides in room 012/7, Bq/(m^2 · day)

^{90}Sr	18
^{137}Cs	11
^{154}Eu	$6.2 \cdot 10^{-2}$
^{238}Pu	$5.9 \cdot 10^{-2}$
$^{239+240}$Pu	0.14
^{241}Am	0.28
^{244}Cm	$4.7 \cdot 10^{-3}$
∑β	47

Table 1. Radionuclide ratios in aerosols, depositions and LFCM in room 012/7

Object, year	$^{137}Cs/^{241}Am$	$^{90}Sr/^{241}Am$	$^{241}Am/^{154}Eu$	$^{241}Am/^{239+240}Pu$	$^{239+240}Pu/^{238}Pu$	$^{137}Cs/\Sigma\beta$
aerosol, 2019	66	51	11	4.8	2.5	0.38
deposition, 2019	17	41	9.1	1.9	2.2	0.17
LFCM, 2019	8.9	15.8	9.6	2.0	2.3	0.22
aerosol, 2020	130	150	16	3.9	2.1	0.32
deposition, 2020	37	88	8.1	2.4	2.4	0.17
LFCM, 2020	8.8	15.6	10.4	2.0	2.4	0.22
aerosol, 2021	190	160	–	4.2	2.2	0.39
deposition, 2021	17	29	9.4	2.5	2.2	0.23
LFCM, 2021	8.7	15.4	11.2	2.0	2.4	0.22
aerosol, 2022	150	230	–	4.0	2.8	0.57
deposition, 2022	9.4	16	14	2.9	2.2	0.37
LFCM, 2022	8.6	15.2	12.1	2.0	2.4	0.22

In table 1, mean annual values are shown of ratios of nuclide activities in aerosol samples and density of radionuclide deposition sampled during 2019–2022, and analogous radionuclide ratios in LFCM of room 012/7. The data on radionuclide composition in LFCM within sub-reactor rooms 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 2019–2022 period, the ratio of radionuclides in aerosols and LFCM of room 012/7 differed substantially due to aerosol enrichment by ^{137}Cs and ^{90}Sr relatively to ^{241}Am, which in 2010–2011 was not observed [3]. At the same time, over time the aerosol enrichment by cesium and strontium is growing. One should also note that americium activity in the aerosols relatively to activities plutonium isotopes has also doubled as compared to their ratio in the lava [6].

In contrast to aerosols, the enrichment of radionuclide deposition by ^{137}Cs and ^{90}Sr being observed in 2019–2021, in 2022 dropped to the level of measurement uncertainty. The ratios of activities of semi-volatile transuranium elements in depositions 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 the both aerosol and radionuclide depositions remained close to value of analogous ratios in LFCM of room 012/7 during 2019–2022, that was also earlier observed [6].

The table 1 data show that owing to continuing erosion of LFCM surface in room 012/7, 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 figs. 2, 3 show, ^{241}Am content in aerosols and radionuclide depositions decreases, given that its content in LFCM grows as result of β^--decay of ^{241}Pu isotope. Therefore, the influence of other radioactive aerosol sources on radionuclide ratio values in room 012/7 aerosol is growing. Such additional sources of radioactive aerosols can, particularly, be the surfaces of bottom sediments after drying of radioactively contaminated water (RCW), and building structures' surfaces, on which so called "condensation cesium" earlier absorbed. The same fact is testified by below results of identification of aerosol particles dispersity.

Room 012/15. Over 2019–2022 period, 29 aerosol samples were taken in the room. Survey results over 2017–2018 period are presented in work [7].

As fig. 4 shows, in 2019–2022, $\Sigma\beta$ VA was varying within the range of 0.032–4.1 Bq/m^3, and ^{241}Am VA was varying within the range of $9.8 \cdot 10^{-5}$ – $1.0 \cdot 10^{-2}$ Bq/m^3. At the same time, trend lines show a drop in volumetric activities of $\Sigma\beta$ and ^{241}Am in room air.

