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Analysis of the Causes of Irregular ^{36}Cl Radionuclide Distribution in Irradiated Nuclear Graphite

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Abstract—The article is dedicated to analyzing the concentration processes of the long-lived ^{36}Cl radionuclide in irradiated graphite of uranium-graphite nuclear reactors. The ^{36}Cl radionuclide is one of the most important isotopes for assessing the safety of radioactive-waste disposal. An analysis of the so-called Nugget effect due to the substantial heterogeneity of the ^{36}Cl content in samples of irradiated graphite of nuclear reactors is carried out. In the same graphite elements, the difference in the ^{36}Cl concentration reaches 100 times regardless of operational factors, such as the neutron flux and temperature. The article discusses the main processes that can affect ^{36}Cl contamination in irradiated graphite. Particular attention is paid to determining the form of ^{36}Cl in graphite, taking into account the features of its manufacturing and thermodynamic modeling of the equilibrium compositions of reaction products during the purification of unirradiated graphite in a gaseous medium.

Keywords: irradiated graphite, Nugget effect, chlorine, decommissioning, radionuclide, leaching, irregularity
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INTRODUCTION

During the operation of uranium-graphite nuclear reactors and after their final shutdown, graphite radioactive waste (RW) is generated. Today, its mass is ≈60 000 t in the Russian Federation only.

The period of potential danger for ≈90% of graphite RW is determined mainly by the presence of long-lived ^{14}C and ^{36}Cl radionuclides in the composition of graphite stack parts and their specific content. The half-life of ^{14}C is 5370 years; its specific activity in graphite of Russian uranium-graphite reactors (UGR) is 10^3–10^6 Bq/g, while for ^{36}Cl it is 3.1×10^5 years and 10–2×10^3 Bq/g, respectively [1].

The main condition for substantiating options for the treatment of graphite RWs is the insulation of radionuclides contained in them for the entire period of their potential danger when being stored. According to assessments, after a characteristic period of ≈10^4–10^5 years, ^{36}Cl will determine the main parameters of the potential impact of irradiated graphite on the environment and population upon burial. Moreover, this radionuclide is barely accumulated by natural materials during migration, while long-lived actinides are effectively sorbed by barrier materials and the materials of host rocks.

Obviously, the guarantee of the insulation of radionuclides for the period of potential danger upon burial or temporary storage of graphite RW will be determined not only by the activity of long-lived radionuclides, but also indicators that determine the durability of radionuclide fixing in graphite itself. Given the fact that graphite is a practically "non-oxidizable" material under normal temperature conditions, the yield of radionuclides (including ^{36}Cl) from graphite RW will be determined by the parameters of their selective leaching, which, in turn, depend on the form of existence and bonding energy of long-lived radionuclides with graphite structure. Therefore, these data will be decisive when performing the predictive modeling of radionuclide migration, the results of which are decisive when substantiating options for the disposal of graphite radioactive waste.

Currently, as a promising approach to solving the problem of handling irradiated graphite, a method of near-surface graphite RW burial with preliminary treatment is considered. The implementation of this approach is possible provided that the problem of the selective removal of the ^{14}C and ^{36}Cl fractions, characterized by a lower strength of fixation in the structure of graphite, is successfully solved. This approach may reduce the potential hazard of graphite during burial

Table 1. Specific activity of ^{36}Cl in different samples of irradiated graphite [2–9]

Country	Location	Reactor type	Activity ^{36}Cl, Bq/g
France	Marcoule G2	UNGG	100–700
France	Chinon A3	UNGG	7–70
France	Saint-Laurent A1	UNGG	3–10
France	Saint-Laurent A2	UNGG	37–100
France	Bugay 1	UNGG	83–850
Great Britain	Hunterston A	Magnox	132–1775
Great Britain	Oldbury	Magnox	≈425
USA	Hanford	Reactor B	≈2460
USA	Hanford	Reactor C	≈705
USA	Hanford	Reactor D	≈1990
USA	Hanford	Reactor DR	≈1530
USA	Hanford	Reactor F	≈1940
USA	Hanford	Reactor KE	≈1998
USA	Hanford	Reactor KW	≈1920
Russia	PO Mayak	AV-1	<13 500*
Russia	PO Mayak	AV-2	<10 800*
Russia	PO Mayak	AV-3	<10 800*
Russia	PO Mayak	AI	<5600
Russia	JSC “ODTS UGR”	I-1	54–110
Spain	Vandellos	UNGG	14–25
Ukraine	CHAES (unit no. 2)	RBMK-1000	700–1500
Lithuania	IgNPP (unit no.1)	RBMK-1500	≈2600
Switzerland	Diorite (PSI)	PHWR	22–414
Switzerland	Proteus (PSI)	Research	0.003–0.031
Denmark	Risoe National Lab.	DR-2	0.18–40
Germany	NPP THTR-300	THTR-300	23–370
Germany	Julich	AVR	≈800

