Investigation of radioactive reactor graphite by gamma-spectrometric method

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Abstract. In this research radioactive contamination of graphite of the 4th block of the Leningrad NPP was determined using the method of gamma-spectrometric analysis. The obtained results of the specific activity of isotopes were compared with the contamination of the graphite masonry of industrial uranium-graphite reactors. It was found that in comparison with graphite of industrial uranium-graphite reactors, the LAES-4 masonry has less contamination with decay products and actinoids, which indicates that, unlike industrial reactors, there were no incidents of fuel Rod destruction in power plants. But at the same time, it was noted that the RBMK masonry has an increased content of $^{60}$Co. It was determined that this is due to the increased content of iron and cobalt impurities in unradiated graphite for RBMK. This research is important in terms of confirming the view that spent graphite from each specific uranium-graphite reactor has important features that need to be taken into account when planning decommissioning measures.

1. Introduction
Carbon is the heaviest of the widely used retarders. It have a fairly low deceleration capacity, but have a very small thermal neutron absorption cross – section and a high deceleration coefficient. Through of these useful properties, graphite began to be used as a retarder in the first nuclear reactors, such as the SR-1, launched by E. Fermi in the United States in 1942, as well as in the first Soviet F-1 reactor, which started working in December 1946. But any material has service life, so there is a reasonable problem of safe decommissioning of nuclear installations with graphite retarder. Have to have sufficient information about the activity and isotopic composition of graphite contamination to achieve this goal.

2. The management of radioactive graphite in the derivation of the reactors out of operation
According to estimates, 192 thousand tons of spent graphite have been accumulated in the world. Currently, 31 nuclear power units with a total capacity of more than 23,000 MW are in operation in the Russian Federation. RBMK reactors will make a big contribution to the accumulation of radioactive graphite in Russia. Currently, 11 units of the RBMK NPP are operating and at the end of their operation, the mass of spent graphite in Russia will reach 60 thousand tons [1].


| Type of reactor | Objects | Total weight of graphite (t) |
|-----------------|---------|-----------------------------|
| Industrial UGR (IUGR): | Mayak: A, AI, AV-1, AV-2, AV-3 | |
| 13 reactors | SHK: H-1, ЭИ-2, АДЭ-3, АДЭ-4, АДЭ-5 | |
| | GHK: AD, ADE-1, ADE-2 | 29 950 |
| | IPPE JSC: AM | |
| Industrial and energy UGR: 7 reactors | BelNPP: AMB-1, AMB-2 | 2 500 |
| | BilNPP: 4 EGI units | |
| | LNPP: 4 RBMK units | |
| | KNPP: 4 RBMK units | |
| RBMK: 11 reactors | SNPP: 3 RBMK units | 27 100 |
| | Total: | 59 550 |

2.1. The output of the reactors out of operation

Nuclear reactors have their own life cycles, including the stages of design, construction, start-up, operation and decommissioning. In the implementation of decommissioning is required to meet the following objectives:

- to ensure radiation safety of personnel, population and the environment;
- to minimize the release of radioactivity and volume of radioactive waste;
- to ensure high technical-economic effectiveness of measures;

Reactor designs and operational histories differ significantly, and the options for decommissioning should differ accordingly. [2]

When withdrawing uranium-graphite reactors, a special problem arises - the need to operate with huge amounts of radioactive graphite products. Therefore, to ensure the implementation of higher-level tasks, it is necessary to have information about the activity and isotopic composition of graphite contamination. [3]

2.1.1. Problems of decommissioning of RBMK reactors

In the Soviet Union in the 60 – 70 years of the last century, the construction of nuclear power plants with channel water graphite reactors was carried out, which was due to the lack of capacity of heavy engineering for the production of hulls and large technological equipment for VVER reactors, as well as the unique properties of channel reactors: the ability to regulate the flow of coolant and fuel[4].

In table 2 [2] some information is given about existing Russian nuclear power plants with RBMK-1000 reactors. In addition, several stopped RBMK units are located abroad: at Chernobyl – 4 RBMK-1000 units, including one emergency one; at Ignalinskaya – 2 RBMK-1500 units.

| NPP | Leningrad | Kursk | Smolensk |
|-----|-----------|-------|----------|
| Unit Year of commissioning | 1973 | 1975 | 1979 | 1981 | 1976 | 1979 | 1983 | 1985 | 1981 | 1985 | 1990 |

After the completion of operation of all RBMK units, about 27 thousand tons of radioactive graphite will remain (table 1). To assess its activity and plan decommissioning measures, it is impossible to directly use the results of studies conducted at the PUGR reactors. First, the operating conditions, terms and modes of operation of RBMK and IUGR are very different. Secondly, the number of incidents with fuel entering the masonry at RBMK is much lower, and, consequently, the content of fission products and actinoids in them is relatively small. Third, the locations of these reactors are different: the IUGR are located in sparsely populated areas away from large cities, and the RBMK is located in densely populated areas[5].
Factors affecting the timing of decommissioning of RBMK reactors may include the need for their timely replacement with other energy sources and high financial costs for decommissioning activities.

