APPROACHES TO GRAPHITE BLOCK CHARACTERIZATION BY GAMMA RADIONUCLIDES AND THEIR PACKAGING DURING UGR STACK DISMANTLEMENT

Pavliuk A. O.[1], Kotlyarevsky S. G.[1], Kan R. I.[1], Volkova A. G.[2], Zakcharova E. V.[2], Komarov E. A.[3]

[1]**Pilot & Demonstration Center for Decommissioning of Uranium-Graphite Nuclear Reactors, JSC, Seversk, Tomsk region, Russia**

[2]**Federal State Budgetary Institution of Science A. N. Frumkin Institute of Physical Chemistry and Electrochemistry of the Russian Academy of Sciences, Moscow, Russia**

[3]**State Corporation Rosatom, Moscow, Russia**

Article received on June 24, 2021

This paper presents technical and methodological approaches developed by PDC UGR to characterize the removed graphite blocks by gamma-emitting radionuclides during graphite stack dismantlement and to package them. It overviews the experimental testing of the characterization method and its findings. It also presents a container design option providing maximum filling efficiency.

Keywords: *radioactive waste, irradiated graphite, non-destructive testing method, characterization, packaging.*

Introduction

Phased final shutdown of power RBMK-1000 and EGP-6 type reactor units was started in Russia in late 2018. Considering the "immediate dismantlement" option adopted for all power reactors undergoing decommissioning in Russia [1, 2], updated were the tasks associated with the development of graphite stack dismantlement methods and the management of graphite radioactive waste removed from the reactors and their testing. Graphite RW characterization by gamma-emitting radionuclides is seen as an essential stage having some important effect on the decommissioning safety of uranium-graphite reactors (UGR) and technical and economic parameters of this process. Deliberate overassessment (considering the application of a conservative approach) can be avoided through the development and implementation of methods allowing to specify the radiation characteristics of the waste with appropriate metrological indicators that would exclude multiple overestimations of the measurement results. This approach can help to optimize the safety measures implemented during RW management and to reduce the costs by avoiding the use of higher-class containers during waste packaging and further handling of waste until its final disposal.

Based on the case study of a shutdown production uranium-graphite reactor ADE-5, methods providing the dismantlement of reactor structures and the characterization of unloaded graphite blocks have been developed by JSC PDC UGR considering the opportunities for further replication of the developed methods at UGRs of another type [3]. PUGR ADE-5 was shut down after 43 years of operation. In 2020, 12 years have elapsed since its final shutdown.

Characterization methods have been developed with an account taken of some specific aspects associated with the management of graphite blocks

from UGR dismantlement. Thus, a layer-by-layer graphite stack dismantlement option was selected as a basic approach involving staged block-by-block dismantlement and their removal through a purposely-designed opening and further containerization [4]. This approach increases the safety of the stack dismantlement process and provides high-precision measurements and segregation, which is impossible in case of graphite removal via grab method (when a group of graphite blocks is grabbed by a ladle) [5].

Characterization of the removed graphite blocks according to the content of the main dose-contributing gamma-emitting radionuclides (^{60}Co, ^{137}Cs) should be implemented to monitor the radiation situation during the dismantlement process, to optimize the packaging process, to specify the passport radiation characteristics and to provide the conditions when the established limits for the equivalent dose rate (EDR) are never exceeded for the gamma radiation emitted by waste packages.

Specific aspects associated with gamma-emitting radionuclide accumulation in UGR graphite blocks and their influence on the dose characteristics

Table 1 presents a list of the main gamma-emitting radionuclides contained in graphite RW.

Table 1. List of gamma-emitting radionuclides being specific for graphite RW after 10 years of cooldown (PA stands for the activation product, PD – for the fission product)

№	Nuclide	Generation source	$T_{1/2}$, years	Energy of gamma radiation, keV
1	^{60}Co	PA	5.27	1 173.22; 1 332.51
2	^{137}Cs + ^{137m}Ba	PD	30.17	661.6
3	^{155}Eu	PD	4.96	86.6; 105.3
4	^{152}Eu	PA, PD	13.5	121.8; 244.7; 964.08; 1 085.87; 1 112.07; 1 408.01
5	^{154}Eu	PD	8.6	123.07; 247.93; 723.30; 873.18; 996.29; 1 004.76; 1 274.43; 1 596.48
6	^{134}Cs	PA, PD	2.06	569.33; 604.72; 795.86; 801.95; 1,365.18
7	^{133}Ba	PA, PD	10.0	81; 276; 303; 356; 384

^{60}Co and ^{137}Cs are considered as the main isotopes governing the equivalent dose rate of gamma radiation after more than 10 years of cooldown. These studies have shown that the blocks removed during graphite stack dismantlement can differ significantly in their content. In addition, each of the considered radionuclides is characterized by some individual features of their spatial distribution over the volume of individual graphite blocks.

In some local areas, graphite stacks appear to be heavily contaminated with ^{137}Cs radionuclide. These areas are characterized by its plume propagation due to the ingress of fuel composition into it, its subsequent irradiation and the spread of "fuel origin" radionuclides under the influence of operational factors (Figure 1).

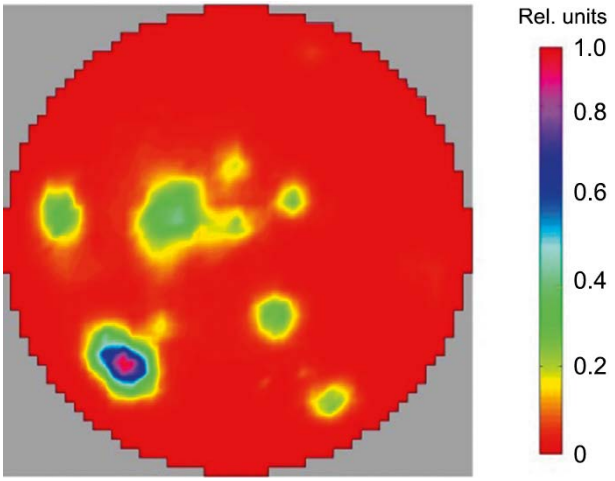

Figure 1. A typical example of a UGR cartogram showing the plumes with radionuclides of "fuel origin" (response of epithermal neutron detectors in the cells of a graphite stack, rel. units)

The nature of block contamination with ^{137}Cs is predominantly superficial. In this case, certain fraction of this radionuclide penetrates into the deep layers of the block wall (Figure 2). This trend is typical both for PUGR and RBMK-1000 reactor units.

