

RADIONUCLIDE CHARACTERIZATION OF GRAPHITE STACKS FROM PLUTONIUM PRODUCTION REACTORS OF THE SIBERIAN GROUP OF CHEMICAL ENTERPRISES

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Abstract. The residual radionuclide concentrations and distributions in graphite from moderator stack of plutonium production reactors at Tomsk-7 have been investigated. It was found that the dominant activity of graphite is ^{14}C . To gain information on surface and volume contamination of graphite blocks from the moderator stack, the special sets of samples were collected and assayed. The schemes are proposed for evaluation of individual radionuclide inventories together with results of the evaluations performed.

1. INTRODUCTION

Till now in Russia, ten plutonium-production uranium-graphite reactors (PUGRs) have been decommissioned. Spent graphite from PUGRs has been contaminated with radionuclides and represents an important fraction of nuclear power industry radiowastes. For proper selection of ways and times for this graphite management, the information is needed about composition and level of the graphite radioactive contamination.

The conditions of PUGRs operation are as follows: duration of the reactors operation was about 30 years, graphite was placed into nitrogen atmosphere, several accidents with destruction of technological channels and multiple leakages of light-water coolant into the graphite stack have taken place. As a consequence, there was formed the radioactive contamination with complex composition and volumetric distribution. Two sources of radioactive contamination existed: activation of impurities and penetration of radionuclides during accidents. Quantitative evaluations of both sources were hampered because available information about impurities content was incomplete and inaccurate while penetrations of radionuclides were of accidental and casual nature.

The report presents results of the studies carried out at the I-1 and EI-2 reactors. Currently, experiments at the ADE-3 reactor are being performed.

The tasks of the studies included:

- to obtain general picture of radionuclide distribution in the reactor stacks, to reveal and to explain relationships in distributions of individual radionuclides;
- to elaborate the approaches to evaluation of radionuclide inventories in individual components and in the stack as a whole, to construct the evaluation schemes and to check them;
- to present the schemes and recommendations on planning of taking the samples for evaluation of radionuclide inventories at other nuclear reactors;
- to determine the possibilities for rapid sorting of the graphite blocks depending on their contamination level under dismantling of the stacks.

The graphite reactor stack consists of graphite blocks stacked to provide a central region for fuel loading and an outer region for a neutron reflector. Total mass of graphite in the reactor cores was about 900 tones.

For sampling from the stacks, the special technology was applied. Using the tool transportable from one channel to another, the graphite cores were taken at specified depth by means of horizontal drilling through full thickness of the block. So, length of the graphite cores covered distance from inner hole of the block to its lateral surface. Using a milling cutter, five samples were cut out from the graphite cores, at 12 mm distance one from another, for subsequent conduction of assays (Fig. 1). The samples represent the disks, 8 mm in diameter, 2 mm thick and mass of about 200 mg.

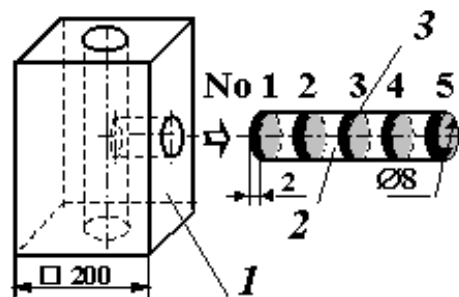


FIG. 1. Scheme of sampling: 1-graphite block; 2-graphite core; 3- sample; No1-5 - numbers of samples.

In the studies the most attention was given to radionuclides - major contributors to contamination, namely to β -emitters (^{14}C and ^3H were the main isotopes of them), to actinides, to fission products and ^{60}Co . The radionuclides from these groups differ with their origination and distribution in graphite.

The complexity of assays on content of individual radionuclides differed very much. For example, assays on content of tritium and actinides required rather long time periods (up to 10 hours or more) while measurements of some fission products or activation products were very simple and low labor-consuming.

In evaluations of radionuclide inventories in the graphite stack, the importance of principle belongs to issue about sufficient scope of sampling (the number of taken and assayed graphite samples). This quantity was limited by duration and complexity of assays as well as by peculiarities in distribution of some radionuclides in volume of the stacks.

Taking into consideration the capabilities available, the problem may be defined as follows: 1. Is it possible to determine radionuclide distribution along height of the cell (7.6 m) with acceptable accuracy using measurement results for 3-7 points at different levels as an input information ? 2. Is it possible to describe radionuclide field in full stack (2000 cells) using measurement results for 20-30 cells as an input information ? Magnitude of errors may depend on the fact whether radionuclide distribution obeys to any certain relationship or not. Such a relationship may be further used for interpolation. If radionuclide distribution does not obey to any certain relationship or if this relationship is distorted, then radionuclide inventory in the stack may be determined as sum of inventories in individual cells with preliminary joining the cells with close values of contamination levels into groups: the cells adjacent to accidental ones, the cells disposed far enough from accidental ones, reflector cells and so on.

To obtain general picture of contamination as a whole, it is necessary to investigate axial and radial distributions of radionuclides in the stack and local distributions in vicinity of accidental cells. Obtaining such an information will require to make assays for no less than 160-200 samples from each reactor, and about 500 graphite samples from three reactors.

The methods used for contamination assays of the graphite samples are presented in Table I.

TABLE I. METHODS FOR MEASUREMENT OF RADIONUCLIDE CONTENT IN GRAPHITE

Radionuclides	Method
Beta-emitters: ^3H , ^{14}C , ^{36}Cl , ^{63}Ni , ^{90}Sr	Liquid scintillation spectrometry (LSS); Radiochemistry and LSS; Measurement of Vavilov-Cerenkov radiation.
Gamma-emitters: ^{60}Co , ^{133}Ba , $^{134,137}\text{Cs}$, $^{152,154,155}\text{Eu}$	γ -spectrometry with application of HPGe- and Ge(Li)-detectors
Alpha-emitters: $^{238,239,240}\text{Pu}$, $^{241,243}\text{Am}$, ^{244}Cm	γ /X-spectrometry with application of LEGe-detectors; Radiochemistry and α -spectrometry with application of Si-detectors.

