Nuclear Fuel Traces Definition in Storage Ponds of Research VVR-2 and OR Reactors in NRC "Kurchatov Institute" – 16022

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ABSTRACT

The Gas Plant complex is the experimental base of the Institute of Nuclear Reactors, which is part of the Kurchatov Institute. In 1954 the commissioning of the first Soviet water-cooled water-moderated research reactor VVR-2 on enriched uranium, and until 1983 the complex operated two research water-cooled water-moderated reactors 3 MW (VVR-2) and 300 kW (OR) capacity, which were dismantled in connection with the overall upgrades of the complex. The complex has three storage ponds in the reactor building. They are sub-surface vessels filled with water (the volume of water in each is about 6 m^3). In 2007-2013 the spent nuclear fuel from storages was removed for processing to "Mayak". Survey of Storage Ponds by Underwater Collimated Spectrometric System shows a considerable layer of slime on the bottom of ponds and traces of spent nuclear fuel in one of the storage pond.

For determination qualitative and the quantitative composition of radionuclide we made complex α-, β- , γ - spectrometric research of water and bottom slimes from Gas Plant complex storage ponds. We found the spent nuclear fuel in water and bottom slime in all storage ponds. Specific activity of radionuclides in the bottom slime exceeded specific activity of radionuclides in the ponds water and was closed to levels of high radioactive waste. Analysis of the obtained data and data from earlier investigation of reactor MR storage ponds showed distinctions of specific activity of uranium and plutonium radionuclides.

INTRODUCTION

The Gas Plant complex is an experimental base of the Institute of Nuclear Reactors, which is part of the Kurchatov Institute. In 1954 the first Soviet water-cooled water-moderated research reactor VVR-2 on enriched uranium was put into commission there. Until 1983 the complex operated two research water-cooled water-moderated reactors: VVR-2 and OR, 3 MW and 300 kW power respectively. They used mainly nuclear fuel assemblies containing fuel rods 16 and 11. WWR-2 used the assembly of eight and four fuel rods as well.

After reboots of the reactor and after discharge of fuel from the reactor during its reconstruction the spent nuclear fuel was placed in a temporary "wet" storage.

In early 2007 300 SFA were kept in the temporary "wet" storage of spent nuclear fuel at the facility part of which was damaged during the operation. (Figure 1) It is

possible that components of spent nuclear fuel could get into the water of storage ponds at that time.

Fig 1. “Wet” storage № 26.

The complex has three storage ponds in the reactor building: №15, №16 and №26. They are sub-surface vessels filled with water (each about 6 m^3) and covered with protective lids (figure 2).

Fig 2. Storage pond № 15.

At the bottom of the vessels there are special constructions that include absorbers providing nuclear safety and cells to accommodate SFA. In 2007-2013 the spent nuclear fuel was removed from the storages for processing at “Mayak” [1]. An examination of the storage ponds made with the Underwater Collimated Spectrometric System shows a considerable layer of slime on the bottom of ponds and traces of spent nuclear fuel in one of the storages (figure 3,4) [2].

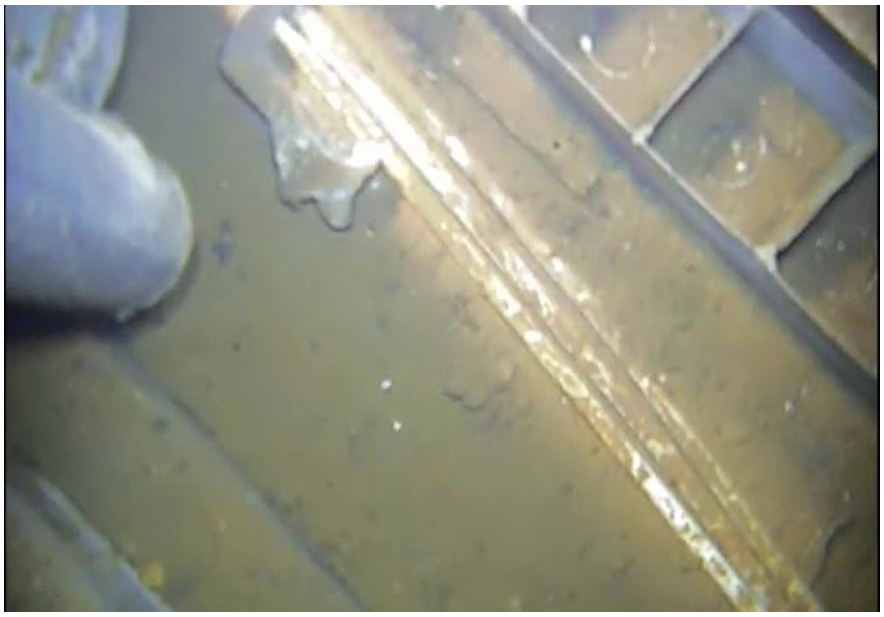

Fig. 3 The bottom of storage № 15.

