# Experimental Simulation of the Radionuclide Behaviour in the Process of Creating Additional Safety Barriers in Solid Radioactive Waste Repositories Containing Irradiated Graphite

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**Abstract.** Results of the experimental modeling of radionuclide behavior when creating additional safety barriers in solid radioactive waste repositories are presented. The experiments were run on the repository mockup containing solid radioactive waste fragments including irradiated graphite. The repository mockup layout is given; the processes with radionuclides that occur during the barrier creation with a clayey solution and during the following barrier operation are investigated. The results obtained confirm high anti-migration and anti-filtration properties of clay used for the barrier creation even under the long-term excessive water saturation of rocks confining the repository.

## 1. Introduction

The majority of the at-reactor repositories of the shutdown Industrial Uranium-Graphite Reactors (IUGR) were in operation for 35÷40 years. They have played their positive part in providing the radiation safety of the personnel, population, and the environment in accordance with the regulatory requirements in effect in 50s-70s of the previous century. There are several dozen at-reactor repositories at the IUGR sites of the Russian Federation where irradiated graphite-containing solid radioactive waste (SRW) of the reactor origin is currently located.

To date the nuclear legacy at-reactor repositories do not entirely pass strict modern requirements imposed on the storage of radioactive materials. Those requirements could be only met if additional engineered safety barriers are created in the existing at-reactor repositories so that in combination with the repository confining rocks (natural barrier) they make a multi-barrier protective system. The developed system of barriers should provide the SRW disposal conditions for the radionuclide migration to be possible merely by means of the diffusion processes. The radionuclide diffusion coefficients in the barrier material should be as low as to ensure the level of activity emerging out of the boundaries of the repository to be within the intervention level, i.e. the regulated parameters defining the boundary between the radioactive and non-radioactive medium.

At the JCS Pilot and Demonstration Center for Uranium-Graphite Nuclear Reactor Decommissioning (JSC «DC») an option of creating the additional protective barriers by means of filling the free repository space by a clayey solution is being considered as a promising solution for the problem [1,2]. The barrier is installed by pumping the clayey solution through the perforated pipes. The pipe location grid size is determined so as to distribute the pumped clayey mass through the entire repository volume. After the clay is dried, the formed clay monolith contains the embedded solid radioactive waste. The implementation of the technique in question is the most relevant for the irradiated graphite-containing repositories as to date the ultimate scenario of handling this waste is not defined yet [3]. The approach suggests a delayed solution of the problem for a period of time long enough to make a decision on whether the SRW is to be retrieved or not.

To prove the efficiency of the technique proposed for the creation of engineered clay barriers, a repository mockup has been fabricated at the JSC «DC». It was used to conduct a series of runs aimed at the simulation of the radionuclide behavior when creating additional protective barriers in the existing repositories.

## 2. The procedure of experimental runs

The repository mockup was fabricated in the shape of a combined metal vessel. The schematic layout of the mockup cross-section is shown in figure 1; the locations of SRW fragments, engineered clay and natural rock barriers are indicated on the layout. The rock for the mockup was drawn nearby the existing at-reactor SRW repository.

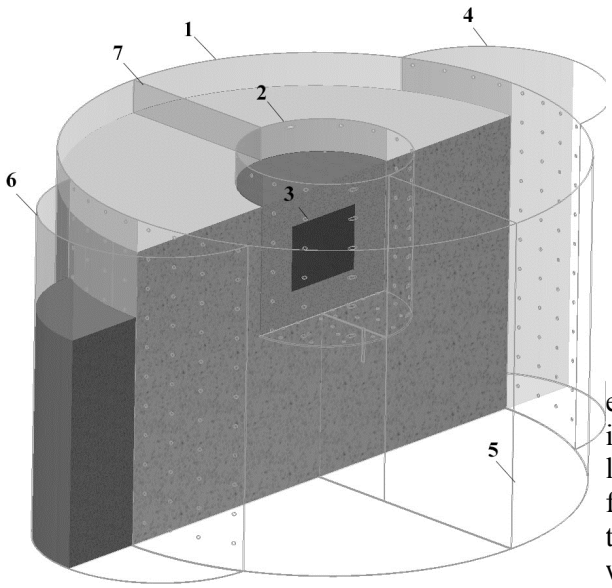


**Figure 1.** The mockup layout: 1 – the external vessel loaded with the rock; 2 – the internal vessel with the holed wall, initially loaded by the clayey solution; 3 – SRW fragments in a basket; 4 – the right tank; 5 – the partition wall; 6 – the left tank with water; 7 – the partition wall.