2. DETERMINATION OF ^3H , ^{14}C , ^{36}Cl , ^{63}Ni AND ^{90}Sr CONTENT

The method used in measurements of ^3H and ^{14}C contents in the reactor graphite samples was presented in Ref.[1]. The method is based upon chemical separation of radionuclides from the graphite sample being assayed in gaseous form with subsequent transition into liquid phase and measurement of activity by means of liquid scintillator. The mass of the sample under investigation was optimised from viewpoint of obtaining reliable data about ^3H and ^{14}C content in the reactor graphite and reducing the expenses for assays. As a result, selected value of the sample mass was equal to about 0.1 g.

In measurements of ^{36}Cl and ^{63}Ni content in graphite it was applied the radiochemical method of radionuclide separation from solution of graphite followed by measurement of its activity by means of liquid scintillation radiometry technique.

^{90}Sr activity was determined via activity measurements of its daughter radionuclide ^{90}Y - emitter of high-energy β -radiation ($E_{\text{max}} = 2.2839 \text{ MeV}$). In equilibrium state, ^{90}Sr activity is equal to ^{90}Y activity. Two different methods were applied to measurement of ^{90}Y activity. One method includes pulverisation of the sample with subsequent measuring the β -activity of thin graphite powder layer by means of plastic scintillation detector. Another method is based upon dissolution of the graphite sample, separation of ^{90}Y from the solution and subsequent measuring the ^{90}Y accumulation by means of Cerenkov counter.

Errors in determination of activity for radionuclides-emitters of β -radiation in graphite via applications of aforementioned methods did not exceed 20%. About 40 graphite samples were taken from the stacks of both reactors and assayed for determination of ^3H and ^{14}C contents. Two hundred samples were assayed for determination of ^{90}Sr content and five samples - for determination of ^{36}Cl and ^{63}Ni contents.

Typical distributions of ^3H , ^{14}C and ^{90}Sr contents in accidental cell 2516 of the I-1 reactor and in the cell 1425 distant from accidental one are presented in Fig. 2.

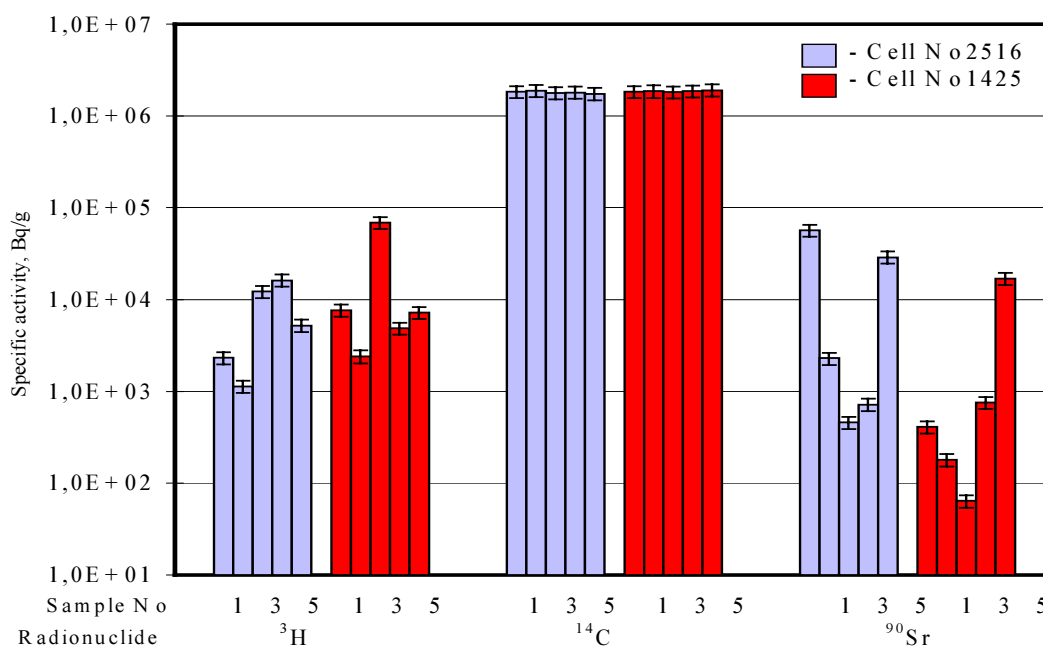


FIG. 2. Specific activity of radionuclides in the samples taken from blocks of the I-1 reactor.

Radionuclide ^{14}C is generated in the reactor graphite due to two main neutron reactions: $^{13}\text{C} (n,\gamma)^{14}\text{C}$ and $^{14}\text{N} (n,p)^{14}\text{C}$. Nitrogen in graphite is in bound state and as a gas filling up the graphite pores. Porosity of the reactor graphite is about 26%. Content of chemically bound nitrogen in unirradiated reactor graphite was evaluated with application of high-temperature extraction technique. This value appeared to be (31 ± 5) ppm in unirradiated graphite of the I-1 reactor. Contributions of these two neutron reactions to build-up of ^{14}C in graphite were evaluated, and the evaluation revealed that neutron reaction with nitrogen gave a major contribution.

In Fig. 3 there are demonstrated the results of ^{14}C activity measurements in the samples taken from the cell 2516 at different axial levels. It can be seen from presented results that there is a linear proportionality between ^{14}C content in graphite and thermal neutron fluence.

