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INVESTIGATION OF RADIOACTIVE CONTAMINATION OF GRAPHITE SAMPLES FROM THE AM REACTOR

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The world's first nuclear power plant operated for almost 48 years. Over this period of time, the neutron fluence on the graphite masonry reached ~10^{22} cm^{-2}, which resulted in activation of the impurities present in the graphite. During operation, incidents occurred with loss of seal and sometimes loss of integrity of the fuel-element claddings in some cells and particles of the fuel and steam-water mixture entered the graphite masonry. This resulted in radiation contamination with a complex radionuclide composition.

Experimental information about the content and distribution of radionuclides in the spent nuclear graphite is needed in order to plan methods and periods of time for disassembly and salvaging of the graphite masonry of the stopped reactor taking account of the dose loads on the workers and the ecological safety norms.

The problems which can be solved on the basis of the present work included the determination of the ^{14}C and ^{3}H contents by liquid-scintillation β spectrometry, analysis of the actinide content by direct γ spectrometry, and neutron-activation analysis followed by γ spectrometry. These invstigation yielded new data on the content of fission products and activation impurities in graphite.

An analysis of the radioactive contamination of the graphite masonry of the reactor in the world's first nucler power plant (Obninsk) was undertaken to check and supplement the previously existing information [1]. As in other similar investigations [2], the data are needed for planning measures on decommissioning a reactor and salvaging radioactive wastes.

We shall list certain features of the construction and operation of the AM reactor [3]:

- the graphite masonry consists of 151 vertical columns arranged in a triangular lattice with spacing 120 mm and assembled from blocks with a hexahedral cross section with a 65-mm in diameter interior opening at the center, and 24 columns assembled from horizontal blocks at the periphery (in the reflector); the mass of the graphite in the masonry is 46.3 tons (10.3 tons at the center);
- the enrichment of the fuel loaded into the core during different periods ranged from 4.4 to 10% ^{235}U;
- the reactor continued to operate for almost 48 yr with interruptions;
- the maximum thermal-neutron fluence was ~$2 \cdot 10^{22}$ cm^{-2}, the nominal thermal power was 30 MW, and the average power was 9.5 MW;
- the working temperature of the graphite reached 700°C.

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TABLE 1. ^{241}Am, ^{137}Cs, and ^{244}Cm Activity in Different Cells at the Center of the Core of the AM Reactor, Bq/g

Sample No.	^{241}Am	^{137}Cs	^{244}Cm
14	$2.4\cdot10^2$	$6.9\cdot10^3$	$1.3\cdot10^4$
24	$1.9\cdot10^2$	$5.7\cdot10^3$	$3.7\cdot10^2$
34	$7.5\cdot10^2$	$2.8\cdot10^4$	$3.2\cdot10^3$
44	$2.3\cdot10^2$	$1.0\cdot10^4$	–

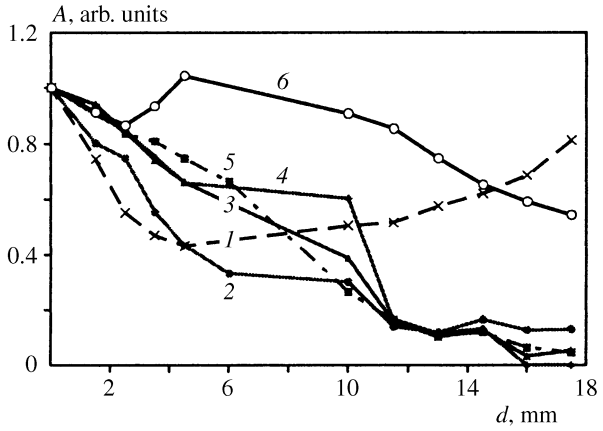

Fig. 1. Distribution of ^{60}Co (*1*), ^{137}Cs (*2*), ^{144}Ce (*3*), ^{106}Ru (*4*), ^{241}Am (*5*), and ^{65}Zn (*6*) along a core.

It should also be noted that during reactor operation there were incidents in which the seal was broken and sometimes the intgerity of the fuel-element cladding in certain cells was disturbed and particles of the fuel and steam-water mixture entered the graphite masonry. The neutron irradiation activated impurities present in the graphite, resulting in multicomponent contamination.

The investigations were performed using the following methods:

- γ spectrometry for measuring the content of γ-emitting radionuclides (fission products, ^{241}Am, products of the activation of impurities);
- liquid scintillation β spectrometry for determining the content of ^{14}C and ^{3}H;
- neutron activation analysis for determining the content and isotopic composition of uranium.