and, as a result, reduce the requirements for the method and technology of burial, which results in a significant reduction in the cost of the entire process. The effectiveness of using this approach is also determined by the forms of location and the binding energy of long-lived radionuclides with the structure of graphite.

Thus, the aim of this work is to evaluate possible compounds in which ^{36}Cl radionuclide can be localized, as well as to analyze the causes of its formation and accumulation in irradiated nuclear graphite.

ANALYSIS OF THE HETEROGENEITY OF THE ^{36}C CONTENT IN THE GRAPHITE OF A URANIUM-GRAPHITE REACTOR

Generalized information on the specific activity of the ^{36}Cl radionuclide in irradiated graphite selected from the stack of various UGR is presented in Table 1.

In this work, we assessed the ^{36}Cl content in the graphite of some industrial UGR (IUGR) and RBMK-1000 reactors. The results are summarized in Table 2.

The results of these studies, as well as a number of others, manifest a fairly wide spread in the content of microimpurities leading to ^{36}Cl radionuclide formation, as well as the specific activity of ^{36}Cl in UGR graphite (Tables 1, 2), obtained both from calculations and experiments. Figure 1 shows the results of determining the specific activity of ^{36}Cl and ^{14}C (for comparison) of block graphite at three points by the height of two cells of the IUGR graphite stack, and by the wall thickness of graphite blocks at the points of sampling. The neutron-physical and thermodynamic parameters of graphite operation in both cells are the same.

It is obvious that the distribution of the specific activity of ^{36}Cl varies over a wide range (substantially wider than the neutron-flux variation, irradiation temperature, etc., taking into account a measurement

Table 2. Content assessment results of ^{36}Cl in Russian graphite of a UGR [10]

Location	Reactor type	Type of graphite parts	Activity ^{36}Cl, Bk/g
FSUE PO "Mayak"	AV-1	Blocks	250–890
FSUE PO "Mayak"	AV-2	Blocks	80–400
JSC "ODTS UGR"	I-1	Sleeves	90–450
JSC "ODTS UGR"	ADE-3	Sleeves	240–670
JSC "ODTS UGR"	ADE-4, ADE-5	Blocks	10–1100
Leningrad NPP-2	RBMK-1000	Mixture of chips from graphite blocks	570
Kursk NPP	RBMK-1000	KTK Rings	260–920

error of ≈10–20%), both in the bulk of the stack as a whole (Fig. 1a), and by the wall thickness of individual graphite blocks (Figs. 1c–1e). General regularities in the shape of the curves of the height distribution and distribution over the thickness of the walls of the blocks are also not observed. It should be noted that, in spite of a more complex formation process (by several independent channels), ^{14}C activity, taking into account a measurement error of ≈20%, is almost uniformly distributed (Figs. 1f–1h) over the cross section of the graphite block. A significant heterogeneous ^{14}C distribution is characteristic only for sizes of graphite regions comparable with crystallite sizes.

The large variation in the specific activity of ^{36}Cl, as well as the absence of general regularities of the ^{36}Cl distribution over the graphite stack volume and over the cross section of the graphite block, is probably due to the fact that ^{36}Cl atoms in graphite are present in the form of localized regions (Nugget effect). The distribution of these regions has a high degree of heterogeneity, which manifests itself both in graphite volumes with characteristic sizes of at least ≈1–2 mm (sample thickness), and in volumes with characteristic dimensions of tens of cm (block size). This distribution can be explained by (presumably): (i) the initial distribution of maternal impurity atoms in the form of local formations formed during process of graphite-block fabrication; (ii) the occurrence of some more complex processes that contribute to the formation of local regions of accumulation of ^{36}Cl atoms during operation.