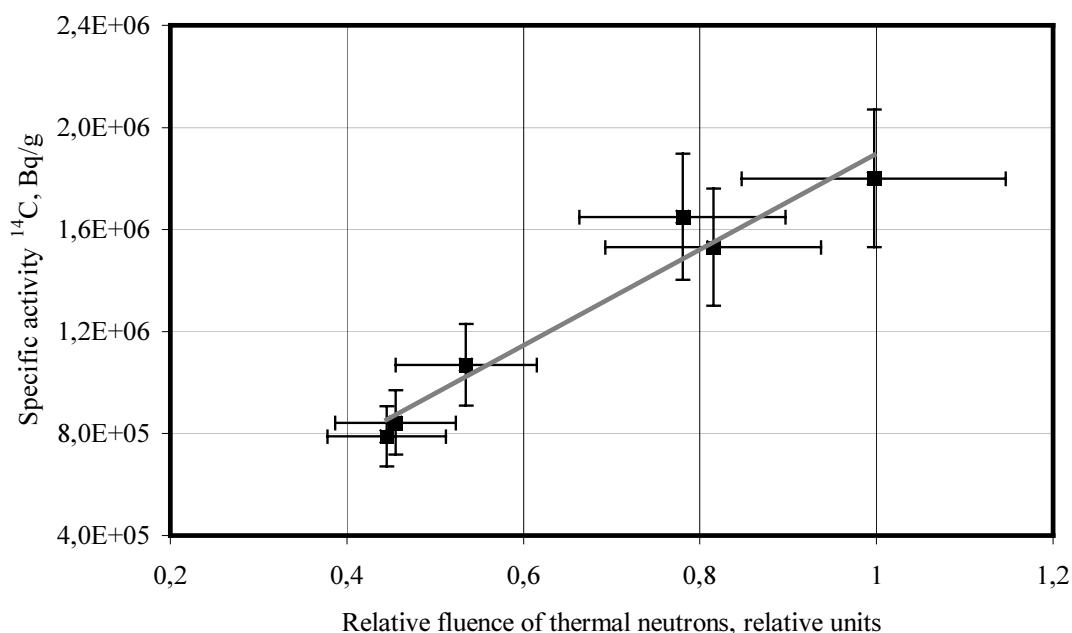


FIG. 3. Dependence of ^{14}C specific activity in the I-1 reactor graphite on relative fluence of thermal neutrons.

The results obtained in assays of the graphite samples enable us to make the following conclusions:

- ^{14}C activity in graphite dominates over activities of all other radionuclides.
- ^{14}C is uniformly distributed in volume of graphite block (within the range of experimental errors).
- Linear proportionality is observed between ^{14}C content in graphite and neutron fluence.

Thus, ^{14}C inventory in the graphite stack of the reactor core can be determined via measurement results for absolute value of ^{14}C specific activity in components of the graphite volume and using the information about neutron fluence distribution in the reactor stack. Such an evaluation performed for the I-1 reactor gave the value $\sim (2.1 \pm 0.6) \times 10^{15}$ Bq, for the EI-2 reactor - $(1.3 \pm 0.4) \times 10^{15}$ Bq.

Main contribution to accumulation of tritium in graphite is given by neutron reactions with ^6Li and ^{14}N nuclei. Generated triton will stay in the graphite matrix with high probability because mean free path of triton is smaller than size of graphite grains and larger than size of pores. However, some fraction of tritons generated near to grain boundaries has no energy high enough to penetrate through pore or gap. Therefore, some fraction of tritium has a possibility to remain in the gas filling up the pores and gaps, and to release with it out of the stack. Tritium initially fixed in graphite may acquire a mobility with time and release into the environment. Apparently, the release of tritium from graphite is caused by graphite oxidation

that leads to oxidation of fixed tritium as well, and tritium is carried by the gas flow blown through the reactor stack. In Ref. [2], it was remarked that content of carbon dioxide in gas circuit for periods of the reactor operation after “wet” accidents has increased. This indicated on the process of graphite oxidation. All these phenomena led to tritium release from the graphite matrix during the reactor operation.

Obtained results demonstrated that tritium was extremely non-uniformly distributed in volume of the stack. It was revealed that distribution of obtained experimental data had a form of logarithmically-normal distribution. So, mean values and their errors were calculated in accordance with such a distribution. Mean specific activity of tritium in the I-1 reactor stack was equal to $(3.4\pm 0.5)\times 10^3$ Bq/g, the same value for the EI-2 reactor was equal to $(5.4\pm 0.8)\times 10^3$ Bq/g. Proceeding from obtained results and graphite mass in the reactor cores, tritium inventories were evaluated for both reactors. These values appeared to be $(3.3\pm 0.5)\times 10^{12}$ Bq and $(5.3\pm 0.8)\times 10^{12}$ Bq for the I-1 and EI-2 reactors, respectively.

In connection with complexity of assays on determination of ^{36}Cl and ^{63}Ni contents in the graphite samples and due to low activity of these radionuclides, only several samples were assayed. Measured values of ^{63}Ni activity in the samples were within the range $(2.5\times 10^2 - 2.2\times 10^3)$ Bq/g, for ^{36}Cl - $(54 - 110)$ Bq/g. Comparison of experimental and calculated data allows to conclude that ^{36}Cl contents in spent reactor graphite are one order of magnitude less than calculated values while ^{63}Ni contents do not contradict to calculated data. Observed difference in values of ^{36}Cl content may be explained by the following consideration. Chlorine in graphite is able to form only weak covalent bonds with carbon or is staying in closed pores (Ref. [3]). During the reactor operation, the chlorine may acquire a mobility and escape from the stack. Proceeding from the data available, the conservative assessment was made, and the following values of radionuclide inventories in the reactor graphite were obtained: ^{36}Cl - $(0.4 - 1.1)\times 10^{12}$ Bq, ^{63}Ni - $(1.9 - 6.9)\times 10^{12}$ Bq.

Table II demonstrates obtained values of radionuclide specific activities in the graphite samples taken from the I-1 reactor stack, calculated predictions (Ref. [4]) as well as some values obtained for graphite from Hanford reactor DR (Ref. [5]). The calculations did not take into account a migration of radionuclides in the reactor graphite. All obtained activity values are presented for a moment of the reactor ultimate shutdown.