Gamma and x-Ray Spectrometric Measurements. Twenty-eight samples were obtained to investigate the content of fission products, products of activation of impurities, and actinides in the graphite. These samples were prepared from cores taken from four cells in different parts of the masonry. The data obtained characterized the distribution of radionuclides over the heigth of the core and the volume of individual graphite blocks.

As established previously, the activity of ^{244}Cm and ^{241}Am on the surface of the blocks was 10–100 times higher than in the volume in the graphite of the reactors at the Siberian Integrated Chemical Plant (I-1, ÉI-2, ADÉ-3) [1]. In the graphite of the AM reactor, the activity of these radionuclides is close to the level of the volume contamination of the graphite in the masonry of the reactors at the Siberian Integrated Plant and much lower than the surface activity in these reactors. The ratio of the ^{137}Cs and ^{241}Am activity in the samples of the graphite from the AM reactor is approximately the same as in the sample from the reactors at the Siberian Integrated Plant. The ^{241}Am, ^{244}Cm, and ^{137}Cs activity of the samples of all four channels from the center of the core was close (Table 1).

TABLE 2. Average ^{241}Am, ^{137}Cs, and ^{244}Cm Activity Along the Height of the Masonry of the AM Reactor, Bq/g (average over four channels)

Height	^{241}Am	^{137}Cs	^{244}Cm
1 (top)	38	$3.6\cdot10^3$	$3.4\cdot10^2$
2	66	$1.4\cdot10^4$	$2.0\cdot10^3$
3	15	$1.4\cdot10^4$	$5.5\cdot10^2$
4	$4.7\cdot10^2$	$1.3\cdot10^4$	$2.3\cdot10^3$
5	$1.3\cdot10^2$	$1.8\cdot10^4$	$5.7\cdot10^3$
6	5	$2.0\cdot10^4$	$9.0\cdot10^2$
7 (bottom)	$3.7\cdot10^2$	$3.8\cdot10^4$	$2.5\cdot10^4$

The distribution of actinides and fission products along the core is similar; their content decreases in the direction from the process channel into the bulk of a graphite block (Fig. 1). Deep penetration of these nuclides into the interior volume of the graphite masonry was found; this was not observed in the graphite of the reactors at the Siberian Integrated Chemical Plant. This difference could be due to the different amount of graphite and the two times higher working temperture of the graphite in the AM reactor, the long operating period of this reactor, and the large number of incidents with disruption of the integrity of the fuel-element cladding and flooding of the masonry of AM, all of which occurred right up to the moment when the reactor was shut down. The distribution of the products of activation of impurities (^{60}Co, ^{65}Zn) in the graphite does not exhibit a regular behavior. Nonetheless, the nonuniformity of the distribution of these radionuclides is much smaller than in the reactors at the Siberian Integrated Chemical Plant. The distribution of all radionuclides over the height of the graphite masonry exhibits a general feature: their content increases substantially at the bottom of the masonry (Table 2). This is probably due to the spillage of particles of radioactive graphite during drilling and repair work done on the channels.

On the basis of the results obtained one can imagine the following mechanism leading to the formation of radioactive contamination in the graphite masonry. Temperature acting over a prolonged period of time resulted in cracking of the graphite blocks. The uranium particles, which enter the coolant as a result of incidents, were disseminated by the steam–water mixture, penetrating deep into the cracks of the graphite moderator. Then, under the action of the neutron flux, the uranium present in the graphite was converted into fission products and heavy actinides. This is confirmed by the decrease in the content of fragment radionuclides in the direction away from the process channel into the interior space of a graphite block, similar to the distribution of fission products which have different migration properties, similar to the distribution of fissiion products and ^{241}Am.

Measurement of the Activity of ^{14}C and ^{3}H in Graphite Samples. Five samples were obtained from four cells at different heights in the masonry to determine the content of these radionuclides in the graphite. Following the procedure used in [4], these samples were burned at 700°C in a melt with a two-component oxidizer containing V_2O_5 and CuO. To perform measurements, approximately 50-mg pieces of graphite are pricked off the graphite cores and placed into a retort for burning. ^{3}H and ^{14}C were separated from the sample in a gaseous form (HTO and $^{14}CO_2$). The gaseous products CO_2 and $^{14}CO_2$ were passed through a trap, filled with NaOH, for complete absorption with formation of water-soluble compounds. After the graphite was completely burned, the contents of the trap were washed off with distilled water. The coefficient of transfer of ^{14}C from the sample into the liquid phase was 0.95 ± 0.05. The tritium-containing gas phase (vapor of tritium water), formed by oxidation of the graphite, condensed in the nitrogen trap. The coefficient of transfer of tritium from the sample into the liquid phase was 0.75 ± 0.1. Some of the solution formed was introduced into a liquid scintillator for subsequent measurement of the activity of ^{14}C and tritium.