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

The ratio of $^{137}Cs/\Sigma\beta$ in 2019–2022 was fluctuating from 0.13 to 0.98 under mean value 0.49. No correlation dependence of this ratio over time was found. From ob-

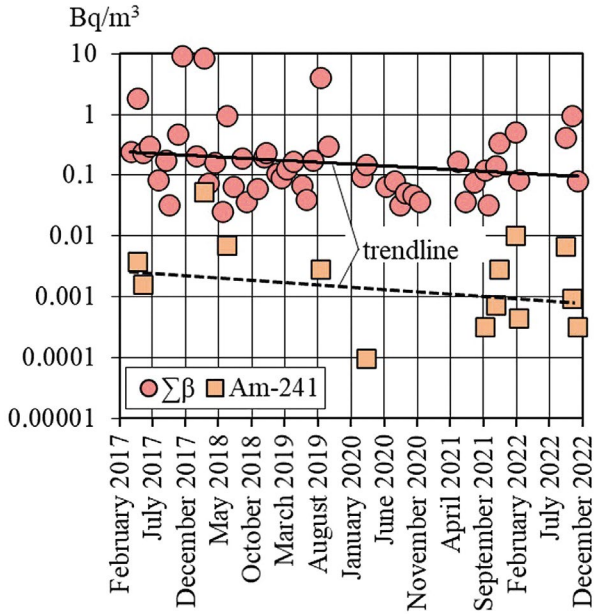

Fig. 4. VA of aerosols carrying Σβ and ^{241}Am in room 012/15 in 2017–2022

tained mean value ^{137}Cs/Σβ follows, that ^{137}Cs input in Σβ made 49 %.

The aerosol samples were analyzed by radiochemical method. Below the analysis results are shown of most active on Σβ aerosol samples, which were taken on September 14, 2022.

Content of radionuclides in aerosols in room 012/15, Bq/m³

^{90}Sr.....$7.9 \cdot 10^{-2}$
^{137}Cs.....0.26
^{154}Eu.....$7.22 \cdot 10^{-4}$
^{238}Pu.....$6.3 \cdot 10^{-4}$
$^{239+240}$Pu.....$1.3 \cdot 10^{-3}$
^{241}Am.....$6.8 \cdot 10^{-3}$
^{244}Cm.....$1.1 \cdot 10^{-4}$
Σβ.....0.42

In table 2, mean annual values of nuclide activity ratios are shown in aerosol samples taken during 2019–2022, and ratios in LFCM localized in room 012/15.

As table 2 demonstrates, during 2019–2022, plutonium isotope ratios in aerosols were close to analogous ratio in LFCM localized in the room. It indicates the fact, that in room 012/15, due to erosion of LFCM surfaces, generation of radioactive aerosols coming in air space is in progress. The divergences in values are related, as already noted above, to measurement uncertainties and inhomogeneity in LFCM composition. As the table demonstrates, the value of ^{137}Cs/^{241}Am ratio had significantly dropped over 2019–2022 period. It is, apparently, due to less air-exchange in room 012/15 and adjacent rooms contaminated by condensation radiocesium, but in which the LFCM is not available.

The value of ratios of ^{241}Am/$^{239+240}$Pu activities in aerosols was higher as compared to analogous ratio in the lava in 1.5–2.3 times. The comparison of ^{137}Cs/Σβ and ^{137}Cs/^{241}Am ratio in aerosols with radionuclide composition of LFCM demonstrates that during 2019–2022, the aerosols in room were enriched by ^{137}Cs relatively to composition of lavas, which also was earlier observed [4, 7]. On top of that, attention should be drawn to ^{90}Sr activity in its ratio to ^{241}Am radionuclide.

Room 210/7. Over 2019–2022 period, 30 aerosol samples and 11 radionuclide deposition samples were taken in the room. Survey results over previous 2017–2018 period are presented in the work [8].

In fig. 5, dynamics of Σβ and ^{241}Am VA in room 210/7 over period 2017–2022 is presented. As the figure shows, in 2019–2022, Σβ VA was varying within the range of 0.044–1.5 Bq/m³ and ^{241}Am VA was varying within the range of $3.2 \cdot 10^{-4}$ – $1.9 \cdot 10^{-2}$ Bq/m³. At the same time, trend lines show a drop in Σβ and ^{241}Am volumetric activities in room air. In most cases, increased aerosol VA was