TABLE II. MEASURED AND CALCULATED CONCENTRATIONS OF RADIONUCLIDES IN THE REACTOR GRAPHITE, BQ/G

Radionuclide	Experiment		Calculation (Ref. 4)
	I-1 reactor	DR reactor (Ref. 5)	
^3H	$(3.4\pm 0.5)\times 10^3$	4.41×10^5	3.2×10^6
^{14}C	$(1.2\pm 0.2)\times 10^6$	1.31×10^5	4.2×10^5
^{36}Cl	$5.4\times 10^1 - 1.1\times 10^2$	$2.8\times 10^2 - 5.3\times 10^2$	8.8×10^3
^{63}Ni	$2.5\times 10^2 - 2.2\times 10^3$	$5.6\times 10^2 - 1.9\times 10^3$	8.9×10^3

It follows from Table II that calculations underestimated accumulation of ^{14}C in graphite by a factor of 4. An inverse situation is observed for tritium: the calculations strongly overestimated its content by a factor of about 700. Value of ^{14}C specific activity in graphite from the I-1 reactor is about 9 times higher than that in graphite from the DR reactor. Most probably, it may be explained by higher content of nitrogen in the I-1 reactor graphite. Nitrogen was used as a protective gas in the I-1 reactor while carbon dioxide was applied for the same purpose in the DR reactor. Comparison of ^{36}Cl specific activities in graphite obtained in experiments and in calculations indicates that calculations overestimated ^{36}Cl contents by two orders of magnitude. This may be explained by two factors: by overestimated value of initial chlorine content used in calculations and by reduction of chlorine content in graphite during the reactor operation. Evaluation of the second factor was made in Ref. 5 and its value was equal to about 10. To refine initial content of chlorine in the reactor graphite, this value was evaluated with application of neutron activation analysis. Obtained results have demonstrated that the value used in calculations was overestimated by the factor higher than 6. Experimental data on ^{63}Ni content in graphite appeared to be several times less than calculated results. Observed difference may be explained by non-uniformity in content distribution for chemical elements of iron group, nickel in particular, in graphite (Refs.[6-7]).

3. DETERMINATION OF ACTINIDES AND FISSION PRODUCTS

Under the accidents which took place at early stages of PUGRs operation, several kilograms of uranium were dispersed in their stacks. Uranium particles were transported with water vapour and deposited on the graphite surface both at side from technological channels and in slits between blocks. In process of long-term neutron irradiation, dispersed uranium was at first converted into ^{239}Pu . The most fraction of ^{239}Pu was burnt-up with accumulation of fission products while some remaining part, through consecutive neutron captures, was transformed into heavier actinides including isotopes of americium and curium.

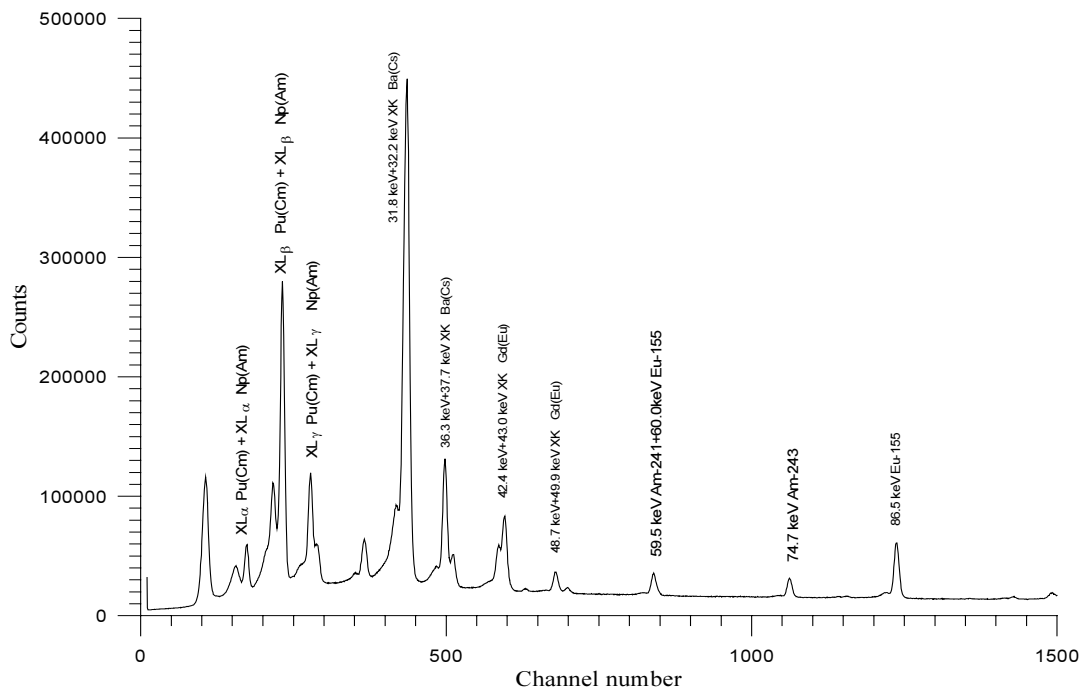


FIG. 4. Gamma- and X-ray radiation spectrum of the graphite sample from block of the EI-2 reactor.

Actinide content in the graphite samples was determined from results of spectrometric measurements with large planar LEGe-detectors. If necessary, continuous measurements were carried out during tens of hours. In spectra of the samples contaminated with actinides, there were observed gamma-lines of ^{241}Am (59.5 keV), ^{243}Am (74.5 keV), and X-ray lines with energies near to 14 keV and 18 keV where XL_α - and XL_β -radiations were summed up which accompany α -decay of Cm, Am and Pu isotopes (Fig. 4). The spectra of highly-contaminated samples were measured by means of small LEGe-detector with higher resolution. It allowed to resolve, completely or partially, XL_β -peaks of Pu, Np and U, and using measured intensities, to determine relative content of Cm, Am and Pu in these samples.

