

**Determination of Components of Fuel Matrix in Water and in Bottom Slimes in the MR Reactor Ponds in NRC “Kurchatov Institute” – 14038**

Alexey Stepanov \*, Iurii Simirskii \*, Ilya Semin \*, Anatoly Volkovich \*  
\* National Research Centre - Kurchatov Institute

**ABSTRACT**

Complex  $\alpha$ -,  $\beta$ -,  $\gamma$ - spectrometric investigation of water, bottom slimes and the wall deposit of the reactor pond and the storage pond of the MR reactor was conducted in this work. Specific activity of radionuclides in the bottom slimes and in the wall deposits considerably exceeded specific activity of radionuclides in the ponds water and was closed to levels of high radioactive waste. It was determined that the main radionuclide contributing to dose was Cs-137. A method for non-destructive distant control of nuclear fuel presence in construction elements of MR reactor was proposed. Water level decrease in the MR reactor ponds caused increase exposure dose rate. It was the result of cesium-137 deposited on the pond walls. The cleaning efficiency of the storage pond walls from the deposit by the high pressure water stream and mechanical cleaning was investigated as well.

**INTRODUCTION**

Specialists of the NRC "Kurchatov Institute" carry out the dismantling of the multiloop research MR reactor. Technical start-up of the reactor was carried out in 1963. The scheme of the MR reactor is represented in figure 1. There are two ponds: the reactor pond and the storage pond connected by the lock-chamber.

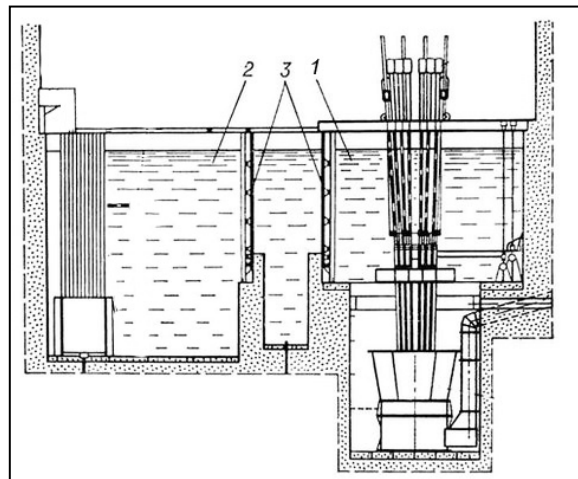


Fig. 1. The scheme reactor MR: 1-reactor pond; 2-storage pond; 3-lock chamber.

During dismantling works the test loops and the fuel channels were removed from the reactor pond to the storage pond (figure 2). Now cutting and removing of the channels from the reactor pond are carried out. Some loops were destroyed partly during experimental work. Nuclear fuel from the damaged loops and channels could get into the water.



Fig. 2. The construction elements on the storage pond bottom.

Removed materials were highly radioactivity, which required development of a method for the distant non-destructive control of fuel presence in them. High levels of the radionuclides Cs-137 and Sr-90 were found in the reactor MR ponds water. Therefore it was necessary to monitor the activity and radionuclide composition  $\alpha$ -,  $\beta$ -,  $\gamma$ -ray radionuclides in the reactor MR ponds water. The visual inspection of the ponds by video camera showed that considerable layers of slime were on the bottom of ponds. Radionuclide composition and specific activity of slime needs to be estimated for choosing how to manage these materials as radioactive waste. Another problem in the dismantling of reactor MR is the surface contamination of the pond walls. Decreasing the water level in the ponds led to increasing exposure dose rate and hence increased radiation dose to the personnel. Therefore it was necessary to estimate the contribution of radioactive surface contamination on the pond walls into exposure dose rate and to develop an efficient method of their removal.

## EQUIPMENT

The concentration of  $\gamma$ -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 typical spectrum of the storage pond bottom slime is shown on figure 3. The analysis of the gamma-spectrum was made by the ISOPLUS-B32 software. The concentration of Sr-90 was detected by the scintillation  $\beta$ -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 K-40 and Sr-90 in equilibrium with Y-90. Radionuclides tritium and Pu-241 with the  $\beta$ - particle energy 18 keV and 20 keV correspondently were detected by the highly sensitive spectrometric complex SKS-07P-B11 with liquid scintillator ULTIMA GOLD AB. The water was bi-distillated for separation the other radionuclides prevented tritium determination. The concentrations of tritium and Pu-241 in probes were calculated by the "Liquid Master" software.