The urgency of solving the problem of handling irradiated graphite in relation to the decommissioning of power units of nuclear power plants with RBMK is enhanced by the following additional circumstances.

First, power units with RBMK do not have a protective containment, which would be an additional barrier to protect the environment from the release of radionuclides, especially at the stage of long-term storage under supervision.

Secondly, when preserving graphite masonry and metal structures of the reactor, it is necessary to reliably predict their mechanical properties and radiation conditions, which will require research on changes in radioactivity over time and the physical and mechanical state of the reactor masonry [2].

3. Radiation contamination of the reactor graphite

In order to determine the radionuclide composition and measure the specific activity of radionuclides contained in the graphite masonry of LAES-4, 23 samples were taken from graphite samples.

The samples were cylinders ~ 10 mm in diameter and ~ 5 mm thick. Weighing of samples was performed on a laboratory analytical balance Sartorius CP 225D.

Determination of the radionuclide composition and measurement of the activity of radionuclides in graphite samples was performed by semiconductor gamma-ray spectrometry.

3.1. Activation radionuclides in reactor graphite

As a result of long-term neutron irradiation, radionuclides with different half-lives, including more than a thousand years, are formed in reactor graphite. Neutron reactions in graphite leading to the formation of detected radionuclides are shown below (table 3) [5].

| The reaction of formation of | Type of decay | The quantum yield (%) | $T_{1/2}$ (days) | Maximum energy and quantum yield of beta particles (Kev) | Energy and quantum yield of gamma radiation (Kev) |
|-----------------------------|-------------|-----------------------|-----------------|----------------------------------------------------------|-------------------------------------------------|
| $^{59}$Fe (n,γ) $^{59}$Fe  | β-          | 98,1                  | 3.01·10$^4$     | 709.55                                                   | 1099.24 (56.5%) 1291.59 (43.2%)                  |
| $^{54}$Fe (n,p)$^{54}$Mn   | ε           | 1,9                   | -               | -                                                        | Annihilation 511                                     |
| $^{54}$Fe (n,p)$^{54}$Mn   | β-          | 0.014                 | 312.4           | 120.14                                                   | -                                                 |
| $^{54}$Fe (n,p)$^{54}$Mn   | ε           | 100                   | -               | -                                                        | -                                                 |
| $^{54}$Fe (n,p)$^{54}$Mn   | β-          | 0.93·10$^{-4}$        | 697.10          | -                                                        | -                                                 |
| $^{59}$Co (n,γ) $^{60}$Co  | β-          | 100                   | 1924.6          | 318.2 (99.88%)                                           | 1173.228                                          |
| $^{92}$Zn (n,β)$^{95}$Nb   | β-          | 100                   | 34.99           | 88.8 (27.27%)                                            | 569.33 (15.4%) 1491.4 (0.12%)                      |
| $^{133}$Cs (n,γ)$^{134}$Cs | β-          | 100                   | 754.2           | 925.6                                                   | 765.8 (99.8%) 1454.1 (2.5%) 604.72 (97.6%)         |
| $^{133}$Cs (n,γ)$^{134}$Cs | β-          | 27.9                  | 4943.7          | 175.4 (1.82%)                                            | 344.28 (26.6%) 1063.4 (90.9%) 1474.5 (8.1%)       |
| $^{151}$Eu (n,γ)$^{152}$Eu | β-          | 100                   | 1924.6          | 318.2 (99.88%)                                           | 1173.228                                          |
$^{60}$Co is one of the main emitters of hard $\gamma$ radiation during the first 40 years after the reactor shutdown. The content of $^{60}$Co affects the choice of terms and method of handling graphite masonry. The main channel of its formation is the $^{59}$Co(n,$\gamma$)$^{60}$Co reaction.

3.2. Fission products and actinides in reactor graphite

Actinoids and fission products are an important component of graphite contamination in uranium-graphite reactors. Information about their content in spent graphite is necessary to ensure nuclear and radiation safety during storage, dismantling and subsequent disposal of graphite masonry.