To determine isotopic composition of plutonium contained in graphite, plutonium was separated from the samples highly contaminated with actinides, and sources for spectrometric measurements were manufactured. Using LEGe-spectrometers, spectra were measured within the range 10-200 keV where lines of ^{238}Pu , ^{239}Pu , ^{240}Pu and ^{241}Pu were observed. These spectra were used for evaluating the relative contents of these isotopes.

The radiation emitted by the graphite was also measured within high-energy range with application of coaxial HPGe-spectrometers. In the spectra obtained the lines of several fission products were observed: ^{134}Cs , ^{137}Cs , ^{106}Ru , ^{154}Eu and some others.

Analysis of obtained results led to the following conclusions:

1. The samples taken from lateral surfaces of the graphite blocks were contaminated with actinides in maximal degree. The experiments carried out with several samples taken from lateral side of one block demonstrated that contamination at different parts and at different sides of one block may be considered as uniformly distributed within the range 40-60%. Actinide content in volume of the graphite blocks was lower by several hundred times than that in superficial layer. It concerns both to the cells placed near to (Table III) and distant enough from accidental ones. Radionuclides weakly penetrated into depth of blocks from their surface. Only in individual cases the penetration due to graphite cracking was observed.

TABLE III. ACTINIDE DISTRIBUTION IN THE GRAPHITE CORE (LOCATION OF SAMPLES IS SHOWN IN FIG. 1)

Sam No	I-1 reactor				EI-2 reactor	
	Cell 0320		Cell 2516		Cell 3326	
	$A(^{241}\text{Am})$ Bq/g	$A(^{244}\text{Cm})$ Bq/g	$A(^{241}\text{Am})$ Bq/g	$A(^{244}\text{Cm})$ Bq/g	$A(^{241}\text{Am})$ Bq/g	$A(^{244}\text{Cm})$ Bq/g
1	$(4.30 \pm 0.13) \times 10^2$	$(1.1 \pm 0.1) \times 10^4$	$(2.0 \pm 0.1) \times 10^3$	$(3.4 \pm 0.4) \times 10^3$	$(1.3 \pm 0.1) \times 10^1$	$(1.5 \pm 0.2) \times 10^3$
2	$(1.21 \pm 0.04) \times 10^2$	$(4.2 \pm 0.4) \times 10^3$	$(1.7 \pm 0.1) \times 10^1$	$(4.4 \pm 0.5) \times 10^3$	$(5.4 \pm 0.6) \times 10^0$	$(5.6 \pm 1.2) \times 10^2$
3	$(1.46 \pm 0.06) \times 10^2$	$(5.4 \pm 0.6) \times 10^3$	$(1.4 \pm 0.1) \times 10^1$	$(3.8 \pm 0.4) \times 10^3$	$(5.1 \pm 0.5) \times 10^0$	$(5.3 \pm 0.8) \times 10^2$
4	$(1.50 \pm 0.06) \times 10^2$	$(5.0 \pm 0.5) \times 10^3$	$(1.2 \pm 0.1) \times 10^1$	$(2.3 \pm 0.3) \times 10^3$	$(7.0 \pm 0.4) \times 10^0$	$(7.3 \pm 0.8) \times 10^2$
5	$(4.85 \pm 0.15) \times 10^4$	$(1.8 \pm 0.2) \times 10^5$	$(3.70 \pm 0.15) \times 10^3$	$(6.3 \pm 0.7) \times 10^5$	$(1.00 \pm 0.03) \times 10^3$	$(8.6 \pm 0.9) \times 10^4$

2. It was revealed that, at present time, the main contribution to activity of actinide mixture is given by ^{244}Cm . The correlations between activities of individual radionuclides are presented in Table IV.

TABLE IV. ISOTOPIC RATIOS OF ACTINIDES IN THE GRAPHITE SAMPLES FROM THE I-1 REACTOR STACK

Cell No	Relative value of fluence	$\frac{A(^{238}\text{Pu})}{A(^{239}\text{Pu})}$	$\frac{A(^{240}\text{Pu})}{A(^{239}\text{Pu})}$	$\frac{A(^{241}\text{Pu})}{A(^{239}\text{Pu})}$	$\frac{A(^{243}\text{Am})}{A(^{241}\text{Am})}$	$\frac{A(^{244}\text{Cm})}{A(^{241}\text{Am})}$
0220	0.19	14.4±0.5	2.66±0.11	310±11	0.090±0.005	11.3±1.1
0320	0.38	10.2±1.0	1.41±0.17	192±19	0.150±0.005	35.4±3.5
2416	0.565	13.3±2.9	2.47±0.57	384±88	0.160±0.001	34.5±3.5

3. Axial distributions of actinides in cells are non-uniform and do not obey to any certain relationship. The distributions in different cells differ each other. Actinide contents in the samples taken from the cells adjacent to accidental ones are by tens times higher than those in the cells distant enough from accidental ones. Contents of ^{241}Am and ^{137}Cs in the cells neighboring to accidental cells of the I-1 reactor were larger in average by a factor of about 6 than those in the EI-2 reactor. Comparison of ^{241}Am and ^{244}Cm contents in the cells distant from accidental ones demonstrated that contamination of graphite in the I-1 reactor is higher by a factor of 2-2.5 than that in the EI-2 reactor.
4. Ratios $^{243}\text{Am}/^{241}\text{Am}$, $^{244}\text{Cm}/^{241}\text{Am}$ in the samples taken at different axial levels differ significantly. This may be explained by variations of neutron flux along height of the cell (Table V). Forms of dependency are similar for both reactors. Values of ratios increase with growth of neutron fluence. Under equal relative fluence, the ratios are of different value in two reactors. This may be explained by difference in exposure time of uranium after its accidental appearance in the reactor stacks.