FEATURES OF ^{36}Cl FORMATION IN GRAPHITE UNDER NEUTRON-FLUX IRRADIATION

The long-lived radionuclide ^{36}Cl is formed in graphite stack elements, regardless of the features and operating conditions of a UGR. This is primarily due to the activation of impurities that are not removed during the manufacture of graphite or are sorbed before it is loaded into the reactor core. Table 3 shows the main nuclear reactions of ^{36}Cl -radionuclide formation. The main nuclear reactions leading to the accumulation of radioactive chlorine in graphite RW are: ^{35}Cl(n, γ)^{36}Cl (reaction cross section is $\sigma = 43$ b); ^{39}K(n, α)^{36}Cl ($\sigma = 0.004$ b); ^{34}S(n, β^-)^{35}Cl(n, γ)^{36}Cl ($\sigma = 0.34$ b). The first two reactions are direct, the last is indirect, the half-life of the intermediate radionuclide ^{35}S is 87 days. It should be noted that the contents of the ^{35}Cl isotope in the natural mixture is 34.97%, ^{39}K–93.26%, ^{34}S–4.21% (Table 3).

FORMATION OF THE IMPURITY COMPOSITION UPON MANUFACTURING GRAPHITE

We consider the process of reactor graphite production in more detail in order to determine the content of impurities (chlorine, potassium, sulfur) and the depth of contamination of graphite construction elements.

The fabrication of Russian reactor graphite (GR-220, GR-280, EGP-6, GRP-2, GR-1, etc.) and European/American reactor graphite (TSGBF, TSX, P3AN, PGA, H-327, H-451, IG-11, P3JHA2N, etc.) is based on the use of petroleum cokes of various microstructures, which are fillers, and a coal-tar pitch, which serves as a binder. Instead of the coal-tar pitch or a substance for graphite-material impregnation, synthetic furan resins are also used, which are obtained using the Paal–Knorr method by cycling 1.4-dicarbonyl compounds in the presence of P_2O_5 and $ZnCl_2$ catalysts. Despite binder purification, up to 0.6% sulfur is present in it.

The main technological stages in the production of artificial nuclear graphite are: petroleum coke grinding and crushing; thermal coke calcination; grinding

Table 3. Nuclear reactions of radioactive chlorine accumulation in graphite RW

Nuclear reaction	Reaction cross section σ, b	Isotope content in the natural mixture, %
^{35}Cl(n, γ)^{36}Cl	43	34.97
^{39}K(n, α)^{36}Cl	0.004	93.26
^{34}S(n, β^-)^{35}Cl(n, γ)^{36}Cl	0.34	4.21