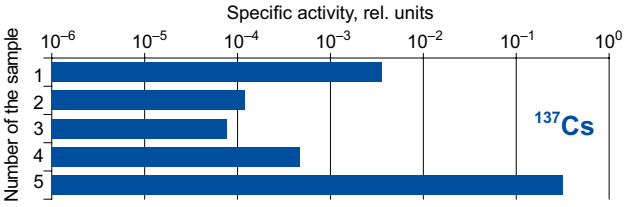

Figure 2. Typical form of ^{137}Cs distribution over the wall thickness of a graphite block located within "fuel origin" radionuclide plume of the LNPP-3 RBMK-1000 reactor unit

The main proportion of ^{60}Co activation product is distributed over the block volume. However, due to corrosion product activation in structural materials and their fixation on graphite blocks, an increased content of this isotope is also observed in the near-surface layer of graphite elements. In addition, the mass content of cobalt impurities ranges widely both within the volume of individual blocks and

from block to block ($0.7 \cdot 10^{-7}$—$3.8 \cdot 10^{-6}$ wt.%) [6]. Together, these factors provide some great variability regarding both ^{60}Co spatial distribution in the volume of some individual blocks (Figure 3) and the average content of this isotope in individual blocks (Figure 4).

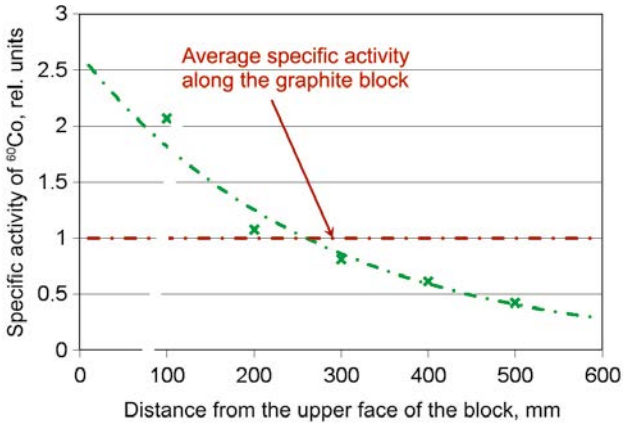

Figure 3. Form of ^{60}Co distribution along the height of a graphite block removed from PUGR ADE-5 graphite stack

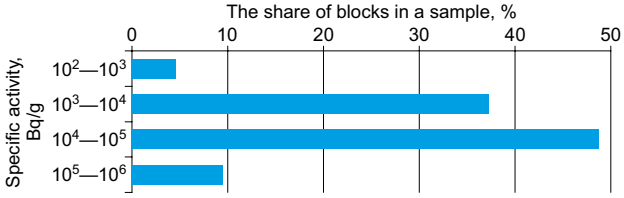

Figure 4. Distribution function in a sample of 43 graphite blocks from LNPP-2 and LNPP-3 RBMK-1000 reactor units according to the average specific activity of ^{60}Co

Distribution function by the average specific activity of ^{60}Co (Figure 4) was calculated based on the evaluated features associated with the radionuclide accumulation in the graphite of RBMK-1000 reactors at the Leningrad NPP in 2020—2021. Based on these studies, one can conclude that during the dismantlement of the graphite RBMK-1000 reactor stacks the expected width of the dose rate variation range from individual graphite blocks would amount to up to 4 orders of magnitude; for the bulk of blocks (~85 %) — up to two orders of magnitude. Figure 5 shows the same distribution function expressed in terms of the gamma radiation dose rate at the time of RBMK-1000 reactor final shutdown at a distance of 10 cm and 1 m from the side surface of the graphite block.

Thus, gamma radiation dose rate can vary significantly from block to block both due to differences in their total activities and due to some specific aspects of their spatial distribution over individual block volumes. Thus, for example, the dose

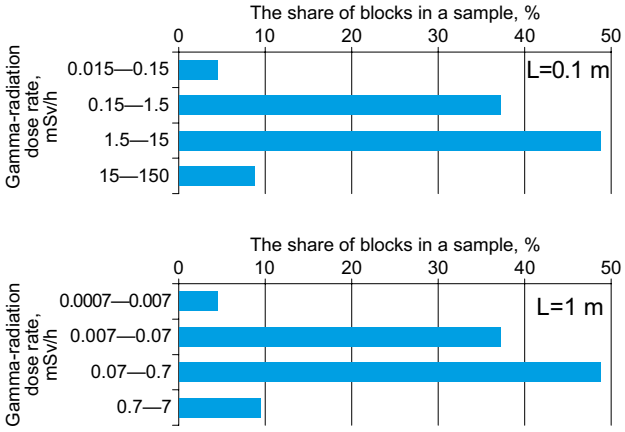

Figure 5. Distribution function for the dose rate of gamma radiation at the time of RBMK-1000 reactor final shutdown at a distance of 10 cm (top) and 1 m (bottom) from the side surface of a graphite block

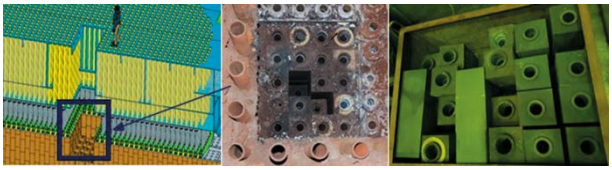

Figure 6. The spot from which the graphite blocks were removed from PUGR ADE-5 through an opening (left and center), container with removed blocks (right)

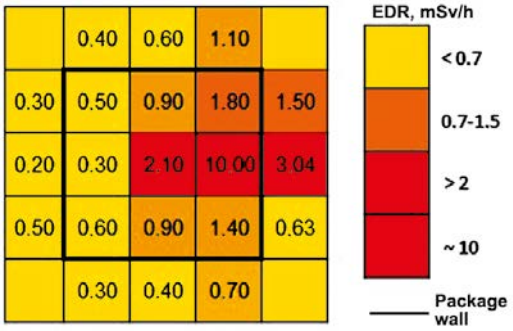

Figure 7. Measurement cartogram for the gamma radiation dose rate around a package with graphite blocks (3 × 3 pcs.), MSv/h (< 10 cm from the surface). Thick line shows the metal wall of a container. Shown in red are the values exceeding 2 mSv/h

rate from PUGR ADE-5 graphite blocks removed through an opening [7] and the one measured in the upper stack layers differed by ~25 times (Figure 6 and Figure 7).

Figure 7 shows measured gamma radiation dose rates around a package filled with graphite blocks (3 × 3 pcs.). As can be seen from the cartogram, the dose rate on the surface from the three sides of the package varies in the range of 0.2—1.1 mSv/h and only due to a single more highly active block located near the wall, the dose rate exceeded 2 mSv/h

on one surface. This EDR limit is seen as a criterion allowing the categorization into the second and third transport categories in accordance with SanPiN 2.6.1.1281-03 [8].