The mockup consists of the two cylinder-shaped nested steel vessels. The external vessel of 1.5 diameter and 0.5 m height is divided with a solid partition into two parts loaded with the layer-by-layer compacted rock. Walls of the internal vessel are holed such as their permeability is significantly higher than that of the concrete walls of the actual repository. SRW fragments of steel, alloys, and irradiated graphite are positioned into this vessel in the metal basket. Taking into account the gamma-activity of the samples and the mass of the SRW fragments the gross gamma-activity of the waste amounted ~$6.2\times10^{8}$ Bq and was defined primarily by $^{137}$Cs and $^{60}$Co radionuclides. The activity of the alpha-emitting trans-uranium (TRU) elements did not exceed $1.0\times10^{5}$ Bq.

The gross activity of radionuclides contained in SRW loaded into the internal mockup vessel are given in table 1.

**Table 1.** Gross activity of SRW positioned in the internal mockup vessel, Bq.

| $^{137}Cs$ | $^{90}Sr$ | $^{60}Co$ | $^{134}Cs$ | $^{152}Eu$ | $^{154}Eu$ | $^{155}Eu$ | TRU |
|---|---|---|---|---|---|---|---|
| $5.9\times10^{8}$ | $3.0\times10^{8}$ | $1.6\times10^{7}$ | $2.9\times10^{6}$ | $2.5\times10^{4}$ | $1.0\times10^{6}$ | $2.3\times10^{5}$ | $<1.0\times10^{5}$ |

The internal vessel and the basket with SRW fragments were filled with clay of the Zavarzinskoe Deposit containing 10% of sand. Clays of the Zavarzinskoe Deposit (located in the Tomsk Region) belong to the illite-kaolinite type characterized as follows: the content of particles sized below 0.01 mm varies from 55 thru 65%; the components in the descending order are illite, kaolinite, and montmorillonite.

To get the maximum filling of free space between SRW fragments we used the clayey solution containing sandy-clayey mix and water at the ratio of 1/1. Once in a while water was pumped out of the internal vessel. Redistribution of the residual moisture between the clay and the rock resulted in the clay drying and eventually in the formation of the clayey monolith with embedded SRW fragments. During the run the mockup was covered by the polyethylene film to prevent the moisture variations in the surface layer.

The tanks located on each side of the mockup were separated from the external vessel by the holed partition. After the clayey monolith was formed, the left tank was filled by the underground water taken from the well located nearby the at-reactor repository. The water level in the tank was higher than the bottom of the basket with SRW fragments positioned into the internal vessel and was maintained constant during the whole run. Thus, a directional water flow was simulated.

The right tank was used for monitoring. In case there was no filtration of water though the clay monolith during the run, the right tank should have remained dry. Conditions in the left tank were simulating the situation when the rocks in the bottom part of the repository were below the ground water level. On the contrary, the conditions in the right tank were simulating the situation when the repository bottom was located higher than the ground water level.

The radionuclide behavior studies were performed by sampling the clay and the rock periodically over a particular time interval. The measurement of γ-emitting $^{60}Co$, Eu isotopes, $^{137}Cs$, and $^{241}Am$ radionuclides was performed using the gamma-spectrometer equipped with GMX25P4 (Ortec) ultra-pure germanium detector. The content of β -emitting $^{90}Sr$ in the samples was measured using the Tri-Carb 3180 TR/SL (Perkin Elmer) liquid scintillation spectrometer. The specific activity of $^{239}Pu$ was analyzed using the extraction-chromatographic technique followed by the electrolytic depositing of plutonium and measuring samples on the Alpha Aria (Ortec) low-background alpha-spectrometer [4].