The activity of ^{14}C and ^{3}H was measured by liquid scintillation spectrometry using a Quantulus 1220 β spectrometer (Finland). The time for one measurement ranged from 10 min to 2 h, depending on the activity of the sample. The determination error using this method does not exceed 20% with confidence probability 0.95.

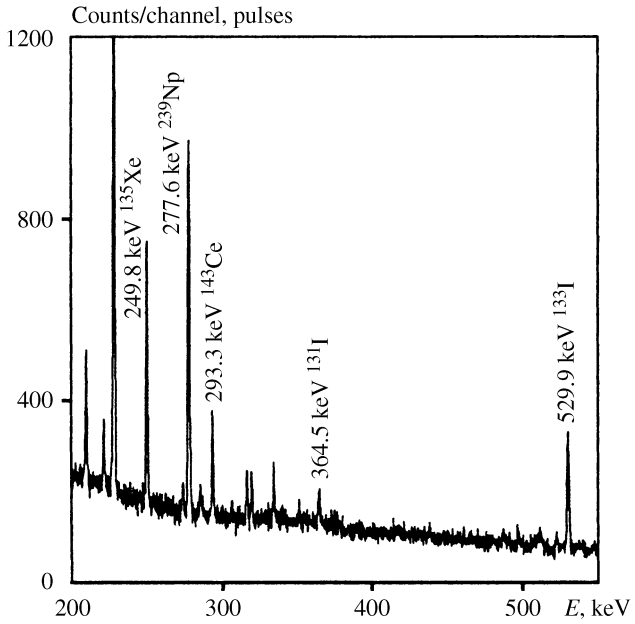

Fig. 2. Radiation spectrum of the products of the reactions of uranium with neutrons after short-time repeated irradiation.

The results obtained show the following:

- the activity of most of the experimental samples does not differ by much, reaching maximum values at the bottom of the masonry at heights +500–+1500 mm;
- the average specific activity of ^{14}C in the experimental graphite samples taken from the AM reactor was about 10^5 Bq/g, which is 10 times lower than in the graphite of the reactors at the Siberian Integrated Chemical Plant; however, it should be noted that the neutron fluence over the operating time of the reactors indicated is not the same and differs by more than a factor of 5;
- the average activity of the tritium in the graphite of the reactors being compared is almost identical (~10^4 Bq/g).

Determination of the Uranium Content and the Ratio $^{235}U/^{238}U$ in Graphite Samples. The uranium content and the isotopic ratio of $^{235}U/^{238}U$ in samples taken from the graphite masonry were determined by neutron-activation analysis. The amount of uranium in the experimental samples with a mass of about 0.2 g was measured according to its content in a comparison sample which was made by evaporating a solution, containing a precisely known amount of natural uranium, onto a graphite substrate.

The experimental samples were irradiated in a vertical experimental channel of the IRT reactor at the Moscow Engineering Physics Institute. The thermal-neutron flux density in the irradiation zone was about $2 \cdot 10^{13}$ $sec^{-1} \cdot cm^{-2}$. The graphite samples placed in aluminum foil were irradiated for 2 h. The measurements performed with a germanium detector were started on the day after irradiation and continued for 3 days. A single measurement took 5 min to 1 h depending on the activity of the sample. One of the spectra obtained is presented in Fig. 2. The following peaks can be identified in this spectrum: 249.8, 293.3, 364.5, and 529.9 keV due to the fission products ^{135}Xe, ^{143}Ce, ^{131}I, and ^{133}I, respectively, the peak 277 keV due to ^{239}Np, and peaks due to various impurities in the graphite.

Comparing the intensities of the peaks in the spectra of the irradiated samples and the comparison sample made it possible to determine the content of ^{238}U and ^{235}U in individual samples and the isotopic ratio $^{235}U/^{238}U$. The amount of ^{238}U in the graphite was calculated from the intensity of the peak ^{239}Np 277 keV and the amount of ^{235}U from the intensities of the peaks due to the fission products.