Specific aspects associated with the dismantled graphite block characterization by gamma-emitting radionuclides during packaging

As noted above, graphite UGR blocks are highly heterogeneous in terms of gamma-emitting radionuclide content both as regards the volume of individual blocks and from block to block (Figures 2—5). This heterogeneity is basically responsible for the deviation in the measured content of gamma-emitting radionuclides from the actual values both during the characterization of individual blocks and the blocks within a filled package (container). The same circumstance can significantly affect the dose characteristics of a filled container (dose rate on the surface) and, accordingly, the container choice.

Figure 8 visually illustrates some effects associated with the non-uniformity of the volumetric gamma-emitting radionuclide distribution over a compact graphite block assembly (imitation of a block layout in a container). The gamma quanta trajectories were modeled using the Monte Carlo method and the Geant 4 software [9].

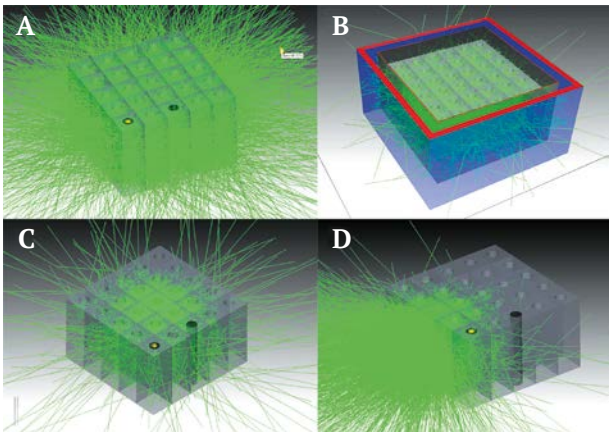

Figure 8. Visualization of numerical modeling results demonstrating scattering and absorption effects of gamma radiation from radionuclides contained in graphite (trajectories of gamma quanta are indicated in green), for: A — irradiated graphite blocks in the form of an assembly of 5 × 5 blocks; B — irradiated graphite blocks in the form of an assembly of 5 × 5 blocks emplaced into containers (inner and outer protective containers); C — an irradiated graphite block installed in the center of a compact assembly involving 5 × 5 unirradiated blocks; D — irradiated graphite block installed in the center of a peripheral row of a compact assembly featuring 5 × 5 unirradiated blocks

The cases shown in Figures 8C and 8D can possibly occur during graphite stack dismantlement since the distribution functions of blocks according to their dose characteristics (Figure 5) indicate that the dose rate for ~ 10 % of the blocks exceeds by 1—2 orders of magnitude the one characteristic for the majority of the blocks. Therefore, after being packed with a fairly high probability, one container will feature several blocks with higher dose characteristics.

Figure 9 shows the modeling results for a gamma-ray detector's relative response depending on different mutual bracing of an irradiated graphite block in a graphite assembly and a source represented by a surface layer of ~ 5 mm.

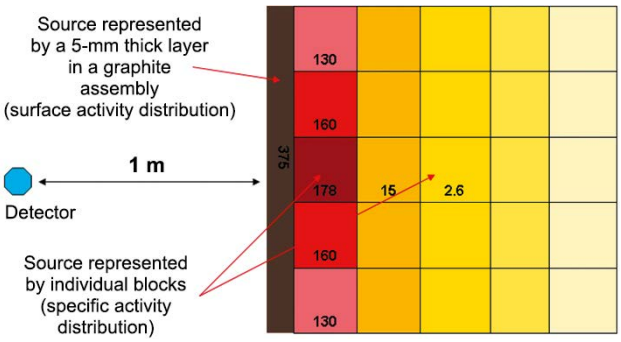

Figure 9. Detector response (recorded radiation intensity) to gamma radiation sources (^{60}Co, 10^7 gamma quanta) represented by a surface graphite layer (5 mm) or a single irradiated graphite block assuming its different orientation within graphite assembly (the values showing the detector response from the corresponding radiation source are indicated in the cells, pulses)

As can be seen in this figure, when the irradiated block was located in the center of the assembly (Figure 8C), the detector's response amounted to 2.6 pulses only, which is ~ 70 times less than the response of the detector in case when this block was located at the periphery (Figure 8D). Given a uniform activity distribution over the entire volume of the assembly (Figure 8A), the response amounted to 30 pulses.

The above examples indicate that due to self-absorption effects and those associated with variations in the distance between the blocks and the detector, it is the exposure coming from the peripheral row of blocks that is basically contributing to the overall response (by over 90 %). These effects can be used to optimize the process of block emplacement into containers (in terms of the gamma-emitting radionuclide activity or DER), thus, minimizing the EDR on the surface of a package (container).

Figure 9 also demonstrates the effect produced on the overall contribution to the surface contamination.

Responses from surface contamination and from the peripheral row of blocks were compared. It was shown that surface contamination can contribute to an overestimated result depending on the ratio between the surface layer activities and the volume.

Under the study, the influence of gamma radiation scattering and absorption effects was evaluated, as well as the uneven distribution of activity due to its surface component assuming the application of a standard (generally accepted) approach to radioactive waste certification (Figure 10).

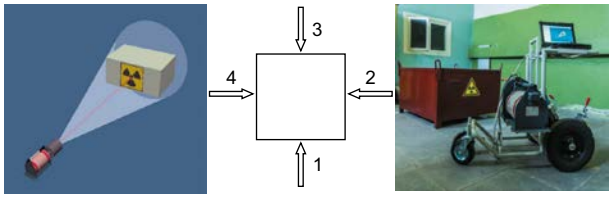

Figure 10. Measurement diagram for a mobile detector (top view) presenting its four positions and involving a ISOCS gamma spectrometric system

According to this approach, the exposure emitted by a filled rectangular container is detected. Measurements are made from four sides with subsequent averaging. Although it allows to reduce the influence of uneven activity distribution on the result, it appears to be insufficient in case of measurements involving items with pronounced unevenness (for example, in case of graphite RW).

Thus, the modeling has shown that if the heterogeneity in the position of a high-activity block was not accounted for in the measurements, the recorded deviation in the ^{60}Co activity would vary in the range between –93 % to +150 %.

As a rule, relevant manuals note that this method cannot be applied in case if container activity is highly heterogenous. The influence of this factor on the results of the packaging certification according to gamma-emitting radionuclides can be significantly reduced if the blocks are characterized individually immediately before packaging.