TABLE V. DEPENDENCE OF RELATIVE ACTINIDE CONTENT ON NEUTRON FLUENCE

Relative value of fluence	Reactor	Cell No	$\frac{A(^{243}\text{Am})}{A(^{241}\text{Am})}$	$\frac{A(^{244}\text{Cm})}{A(^{241}\text{Am})}$
0.997	I-1	1425, 2638	0.50±0.12	434±78
	EI-2	2626, 2639	0.49±0.01	273±14
0.710-0.780	I-1	1425, 2516, 2638	0.38±0.06	233±40
	EI-2	2626, 2627, 2639	0.31±0.01	92±3
0.500-0.550	I-1	0225, 0320, 2638	0.19±0.04	74±22
	EI-2	2626, 2639	0.22±0.01	52±3
0.400-0.470	I-1	0225, 0320, 1425 2516, 2638, 2729	0.15±0.02	42±6
	EI-2	2627, 2639, 3917 4016, 4017, 4018	0.080±0.002	18±1
0.190-0.230	I-1	0225, 0320, 2516	0.07±0.04	14±8

Results of XL_{β} -radiation measurements also demonstrated that correlation Cm/Am was very sensitive to axial variations of neutron flux in the cell. In the ADE-3 reactor the averaged values of activity correlation Cm/Am are equal to 8.7 and 35 for relative neutron fluences 0.49 and 0.735, respectively. Averaged values of activity ratio Pu/Am are equal to 0.21 and 0.17 for the same fluences, i.e. they coincide within the range of experimental errors.

Basing upon obtained results, the following scheme for contamination of the stack with actinides may be presented:

- under accidents, uranium particles were transported through the stack and fixed on surface of the graphite blocks;
 - under subsequent long-term neutron irradiation, composition of contamination was gradually formed. Observed dependence between isotopic ratios and neutron fluence enable us to conclude that superficial displacements of radionuclides during irradiation were small. Transport processes of actinides in graphite would promote their mixing, lead to flattening of their relative concentrations and, probably, contamination levels.
5. The information was obtained about ^{134}Cs , ^{137}Cs , ^{106}Ru , ^{154}Eu contents and about their distribution in the graphite blocks of the I-1 and EI-2 reactor cores. Content of these radionuclides in superficial graphite layer is significantly higher than that in volume of blocks. Radionuclide ^{134}Cs is generated in graphite through activation of ^{133}Cs impurity and as a fission product. The second channel for ^{134}Cs generation defines the superficial contamination level of the graphite blocks with this radionuclide. Table VI demonstrates measurement results for content of ^{137}Cs , ^{134}Cs , ^{106}Ru in superficial layer of the graphite blocks from the I-1 and EI-2 reactor cores as well as the information about ^{90}Sr content obtained in β -spectrometric measurements. Isotopic ratios $^{134}\text{Cs}/^{137}\text{Cs}$, $^{106}\text{Ru}/^{137}\text{Cs}$ and $^{90}\text{Sr}/^{137}\text{Cs}$ are almost constant that indicates on absence of fission products migration over surface of the graphite stack blocks after generation of fission products. Obtained results enable us to conclude that contamination of the stack with fission products has occurred according to the same scheme as contamination with actinides.
6. The following scheme was applied to evaluation of radionuclide inventories in the reactor stacks. It was assumed that actinides (and fission products) were contained in superficial layer (2 mm thick) of the graphite blocks. Absolute value of contamination was determined from measurement results of ^{241}Am content in the samples and from correlations between content of this nuclide and other actinides.

All the cells in the stack were divided on two groups: adjacent to accidental cells (including accidental cells) and distant from accidental cells. For each group of the cells, mean values of ^{241}Am and ^{137}Cs content were calculated under assumption that measurement results obey to logarithmically-normal distribution. For obtaining the ratios between contents of different actinides, averaging procedure was performed with results of measurements for the samples taken from different axial levels.

According to preliminary assessment, ^{241}Am inventory in graphite of the I-1 reactor core is equal to 6.3×10^{10} Bq, in the EI-2 reactor – 2.0×10^{10} Bq. Inventories of ^{137}Cs are equal to 6.3×10^{12} Bq and 3.6×10^{12} Bq in the I-1 and EI-2 reactors, respectively.

7. Ratios between content of actinides (^{241}Am , ^{244}Cm) and ^{137}Cs in the graphite stack were determined. Observed dispersion of ratio values does not exceed a factor of 2-3 while contamination levels of individual blocks may differ by several tens and hundred times. It opens a possibility to evaluate approximate content of actinides in individual blocks from results of simple measurements for ^{137}Cs gamma-radiation. Such a method may be applied in future for rapid sorting of the graphite blocks depending on contamination level prior to their treatment.

4. DETERMINATION OF ^{60}Co CONTENT

^{60}Co is one of the main γ -emitters in both reactors during 40 years after their ultimate shutdown. The major channel for ^{60}Co generation is the neutron reaction $^{59}\text{Co}(n,\gamma)^{60}\text{Co}$. Inside of the graphite block volume, ^{60}Co is generated through activation of impurities while in superficial layer - both through impurities activation and by transport of cobalt with water-vapour mixture under accidents. Due to the reasons mentioned above, contamination of spent graphite with cobalt may be divided on superficial and volumetric contamination for further investigation of both types separately.

For evaluating the volumetric component of contamination it is necessary to know initial content of cobalt impurity in graphite. These data are inaccurate and contradictory. It is known that cobalt is non-uniformly distributed in the reactor graphite (Ref. [7]).

^{60}Co content in the graphite samples was measured by method of γ -spectrometry with application of coaxial HPGe-detector. Experimental errors in measurements of ^{60}Co activity in the samples were within the range 3-5%. Total numbers of assayed graphite samples were about 250 from the I-1 reactor and about 200 from the EI-2 reactor.