Table 2. Radionuclide ratios in aerosols and LFCMs in room 012/15

Object, year	^{137}Cs/^{241}Am	^{90}Sr/^{241}Am	^{241}Am/$^{239+240}$Pu	$^{239+240}$Pu/^{238}Pu	^{137}Cs/Σβ
aerosol, 2019	1,380	16	2.7	2.5	0.98
LFCM, 2019	9.6	21.7	1.7	2.3	0.23
aerosol, 2020	1,100	220	3.0	3.0	0.72
LFCM, 2020	9.5	21.4	1.7	2.4	0.23
aerosol, 2021	130	52	3.1	2.1	0.55
LFCM, 2021	9.4	21.1	1.7	2.4	0.23
aerosol, 2022	270	28	3.9	2.1	0.58
LFCM, 2022	9.3	20.8	1.7	2.4	0.23

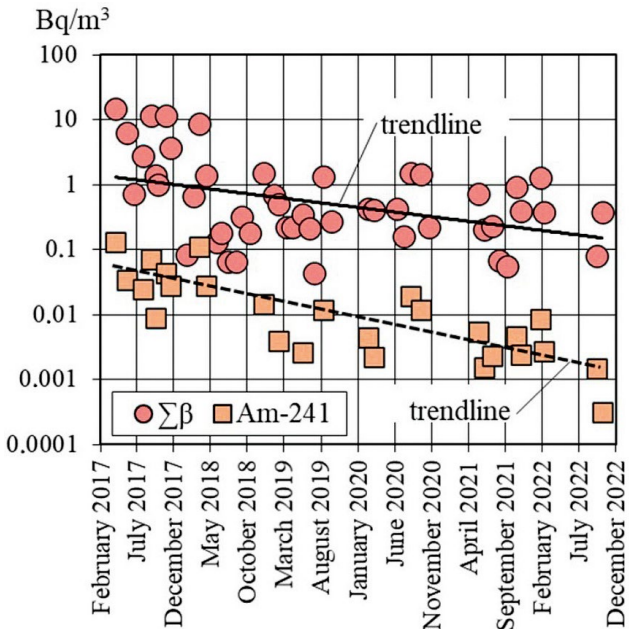

Fig. 5. VA of aerosols carrying Σβ and ^{241}Am in room 210/7 in 2017–2022

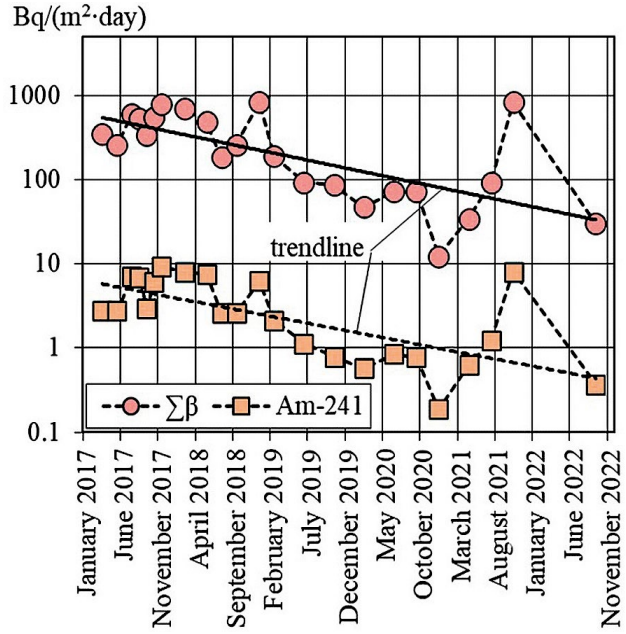

Fig. 6. Deposition density of Σβ and ^{241}Am in room 210/7 in 2017–2022

observed under relatively low air moisture in the room, and decreased VA coincided in time with high moisture value.

The ratio ^{137}Cs/Σβ in aerosols of room 210/7 during 2019–2022 was fluctuating from 0.22 to 0.78 under mean value 0.54. Correlation dependence of this ratio over time was not revealed. From obtained mean value ^{137}Cs/Σβ follows, that ^{137}Cs input in Σβ made 54 %.

In fig. 6, dynamics of Σβ and ^{241}Am deposition density in room 210/7 is presented over 2017–2022 period. In 2019–2022, Σβ was varying within the range of 12–830 Bq/(m^2 · day). Deposition density of ^{241}Am was varying within the range of 0.19–13 Bq/(m^2 · day). Trend lines show a drop in deposition density of Σβ and americium in the room. As the figure shows, the graphs with radionuclide deposition density are similar in their symbasis, which makes possible to assert that the radionuclide are located on the same particles. Such particles are, apparently, LFCM fragments localized in room 210/7.