Thus, to take advantage of the demonstrated effects (Figures 8, 9) and to minimize the radiation dose rate from the filled packages (containers), it seems feasible to characterize the graphite RW at the stage of its generation. At the same time, it is advisable to apply measurement techniques (MT), which would guarantee that the results are never underestimated, but the influence of factors causing the overestimation of actual values is minimized. An overly conservative approach to measurements may entail unreasonable costs due to the application of higher class package (container) designs.

Optimal procedure for container filling and certification of packages with dismantled graphite blocks according to gamma-emitting radionuclides

The experience of RW management accumulated to date both in Russia and globally indicates that the radionuclide composition and the activity characteristics of filled containers are most commonly identified based on the measured and evaluated spectrum of gamma radiation emitted by an already filled container (see Figure 9). However, the deviation in the evaluated average volume and the total activity of the container from the actual values depends significantly on the uniformity of distribution over the volume of the measured item, which is seen as the main disadvantage of this method. Some inhomogeneities (azimuthal and axial) can be accounted for via some measurement techniques: preliminary segregation by dose rate, multi-detector layouts, investigation from several sides, rotation of the object.

Commonly such a certification flowchart for containers with SRW based on gamma-emitting radionuclides is considered as the simplest and most effective option if the RW fragments are available in the form of chaotic scrap and they are packed into a container in bulk. In this case, the waste is accepted at the containerization station in separate batches along the execution of dismantlement or other operations. Prior to packaging, RW is subject to some preliminary control according to the EDR levels: RW fragments with EDR higher than the established level are removed from the main waste volume and are subject to further containerization in packages of a higher class.

Features analyzed in the previous section associated with the specific aspects of graphite stack dismantlement technology, the geometric shape of the blocks, the distribution of gamma-emitting radionuclides over the volume of graphite stacks and some individual blocks, as well as physical characteristics of graphite responsible for the attenuation of gamma radiation fluxes give reason to recommend a special procedure for container filling and the certification of packages with dismantled graphite blocks according to gamma-emitting radionuclides.

The method enabling the characterization, segregation and containerization of graphite blocks extracted during UGR dismantlement developed by JSC PDC UGR [10] is based on a three-coordinate device that provides remote gripping, removal of irradiated graphite elements directly from the reactor stack or an intermediate reservoir excluding any risks associated with any damage and their

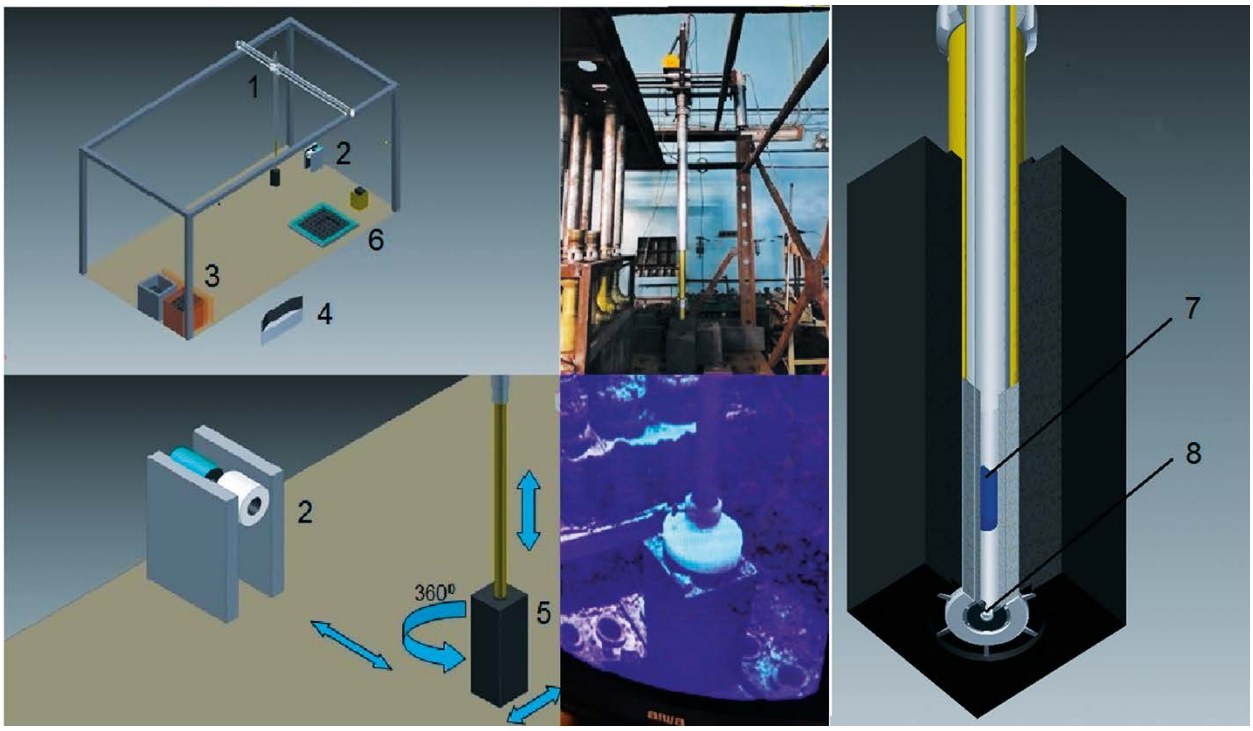

Figure 11. Flowchart providing the measurements of gamma radiation emitted by a graphite block [10]: 1 — device enabling gripping, lifting, rotation and transportation; 2 — spectrometric complex based on a high-resolution HPGe detector; 3 — containers (packages); 4 - remote control; 5 — removed graphite block; 6 — penetration; 7 — low resolution detector (NaI or CdZnTe); 8 — grip tip with a video camera

transportation to a chosen site (Figure 11). The device was successfully tested during graphite block removal from PUGR ADE-5 stack.

During block removal operations, the load is monitored using a digital dynamometer, which is also used for mass measurements. Operator using a video image from a digital video camera provides accurate gripper alignment towards the joint between the blocks. The design of a purposely designed gripper and the automation of the process at all stages of graphite block management provides safety, in particular, when it comes to them management of cracked and highly active blocks.

The activity of gamma-emitting radionuclides is measured in two stages (Figure 11):

Stage 1. Preliminary measurements of radionuclide composition are performed via NaI or CdZnTe detector located inside the head of a block gripping device (see Figures 11, 12) immediately after its removal from the stack. During these measurements, the point detector is located within a hole drilled inside the graphite block. In this case, the head of the gripping device acts as a filter for the interfering low-energy radiation. The estimated result from this stage is used to adjust the geometry and/or the length of measurements implemented at the second stage.