Secondly, when preserving graphite masonry and metal structures of the reactor, it is necessary to reliably predict their mechanical properties and radiation conditions, which will require research on changes in radioactivity over time and the physical and mechanical state of the reactor masonry. [2]

Over a long period of operation, accidents have occurred at all IUGR related to the destruction or breakage of the technological channel, the subsequent destruction of uranium blocks and the ingress of fragments of fuel and water into the hot graphite masonry. Then the uranium particles were carried by water vapor and deposited on the graphite surface both from the side of the process channels and in the cracks between the blocks. During long-term neutron irradiation, the uranium adsorbed on the surface of the blocks was first converted into $^{239}$Pu, which mostly burned out with the formation of fission products, and partially, by successive neutron capture, turned into heavier actinoids, including americium and curium (Fig. 3.). This has led to a significant contribution of actinoids and fission products to the total radioactivity of graphite in PP. For RBMK reactors, this component of contamination is not so significant, but it should be taken into account that actinoids and fission products can be formed not only in accidents, but also from impurity uranium [6].

During nuclear fission, 30 pairs of different fragment nuclides are formed. They experience an average of three consecutive beta decays, thus becoming increasingly long-lived fission products. Among them, the most significant contribution to radioactive contamination of graphite is made by $^{137}$Cs, which has a high probability of formation during fission and a half-life of about 30 years, comparable to the duration of decommissioning activities [7].

![Figure 1. Isotope transformations during neutron irradiation of uranium.](image-url)

Thus, the radioactivity of graphite is created by dozens of radionuclides and depends on the flux density and neutron spectrum, the time and history of operation of the reactor, the chemical composition and structure of the source material. It should be noted that the distribution of many nuclides in the masonry volume and its elements is random. This is due to variations in the impurity content and the random location of emergency cells.

The complex unpredictable process of pollution formation, which determines the level and composition of accumulated radioactivity, requires mandatory research, the results of which serve to
create a database used to assess the amount of RW and dose loads during radiation-hazardous work (dismantling of masonry, etc.). [2]

4. **Comparison of radiation contamination of reactor graphite in industrial and power reactors**
   This section presents the results of comparing the results of studies of radioactive contamination of the masonry of the I-1, EI-2 and ADE-3 reactors of the Siberian chemical combine in 1996-2001 [8, 9, 10, 11, 12] and the results of contamination of graphite masonry of the 4th block of the Leningrad NPP in 2019.
   23 graphite samples from 10 cores were examined to obtain data on the content of radionuclides in the LAES 4 masonry.
   Data on the distribution of radionuclides in the graphite masonry and their content near the emergency cells were taken from a study that took 160-200 graphite samples from each reactor. The kernels were taken from blocks located near and far from the emergency cells, at different heights in the selected cells, and in different cells at half the height of the masonry.

4.1. **Gamma-ray spectrometry method for determining radiation contamination of graphite**
   Gamma-ray spectrometry is the most commonly used method for passive and active analysis of the nuclide composition of materials performed without chemical preparation of samples (i.e., without destruction) [7].
   In the study of graphite contamination, semiconductor detectors with high energy resolution were used to isolate the radiation of the analyzed nuclides in complex multicomponent spectra.
   Radioactive nuclei emit strictly individual discrete gamma-ray spectra. Detection of characteristic peaks in the spectrum makes it possible to identify radiating radionuclides, and the intensity of their lines is proportional to the content of these nuclides in the sample under study. In addition, gamma radiation has a relatively high penetrating power, which makes it possible to perform measurements with relatively large graphite samples [2].
   The measurements were performed on a gamma-spectrometric complex that includes an ORTEC GMX–10 end coaxial extended energy range detector based on an n-type high-purity germanium crystal with a beryllium window and Adspecjr 2.0 digital multichannel analyzer with 16k channels.
   Control of the spectrometric complex and analysis of the obtained gamma-ray spectra were performed using the SpectraLineGP software package designed for spectrometric measurements and precision processing of gamma-ray spectra from semiconductor spectrometers.
   Calibration of the gamma-ray spectrometer was carried out using the sample spectrometric gamma-ray SOURCES OSGi$^{54}$Mn, $^{133}$Ba, $^{60}$Co, $^{137}$Cs, $^{152}$Eu. The activities of sources from the OSGi SET have an extended uncertainty of 1.7% to 3.0% with a coverage ratio of k=2. The processing of the measurement results of the OSGi sources and the construction of the calibration curve were performed using the SpectraLineGP program.
   The activity of each radionuclide in the samples was determined based on the measured peak areas of total absorption using GammaVision software. An example of the measured gamma-Ray spectrum from the samples is shown in figure 2.