TABLE VI. MEASUREMENT RESULTS FOR ^{137}CS , ^{106}RU , ^{90}SR SPECIFIC ACTIVITIES IN THE GRAPHITE SAMPLES TAKEN FROM SURFACE OF THE GRAPHITE BLOCKS (ON DECEMBER 10, 1998)

Reactor	Cell No	Relative value of fluence	$A(^{137}\text{Cs})$, 10^5Bq/g	$A(^{106}\text{Ru})$, 10^3Bq/g	$A(^{90}\text{Sr})$, 10^4Bq/g	$\frac{A(^{106}\text{Ru})}{A(^{137}\text{Cs})} \times 10^3$	$\frac{A(^{90}\text{Sr})}{A(^{137}\text{Cs})}$
EI-2	3425	0.46	1.38	1.3	7.4	4.8	0.54
		0.96	1.23	1.2	4.7	4.9	0.38
		0.61	32.3	29	160	4.5	0.48
I-1	3644	0.55	5.62	1.9	29	3.5	0.52
		0.82	3.00	1.2	17	3.8	0.57
		0.80	1.00	0.30	7.5	3.0	0.75

Analysis of obtained results led to the following conclusions:

1. It was revealed that distributions of experimental data are close to logarithmically-normal distribution. So, treatment of results, calculations of mean values and their errors were performed in accordance with this distribution.
2. In some cases, radioactivity of inner samples taken from different graphite core or even from one graphite core differed one from another by several times. This may be explained by non-uniform distribution of cobalt impurity in graphite (Table VI).

3. Contamination level of the graphite block surfaces is near the same for the whole stack. However, ^{60}Co activity in some samples differed by several times. It may be related with peculiarities in cobalt transportation with water-vapour phase.
4. ^{60}Co activity in the samples taken from surface of the graphite blocks was higher than that in the samples taken from volume of the blocks. It was revealed that outer graphite layer of 2 mm thick defines superficial contamination of the blocks with ^{60}Co . Proceeding from this fact, a ratio was calculated between inner volume¹ of a graphite block and volume of superficial layer ($V_{\text{vol}}/V_{\text{sup}}$). This ratio was equal to 13.5 and 15.1 for the I-1 and EI-2 reactors, respectively. Mean activity ratio for superficial samples to inner samples for all graphite cores was equal to 4.7 and 3.2 in the EI-2 and I-1 reactors, respectively, i.e. difference in levels of superficial and volumetric contamination was rather small. Therefore, ^{60}Co contained in inner volume of the graphite blocks gives the main contribution to inventory of this radionuclide in the graphite stacks of both reactors.
5. The cells adjacent to and distant enough from accidental ones do not differ in levels of contamination with ^{60}Co . ^{60}Co content in central and in peripheral graphite blocks of the reactor stack was the same within the range of experimental errors. ^{60}Co content in volume of the reflector blocks from the EI-2 reactor (the graphite cores taken from two cells were assayed) was about twice smaller than that in blocks of the reactor core.
6. In the I-1 reactor, at level of 1 m upward from centre of the reactor core, content of ^{60}Co reaches a maximal value (approximately, twice higher than mean level) both in central part and at periphery of the reactor core. In the EI-2 reactor, maximal content of ^{60}Co was observed in lower part of the stack. This effect can't be explained neither by variations of neutron flux nor by difference in amount of transported cobalt. A possible cause is a different content of cobalt impurity in different layers of the graphite stack.

TABLE VII. ^{60}CO SPECIFIC ACTIVITY IN THE GRAPHITE SAMPLES TAKEN FROM DIFFERENT REACTOR CELLS, KBQ/G (ERRORS OF PRESENTED RESULTS WERE LESS 5%)

Sam No	Place of sampling					
	EI-2			I-1		
	Cell 2639		Cell 3225	Cell 2638	Cell 1425	
	2 m upward from core centre	Core centre	3 m downward from core centre	3 m downward from core centre	2 m upward from core centre	Core centre
1	9.50	13.0	7.50	20.6	6.10	2.70
2	6.90	1.36	1.10	0.195	0.53	1.86
3	1.48	0.63	2.12	1.99	1.74	1.96
4	2.51	0.47	1.40	0.60	0.43	1.43
5	7.30	11.0	11.0	2.90	1.74	9.8

According to the results obtained, superficial contamination level of the graphite blocks may be considered as the same for the reactor core as a whole within the range of presented errors (Table VII). Mean superficial contamination with ^{60}Co of the graphite blocks from the I-1 reactor core was estimated as 6.9 ± 0.7 kBq/g; from the EI-2 reactor core - as

¹ Inner volume of a graphite block (V_{vol}) is a difference between total volume of a graphite block and volume of superficial layer, 2 mm thick.

14.0±2.4 kBq/g. ⁶⁰Co content inside of the graphite block volume is kept constant over whole reactor core (2.2±0.5 kBq/g and 3.0±0.6 kBq/g for the I-1 and EI-2 reactors, respectively).

Basing upon these conclusions, it is possible to obtain approximate evaluation for inventory of ⁶⁰Co in graphite of the reactor core² as a sum of contributions from superficial and volumetric contamination:

$$A = \frac{m \times (V_{sup} A_{sup} + V_{vol} A_{vol})}{V_{sup} + V_{vol}},$$

where *m* - total mass of the graphite blocks in the reactor core; *V_{sup}* - volume of superficial layer; *V_{vol}* - inner volume of the graphite block; *A_{sup}* - mean specific activity of ⁶⁰Co on surface of the graphite block; *A_{vol}* - mean specific activity of ⁶⁰Co in volume of the graphite block.

Activity of ⁶⁰Co contained in all blocks of the reactor core was estimated as 2.3×10¹² Bq and 3.3×10¹² Bq in the I-1 and EI-2 reactors, respectively.

In Ref. 4, the results are presented for calculated evaluation of ⁶⁰Co specific activity in spent reactor graphite. Calculated specific activity of ⁶⁰Co appeared to be about 420 kBq/g that exceeded our results obtained for the graphite block volume of the I-1 reactor by a factor of 190, and for the EI-2 reactor - by 140 times³. It is most probably related with use in calculations of overestimated ⁵⁹Co content (0.3 ppm) in the reactor graphite. According to results on neutron activation analyses, cobalt content in unirradiated graphite is about (0.0094±0.0017) ppm.