Fig. 4 The bottom of storage № 16.

Surely, a plan of the storages’ decontamination should include investigation of the radionuclide composition and evaluation of the specific activity of the water and the

bottom slime. The results will show ways to transfer them to radioactive waste. To get the data we made a comprehensive alpha-, beta-, and gamma-spectrometric analysis water and bottom sludge from storage №15, №16 and №26 Complex "Gas Plant".

EXPERIMENT

The concentration of gamma-ray radionuclides was estimated by the spectrometric complex InSpector-2000 of the Canberra Company that included a semi-conductor detector made of HP Germanium GC-4018. The analysis of the gamma-spectrum was made by the GENIE 2000 software. The concentration of ^{90}Sr was detected by the scintillation beta-ray spectrometer “Progress-beta” with the plastic scintillation detector BDEB-3-2U. Using The spectra were analyzed by the “Progress-5” software in 0.9 MeV to 30 MeV range with the assumption of ^{40}K and ^{90}Sr in equilibrium with ^{90}Y present in the sample. The radionuclides tritium and ^{241}Pu with the beta- particle energy 18 keV and 20 keV correspondently were detected by the highly sensitive spectrometric complex SKS-07P-B11 with the liquid scintillator ULTIMA GOLD AB. The water was bi-distillated to separate other radionuclides which prevent tritium determination. To determine ^{241}Pu we used an aliquot of the solution purified from radionuclides ^{137}Cs and ^{90}Sr by extraction method. The concentrations of ^{3}H and ^{241}Pu in probes were calculated by the “Liquid Master” software. The concentrations of the alpha-emitting radionuclides uranium and plutonium ware determined by the alpha spectrum of the targets prepared electrochemically after radiochemical purification and concentration of the investigated samples. The alpha-spectra were obtained from the vacuum Alpha Analyst Integrated Alpha Spectrometer of the Canberra company with a semi-conductor passivated implanted planar silicon detector.

A 500ml water sample was acidified with nitric acid, reduced to 50 ml by distillation and then evaporated to dryness. To examine the bottom slime a 20-30mg sample was leached with 5ml of hot concentrated nitric acid for 1 hour, then the procedure was repeated using 5 ml of aqua regia. The resulting solutions were combined and evaporated to dryness. The dry residue was dissolved in 10 ml of 5M nitric acid, and uranium-plutonium fraction was extracted by 30% solution of tributyl phosphate in toluene. Earlier 1 Bq of ^{232}U and 1 Bq ^{242}Pu were added to samples as tracers. As a result, the chosen method allowed to determine in the probe both spent and non-irradiated nuclear fuel. Plutonium was reextracted from the organic phase by the equal volume of the mixture of 0.25M nitric acid and 0.025M hydrofluoric acid. Uranium was reextracted by the 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.

1 ml of aliquot was taken from the solution to determine the initial amount of uranium and plutonium. Then1 ml of saturated solution of ammonium oxalate and 1 ml of 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 deposed on a stainless steel cathode during 45 minutes under the current density of 0.3 A/cm^2. During the electrolytic process we kept the pH level 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 we analyzed alpha-spectra of the prepared target with the vacuum Alpha Analyst Integrated Alpha Spectrometer. The examples of alpha-spectra are represented in figures 5, 6.