![Figure 2. Example of the gamma spectrum from a sample with LNPP-4.](image-url)
Radiation in the range from 100 Kev to 2.5 MeV was analysed using a coaxial HPGe detector made of high-purity germanium. The energy resolution of the coaxial detectors is 1.7 Kev on the 1332.5 Kev (60Co) gamma line. As the size of the detector crystal increases, its relative efficiency and the peak/Compton ratio increase, so this study used a sufficiently large coaxial detector to determine the peaks of total absorption in a complex Compton-loaded background spectrum. However, as the crystal size increases, the energy resolution deteriorates, which made it difficult to analyse the radiation spectrum below 100 Kev. Using this coaxial detector, such isotopes as 137Cs, 154Eu, 46Sc, 133Ba, 134Cs, 60Co, 54Mn, 160Tb, 59Fe, 181Hf, 139Se, 141Se, 95Nb were detected.

Gamma-ray spectrometry is a non-destructive measurement method, so it can be widely used for sorting graphite components when disassembling masonry and forming batches for disposal or burial of graphite.

4.2. Activation radionuclides in the graphite masonry in an RBMK PPR and IUGR

Radioactive cobalt is one of the main emitters of $\gamma$-radiation for 40 years after the reactor is discontinued. Inside the volume of graphite blocks, 60Co was formed by activation of impurities. In the surface layer, 60Co was formed by activation and as a result of the introduction of cobalt of a corrosive origin with a vapor-gas mixture during incidents. Cobalt contamination of spent graphite can be divided into surface and bulk contamination, and each of them can be investigated separately.

Surface contamination with radioactive cobalt refers to a layer 2 mm thick. All graphite samples from LAES-4 do not belong to the surface layer, so only bulk cobalt contamination of the reactor masonry of different types will be compared.

The 60Co activity in the LAES-4 masonry varies greatly within a single core from sample to sample randomly (from 11.4 kBq to 245 kBq). Since 60Co is formed as a result of activation of the 59Co impurity, the spread of 60Co activity values may be due to the uneven content of these impurities in graphite.

In support of this hypothesis, a study of radioactive contamination of graphite masonry in the IUGR shows a significant heterogeneity in the distribution of 60Co over the volume of graphite blocks. As an example of this inhomogeneity, table 4 shows the results of measurements of the specific activity of 60Co from kerns from five cells of the IUGR. [9]

| Sample number in the kern | Reactor | EI-2 | I-1 | ADE-3 |
|--------------------------|---------|------|-----|-------|
|                          | Distance from the core center | 2 m up | 0 m | 3 m down | 3 m down | 2 m up | 0 m | 1 m up |
| 1 | 9,50 | 13,0 | 7,50 | 20,6 | 6,10 | 2,70 | 5,00 |
| 2 | 6,90 | 1,36 | 1,10 | 0,195 | 0,53 | 1,86 | 0,67 |
| 3 | 1,48 | 0,63 | 2,12 | 1,99 | 1,74 | 1,96 | 1,16 |
| 4 | 2,51 | 0,47 | 1,40 | 0,60 | 0,43 | 1,43 | 1,10 |
| 5 | 7,30 | 11,0 | 11,0 | 2,90 | 1,74 | 9,8 | 2,80 |

*The error of a single measurement does not exceed 5%*

The average value of the specific activity of 60Co in the LAES-4 masonry is (44.9±1.4) kBq/g.
The average values of $^{60}$Co activity for a large number of samples from the upper, Central, and lower graphite blocks of IUGR masonry, as well as samples taken near and far from emergency cells, are shown in table 5 [9].

### Table 5. Average specific activity of $^{60}$Co in graphite blocks (kBq/g).

| Sampling location                        | I-1 Inside the brick | I-1 The surface of the bricks | EI-2 Inside the brick | EI-2 The surface of the bricks |
|------------------------------------------|----------------------|-------------------------------|-----------------------|-------------------------------|
| Upper part of the reactor masonry       | 1,3±0,5              | 6,3±1,2                       | 2,6±1,2               | 6,7±1,8                       |
| The center column of reactor             | 2,0±1,0              | 6,8±1,6                       | 3,0±1,0               | 9,7±2,6                       |
| Lower part of the reactor masonry       | 1,0±0,4              | 6,2±1,4                       | 6,3±3,8               | 26±18                         |
| Cells close to the emergency             | 1,4±0,6              | 6,2±0,8                       | 2,8±0,6               | 14,0±2,7                      |
| Cells that are remote from emergency     | 2,5±0,7              | 7,2±1,0                       | 3,4±1,0               | 13,9±3,3                      |
| Average by masonry                       | 2,2±0,5              | 6,9±0,7                       | 3,0±0,6               | 14,0±2,4                      |