Stage 2. High-precision measurements of the block are carried out using HPGe detector. At the same time, considering previously obtained information, such operations as weighing, installation of a graphite block at a certain distance from the spectrometer, as well as its rotation and positioning in height are performed remotely. The result from this stage can be used to optimize block positioning in a container and during package certification after its filling.

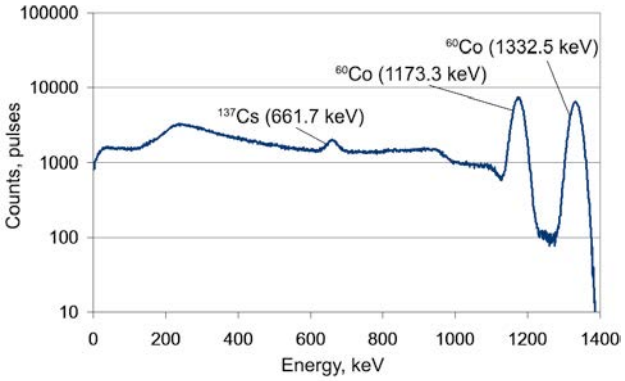

Figure 12. Gamma radiation spectrum from a graphite block calculated using a low-resolution detector during the measurements performed inside the block

This device provides significant reduction in the block segregation time, their remote removal, characterization and subsequent packaging.

An ordered staged block removal during layer-by-layer dismantlement of a graphite stack using

a three-coordinate device (Figure 11) allows to arrange the process of their sequential characterization assuming their further emplacement into containers [10]. It is performed according to a method and a flowchart described and experimentally tested under this study (see the section "Experimental testing ...").

In this case, the passport characteristics according to gamma-emitting radionuclides inside a filled package can be identified based on the following the expression:

$$A^{\Sigma} = \sum_{i=1}^{L} A_i, \quad (1)$$

where L is the number of removed blocks in a package (container);

A^{Σ} is the total radionuclide activity of blocks in a package (container), Bq;

A_i is the radionuclide activity in the i-th block emplaced into a package (container), Bq.

The average specific activity of a package is calculated based on A^{Σ} and the total mass of the blocks in the container.

The proposed method does not require any significant time input associated with the measurement of each block and arrangement of a separate containerization and certification station. The entire sequence of operations can be arranged for using protective enclosures directly installed in the central hall of a reactor, since such manipulations as block gripping, its extraction from the stack, transportation to a research site, measurement and unloading are performed using a three-coordinate device (Figure 11) and are viewed as stages of a single process operation [10].

Opportunities for arranging an optimal spatial layout of blocks in a container depending on their individual radiation characteristics and providing EDR minimization are seen as an important advantage of the recommended flowchart.

As shown above (Figures 4, 5), the content of gamma-emitting radionuclides and, accordingly, the EDR of graphite blocks in RBMK-1000 stack varies in a fairly wide range. At the same time, physical indicators demonstrating the attenuation of gamma radiation in graphite can be effectively used as a screen shielding particular blocks with the highest activity if they are spatially located within the package volume. Their share is nevertheless relatively low (~ 10 %, see Figure 4, 5), i.e., on average, a package of 50 blocks will account for ~ 5 such blocks. When 50 blocks are placed in 2 layers of 5 × 5 pcs. into a container with ~ 5 blocks with high EDR level being located in the center, the contribution of the radiation emitted by the latter ones to the resulting EDR on the package surface can be reduced by up to ~ 70 times.

The problem referring to a most optimal block layout inside a container and its identification can be most effectively addressed through the use of digital technologies and numerical algorithms. These can be used to minimize the EDR from a package given a certain sample (number) of graphite blocks with known radiation properties (identified during block characterization). It also helps to automate container installation process according to some coordinates specifying the position of each block that are predetermined based on optimization calculations. If the most acceptable block positioning does not provide an adequate EDR being below the established limits, the most active blocks are removed from the selected sample and are emplaced into the next package. The configuration and the number of blocks that are put aside is calculated based on a certain algorithm involving sequential (one by one) removal of most active blocks from the entire sample until the EDR no longer exceeds the established limit. If the dismantlement process results in the accumulation of such "put aside" blocks which exceeds a certain level, then it is necessary to provide for their emplacement into "higher" class containers (providing more effective radiation shielding). In practice, the process can be implemented according to the below block diagram (Figure 13).

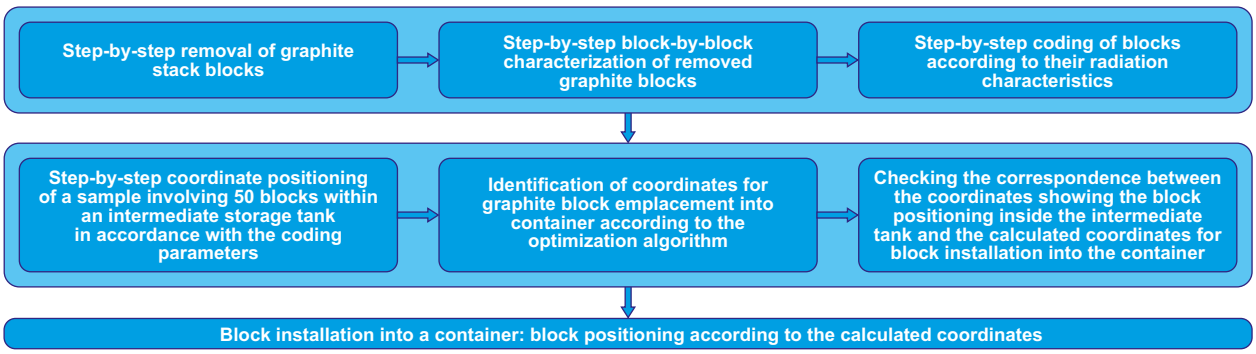

Figure 13. Block diagram providing the automation of processes associated with container filling and certification of packages with dismantled graphite blocks according to gamma-emitting radionuclides

Experimental testing of a method providing the characterization of graphite blocks removed from UGR

Radiation properties of some blocks removed from different heights of the PUGR ADE-5 graphite stack were studied in detail. These were considered as objects of measurements providing the experimental refinement of the characterization technique. The studies involved some sampling operations (Figure 14).