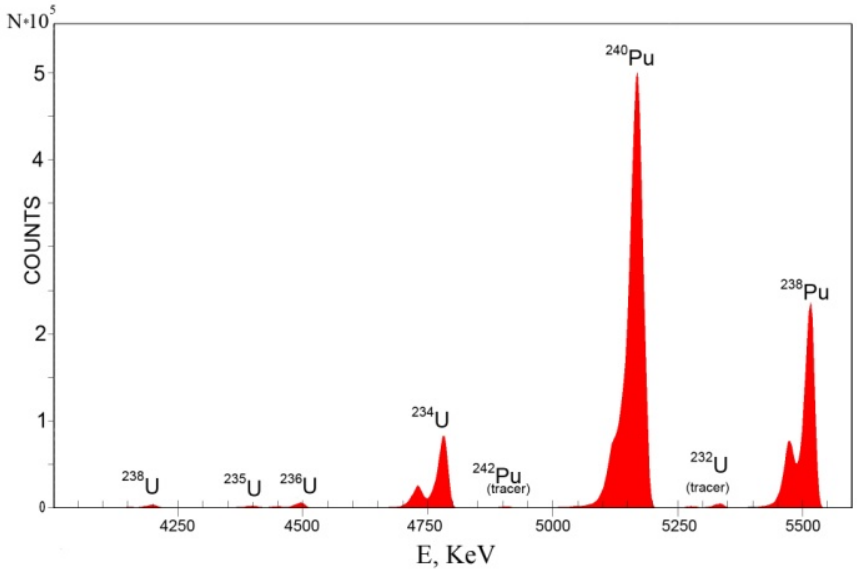

Fig. 5. The alpha-spectra of water from storage № 16.

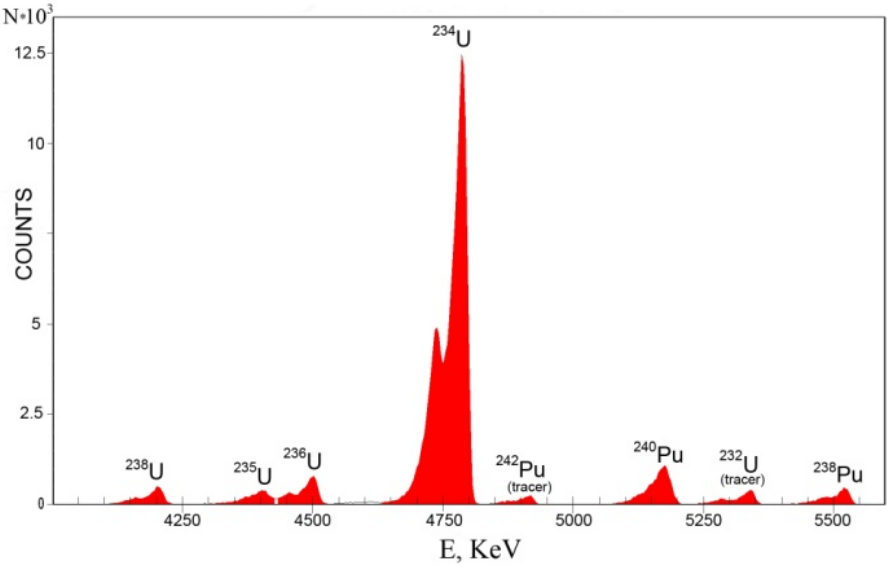

Fig. 6. The alpha-spectra of bottom slime from storage № 16.

RESULTS

Results of the complex spectrometric analysis of bottom slime and water are shown in Tables I and II respectively.

Table I. The specific activity of radionuclides in water from the 16, 15, and 26 storages of “Gas factory” and storage pond of reactor MR.

radionuclide	Specific Activity, Bq/kg			
	16 storage	26 storage	15 storage	MR reactor storage pond [2]
^{3}H	$1,3\cdot10^{4}$	$7,9\cdot10^{3}$	$5,8\cdot10^{3}$	$1,0\cdot10^{7}$
^{134}Cs	-	-	-	$1,0\cdot10^{2}$
^{137}Cs	$1,3\cdot10^{6}$	$3,0\cdot10^{5}$	$3,5\cdot10^{5}$	$5,2\cdot10^{5}$
^{60}Co	$4,1\cdot10^{2}$	$3,3\cdot10^{2}$	$3,1\cdot10^{2}$	$1,3\cdot10^{3}$
^{90}Sr	$1,1\cdot10^{6}$	$3,1\cdot10^{6}$	$1,8\cdot10^{5}$	$8,9\cdot10^{4}$
^{234}U	59	37.4	8.5	0.9
^{235}U	2,1	1.3	0.4	0.02
^{236}U	3,5	1.4	0.5	0.12
^{238}U	2,2	0.7	0.3	0.07
^{238}Pu	4,2	1.8	0.3	1.1
$^{239+240}Pu$	10,3	6.3	1.3	0.6
^{241}Pu	14	9.4	5.8	-

Table II. The specific activity of radionuclides in bottom slime from the 16, 15, and 26 storages of “Gas factory” and storage pond of reactor MR.

