Reactor Pond Walls Deposit Investigation during Dismantling MR Reactor – 15028

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ABSTRACT

Specialists of the NRC "Kurchatov Institute" now carry out the dismantling of the multiloop research MR reactor. During this works the water level was decreased in the MR reactor pond on 4 meter. Dependence of exposure dose rate above the pond from deposits on the pond walls was confirmed. Detailed research of deposits on MR reactor pond walls was made, using α -, β -, γ - spectrometric methods. Correlation coefficients between concentration Pu and U radionuclides and concentration ¹³⁷Cs and ²⁴¹Am was calculated. The surface activity of the main dose-forming radionuclide ¹³⁷Cs on the walls of the pond was measured.

INTRODUCTION

The dismantling work of the multiloop research MR reactor was carried out since 2011. These works are performed according to the project of a decommissioning approved by government authorities in frames of the Federal Target Program "Nuclear and radiation safety for 2008 and for the period till 2015". According to the project of decommissioning MR reactor, dismantling work of the equipment in process compartment and MR reactor component were carried out in 2013-2014. First after reactor shutdown working channels and channels of control and protection system (fig. 1) was evacuated from active zone of reactor.



Fig.1 Working channels and channels of control and protection system of reactor MR.

Water level in reactor pond was decreased on 2 meters, that allows dismounting truck of control and protection system and movable fuel assembly (fig. 2).



Fig.2 Reactor MR pond after dismantling truck of control and protection system and movable fuel assembly.

Next decreasing water level in reactor pond on 4 meters gave a possibility dismounting upper tie plate and upper core basket, ducts and collectors (fig 3). At the final stage of work the reactor shell and a lower tie plate were dismantled.



Fig. 3 Upper tie plate and upper core basket of reactor MR.

During the dismantling work the radiation situation in the reactor hall was changed constantly. To minimize the radiation dose to personnel during this work it was necessary to monitor exposure dose rate (EDR) in reactor hall. The main task was to estimate contribution of wall surface activity to EDR and find the way to decrease it. Radionuclide composition and specific activity of deposits on the walls was to be estimated for choosing how to manage these materials as radioactive waste.

EXPERIMENTAL WORK

Initially exposure dose rate (EDR) in reactor hall was 30-40 μ Sv/h. After water level in reactor pond was decreased on 2 meters EDR was increased to 170-200 μ Sv/h above the center of pond. For investigation of the surface gamma activity of the wall deposit of the storage pond was used the spectrometer complex "Kolibry" SKS-08P with the detecting block of "SKD" based on optocoupler scintillator-photodiode with CsI(Tl) crystal diameter 30 mm and thickness 10 mm. Calibration of the detecting device was conducted with relevant plane sources with a known surface activity. Two measurements were made in order to estimate background influence with a lead screen and without one. The scheme of experiment is submitted in fig. 4.



Fig.4. Scheme of experiment.

Surface activity of ¹³⁷Cs on reactor pond walls was measured after each decreasing water level in pond at 10 - 20 different points (fig 5). It was found that the distribution of ¹³⁷Cs on the reactor pond walls is characterized by considerable heterogeneity. The average value of the surface activity of ¹³⁷Cs after decreasing water level in reactor pond on 2 meters was (16.3 ± 7.2) kBq/cm².



Fig. 5 Measuring of surface activity by "Kolibry" spectrometer complex.

Calculation of contribution ¹³⁷Cs, deposed on walls and on constructive elements in pond (~40-50% from pond walls area), to exposure dose rate above the centre of pond by program MicroShield-5 estimated the value as 166 μ Sv/h. This value has a good agreement with measured value of EDR 170-200 μ Sv/h above the center of pond.

To reduce exposure dose rate after decreasing water level in reactor pond on 4 meters the walls was deactivated by high pressure (60 barr) water stream. Repeated measurements surface activity of ¹³⁷C deposed on pond walls gave average value (8.1 ± 5.7) kBq/cm². This confirms the earlier conclusion [1] that the deactivation pond walls by high pressure water stream decreased the surface contamination at 1.5-2 times. The exposure dose rate above the pond after decreasing water level on 4 meters, cleaning the walls, and removing constructive elements was 70-80 µSv/h. The exposure dose rate calculated in MicroShild-5, using the new data of the surface activity gave 58 µSv/h.

Simultaneously with measurement of surface activity of 137 C we carried out the complex spectrometric analysis of qualitative and quantitative radionuclide composition of deposits on reactor pond walls. Nine samples was taking on different distance (0.45 m, 1.15 m, 1.95 m) from level or reactor hall floor to make this analysis (fig. 5).