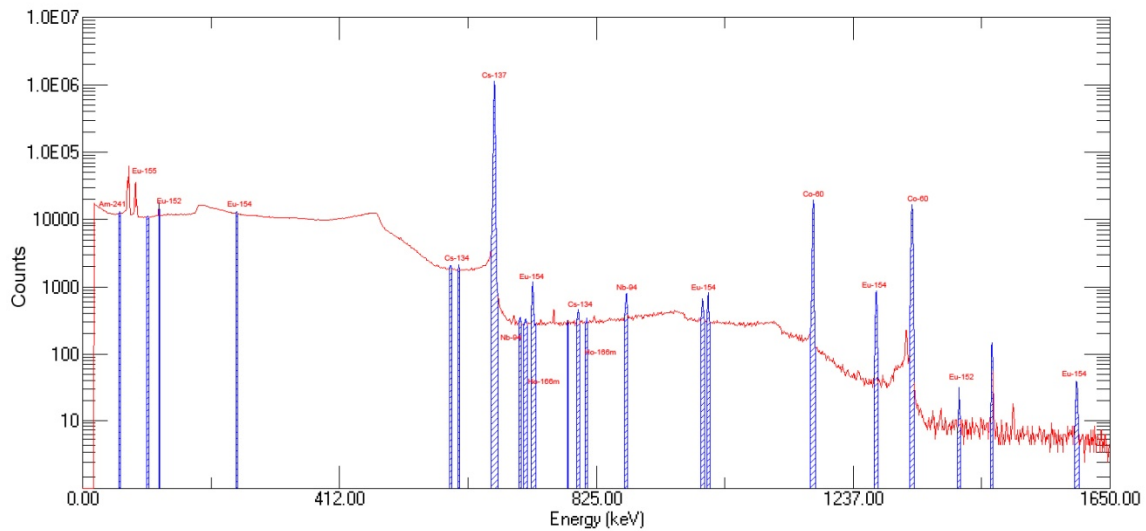


Fig. 3. The gamma-spectrum of bottom slime.

The concentrations of uranium and plutonium radionuclides in the water were determined by alpha-spectra of the targets, prepared by the electrochemical method after radiochemical separation of the investigated probes. The concentration of plutonium-241 was determined in the aliquot selected from solution prepared for electrolytic deposition. The typical spectrum of the aliquot of the reactor pond slime solution obtained by highly sensitive spectrometric complex SKS-07P-B11 is shown in fig. 4.

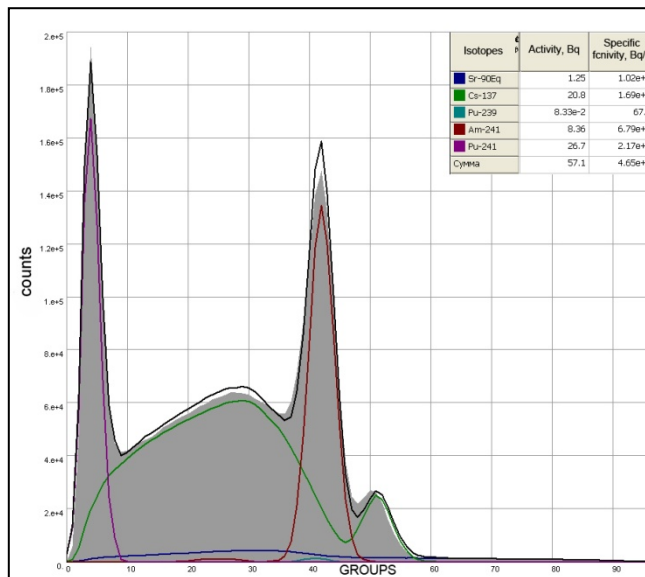


Fig. 4. The spectrum obtained Liquid Scintillation Analyzer SKS-07P-B11.