radionuclide	Specific Activity, Bq/kg			
	16 storage	26 storage	15 storage	MR reactor storage pond [3]
^{134}Cs	-	-	-	$6.9\cdot10^{5}$
^{137}Cs	$1.6\cdot10^{9}$	$1.5\cdot10^{10}$	$8.4\cdot10^{8}$	$3.6\cdot10^{9}$
^{60}Co	$2.4\cdot10^{6}$	$4.2\cdot10^{6}$	$1.8\cdot10^{6}$	$4.6\cdot10^{7}$
^{152}Eu	$4.0\cdot10^{5}$	$8.4\cdot10^{5}$	$2.8\cdot10^{5}$	$1.2\cdot10^{6}$
^{154}Eu	$2.9\cdot10^{6}$	$1.6\cdot10^{6}$	$1.2\cdot10^{6}$	$1.3\cdot10^{7}$
^{155}Eu	-	-	-	$2.0\cdot10^{6}$
^{94}Nb	-	-	-	$1.4\cdot10^{5}$
^{166m}Ho	-	-	-	$3.4\cdot10^{5}$
^{90}Sr	$1.4\cdot10^{9}$	$4.9\cdot10^{9}$	$9.1\cdot10^{8}$	$2.2\cdot10^{8}$
^{241}Am	$1.0\cdot10^{7}$	$3.3\cdot10^{6}$	$3.3\cdot10^{6}$	$7.7\cdot10^{6}$
^{234}U	$3.7\cdot10^{5}$	$7.5\cdot10^{5}$	$2.5\cdot10^{5}$	$1.0\cdot10^{4}$
^{235}U	$1.1\cdot10^{4}$	$3.8\cdot10^{4}$	$9.0\cdot10^{3}$	$5.0\cdot10^{2}$
^{236}U	$2.4\cdot10^{4}$	$5.8\cdot10^{4}$	$1.6\cdot10^{4}$	$1.1\cdot10^{3}$
^{238}U	$1.7\cdot10^{4}$	$3.9\cdot10^{4}$	$1.3\cdot10^{4}$	$1.0\cdot10^{3}$
^{238}Pu	$3.5\cdot10^{6}$	$8.8\cdot10^{6}$	$4.7\cdot10^{6}$	$1.2\cdot10^{6}$
$^{239+240}Pu$	$8.3\cdot10^{6}$	$2.1\cdot10^{6}$	$1.4\cdot10^{6}$	$2.3\cdot10^{5}$
^{241}Pu	$3.4\cdot10^{7}$	$8.9\cdot10^{6}$	$1.2\cdot10^{7}$	$4.8\cdot10^{6}$

From the results shown in Table l it is clear that the specific activity of the main dose-forming radionuclide ^{137}Cs in the water of all investigated ponds is practically identical. At the same time the water of the MR reactor storage pond is marked by high activity of ^{3}H (10^{7} Bq / liter). It is known that tritium produced by ternary fission of uranium and plutonium irradiated by neutrons, penetrates into the coolant through the fuel cladding and can get into the reactor pond water. A gateway connecting the reactor and the storage ponds is a design feature of the MR reactor. While dismantling the MR reactor tightness of the gateway was broken and radionuclide concentrations in both ponds alined [3], which led to a significant increase in the specific activity of tritium in the storage pond. The storage ponds located in the reactor hall of the Complex "Gas Plant" are isolated from the reactor pond, what explains the low concentration of tritium in them. (10^{4} Bq / liter).The high specific activity of ^{60}Co in the water and bottom slime of storage pond of MR reactor probably results from the fact that the reactor was used for studying nuclear fuel elements of varying designs, including those with claddings made of zirconium alloy and stainless steel. Analysis of the results presented in Table l and Table ll shows that the specific activity of radionuclides of uranium and plutonium in the water and bottom slime of the storages of the Complex "Gas Plant" exceeds 5-10 times their specific activity in the MR reactor storage pond. This may be due to the fact that nuclear fuel assemblies with damaged claddings were stored in the ponds of the Complex "Gas Plant". In accordance with the established norms [4] the bottom slime of the storage ponds in the Complex "Gas Plant" is regarded as an intermediate level radioactive waste coming close to a high-level radioactive waste that must be considered before disposal of the bottom slime.

CONCLUSIONS

Special design of storages in the Complex "Gas Plant" resulted in lower specific activity of tritium in water. ^{137}Cs is the main dose-forming radionuclide in the MR reactor storage pond. A specific activity of radionuclides of uranium and plutonium in the water and bottom slime of the storage ponds is high. Before the bottom slime disposal it must be considered that the specific activity of ^{90}Sr and plutonium radionuclides is close to high-level radioactive waste.

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ACKNOWLEDGEMENTS

We thank the staff of the Complex "Gas Plant" Drozdov A.A., Chumakov V.N. and Romanov V.I. for kindly providing samples for research.