The concentration of γ -ray radionuclides was estimated by the spectrometric complex ISO-CART of the ORTEC Company that included a semi-conductor detector using HP Germanium GEM40P4. The analysis of the gamma-spectrum was made by the ISOPLUS-B32 software. The concentration of ⁹⁰Sr was detected by the scintillation β -ray spectrometer "Progress-beta", with the plastic scintillation detector BDEB-3-2U. Using the "Progress-5" software the spectra were analyzed in 0.9 MeV to 30 MeV range with the assumption that the sample contained ⁴⁰K and ⁹⁰Sr in equilibrium with ⁹⁰Y. The concentrations of uranium and plutonium radionuclides in the samples were determined by alpha-spectra of the targets, prepared by the electrochemical method after radiochemical separation of the investigated samples. Presence of ²⁴¹Pu in the aliquot, selected from prepared solution for electrolytic deposition, was detected by the highly sensitive spectrometric complex SKS-07P-B11 with liquid scintillator ULTIMA GOLD AB. The concentration ²⁴¹Pu in samples was calculated by the "Liquid Master" software. The results of spectrometric analysis of deposits on walls of the reactor MR pond and bottom slime from [1] are presented in table 1a, 1b, 1c.

Isotope	Specific activity, Bq/kg						
	sample 1(0.45 m)	sample 2(1.15 m)	sample 3(1.95 m)	bottom slime[1]			
^{134}Cs	$9.5*10^5$	$3.9*10^{6}$	$2.8*10^{6}$	$1.5*10^{6}$			
¹³⁷ Cs	9.2*10 ⁹	$2.8*10^{10}$	$2.1*10^{10}$	$1.3*10^{10}$			
⁶⁰ Co	$4.3*10^{7}$	$1.1*10^{8}$	$1.3*10^{8}$	$1.1*10^{8}$			
¹⁵² Eu	$2.0*10^{6}$	$8.3*10^{6}$	$2.0*10^{7}$	$3.6*10^{6}$			
¹⁵⁴ Eu	$4.3*10^{6}$	$5.4*10^{6}$	$1.3*10^{7}$	$1.5^{*}10^{6}$			
¹⁵⁵ Eu	$1.2*10^{6}$	-	-	-			
⁹⁴ Nb	8.9*10 ⁵	$1.5*10^{6}$	$1.2*10^{5}$	$3.0*10^5$			
^{166m} Ho	$1.2*10^{6}$	$6.2*10^5$	$9.2*10^{5}$	$1.2*10^{6}$			
²⁴¹ Am	$1.1*10^{6}$	-	$7.9*10^{6}$	$3.8*10^{6}$			
⁹⁰ Sr	$2.1*10^{8}$	$6.0*10^8$	$2.6*10^8$	$4.7*10^{8}$			
²³⁴ U	$3.5*10^{3}$	$1.6*10^4$	$4.2*10^4$	$1.9*10^4$			
²³⁵ U	$4.0*10^2$	$3.0*10^2$	$1.5*10^{3}$	$7.0*10^2$			
²³⁶ U	$4.0*10^2$	$6.0*10^2$	$2.6*10^{3}$	$2.5*10^{3}$			

Table 1a. The spec	ific activity of	f radionuclides	of deposits	on walls of the	reactor MR 1	pond.
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²³⁸ U	$2.0*10^2$	$1.5*10^{3}$	$1.6*10^{3}$	$1.4*10^{3}$
²³⁸ Pu	$1.4*10^{6}$	$1.1*10^{6}$	$5.5*10^{6}$	$6.0*10^{6}$
²³⁹⁺²⁴⁰ Pu	$2.3*10^5$	$4.2*10^{5}$	$7.0*10^5$	$9.7*10^5$
²⁴¹ Pu	$5.5*10^{7}$	$6.9*10^{6}$	$2.2*10^{7}$	$2.2*10^{7}$

Table 1b. The specific activity of radionuclides of deposits on walls of the reactor MR pond.