During 2019–2022, the ratio ^{137}Cs/Σβ in radionuclide deposition density was changing from 0.29 to 0.59 under mean value 0.41. Correlation dependence of this ratio over time was not established. From obtained mean value of ^{137}Cs/Σβ follows, that ^{137}Cs input in Σβ radionuclide deposition made 41 %.

Aerosol samples and radionuclide deposition were analyzed by radiochemical method. The analysis results of most active on Σβ aerosol sampled on September 15, 2020, are shown below.

Content of radionuclides in room 210/7 aerosols, Bq/m^3

^{90}Sr.....0.36
^{137}Cs.....0.74
^{154}Eu.....$7.9 \cdot 10^{-4}$
^{238}Pu.....$3.2 \cdot 10^{-3}$
$^{239+240}$Pu.....$7.2 \cdot 10^{-3}$
^{241}Am.....$1.2 \cdot 10^{-2}$
^{244}Cm.....$1.7 \cdot 10^{-4}$
Σβ.....1.5

The analysis results of samples with highest deposition density of Σβ obtained when exposing deposition collector from June 11 to September 15, 2020, are shown as well.

Deposition density of radionuclides in room 210/7, Bq/(m^2 · day)

^{90}Sr.....24
^{137}Cs.....26
^{154}Eu.....$5.6 \cdot 10^{-2}$
^{238}Pu.....0.15
$^{239+240}$Pu.....0.36
^{241}Am.....0.78
^{244}Cm.....$1.2 \cdot 10^{-2}$
Σβ.....73

Table 3 shows mean values of ratio of nuclide activities in aerosol samples and depositions sampled during 2019–2022, and ratio in LFCM located in room 210/7.

Table 3. Radionuclide ratios in aerosols, depositions and LFCMs in room 210/7

Object, year	$^{137}Cs/^{241}Am$	$^{90}Sr/^{241}Am$	$^{241}Am/^{154}Eu$	$^{241}Am/^{239+240}Pu$	$^{239+240}Pu/^{238}Pu$	$^{137}Cs/\Sigma\beta$
aerosol, 2019	72	26	11	1.8	2.5	0.57
deposition, 2019	35	29	12	1.9	2.4	0.36
LFCM, 2019	8.8	17.6	9.9	1.6	2.3	0.20
aerosol, 2020	47	25	16	1.8	2.4	0.47
deposition, 2020	30	26	12	1.9	2.3	0.37
LFCM, 2020	8.7	17.3	10.8	1.6	2.4	0.20
aerosol, 2021	83	32	–	1.7	2.6	0.54
deposition, 2021	34	22	14	2.6	2.7	0.43
LFCM, 2021	8.6	17.1	11.7	1.6	2.4	0.20
aerosol, 2022	360	56	8.5	2.2	2.4	0.64
deposition, 2022	48	17	14	3.2	2.8	0.59
LFCM, 2022	8.5	16.8	12.7	1.6	2.4	0.20

Data analysis given in table 3 shows that in 2019–2022 period, aerosol and radionuclide depositions were enriched by ^{137}Cs, as compared to LFCM composition, that was also earlier observed [5, 8]. Moreover, in the aerosols, increased ^{90}Sr activities in relation to ^{241}Am activities takes place.

Before 2021, the ratio of transuranium element activities in aerosols and depositions corresponded to analogous ratios of room 210/7 LFCM. The discrepancies, as noted above, are associated with heterogeneity of LFCM composition, and measurement uncertainty. In 2021–2022 period, a growth is observed of ^{241}Am input in alpha-activity of transuranium elements in radionuclide depositions.

As figs. 5, 6 show, volumetric activity of nuclides drops, that is, evidently, due by lessening of LFCM ability to generate aerosol particles. Therefore, the influence on the values of radionuclide ratios of room 210/7 aerosols grows for the other radioactive aerosol sources. Such a conclusion is confirmed by below results of identified disperse composition of aerosol particles.

Room 304/3. Over 2017–2022 period, 44 aerosol samples were taken in the room.