For an extracted block (see Figure 14), the specific activity of ^{60}Co was found to be equal to ~ $6.9 \cdot 10^3$ Bq/g. This value was calculated by averaging the distribution shown in Figure 3. Considering the increased concentration of the isotope in the near-surface layer (~ 10^5 Bq/g), the ^{60}Co content averaged over the block volume amounted to ~ $7.5 \cdot 10^3$ Bq/g. The specific activity of ^{137}Cs (~ 30 Bq/g calculated as the arithmetic mean over the samples taken from the block volume) is low compared to ^{60}Co. However, in the near-surface layer (< 1 mm) its content appears to be statistically significant amounting to up to $2 \cdot 10^3$ Bq/g. When the surface and volumetric contents are averaged over the entire block volume, the specific activity for ^{137}Cs averaged to ~ 70 Bq/g. In addition to ^{60}Co and ^{137}Cs, the studied samples were also found to contain some minor amounts of ^{134}Cs and ^{133}Ba.

It should be noted that the radionuclide composition, activity levels and features of the volumetric radionuclide distribution in a selected block are considered characteristic for the spills of irradiated fuel element material being located outside the containment areas, i. e., for the absolute majority of graphite blocks (both PUGR and RBMK-1000 reactor units).

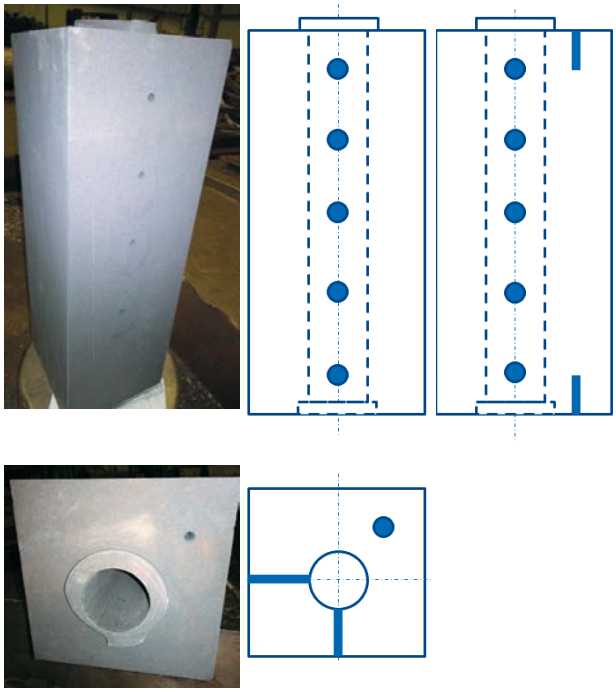

Figure 14. Graphite block removed from the graphite stack of PUGR ADE-5 after sampling: side wall and end surface (left); sampling flowchart (right)

To refine the methodology applied to characterize individual graphite blocks by gamma-emitting radionuclides, the block described above was used. The ISOCS gamma-spectrometric system was used to measure and process the spectra. Figure 15 presents the measurement flowchart.

The emission spectra from a block extracted from a PUGR ADE-5 graphite stack were measured

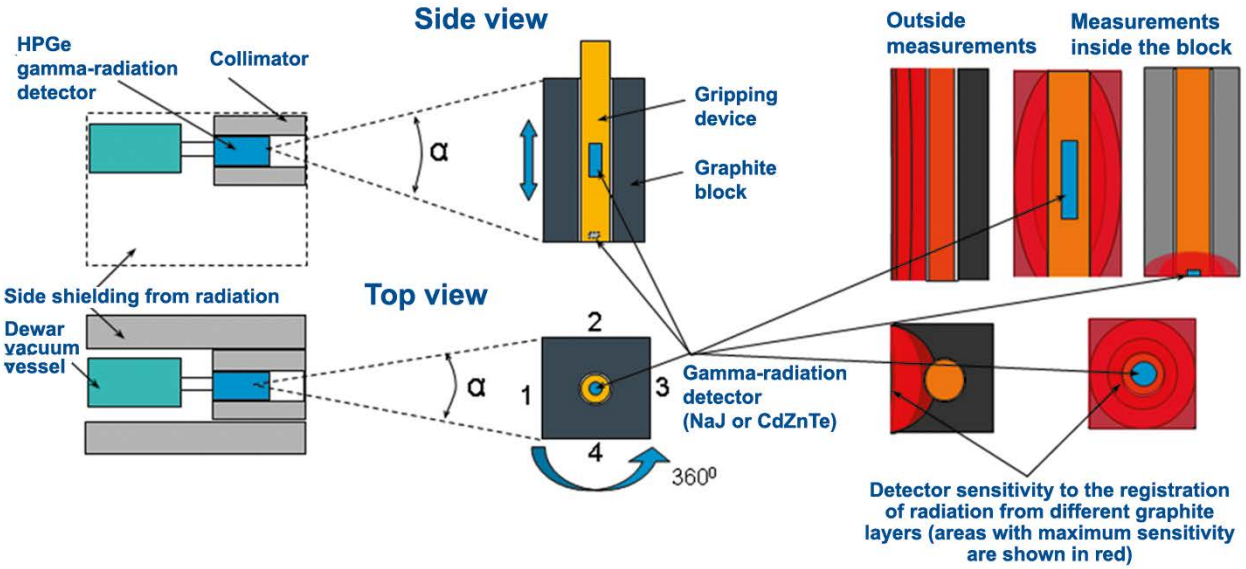

Figure 15. Flowchart for the measurement of gamma radiation spectra from a graphite block involving high (HPGe) and low (NaI or CdZnTe) resolution detectors

considering four fixed positions (the detector axis was perpendicular to each face of the block) and continuous rotation (~10 full revolutions of the block during the measurement time), their duration accounted for ~5 min. At the same time, the intensity of the spectrum gain allows to reduce this time to 1 minute without any significant accuracy decrease. Figure 16 presents the characteristic gamma radiation spectrum from the block under given measurement conditions.

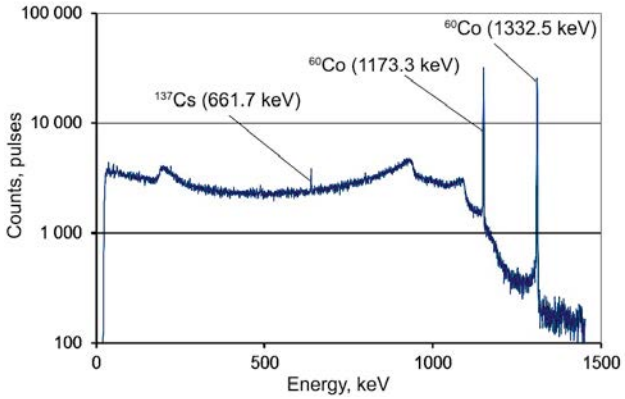

Figure 16. Characteristic gamma radiation spectrum for a graphite block removed from PUGR ADE-5 graphite stack

Table 2 summarizes measured average specific activities for a graphite block extracted from PUGR ADE-5 graphite stack. These values were obtained from the characterization performed using the ISOCS spectrometric complex.