The following conclusions can be drawn from the results of measurements of $^{60}$Co activity in the masonry of the IUGR and LNPP:

- the content of $^{60}$Co inside the volume of graphite blocks of the IUGR is approximately the same throughout the core and does not differ much for different reactors (I-1 – 2.2±0.5 kBq/g, EI-2 – 3.0±0.6 kBq/g, ADE-3 – 4.4±1.2 kBq/g);
- the content of $^{60}$Co inside the volume of graphite blocks of LNPP-4 is an order of magnitude higher than in the volume of graphite blocks of IUGR and is 44.9±1.4 kBq.

This difference in results is due to the different operating times of the reactors: the IUGR is about 30 years old, the RBMK is about 40 years old, and the samples have different holding times after being removed from the masonry. Samples with IUGR were kept for about 5 years before measurement, and samples with RBMK for about six months. In addition, as shown in [13,14], which describes the study of impurity concentrations in the non-irradiated graphite of the IUGR and RBMK reactors, the impurity content in the graphite of the masonry of the three IUGR is approximately the same. There is a 1.5–20 times higher concentration of iron and cobalt in RBMK graphite compared to IUGR graphite. For zinc, this difference is even more significant (10-100 times).

- the IUGR cells adjacent to the emergency ones and remote from the incident sites do not differ in the level of contamination of $^{60}$Co.;
- Comparison of the content of activation radionuclides in graphite samples from cell blocks located near and far from the emergency cell does not reveal any significant difference. Consequently, accidents that occurred during the operation of the reactors did not affect their distribution. Therefore, based on the data on the distribution of $^{60}$Co activity, it is impossible to draw a conclusion about the existence of possible incidents at LNPP-4.

### 4.3. Actinoids and fission products in the graphite masonry of IUGR and RBMK

The source of graphite contamination with actinoids and decay products can be accidents with destruction of process channels and Fuel element shells. Also, the source of their formation in the masonry may be an admixture of uranium in graphite. According to [13] the content of impurity uranium in graphite is from 0.15 to 6.5 ppm.

The study of changes in the average level of contamination of graphite by the masonry radius showed that it is determined, first of all, by the position of the investigated cell relative to the emergency ones (table 6) [2]. The content of actinoids and fission products in samples taken from
cells adjacent to the emergency cells is hundreds of times higher than from cells remote from the
emergency cells.[15]

Table 6. The average content of $^{137}$Cs and $^{241}$Am from graphite blocks of IUGR cells, neighboring
and remote from emergency ones.

| Cell          | Act.$^{(137\text{Cs})}$ (Bq/g) | $\sigma$ Act. ($^{137}\text{Cs}$) (%) | Act.$^{(241}\text{Am})$ (Bq/g) | $\sigma$ Act. ($^{241}\text{Am}$) (%) |
|---------------|--------------------------------|-----------------------------------|-----------------------------|---------------------------------|
| 0320          | $1,3\times10^6$                | 41                               | $2,5\times10^4$            | 44                              |
| 2516          | $6,0\times10^4$                | 33                               | $4,2\times10^3$            | 35                              |
| 3644          | $3,2\times10^5$                | 40                               | $1,8\times10^3$            | 43                              |
|               | Cells close to the emergency   |                                   |                             |                                 |
| 2647          | $2,1\times10^3$                | 29                               | 16,6                        | 81                              |
| 2638          | $2,3\times10^3$                | 34                               | 17                          | 52                              |
| 2651          | $3,9\times10^2$                | 25                               | Less than 1.6               |                                 |
|               | Cells that are remote from emergency |                             |                             |                                 |

In the samples from the Leningrad NPP, there are no large differences in specific activities for
$^{137}\text{Cs}$. The average value is 1.78±0.13 kBq, which within the margin of error coincides with the values
of the average content of $^{137}\text{Cs}$ in the cells of the IUGR remote from the emergency ones. From this we
can conclude that the source of formation of fission products in the LAES-4 masonry was impurity uranium. There were no signs of emergency situations with the destruction of Fuel elements in the
LAES-4 masonry.

5. Conclusions

This article is important in terms of confirming the view that spent graphite from each specific
uranium-graphite reactor has important features that need to be taken into account when planning
decommissioning measures. This is due to differences in their design, materials used, and operational
history. Therefore, justifications are required for direct transfer of the results obtained from graphite
studies of one reactor to another reactor. A detailed study of the radioactive contamination of spent
graphite from each UGR is a necessary part of the decommissioning process.

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