Isotope	Specific activity, Bq/kg					
	sample 4(0.45 m)	sample 5(1.15 m)	sample 6(1.95 m)	bottom slime[1]		
^{134}Cs	$1.1*10^{6}$	$3.6*10^{6}$	$1.5^{*}10^{6}$	$1.5*10^{6}$		
¹³⁷ Cs	$1.1*10^{10}$	$1.3*10^{10}$	$1.5*10^{10}$	$1.3*10^{10}$		
⁶⁰ Co	$4.7*10^{7}$	$7.5*10^{7}$	$8.7*10^{7}$	$1.1*10^{8}$		
¹⁵² Eu	$2.0*10^{6}$	$8.1*10^{6}$	$1.0*10^{8}$	$3.6*10^{6}$		
¹⁵⁴ Eu	$4.2*10^{6}$	$5.2*10^{6}$	$2.2*10^{7}$	$1.5*10^{6}$		
¹⁵⁵ Eu	-	-	$1.8*10^{6}$	-		
⁹⁴ Nb	-	$1.6*10^5$	$2.8*10^{5}$	$3.0*10^5$		
^{166m} Ho	$8.5*10^5$	$2.4*10^{6}$	$1.1*10^{7}$	$1.2*10^{6}$		
²⁴¹ Am	8.9*10 ⁵	$4.2*10^{6}$	$1.9*10^{6}$	$3.8*10^{6}$		
⁹⁰ Sr	$2.4*10^{8}$	$3.6*10^8$	$1.2^{*}10^{8}$	$4.7*10^{8}$		
²³⁴ U	$5.1*10^3$	$1.0*10^4$	$1.9*10^4$	$1.9*10^{4}$		
²³⁵ U	$2.0*10^2$	$3.0*10^2$	$9.0*10^2$	$7.0*10^2$		
²³⁶ U	$5.0*10^2$	$7.0*10^2$	$1.8*10^{3}$	$2.5*10^{3}$		
²³⁸ U	$5.0*10^2$	$9*10^{2}$	$1.0*10^{3}$	$1.4*10^{3}$		
²³⁸ Pu	$1.1*10^{6}$	$1.2*10^{6}$	$3.0*10^{6}$	$6.0*10^{6}$		
²³⁹⁺²⁴⁰ Pu	$2.5*10^{5}$	$3.6*10^5$	$3.7*10^5$	9.7*10 ⁵		
241 Pu	$5.2*10^{7}$	$8.5*10^{7}$	$1.0^{*}10^{8}$	$2.2*10^{7}$		

Table 1b. The specific activity of radionuclides of deposits on walls of the reactor MR pond.

Isotope	Specific activity, Bq/kg						
	sample 7(0.45 m)	sample 8(1.15 m)	sample 9(1.95 m)	bottom slime[1]			
¹³⁴ Cs	$6.1*10^5$	$1.6*10^{6}$	$5.4*10^{6}$	$1.5*10^{6}$			
¹³⁷ Cs	$5.4*10^9$	$1.2*10^{10}$	$1.2*10^{10}$	$1.3*10^{10}$			
⁶⁰ Co	$3.3*10^{7}$	$4.2*10^{7}$	$1.1*10^{8}$	$1.1*10^{8}$			
¹⁵² Eu	$2.2*10^{6}$	$1.8*10^{6}$	$7.0*10^{6}$	$3.6*10^{6}$			
¹⁵⁴ Eu	$2.6*10^{6}$	$1.8*10^{6}$	$5.3*10^{6}$	$1.5*10^{6}$			
¹⁵⁵ Eu	$1.4*10^5$	$8.7*10^5$	$6.0*10^{6}$	-			
⁹⁴ Nb	$3.0*10^4$	-	-	$3.0*10^5$			
^{166m} Ho	8.3*10 ⁵	$3.9*10^{6}$	$2.0*10^{6}$	$1.2*10^{6}$			
²⁴¹ Am	$6.5*10^5$	$1.2^{*}10^{6}$	$4.3*10^{6}$	$3.8*10^{6}$			
⁹⁰ Sr	$7.4*10^{7}$	$8.0*10^{7}$	$3.2*10^{8}$	$4.7*10^8$			
²³⁴ U	$5.0*10^3$	$8.4*10^{3}$	$1.3*10^4$	$1.9*10^4$			
²³⁵ U	$3.0*10^2$	$6.0*10^2$	$7.5*10^2$	$7.0*10^2$			
²³⁶ U	$5.0*10^2$	$8.0*10^2$	$1.4*10^{3}$	$2.5*10^{3}$			
²³⁸ U	$5.0*10^2$	$8*10^{2}$	$2.3*10^{2}$	$1.4*10^{3}$			
²³⁸ Pu	$5.1*10^5$	$1.1*10^{6}$	$1.1*10^{6}$	6.0*10 ⁶			

²³⁹⁺²⁴⁰ Pu	$1.5*10^5$	$2.2*10^{5}$	$3.6*10^5$	9.7*10 ⁵
²⁴¹ Pu	$3.3*10^{7}$	$7.2*10^{7}$	$1.0*10^{8}$	$2.2*10^{7}$

ANALYSIS

Analysis of the obtained data was show that qualitative and quantitative radionuclide compositions of the wall deposits and bottom slime are practically identical. This confirm our earlier [1] suggestion that the wall deposits of the pond were formed by fine particles of the bottom slime during the deposition. Obtained data on radionuclide composition of deposits on the walls of the reactor MR pond allow us to calculate the correlation coefficients between the α - and γ - emitting radionuclides contained in the spent nuclear fuel. The calculated correlation coefficients are given in Table 2 and 3.