The speciation of radionuclides adsorbed by clays was determined by the selective desorption technique. The species are subdivided into groups based upon the distinctions in the solubility thereof. The selective desorption techniques are widely used in the radionuclide behavior studies in the natural and natural-technogenic systems, i.e. the protective barriers [5, 6]. Using the results of the selective desorption (see table 2) one can evaluate the content of radionuclides existing as mobile species (Stages I and II), potentially mobile species (Stage III), and the species fixed on rocks (Stages IV and V).

**Table 2.** The successive desorption procedure used to determine the sorption mechanism (S/L=1/10).

| Stage | Composition of the desorbing solution | Desorbed species |
|---|---|---|
| **I** | Distilled water, 20°C | Water soluble |
| **II** | $NH_4Ac$, 1 mole/L, pH 4,8 (HAc), 20°C | Exchangeable |
| **III** | 1 mole/L HCl, 20°C | Surface complexation |
| **IV** | 6 mole/L HCl, 20°C | Soluble in acids |

| Stage | Composition of the desorbing solution | Desorbed species |
|---|---|---|
| **V** | 6 mole/L $HNO_3$, 85°C | Soluble in acids |

## 3. The experimental modeling of the radionuclide behaviour - results

In 8 months of filling the internal and external vessels the rock surrounding the internal vessel was sampled as follows: IB-1 ÷ IB-4 and IC-1 ÷ IC-5 samples were taken in the vicinity of the central SRW containing vessel and IIC-1 ÷ IIC-5 samples were taken at 100 mm distance from it. The sampling was performed using the piston sampler. The samples were taken along the flow, IB-1 ÷ IB-4 samples before the internal SRW containing vessel and IC-1 ÷ IC-5 samples - after it.

The sampling layout is shown in figure 2. The number assignment in the sample labels corresponds to the downward direction from top to bottom. The radionuclide content in the samples is given in tables 3-5.

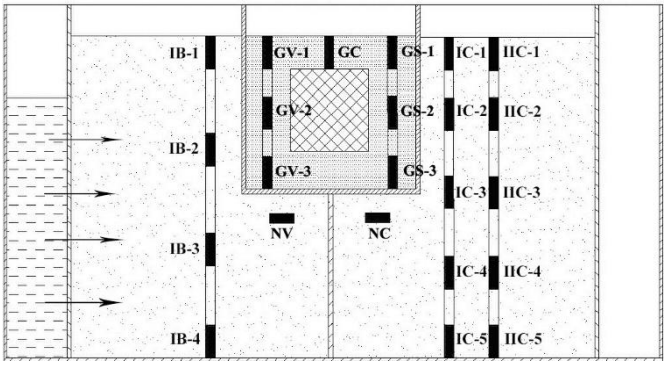


**Figure 2.** The sampling layout: IB-1 ÷ IIC-5 - the sampling points; → -the direction of the water flow through the holed wall of the left tank.

**Table 3.** The radionuclide content in the rock samples taken in the vicinity of the central SRW containing vessel and at 100 mm distance from it, Bq/g.

| S. No. | Sample | $^{137}Cs$ | $^{90}Sr$ | TRU |
|---|---|---|---|---|
| **1.** | IB-1 | 0,6 | <0,02 | <0,02 |
| **2.** | IB-2 | 0,6 | <0,02 | 0,02 |
| **3.** | IB-3 | 0,2 | <0,02 | <0,02 |
| **4.** | IB-4 | <0,02 | <0,02 | <0,02 |
| **5.** | IC-1 | 1,5 | <0,02 | <0,02 |
| **6.** | IC-2 | 0,4 | <0,02 | <0,02 |
| **7.** | IC-3 | 0,4 | <0,02 | 0,03 |
| **8.** | IC-4 | <0,02 | <0,02 | <0,02 |
| **9.** | IC-5 | <0,02 | <0,02 | <0,02 |
| **10.** | IIC-1 | <0,02 | <0,02 | <0,02 |
| **11.** | IIC-2 | <0,02 | <0,02 | <0,02 |

There were no Eu isotopes, $^{60}Co$, and $^{90}Sr$ radionuclides detected in the samples. From the data obtained it follows that a negligible quantity of $^{137}Cs$ and TRU radionuclides has been detected in the immediate vicinity of the SRW containing vessel. There is a possibility the sample was contaminated in the process of sampling. At the 100 mm distance from the wall of the vessel the concentration of all radionuclides was below the detection limit.