As fig. 7 shows, in 2017–2018, Σβ VA was varying within the range of 0.03–3.9 Bq/m^3, and ^{241}Am VA was varying within the range of $7.1 \cdot 10^{-3}$ – $3.8 \cdot 10^{-2}$ Bq/m^3. Over the next 2019–2022 period, Σβ VA was varying within the range of 0.05–0.76 Bq/m^3, and VA ^{241}Am was varying within the range of $3.2 \cdot 10^{-4}$ – $5.6 \cdot 10^{-3}$ Bq/m^3. At the same time, trend lines indicate a drop in volumetric activities of Σβ and ^{241}Am in room air. In most cases,

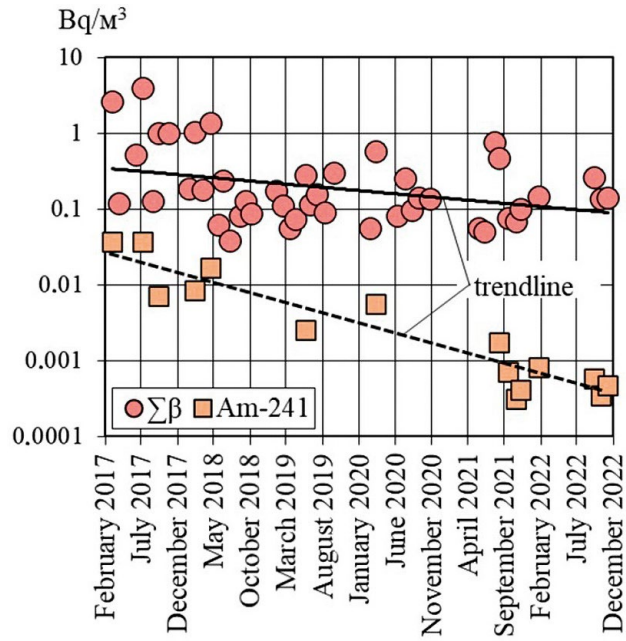

Fig. 7. VA of aerosols carrying Σβ and ^{241}Am in room 304/3 in 2017–2022

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

The ratio of $^{137}Cs/\Sigma\beta$ in 2017–2018 was fluctuating from 0.3 to 0.89 under mean value 0.6. In the next 2019–2022 period, $^{137}Cs/\Sigma\beta$ value was varying from 0.17 to 0.88 under mean value 0.61. From obtained mean value follows, that ^{137}Cs input in Σβ made not less than 60 %. No correlative dependence of the ratio over time was detected.

The aerosol samples were analyzed by radiochemical method. Below the analysis results are shown of most active on Σβ aerosol samples taken on November 18, 2021.

Content of radionuclides in room 304/3 aerosols, Bq/m³

^{90}Sr.....$1.1 \cdot 10^{-2}$
^{137}Cs.....$7.7 \cdot 10^{-2}$
^{154}Eu.....$1.2 \cdot 10^{-4}$
^{238}Pu.....$2.8 \cdot 10^{-4}$
$^{239+240}$Pu.....$4.1 \cdot 10^{-4}$
^{241}Am.....$1.2 \cdot 10^{-5}$
^{244}Cm.....$1.2 \cdot 10^{-5}$
Σβ.....0.10

In table 4, mean values of nuclide activity ratios in aerosol samples taken during 2017–2022, and ratios in LFCM localized in room 304/3, are given.

When comparing the ratios of ^{137}Cs/Σβ and ^{137}Cs/^{241}Am in room 304/3 LFCM with radionuclide composition of aerosol, we see that during 2017–2022, aerosol was enriched by ^{137}Cs relatively to lava composition (see table 4). Starting from 2020, growth in ^{90}Sr activities in relation to ^{241}Am activities has been observed. The table also shows that in contrast to the cesium ratios, transuranium element activities in room aerosol during 2017–2022 were remaining identical to the values of analogous ratios in the LFCM. The discrepancies in values can be associated, as mentioned above, with measurement uncertainty and heterogeneity of lava. Thus, due to LFCM surface erosion, generation of radioactive dust occurs, which penetrates in the air in room 304/3.

Dispersity of radioactive aerosols

The procedure to determine aerosol size is described in [13]. Assuming a priori lognormal distribution of aerosol particle size, we calculated activity median aerodynamic diameter (AMAD) and geometric standard deviation (σ).