The alpha-spectra of the reactor pond slime and water obtained by vacuum Alpha Analyst Integrated Alpha Spectrometer of the Canberra company with a semi-conductor passivated implanted planar silicon detector are represented in figure 5.

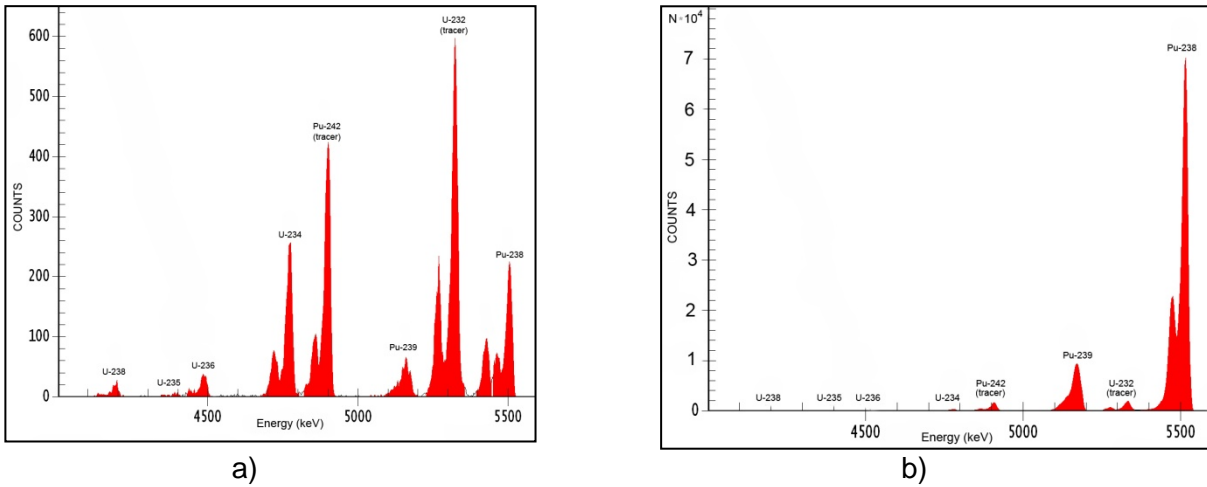


Fig. 5. The alpha-spectra obtained by vacuum spectrometer: a) reactor pond water; b) reactor pond slime.

## EXPERIMENTAL METHODS

The presence of residual fuel in the construction elements removing from the MR reactor ponds was controlled by gamma-spectrometric method with using the characteristic radiation of uranium which was excited by the fission products self-radiation, mainly cesium-137, after photo-absorption by uranium atoms [1]. In table 1 the intensities of the K-series of the characteristic radiation of uranium are shown.

Table 1. The intensities of the K-series of the characteristic radiation of uranium.

lines	E, keV	Intensity, %
UK <sub>α2</sub>	94.65	28.3
UK <sub>α1</sub>	98.43	45.8
UK <sub>β3</sub>	110.42	5.64
UK <sub>β1</sub>	111.30	10.7
UK <sub>β5</sub>	111.96	0.396
UK <sub>β2</sub>	114.45	4.14
UK <sub>β4</sub>	114.84	1.48

The presence of the nuclear fuel components in the water and slime of the reactor MR ponds was checked by detecting radionuclides of uranium and plutonium. The problem of detecting α-ray

radionuclides in the presence high radioactivity of Cs-137 and Sr-90 is very complicated. The extraction method was used for concentration and separation of the uranium-plutonium fraction from other radionuclides. 30% solution of tributyl phosphate in toluene was chosen as extractant. This allowed quantitatively extraction of both uranium and plutonium. As a result, the chosen method allowed determination of the presence in the probe for both spent and non-irradiated nuclear fuel.