TABLE 3. ^{235}U and ^{238}U Mass per 1 g of the Graphite Sample

Cell	Height	^{235}U, ng	^{238}U, ng	$^{235}U/^{238}U$, %
13-12	+1500	4.2 ± 0.3	46 ± 3	8.5 ± 0.8
13-12	+500	4.2 ± 0.3	62 ± 3	6.3 ± 0.5
13-12	0	158 ± 5	3110 ± 80	4.8 ± 0.2
13-12	0	62.4 ± 1.2	2580 ± 40	2.36 ± 0.05
13-12	– 500	27.3 ± 1.7	639 ± 14	4.1 ± 0.3
13-12	–1000	63.1 ± 1.8	2700 ± 200	2.29 ± 0.18
13-12	–1000	12.0 ± 0.6	324 ± 13	3.6 ± 0.2
13-12	–1500	70 ± 2	1330 ± 100	5.0 ± 0.4
13-12	–1500	24.0 ± 0.8	750 ± 30	3.11 ± 0.17
11-06	+1500	185 ± 19	4000 ± 400	4.4 ± 0.6
11-06	+1500	50 ± 4	1060 ± 30	4.5 ± 0.4
11-06	+1500	165 ± 5	3090 ± 60	5.07 ± 0.17
11-06	+500	36 ± 3	640 ± 30	5.3 ± 0.6
11-06	0	155 ± 9	7280 ± 130	2.09 ± 0.13
11-06	0	10.5 ± 0.4	363 ± 10	2.80 ± 0.14
11-06	0	109 ± 2	5490 ± 90	1.95 ± 0.05
11-06	– 500	91.4 ± 1.9	2020 ± 130	4.3 ± 0.3
11-06	–1000	25.3 ± 1.3	465 ± 40	5.2 ± 0.5
11-06	–1500	46 ± 2	1417 ± 90	3.1 ± 0.2

The amount of ^{238}U in the graphite samples was calculated from the relation

$$M(^{238}U)_g = M(^{238}U)_c S_g^{Np}/S_c^{Np},$$

where $M(^{238}U)_c$ is the mass of ^{238}U in the comparison sample, which was $55\cdot10^{-3}$ mg; S_g^{Np} and S_c^{Np} are the areas of the ^{239}Np peak in the spectrum of the graphite sample and comparison sample, respectively.

The amount of ^{235}U was calculated using the analogous formula

$$M(^{235}U)_g = M(^{235}U)_c S_g^f/S_c^f,$$

where $M(^{235}U)_c$ is the mass of ^{235}U in the comparison sample, which was $0.396\cdot10^{-3}$ mg; S_g^f and S_c^f are the area of the peak of one of the fission products in the spectrum of the graphite sample and the comparison sample, respectively.

The average of the values obtained for the ratio of the areas of the peaks for four fission products was calculated and the result was used to calculate the mass of ^{235}U in the graphite sample (Tables 3 and 4).

The uranium content was analyzed in 22 graphite samples from cells 13-12 and 11-06. The average content of uranium in the graphite of the cells studied differs by approximately a factor of 2. The isotopic composition of the uranium is almost the same (see Table 4). The content of uranium on different sides was compared for the two cores taken from cell 11-06: on the channel side and from the interior of a graphite block. In one core, the uranium content differed by a factor of 3 and in the second one by a factor of 10.

Conclusions. The investigation confirmed a previously drawn conclusion that the amount of ^{14}C in spent graphite masonry can be estimated from the results of a small number of analyses and computational data on the distribution of neutrons in the volume of the reactor. The uranium contamination of the graphite in the AM reactor is substantial and "younger,"

TABLE 4. Average ^{235}U and ^{238}U Content per 1 g of Graphite

Mass	Cell	
	13-12	11-06
^{235}U, ng	47.3	87.3
^{238}U, ng	1280	2580
$^{235}U/^{238}U$, %	3.6	3.3

and the ratio $^{235}U/^{238}U$ is higher than in the commercial uranium–graphite reactors which were shut down at the Siberian Integrated Chemcial Plant. Judging from the small sample obtained, the total mass of ^{235}U in the masonry of the AM reactor does not exceed 3 g.

The processes by which radionuclides penetrate into the masonry of the reactors being compared differed substantially: in the first case, surface contamination of the graphite blocks prevailed on the side of the process channel and at the end surface; in the second case, the dissemination of the fuel particles by the steam–water mixture along the cracks in the graphite led to volume contamination of the blocks by uranium with maximum contamination on the side of the process channels.

A correlation was established between the distribution of separate fission products and actinides. This shows that they are secured on sections where a fission event has occured. In our opinion, this makes it possible to estimate the content of radionuclides which are difficult to measure, for example, ^{99}Tc, ^{129}I, and others, from the activity of radionuclides for which measurements are easy to perform, for example, ^{137}Cs, by using data on their yield and half-life.

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