If in the samples taken from the rock in the external vessel there was practically no radionuclides

detected, in the internal vessel a transition thereof into the clay monolith was observed after the contact of clay with the SRW fragments (see table 4). The process of the radionuclide desorption from the SRW and sorption thereof on clay occurred with the participation of water remained after the installation of the clay barrier. The radionuclide distribution along the height of the internal vessel is irregular; probably, it is attributed to the different level of the contamination of various SRW fragments located in the basket. As compared to other radionuclides, the content of $^{137}$Cs in clay is considerably higher.

**Table 4.** The radionuclide content in clay samples taken from the internal vessel simulating the SRW repository and in the rock under the vessel, Bq/g.

| S.No. | Sample | $^{137}$Cs | $^{60}$Co | $^{90}$Sr | TRU |
|---|---|---|---|---|---|
| **1.** | GS-1 | $5.7\cdot10^2$ | 0.8 | 30 | 4.5 |
| **2.** | GS-2 | $7.8\cdot10^2$ | 0.3 | 19 | 2.0 |
| **3.** | GS-3 | $4.7\cdot10^2$ | 0.2 | 32 | 1.6 |
| **4.** | NV | 0.9 | <0.01 | 0.5 | 8.3 |
| **5.** | NC | <0.01 | <0.01 | <0.02 | <0.02 |

In the rock under the internal vessel (NV sample) the radionuclide activity level is lower, TRU being the only exception.

In 10 months one more sampling has been conducted; hence the total duration of the run was 18 months. During this time the rock in the left part of the vessel was saturated with water so that water appeared on the rock surface; however there was no penetration of water into the right part of the vessel. In the rock samples taken in the vicinity of the internal vessel the presence of radionuclides was not detected, the only exception being IV-1 sample with 1.3 Bq/g of $^{137}$Cs.

The difference in the content of $^{137}$Cs in the samples taken at various times is associated with the fact that at the initial stage the distribution of the excessive water arisen from the barrier installation process due to the contact with SRW caused the non-uniform contamination of the rock. Besides, the samples in the course of time were not taken from the same point rather from the various mockup areas. During the second sampling the set clay monolith barrier prevented the filtration of the directed water flow through the contaminated zone.

The character of the radionuclide distribution in clay was unchanged since the first sampling, the only difference being the higher contamination level (see tables 4 and 5).

**Table 5.** The radionuclide content in the clay in the internal vessel simulating the SRW repository, Bq/g.

| S.No. | Sample | $^{137}$Cs | $^{60}$Co | $^{90}$Sr | TRU |
|---|---|---|---|---|---|
| **1.** | GV-1 | $7.5\cdot10^3$ | 1.8 | 40 | 0.6 |
| **2.** | GV-2 | $6.3\cdot10^3$ | 1.0 | 20 | 0.5 |
| **3.** | GV-3 | $0.2\cdot10^3$ | 1.3 | 1.0 | 1.15 |
| **4.** | GC | $52.6\cdot10^3$ | 3.2 | $0.7\cdot10^3$ | 2.4 |
| **5.** | GS-1 | $0.7\cdot10^3$ | 1.1 | 16 | 5.7 |
| **6.** | GS-2 | $1.4\cdot10^3$ | 1.0 | 10 | 1.4 |
| **7.** | GS-3 | $0.6\cdot10^3$ | 0.05 | 16 | 1.5 |

The highest content of $^{137}$Cs and $^{90}$Sr was found in GC sample of clay taken from the top part of the internal vessel. Probably it is associated with the SRW fragments positioned in the top part of the basket and possessing the high level of the surface contamination by $^{137}$Cs and $^{90}$Sr.

The third sampling was conducted in 10 months, the total duration of the run by then was 28 months. During all that time the soil was watered, but there was no water in the right part of the vessel.