For the extraction probe 500 ml of water was acidified with nitric acid and concentrated to 50 ml by distillation after addition 1 Bq of U-232 and Pu-242 as tracers. Then water was evaporated to dryness. Dry residue was dissolved in 10 ml of 5M nitric acid and uranium-plutonium fraction was extracted by 3 ml of 30% solution tributyl phosphate in toluene twice for 5 min. The organic phases were combined and washed six times with equal volumes of 5M nitric acid. Plutonium was reextracted from the organic phase with an equal volume of the mixture 0.25M nitric acid and 0.025M hydrofluoric acid. Uranium was reextracted by equal volume of distilled water. Reextraction was repeated twice for 3 minutes. The aqueous phases were combined and evaporated to dryness. Residue was dissolved in 10 ml of 0.5M solution of nitric acid. A 1 ml saturated solution of ammonium oxalate and 1 ml 25% solution of ammonium chloride were added to solution and pH was adjusted to 9 by an ammonia solution. The resulting solution was put in electrolytic cell. Uranium and plutonium were deposited on the stainless steel cathode during 45 minutes under the current density of  $0.3 \text{ A/cm}^2$ . During the electrolytic process the pH level we kept at 9 adding the ammonia solution if necessary. When the electrolytic process was completed the aliquot was taken from the cell to define the electrochemical yield of uranium -plutonium fraction. The cathode was washed by distilled water and the  $\alpha$ -spectrum of the prepared target was analyzed with the vacuum Alpha Analyst Integrated Alpha Spectrometer of the Canberra company with a semi-conductor passivated implanted planar silicon detector. For the analysis of slimes and the wall deposits 20-30 mg probe after addition 2 Bq U-232 and 2 Bq Pu-242 as tracers was lightened in 5 ml of hot concentrated nitric acid during 1 hour, after which the procedure was repeated using 5 ml aqua regia. The solutions were combined and evaporated to dryness. The residue was dissolved in 10 ml of 5M nitric acid and uranium-plutonium fraction was separated by extraction.

For investigation of radionuclides composition and determination of the surface gamma activity of the wall deposit of the storage pond was used the spectrometer complex "Kolibrý" SKS-08P with the detecting block of "SKD" based on optocoupler scintillator -photodiode with CsI(Tl) crystal diameter 30mm and thickness 10mm. 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. 6.

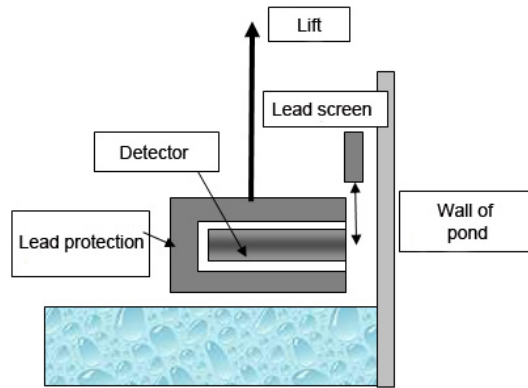


Fig.6. Scheme of experiment.

Calibration of the detecting device was conducted with relevant plane sources with a known surface activity.

## RESULTS

The typical spectrum obtained for the construction element with traces of fuel is shown in the fig. 7. It is seen that the spectrum allocate only four peaks as the spectrometer does not allow the lines  $UK_{\beta 1}$ ,  $UK_{\beta 3}$ ,  $UK_{\beta 5}$ , as well as lines  $UK_{\beta 2}$  and  $UK_{\beta 4}$ .

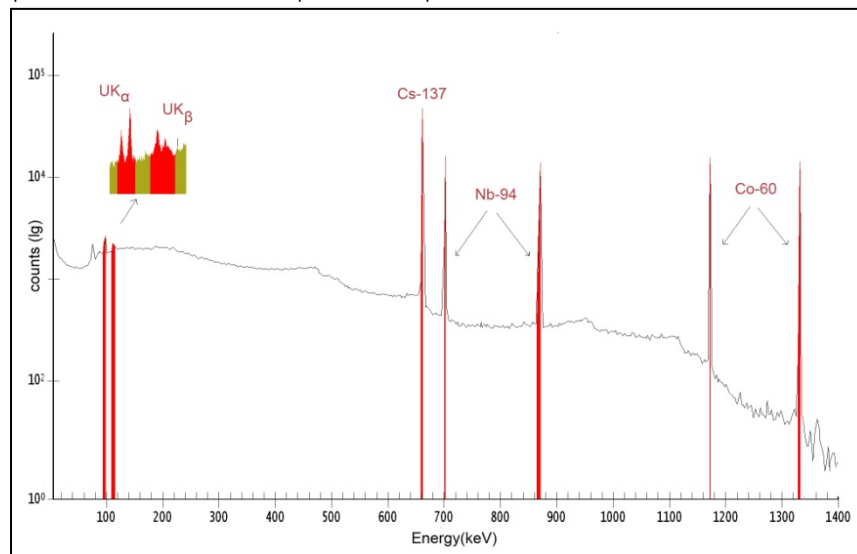


Fig.7. Typical spectrum of the construction element with of fuel traces.