Table 2. Average 60Co specific activity in a graphite block with relevant measurements performed using the ISOCS spectrometric complex

№	Block position in relation to the detector	Average specific activity of the block, Bq/g	Deviation from the level obtained for the samples, ±%
1	Face 1	$8.6 \cdot 10^3$	+15
2	Face 2	$9.1 \cdot 10^3$	+21
3	Face 3	$9.6 \cdot 10^3$	+28
4	Face 4	$7.9 \cdot 10^3$	+5
5	Continuous block rotation	$8.5 \cdot 10^3$	+14

The deviations of the average ^{60}Co specific activity in the block from the levels measured ranged from +5 to +28% with a single measurement error of <5%. This result (statistically significant systematic positive deviation) is explained by the fact that the calibration of the ISOCS system according to the registration efficiency is based on a uniform radionuclide distribution over the volume of the measured item (block). The increased surface content of ^{60}Co recorded in the samples (see above) due to the absence of the attenuation effect in the block material can potentially contribute to the measurement result. Influence of this effect is also traced in case of specific activities measured during the analysis of the selected samples, namely, the ratio between the specific activity in the near-surface layer and the average ^{60}Co specific activity amounted to ~15.

^{134}Cs and ^{133}Ba isotopes were not identified in the measurements performed using the ISOCS spectrometric complex since the spectrum from the entire block was characterized by a higher fraction of scattered radiation ("Compton substrate") than the one from a graphite sample taken from the same block with a mass of ~0.1 g. These radionuclides are distributed over the entire volume of the block, and their radiation contribution is negligible as compared against the scattered radiation background.

Thus, the experimental characterization of irradiated graphite blocks using the ISOCS spectrometric complex has demonstrated the potential for the practical application of this method. The calculated deviations in the ^{60}Co specific activity indicate that this method provides certain conservative margin for their activity. At the same time, the level of conservative overestimation is considered acceptable and would not affect RW categorization process. The degree of conservatism depends on the ratio between the surface contamination and the average volumetric content.

This effect may appear to be most pronounced in case of "fuel origin" isotopes (in particular, 137Cs), which is considered as a general tendency characteristic for the UGR graphite contamination with radionuclides during reactor operation [11]. It should be borne in mind that when the uniform radionuclide distribution model is applied over the measured block volume (as in case of ^{60}Co) for ^{137}Cs, the deviation between the average specific activities in the block and those measured in the samples will range from +43 to +160% with a single measurement error of <10%. The latter is explained by the fact that due to the absence of the attenuation effect in the block material, the increased surface content provides positive contribution to the measurement result (to a lesser extent in case of ^{60}Co, to a greater extent in case of ^{137}Cs).

Considering relatively low 137Cs content in the graphite block, as in our case, the conservative estimate seems to be quite acceptable. However, in cases of a high ^{137}Cs content in graphite (from the zones of "fuel" origin radionuclide localization, Figure 1), this problem requires some research to

increases the measurement accuracy by taking into account the nature of the radionuclide localization.

Optimal physical parameters for graphite RW containers

Under disposal conditions, potential hazard associated with graphite RW depends on the residual content of long-lived radionuclides (^{14}C, ^{36}Cl, actinides) and the parameters characterizing their potential leaching rate from the RW material in mobile forms. Under these conditions the safety relies on the level of protective functions provided by the RWDF itself and the external conditions (geophysical, geochemical, hydrological, etc.). Therefore, it would be incorrect to consider the containers as a factor providing the delay of radionuclides for the entire period while the RW potentially remains hazardous. For this reason, during graphite stack dismantlement followed by the RW management, the main purpose of the containers is to ensure:

- tightness avoiding any releases of radionuclides during a time period that starts from the point of container filling and lasts up to its emplacement into the disposal facility;
- radiation protection (EDR level on the container surface should not exceed the established limits) provided for the same period of time.

According to the experimental data provided by NRC Kurchatov Institute [12] and JSC PDC UGR (studies performed in 2020—22021), by the time of RBMK-1000 reactor final shutdown, ^{60}Co content variability in graphite stack blocks will amount to 10^2—10^6 Bq/g (Figure 4). For the main portion of blocks (~85 %) it will be equal to 10^3—10^5 Bq/g (Figure 4). Abnormally high levels of 10^5—10^6 Bq/g would be typical for ~10 % of blocks located within the stack volume. The average value over the entire distribution function will amount to $4.8 \cdot 10^4$ Bq/g. In ~10–15 years after the reactor final shutdown, which is considered as a characteristic time, these values will decrease by ~4—8 times. For obvious reasons, practical dismantlement efforts can be launched directly at the reactor site not earlier than this date. Thus, by the start of the dismantlement operations, the average ^{60}Co specific activity in RBMK-1000 reactor graphite stacks will decrease to $6.0 \cdot 10^3$—$1.2 \cdot 10^4$ Bq/g. It should be noted that most of the blocks do not appear to be contaminated by "fuel" origin radionuclides to any significant level, and, therefore, the EDR level will be depend solely on the ^{60}Co isotope, whereas the ^{137}Cs contribution will be negligible.

[13] provides calculations for the EDR on the surface of a container accounting for the RW geometry corresponding to the useful capacity of NZK-150-15P containers. In these calculations, simulated RW density was taken equal to ~2 g/cm^3 (similar to the graphite density ranging from 1.65 to 1.7 g/cm^3). According to the results obtained, to comply with a condition suggesting that the EDR on the surface does not exceed 2 mSv/h, no additional radiation shielding is required when the uniformly distributed RW activity (^{60}Co) over the container volume falls within a range between $6.0 \cdot 10^3$—$1.2 \cdot 10^4$ Bq/g. Thus, in this case, it is sufficient to provide the compliance with the requirements on container tightness and performance. Provided that the container design features an insert and an external container, the thickness of the steel walls should be at least 5 mm to ensure sufficient container strength and rigidity. Considering such steel thickness (10 mm), the permissible averaged specific activity of ^{60}Co in graphite RW will amount to ~$1 \cdot 10^5$ Bq/g.

Various graphite block packaging options are currently considered, which is due to a low filling factor of NZK-150-1.5 container with a CM500 insert that has been developed specifically for graphite RW. The internal net volume of the NZK container is only 1.3 m^3 with an external volume of 3.74 m^3. At the same time, the disposal (storage) cost depends on the external dimensions.