The samples were taken along the flow in the vicinity of the central SRW containing vessel and at 100 mm distance from it. In the rock samples before the internal vessel the content of $^{137}$Cs increased as follows: IB-1 – 1.5; IB-3 – 0.5; IB-4 – 0.5 Bq/g. In the samples taken along the flow after the internal vessel the content of $^{137}$Cs did not exceed 0.4-0.6 Bq/g. Other fission products were not detected also. In two samples, namely, IC-2 and IC-3 the content of TRU was 0.05 and 0.07 Bq/g. In the samples taken at the 100 mm distance from the internal vessel the content of $^{137}$Cs was either below its detection limit or slightly higher. Along the way there was no marked variation in the activity level of clay. As before, the maximum contamination of samples was with $^{137}$Cs.

When evaluating the protective properties of the barrier material, the important factors are, along with its sorption properties, the radionuclide speciation on the barrier material and the trend of the redistribution thereof in time taking into account the barrier has to keep its protective properties for a long period of time. The results of the selective radionuclide desorption from the clay samples taken during the run are given in figure 3. The results were obtained in the study of the GS-1 samples taken in the vicinity of the internal vessel along the flow.

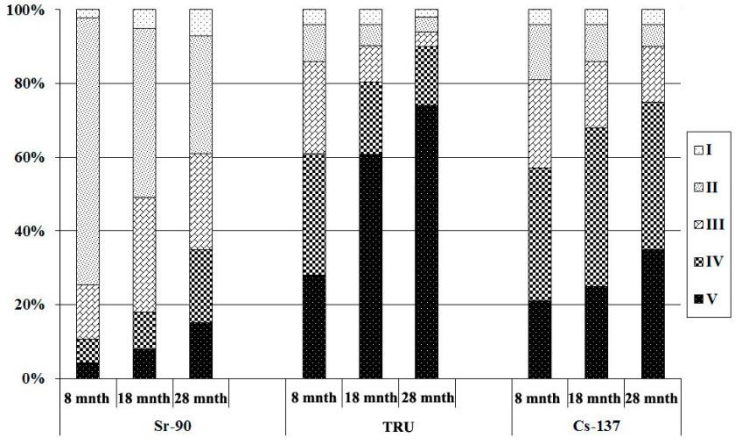


**Figure 3.** The distribution of the radionuclide speciation in clay (I – water soluble; II – exchangeable; III – mobile; IV and V – strongly fixed) depending on the time of the radionuclide interaction with the barrier material.

As it follows from the results of radionuclide speciation studies, in 8 months from the beginning of the run merely 10% of $^{90}$Sr existed in the strongly fixed species (Stages IV and V). The corresponding values for $^{137}$Cs and TRU are 60%. As the interaction time increases, the radionuclide speciation is redistributed towards the growth of the strongly fixed species content. In 28 months of interaction of clay with radionuclides the content of the strongly fixed species for $^{90}$Sr amounted 35%, and for $^{137}$Cs and TRU – 75% and 90%, respectively.

## 4. Conclusion

1. In order to evaluate the efficiency of the protective barrier installation in the existing SRW repositories the radionuclide behavior has been investigated in the combined barrier made of the illite-kaolinite clay and the repository confining rocks. The experiments were run for 28 months using the repository mockup containing solid radioactive waste fragments including irradiated graphite.

2. In the internal vessel of the mockup simulating the SRW repository the contamination of clay has occurred on the barrier installation stage, when the clayey solution used for the installation desorbed the radionuclides from the surface of SRW fragments. The following formation of the clay monolith effectively prevented the further distribution of $^{137}$Cs, gamma-emitters, $^{90}$Sr, and alpha-emitting TRU radionuclides through the rock. For the most readily desorbed $^{137}$Cs radionuclide the difference in its concentration between the clay and the rock in the vicinity of the repository simulating vessel reached four orders of magnitude.

3. Thus, during more than two years of observation no significant distribution of radionuclides in the rock has occurred, even though under the maintained conditions of the constant wetting. It proves the high anti-migration and anti-filtration properties of clay used for the barrier material.

## 4. References

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