The results of the investigation of the radionuclide composition of the MR reactor ponds water are represented in table 2.

Table 2. The specific activity of radionuclides in the reactor MR ponds water.

Isotope	Specific activity, Bq/kg	
	Reactor pond	Storage pond
H <sup>3</sup>	1.1*10 <sup>7</sup>	1.0*10 <sup>7</sup>
Cs <sup>134</sup>	1.1*10 <sup>2</sup>	1.0*10 <sup>2</sup>
Cs <sup>137</sup>	5.1*10 <sup>5</sup>	5.2*10 <sup>5</sup>
Co <sup>60</sup>	1.5*10 <sup>3</sup>	1.3*10 <sup>3</sup>
Sr <sup>90</sup>	9.0*10 <sup>4</sup>	8.9*10 <sup>4</sup>
U <sup>234</sup>	0.9	0.9
U <sup>235</sup>	0.02	0.02
U <sup>236</sup>	0.1	0.12
U <sup>238</sup>	0.06	0.07
Pu <sup>238</sup>	1.1	1.1
Pu <sup>239+240</sup>	0.6	0.6

From the results given in table 2 we can conclude that the qualitative and quantitative radionuclides composition is identical for both ponds, and the main radionuclide contributing to dose is Cs-137.

Results of the complex spectrometric analysis of the slime and the wall deposit probes taken from MR reactor ponds are given in table 3.

Table 3. The specific activity of bottom slime and the wall deposit of reactor and storage ponds.

Isotope	Specific activity, Bq/kg			
	Reactor pond		Storage pond	
	Bottom slime	Wall deposit	Bottom slime	Wall deposit
Cs <sup>134</sup>	1.5*10 <sup>6</sup>	2.0*10 <sup>6</sup>	6.9*10 <sup>5</sup>	8.9*10 <sup>5</sup>
Cs <sup>137</sup>	1.3*10 <sup>10</sup>	1.8*10 <sup>10</sup>	3.6*10 <sup>9</sup>	5.4*10 <sup>9</sup>
Co <sup>60</sup>	1.1*10 <sup>8</sup>	0.7*10 <sup>8</sup>	4.6*10 <sup>7</sup>	3.9*10 <sup>7</sup>
Eu <sup>152</sup>	3.6*10 <sup>6</sup>	4.1*10 <sup>6</sup>	1.2*10 <sup>6</sup>	5.8*10 <sup>6</sup>
Eu <sup>154</sup>	1.5*10 <sup>6</sup>	3.4*10 <sup>7</sup>	1.3*10 <sup>7</sup>	1.6*10 <sup>7</sup>
Eu <sup>155</sup>	-	-	2.0*10 <sup>6</sup>	5.1*10 <sup>6</sup>
Nb <sup>94</sup>	3.0*10 <sup>5</sup>	-	1.4*10 <sup>5</sup>	2.4*10 <sup>5</sup>
Ho <sup>166m</sup>	1.2*10 <sup>6</sup>	0.5*10 <sup>6</sup>	3.4*10 <sup>5</sup>	6.6*10 <sup>5</sup>
Am <sup>241</sup>	3.8*10 <sup>6</sup>	4.0*10 <sup>6</sup>	7.7*10 <sup>6</sup>	3.7*10 <sup>6</sup>
Sr <sup>90</sup>	4.7*10 <sup>8</sup>	3.2*10 <sup>8</sup>	2.2*10 <sup>8</sup>	3.1*10 <sup>8</sup>
U <sup>234</sup>	1.9*10 <sup>4</sup>	9.0*10 <sup>3</sup>	1.0*10 <sup>4</sup>	8.0*10 <sup>3</sup>
U <sup>235</sup>	7.0*10 <sup>2</sup>	1.4*10 <sup>2</sup>	5*10 <sup>2</sup>	2*10 <sup>2</sup>
U <sup>236</sup>	2.5*10 <sup>3</sup>	1.0*10 <sup>3</sup>	1.1*10 <sup>3</sup>	6*10 <sup>2</sup>
U <sup>238</sup>	1.4*10 <sup>3</sup>	1.0*10 <sup>3</sup>	1.0*10 <sup>3</sup>	5*10 <sup>2</sup>
Pu <sup>238</sup>	6.0*10 <sup>6</sup>	6.8*10 <sup>5</sup>	1.2*10 <sup>6</sup>	1.8*10 <sup>6</sup>
Pu <sup>239+240</sup>	9.7*10 <sup>5</sup>	2.5*10 <sup>5</sup>	2.3*10 <sup>5</sup>	4.5*10 <sup>5</sup>
Pu <sup>241</sup>	2.2*10 <sup>7</sup>	4.8*10 <sup>6</sup>	4.8*10 <sup>6</sup>	6.4*10 <sup>6</sup>