Room 012/7. In 2019–2022 period, AMAD value of ^{137}Cs-bearing particles was varying within the range of 10.5–11.9 μm, and AMAD of ^{241}Am-bearing particles were 10.7–10.9 μm. Within the measurement errors, the data on nuclide activities of AMAD values can be considered as matching ones, and at the same time, significantly different from previously observed values. In 2010–2011, Σβ carriers in room air were aerosol particles with AMAD 2–8 μm [3]. In the same work it was also shown that the particles containing ^{241}Am, based on presented results of radiochemical studies, should be considered as products of LFCM surface degradation, which are localized in room 012/7. 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, which was not earlier observed. Taking into account the values σ>3, one can assume that the aerosols in room 012/7 in 2019–2022 period were produced from different sources unlike to 2010–2011 aerosols.

Room 012/5. In 2019–2022 period, the AMAD value of ^{137}Cs-bearing particles was varying within the range of 7.2–10.1 μm, and AMAD of ^{241}Am-bearing particles were 6.5–10.8 μm. Within the measurement errors,

Table 4. 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/^{238}Pu	^{137}Cs/Σβ
aerosol, 2017	60	20	1.7	2.1	0.55
LFCM, 2017	8.0	22.5	1.6	2.3	0.21
aerosol, 2018	41	21	1.6	2.6	0.49
LFCM, 2018	7.9	22.3	1.6	2.3	0.21
aerosol, 2019	68	21	2.0	2.5	0.62
LFCM, 2019	7.8	22.0	1.6	2.3	0.21
aerosol, 2020	54	26	1.7	2.9	0.51
LFCM, 2020	7.7	21.7	1.6	2.4	0.21
aerosol, 2021	130	40	2.0	2.3	0.64
LFCM, 2021	7.6	21.4	1.6	2.4	0.21
aerosol, 2022	270	38	1.9	2.5	0.77
LFCM, 2022	7.5	21.1	1.6	2.4	0.21

the data on nuclide activities of AMAD values can be considered as matching ones and significantly different from previously observed values. In 2014, most often Σβ carriers in room air were the aerosol particles with AMAD from 0.8 to 2 μm [4]. The value of $^{137}Cs/^{241}Am$ ratios has shown that at all impactor cascades, ^{137}Cs was present in excess amount relatively to its content in the lava, which was also earlier observed. The magnitude of value σ for majority of samples was more than 3.

Room 210/7. In 2019–2022 period, the AMAD value ^{137}Cs-bearing particles was varying within the range of 8.9–11.6 μm, and AMAD of ^{241}Am-bearing particle were10–11.7 μm. Within the measurement errors, the data on nuclide activities of AMAD values can be considered as matching ones and significantly different from previously observed values. In 2013, most often Σβ carriers in room air were the aerosol particles with AMAD 1–7 μm. [5]. The value of $^{137}Cs/^{241}Am$ ratios has shown that at all impactor cascades, ^{137}Cs was present in excess amount relatively to its content in the lava, which was also earlier observed. The magnitude of σ value in most cases was more than 3.

Discussion

The above materials demonstrate that in rooms 012/7, 012/15, and 210/7, volumetric activities, radionuclide composition and aerosol dispersity have changed significantly over the past ten years.

Before the last days of 2022, maximum and mean Σβ and ^{241}Am VA dropped in room 012/7 air an order of value as compared to the results of previous surveys [3]. At the same time, ^{137}Cs input in Σβ has tripled and significantly exceeded the value of analogous input in 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 the aerosols of radiocesium-bearing particles 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 in room 012/7 air by ^{90}Sr relatively to LFCM composition in 2019–2022 period (see Content of radionuclides in aerosol in room 012/7, Bq/m^3) was, apparently, entailed by dust generation on bottom sediment surfaces, which produced as result of complete drying in the early days of 2019 of radioactively contaminated water accumulation in room 009/4, which is connected to room 012/7. Regular surveys have shown that the value of ratios of $^{90}Sr/^{241}Am$ activities in this accumulation in 2018 made around 170 [14], that is much more of analogous ratio in room 012/7 LFCM. At the same time, the value of ratios of $^{241}Am/^{239+240}Pu$ activities in water accumulation made not less than an order of value, which could serve as a reason for increased ratio of americium to plutonium activities in room 012/7 air in 2019–2022. Thus, bottom sediments in room 009/4 can be seen as a source more in forming aerosol situation in room 012/7, which influence, primarily, the dynamics of Σβ VA (see fig. 2). The availability of more than one source, which generates aerosols, is also confirmed by the results of analyzed aerosol dispersity in room 012/7.