TABLE VIII> MEAN SPECIFIC ACTIVITY OF ⁶⁰CO IN THE GRAPHITE BLOCKS FROM THE REACTOR STACKS, KBQ/G (ON DECEMBER 10, 1998)

Place of sampling	I-1		EI-2	
	Volume of block	Surface of block	Volume of block	Surface of block
Upper part of the reactor stack	1.3±0.5	6.3±1.2	2.6±1.2	6.7±1.8
Centre of the reactor stack	2.0±1.0	6.8±1.6	3.0±1.0	9.7±2.6
Lower part of the reactor stack	1.0±0.4	6.2±1.4	6.3±3.8	25.6±17.7
Cells adjacent to accidental ones	1.4±0.6	6.2±0.8	2.8±0.6	14.0±2.7
Cells distant from accidental ones	2.5±0.7	7.2±1.0	3.4±1.0	13.9±3.3
Stack as a whole	2.2±0.5	6.9±0.7	3.0±0.6	14.0±2.4

² There is no sufficient information to evaluate ⁶⁰Co content in the reactor reflector. These results will be presented later.

³ These data correspond to 8-year period of the graphite cooling time when the experiments have been carried out.

5. CONCLUSION

According to previous investigations, it might be anticipated that radionuclide distributions will be affected by the processes related with temperature field, multiple water leakages into the stack, nitrogen blowing down, difference in migration properties of individual nuclides etc, and this will lead to changes in radionuclide distributions. However, according to the results obtained, initially formed distributions of many radionuclides (except of tritium) kept practically time-independent later on. It may be explained by fixation of uranium-containing particles on the graphite surface.

On basis of obtained information about radionuclide distributions of different origination, the schemes were elaborated and tested for evaluation of radionuclide inventories in the graphite stack. The following approaches were used:

main attention should be given to long-lived radionuclides and, in the first turn, to major contributors to radioactivity of spent graphite;

- the number of analysed radionuclides may be reduced via application of the correlations worked out on base of obtained results;
- the number of assays on content of some radionuclides may be reduced via their replacement by the calculations based on application of reliable data about impurities content in graphite and about neutron fluence.

In evaluation schemes the stack is represented as a set of cells. Radionuclide inventory is determined as a sum of its contents in each cell taking into account the classification of the cells depending on contamination level. Mean content of radionuclide in a cell was determined from measurement results for five graphite cores taken at different axial points.

As a rule, error of individual measurements was equal to several per cents. In some cases, there were observed the high discrepancies between results obtained for different samples. As a consequence, it led to increase in evaluation error for mean content of radionuclides in the cells and, to the less extent, radionuclide inventories in the stack.

Uncertainty in mean content of radionuclides could achieve 50%, 100% and even more in some cases. Dispersion of radionuclide contents on surface of the graphite blocks (fission products, actinides) may be explained by accidents and local conditions for fuel particles deposition: variations in width of slits between blocks, surface state of the individual blocks and so on. Difference of radionuclide concentrations in volume of the blocks appeared both as a natural result of variations in neutron fluence and due to random causes (variations in impurities content; it concerns ^{60}Co , in the first turn).

The evaluations of radionuclide inventories in the reactor stack may be used for decision making in spent graphite management and in selection of utilization technologies. In these cases, uncertainty of 2-3 times has no importance of principle, and accuracy of such an evaluation may be considered as satisfactory.

Presently, development of schemes for evaluation of individual radionuclide inventories was not completely finished. The scheme for evaluation of ^{14}C inventory was the most advanced. This scheme will be based upon calculations of neutron field in the reactor and upon normalisation of the results obtained in 10-20 assays. It may be anticipated that accuracy

of the scheme will be better than that for other radionuclides. Elaboration of similar scheme for evaluation of ^{60}Co inventory is near to the end. Probably, evaluations for non-investigated reactors will be based upon the results obtained for ^{60}Co inventories in the SGCE reactors but properly corrected with taking into consideration the distinctions in neutron flux and modes of the reactor operation. The most difficulties appeared in evaluations of actinide and fission product inventories. In all cases, results of current investigations being performed at the ADE-3 reactor will be used for examination and correction of the schemes being proposed.

It should be expected that radionuclide inventories in the reactor stacks evaluated with use of newly obtained information might significantly differ, in some cases, from values previously predicted by calculations.

Radionuclide content in individual blocks may differ by many times. So, to make proper decision on their utilization, the blocks, probably, should be sorted depending on contamination level. Obtained results may be used in development of rapid sorting method.

Problem of evaluating the radionuclide inventories in the graphite stacks of the RBMK-type reactors is rather simpler due to absence of accidents with fuel penetration into graphite. Therefore, the number of radionuclides which present in graphite is smaller, description of radionuclide distributions in the graphite volume is simpler, possibility to apply calculations is larger, and, as a consequence, needs in experiments are less. In this case, information about impurities content in graphite will be of special significance.

One else type of radioactive wastes accumulated for period of PUGRs operation is the graphite sleeves. Total mass of the graphite sleeves is near to mass of the graphite stacks. The sleeves are disposed in the near-to-reactor repositories of different type. Conditions of their storage do not meet the contemporary requirements. Since exposure time of the sleeves in the reactors was longer by a factor of 10 or more than that for the reactor stacks, and since about 70% of the graphite sleeves were loaded into reactors after the last accidents accompanied by fuel release from channels, it may be anticipated that contamination level of the sleeves is substantially less, and the most part of the sleeves is not contaminated with actinides and fission products.

In this case, another times and another technologies might be selected for utilization of the graphite sleeves as compared with the graphite stacks. Due to this consideration, it seems reasonable and important for us to dedicate a special study to investigations of the graphite sleeves contamination in repositories. The results of these investigations may be further used for substantiation of measures for management of spent graphite sleeves.

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