Analysis of the obtained data was show that qualitative and quantitative radionuclide compositions of the bottom slime and the wall deposit are practically identical in both the reactor pond and the storage pond. The suggestion can be made that the wall deposits of the pond were formed by fine particles of the bottom slime during the deposition. Specific activity of the main radionuclides (cesium-137, cobalt-60 and strontium-90) in the reactor pond was 2-3 times higher than the specific activity in the storage pond. At the same time the specific activity of radionuclides of the bottom slime and the wall deposit was the much higher than their specific activity in MR reactor ponds water. This will be taken into account during slime conversion to radioactive waste, because its parameters are closed to that of highly active waste.

The radionuclide composition and the surface activity of deposit on the storage pond wall were measured in 7 randomly chosen points (fig.8). The results are presented in table 4.

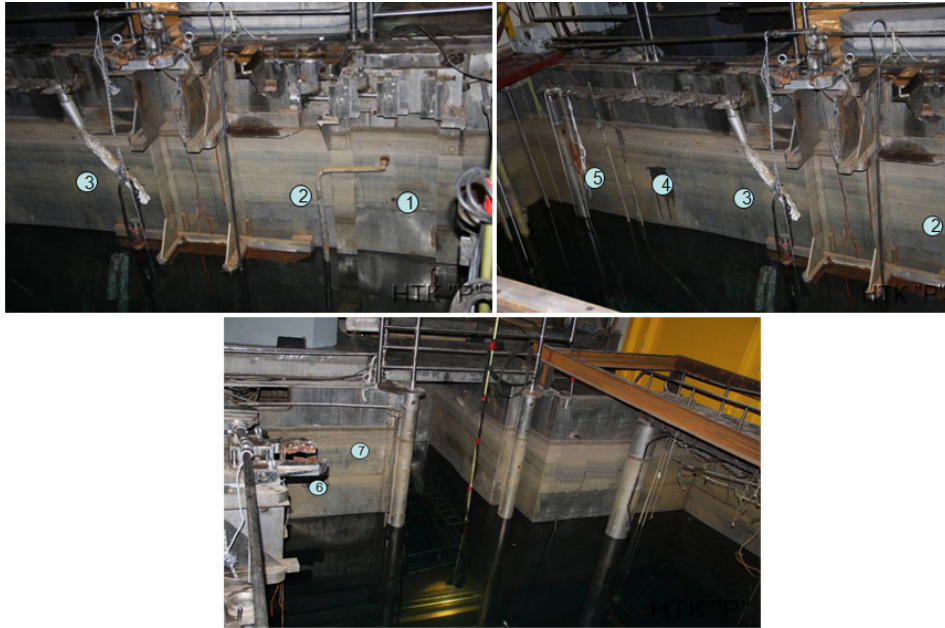


Fig.8. Measurements position of surface activity.



Table 4. The surface activity of the storage pond wall.