Before 2019, volumetric activity of radionuclides in rooms 012/15 and 210/7 in most cases was lower, than in room 012/7, that was also earlier observed [3–8]. So, ten years ago in rooms 012/15 and 210/7, Σβ value seldom exceeded 10 Bq/m^3, and in room 012/7, almost always this value was higher and even reached 600 Bq/m^3. Such a difference was explained by less stability of lava in room 012/7 as compared to the lavas in rooms 012/15 and 210/7 [4–5].

In 2019–2022, the enrichment of aerosols in rooms 012/15, 210/7, and 304/3 air by ^{137}Cs and ^{90}Sr relatively to LFCM composition in these rooms and in room 012/7 was provoked by the presence of carrier radiocesium-bearing aerosols and dust rise from the surface of bottom sediments produced as result of RCW drying.

Figs. 2, 4, 5, and 7 show, that over the past five years, ^{241}Am VA in all four controllable rooms dropped at 10–100 times. From this it can be concluded that the capability of LFCM accumulations to generate the aerosols in these rooms is reducing. At the same time, the influence is growing of two mentioned above additional aerosol sources at the values of radionuclide ratios in the aerosols. Over the same period, Σβ VA in three rooms dropped by no more than an order of value, and in room 012/7, it remained at the same level.

The AMAD of carriers of radioactive products of accident in rooms 012/7, 012/15, and 210/7 significantly grew over the past 8–10 years and reaches the values of 10–11 μm. It testifies the fact that the origination of aerosol particles-carriers of accident products occurs, mainly, due to dispersion processes [15]. At the same time, the values of dispersity parameters of aerosols also indicate that the radionuclide composition of aerosols is, apparently, formed of more than one source, which generates aerosol particles.

The results of researches of radionuclide depositions in rooms 012/7 and 210/7 show that they produced, mainly, owing to particle-fragments of LFCM surfaces

degradation. At the same time, the rate of enrichment of radionuclide deposition on ^{137}Cs and ^{90}Sr relatively to LFCM composition, is much lower than in the aerosol. 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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Радіоактивний аерозоль у підреакторних приміщеннях об'єкта «Укриття» в умовах нового безпечного конфайнмента

Представлено радіонуклідний склад, об'ємні активності, щільність випадінь радіонуклідів та дисперсність аерозолів, відібраних у 2019–2022 рр. у приміщеннях 012/7, 012/15, 210/7, а також у 2017–2022 рр. у приміщенні 304/3 об'єкта «Укриття», куди після аварії протікали лавоподібні паливовмісні матеріали (ЛПВМ). Встановлено, що медіанній за активністю аеродинамічний діаметр (АМАД) носіїв радіоактивних продуктів аварії в приміщеннях 012/7, 012/15, 210/7 суттєво збільшився за останні 8–10 років і досягає 10 μm. Це свідчить про їхнє диспергаційне походження. Наявність трансуранових елементів у радіонуклідному складі аерозолів у всіх приміщеннях у співвідношеннях, які близькі до аналогічних співвідношень ЛПВМ, показує, що аерозолі продовжують виникати внаслідок руйнування лави. Істотне збагачення аерозолю у приміщенні 304/3 ^{137}Cs і ^{137}Cs та ^{90}Sr у приміщеннях 012/7, 012/15, 210/7 щодо складу лав вказує на наявність ще двох джерел, що впливають на радіонуклідний склад аерозолів. Це підтверджується значеннями параметрів дисперсного складу аерозолів. За останні п'ять років об'ємна активність ^{241}Am в повітрі чотирьох приміщень знизилася у 10–100 разів. З цього можна зробити висновок, що знижується здатність до генерації аерозолю скупчень ЛПВМ у цих приміщеннях. 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 трьох приміщеннях зменшилася не більше ніж на порядок величини, а в приміщенні 012/7 залишалася на одному рівні.

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

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