Table 2. Correlations coefficients between U isotopes and ¹³⁷Cs, ²⁴¹Am.

N⁰	$^{234}\text{U}/^{137}\text{Cs}$	$^{236}\text{U}/^{137}\text{Cs}$	$^{234}\text{U}/^{241}\text{Am}$	$^{236}\text{U}/^{241}\text{Am}$
1	3,8*10-7	$0,4*10^{-7}$	3,2 *10-3	3,6 *10 ⁻⁴
2	$5.7*10^{-7}$	$0.2*10^{-7}$		-
3	19.8*10-7	$1,2*10^{-7}$	5,3*10-3	3.3*10 ⁻⁴
4	4.6*10-7	$0.5*10^{-7}$	$5.7*10^{-3}$	5.6*10-4
5	$7.7*10^{-7}$	$0.5*10^{-7}$	$2,4*10^{-3}$	$1.7*10^{-4}$
6	$12.6*10^{-7}$	$1,2*10^{-7}$	$10.0*10^{-3}$	9.5*10 ⁻⁴
7	$9.2*10^{-7}$	0,9*10 ⁻⁷	$7.7*10^{-3}$	7,7*10 ⁻⁴
8	7,0*10-7	$0,7*10^{-7}$	7,0*10 ⁻³	6,7*10 ⁻⁴
9	$2.9*10^{-7}$	0.3*10-7	$3.0*10^{-3}$	$2.3*10^{-4}$
Σ	$(8,1\pm3,4)*10^{-7}$	$(0,7\pm0,25)*10^{-7}$	$(5,5\pm1,8)*10^{-3}$	$(5,1\pm1,9)*10^{-4}$

Table 3. Correlations coefficients between Pu isotopes and ¹³⁷Cs, ²⁴¹Am.

$\mathbb{N}_{\underline{0}}$	$^{239+240}$ Pu/ 137 Cs	238 Pu/ 137 Cs	241 Pu/ 137 Cs	$^{239+240}$ Pu/ 241 A	238 Pu/ 241 Am	241 Pu/ 241 Am
1	$2.5*10^{-5}$	$1.5*10^{-4}$	$6.0*10^{-4}$	0.21	1.3	5.0
2	$1.5*10^{-5}$	$0.4*10^{-4}$	$2.5*10^{-4}$	-	-	-
3	3.3*10 ⁻⁵	$2.6*10^{-4}$	$10.3*10^{-4}$	0.09	0.7	2.8
4	$2.2*10^{-5}$	$1.0*10^{-4}$	$4.7*10^{-4}$	0.28	1.2	5.8
5	$2.8*10^{-5}$	$0.9*10^{-4}$	$6.5*10^{-4}$	0.09	0.3	2.0
6	$2.4*10^{-5}$	$2.0*10^{-4}$	$6.7*10^{-4}$	0.2	1.6	5.3
7	$2.7*10^{-5}$	$0.9*10^{-4}$	$6.1*10^{-4}$	0.23	0.8	5.1
8	$1.9*10^{-5}$	$0.9*10^{-4}$	$6.0*10^{-4}$	0.2	0.9	6.0
9	$0.8*10^{-5}$	$0.3*10^{-4}$	$2.2*10^{-4}$	0.08	0.3	2.3
Σ	$(2.2\pm0.5)*10^{-5}$	$(1.2\pm0.5)*10^{-4}$	$(5.7\pm1.6)*10^{-4}$	0.17 ± 0.05	0.9±0.3	4.3±1.1

These coefficients allow applying express assessment of α - radionuclides on a surface of constructional elements of the reactor MR without radiochemical operations. But it should be taking into account, that in reactor MR was experiments with reactor fuel assembly with different composition of nuclear fuel. Therefore, data of radionuclide composition of deposits on walls have effective character, and the received correlative coefficients can be used only for the MR reactor.

CONCLUSIONS

We proposed a method of measuring the surface activity of radionuclides on the walls of the reactor pond. It was found that the main contribution to the exposure dose rate over the reactor pond is made deposits on the walls of the pond. Radionuclide compositions of deposits on walls of the reactor pond and bottom slime are identical.

We calculate correlative coefficients the setting ratios between α -, and γ -emitting radionuclides for reactor MR. This allow carrying out an assessment of α -contamination without complex radiochemical analysis.

REFERENCES

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