№	$A_{Cs-137}, Bq/m^2$	$A_{Co-60}, Bq/m^2$
1	$1.4 \cdot 10^8$	$5.9 \cdot 10^5$
2	$0.9 \cdot 10^8$	$15.5 \cdot 10^5$
3	$1.3 \cdot 10^8$	$7.6 \cdot 10^5$
4	$2.0 \cdot 10^8$	$0.9 \cdot 10^5$
5	$1.3 \cdot 10^8$	$22.0 \cdot 10^5$
6	$1.0 \cdot 10^8$	$5.9 \cdot 10^5$
7	$1.5 \cdot 10^8$	$34.4 \cdot 10^5$
$\Sigma$	$1.4 \cdot 10^8$	$13.2 \cdot 10^5$

From the table 4 we can see that average surface activity of Cs-137 in deposit on the storage pond wall is  $1,4 \cdot 10^8 Bq/m^2$  and a Co-60 -  $1,32 \cdot 10^6 Bq/m^2$ . This confirms the earlier assumption that main contribution to exposure dose rate was cesium-137. Total activities of Cs-137 and Co-60 in the wall deposit above the water level ( $S=21 m^2$ ) are  $2.9 \cdot 10^9 Bq$  и  $2.8 \cdot 10^7 Bq$  correspondently. Average exposure dose rate in reactor hall is  $30 \mu Sv/h$ . After decreasing the water level in reactor ponds by 1 meter the average exposure dose rate doubled. Calculation of the cesium-137 contribution to exposure dose rate at 1 meter distance from the pond by program MicroShield- 5 estimated the value as  $30 \mu Sv/h$ . This value was comparabled with exposure dose rate in a reactor hall before decreasing of water level and explained the increased exposure dose rate. To estimate the efficiency cleaning from deposit by the high pressure water stream and mechanical methods the initial surface activity of Cs-137 on the wall section area of  $0.5 m^2$ : before and after cleaning were measured [2]. The experimental results are given in table 5.

Table 5. The surface activity of the storage pond wall.

	Initial	Water stream 60 barr	Water stream 180 barr	Mechanical cleaning
Ev. $A_{Cs-137}, Bq/m^2$	$1.7 \cdot 10^8$	$1.0 \cdot 10^8$	$7.2 \cdot 10^7$	$3.3 \cdot 10^7$

Using data from table 5 we can make the conclusion that cleaning of the wall by water stream under pressure 60 bars reduces the surface activity of Cs-137 by 1.5 times. Increasing the water stream pressure to 180 bars has no significant improvement on degree of a cleaning the walls of the storage pond. At the same time the single mechanical cleaning of the pond walls surface was reduced the surface activity of cesium-137 in 5 times, i.e. was more effective.

## CONCLUSIONS

We proposed a distant non-destructive method of nuclear fuel control in the constriction elements removed from ponds of MR reactor. Complex  $\alpha$ -,  $\beta$ -,  $\gamma$  - spectrometric research of water, bottom

slimes and the wall deposits of the reactor pond and the storage pond of the MR reactor was made. It was found out that cesium-137 was main dose- forming radionuclide. It was determined that the specific activity of bottom slime was significantly higher than the specific activity of water and closed highly radioactive waste. The twice increasing of the expose dose rate after decreasing water level in storage pond about 1 meter can be explained by contribution Cs-137 containing in the wall deposit. The presence of significant amounts of radionuclides of the U and Pu in slime showed that components of the fuel matrix inserted to the reactor ponds.

## **REFERENCES**

1. Potapov, V. N.; Volkovich, A. G.; Simirskii, Yu. N., «Spectrometric method of evaluating spent nuclear fuel characteristics», Atomic Energy, V. 106, №5, p.348-354, 2009
2. A. Stepanov, Y. Simirskiy, I. Semin, A. Volkovich, V. Potapov, V. Stepanov. Characterization of MR Reactor Pond in NRC “Kurchatov Institute” Before Dismantling Work – ICEM2013 Conference proceedings – 96112, September 8-12, 2013, Brussels, Belgium

## **ACKNOWLEDGEMENTS**

The authors wish to acknowledge the valuable help provided by staff of the “Rehabilitation” department of NRC “Kurchatov Institute” during measurements in high dose rate conditions.