Different container options were evaluated [14] from an economic point of view and it was concluded that in case of graphite waste it is advisable to use returnable metal containers with non-returnable removable parts (inserts) required to provide the radiation safety during waste transportation to the disposal site. Whereas the insert is required for further graphite RW emplacement into the disposal facility. At the same time, the designs should provide opportunities for the application of already existing container handling infrastructure. In order to increase the filling factor, it is also advisable to stack the graphite blocks in two layers (Figure 17).

If RBMK-1000 graphite blocks are positioned according to the above layout (2 layers, 5 × 5 pcs.), the net RW volume would amount to 1.73 m^3. Taking into account these recommendations and the results of experimental block-by-block characterization (see the previous section), a metal container featuring an external container BUK-3.1 and an insert BUK-1.9 was used under this study.

BUK-3 container design involves some fitting elements providing the installation of BUK-1.9. BUK-3.1 (Table 3) designs also involve some elements required for its transportation and necessary loading-unloading operations. Considering its dimensions, the BUK-1.9 insert can be used to accommodate the following number of graphite blocks: 72 pcs. of PUGR blocks or 50 pcs. of RBMK-1000 blocks.

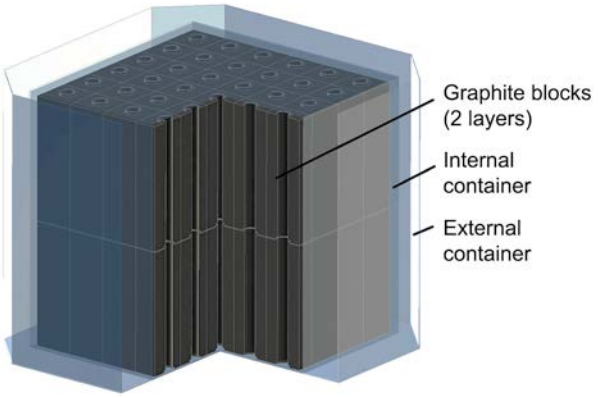

Figure 17. Layout providing most optimal positioning of graphite blocks in containers

Table 3. Design parameters of the outer and inner containers

Container parameters	BUK-3.1	BUK-1.9
Overall dimensions, no more than, mm:		
height	1 375	1 240
length	1 640	1 640
width	1 640	1 640
Wall thickness, mm	5	5

To ensure certain conservative EDR margin on the container surface, the design provides opportunities for the application of thicker steel sheets in the designs of BUK-1.9 and/or BUK-3 .1 body frames.

Following the segregation stage, to provide the characterization of BUK containers with graphite RW in the form of scrap blocks, bushings, graphite rings, graphite chips, a standard approach can be applied (Figure 9). It can also provide for the application of spectrometric systems such as ISOCS and domestic SKS-07P according to both single-detector and multi-detector measurement flowcharts. Rotation during the certification can be provided by the POST-K10 turntable (Figure 18).

Figure 18. Containers BUK-1.9 (left), BUK-3.1 (center), turntable POST-K10.5 (right)

Conclusion

The proposed and experimentally tested methodological approaches and an algorithm providing graphite RW characterization by gamma-emitting radionuclides during UGR dismantlement have demonstrated the adequacy of their application and satisfactory metrological characteristics allowing the categorization of individual graphite blocks under the package (container) certification process implemented during container filling.

The evaluated specific features of gamma-emitting radionuclide distribution in graphite UGR stack blocks, as well as the results obtained from the testing of the proposed graphite RW containerization and package certification process have indicated the feasibility of block characterization at the packaging (containerization) stage. Application of the proposed flowchart and measurement techniques allows to avoid unreasonable overestimation of the actual activity and EDR levels, which may result in unreasonable expenses associated with the application of higher-class packages (containers). In this case, physical foundations of the methods applied and the characteristic features governing spatial distribution of radionuclides in dismantled graphite blocks ensure that the measurement results are never underestimated. Block categorization by the content of gamma emitters provides their optimal layout inside packages minimizing the EDR from the container. For the selected packages, it also allows to avoid potential exceedance of radiation levels according to the limits set forth in the regulatory provisions.

Under this study, considered were the container designs being similar to the optimal ones and providing the compliance with such requirements.

The presented approach was integrated into a system designed for graphite block handling (segregation, containerization, etc.) and is considered as an important element of engineering solutions providing the dismantlement of graphite stacks and the characterization of graphite RW patented by Rosatom.

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Information about the authors

Pavliuk Alexander Olegovich, PhD, head of the group, "Pilot & Demonstration Center for Decommissioning of Uranium-Graphite Nuclear Reactors", JSC (Building 179, 13 Avtodoroga, Seversk, Tomsk Region, 636000, Russia), e-mail: pao@dnrc.ru.

Kotlyarevsky Sergey Gennadievich, leading engineer, "Pilot & Demonstration Center for Decommissioning of Uranium-Graphite Nuclear Reactors", JSC (Building 179, 13 Avtodoroga, Seversk, Tomsk Region, 636000, Russia), e-mail: ksg@dnrc.ru.

Kan Roman Igorevich, engineer-physicist, "Pilot & Demonstration Center for Decommissioning of Uranium-Graphite Nuclear Reactors", JSC (Building 179, 13 Avtodoroga, Seversk, Tomsk Region, 636000, Russia), e-mail: kri@dnrc.ru.

Volkova Anna Genrihovna, PhD, Senior Researcher, Federal State Budgetary Institution of Science A. N. Frumkin Institute of Physical Chemistry and Electrochemistry of the Russian Academy of Sciences (31, Leninsky Av., Moscow, 119071, Russia), e-mail: anna.agv@yandex.ru.

Zakharova Elena Vasil'evna, PhD, Head of the Laboratory, Federal State Budgetary Institution of Science A. N. Frumkin Institute of Physical Chemistry and Electrochemistry of the Russian Academy of Sciences (31, Leninsky Av., Moscow, 119071, Russia), e-mail: zakharova@ipc.rssi.ru.

Komarov Evgenij Alekseevich, Senior Manager of the Decommissioning department, State Corporation "Rosatom" (24, Bolshaya Ordynka St., Moscow, 119017, Russia), e-mail: EAKomarov@rosatom.ru.

Bibliographic description

Pavliuk A. O., Kotlyarevsky S. G., Kan R. I., Volkova A. G., Zakcharova E. V., Komarov E. A. Approaches to graphite block characterization by gamma radionuclides and their packaging during UGR stack dismantlement. *Radioactive Waste,* 2021, no. 3 (16), pp. 30—43. DOI: 10.25283/2587-9707-2021-3-30-43. (In Russian).