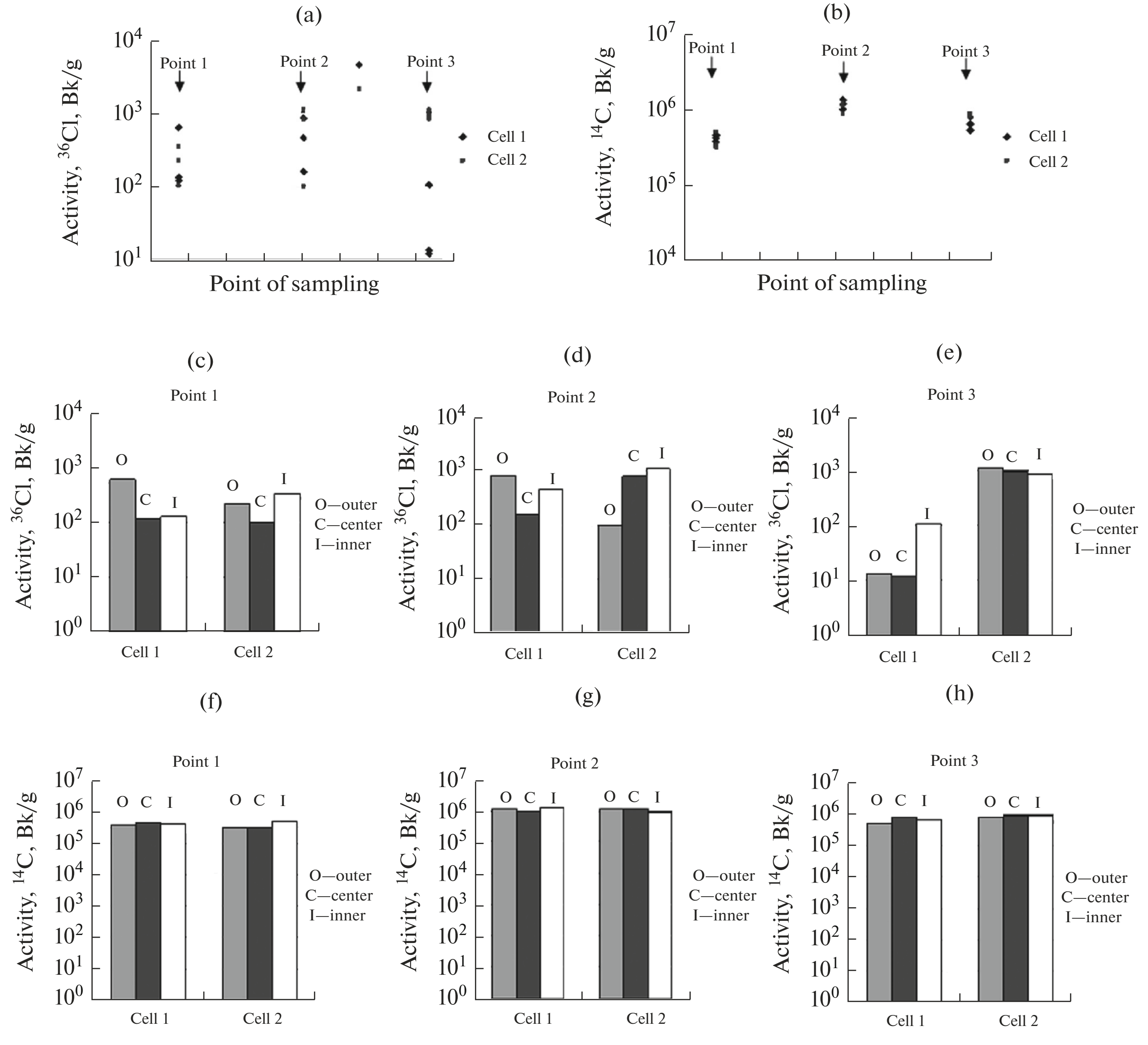

Fig. 1. Specific activity distributions of ^{36}Cl and ^{14}C in graphite the stack of a PUGR: (a, b) by the height of cells; (c–h) by the thickness of the graphite-block walls at the selection marks (the outer surface of the block, the block-wall center, the inner surface of the hole in the block).

and fractionation; binder preparation; dosing and mixing of coke with the binders; forming unfired blanks; high temperature firing; graphitization; creating UGR structural elements. At almost every stage of manufacturing, various microimpurities are accumulated, which are activated during graphite operation in a nuclear reactor.

The structure and properties of carbon-graphite materials are formed in the process of the thermal calcination of coke. In Europe and in the USA, this process is carried out at a temperature of ≈1300°C [11], in Russia (previously in the USSR), petroleum coke is treated in the temperature range of 1100–1300°C without contact with air [12, 13]. An increase in the calcination temperature, on the one hand, increases the structure ordering of two-dimensional crystals, and on the other hand, increases the adsorption capacity of the cokes. This leads to the accumulation of microimpurities in the material after extraction from the furnace, as well as in the process of forming unfired blanks. The average potassium concentration in unfired blanks before the high-temperature cleaning process is ≈6.08 μg/g [14].

The maximum amount of microimpurities in the material is accumulated directly during graphitization, as well as after process completion when removing graphite from electric-resistance furnaces [15]. It is known that the space between the blanks in the fur-

Table 4. Impurity content in various brands of reactor graphite [ppm]

Impurity	USA [19]	Great Britain [20]	France [21]	USSR/Russia [10]
Cl	(0.6–35)	≈2	(8.1–8.3)	(0.1–10.4)
K	(1.4–320)	(0.24–0.36) [22]	(5–50)	≈1.9
S	(10–175)	50	(5–10)	5–52

nace is filled with a powder, which consists of foundry coke containing 0.45–0.50 wt % sulfur [16]. In the process of graphitization, carbon atoms which have free bonds at the edge are able to join various elements (including Cl). Some atoms are able to replace carbon atoms at crystal-lattice sites. Individual molecules can penetrate between graphite layers to form rare-earth- and transition-metal chlorides. Chlorine molecules are capable of forming covalent bonds with carbon atoms, potassium can be in graphite in the form of a substitutional solid solution, and sulfur can be in the free state.

The removal of microimpurities after graphitization is usually carried out by keeping the obtained graphite samples in a gas medium or in vacuum at a temperature of 2500–3000°C [18]. Cleaning in a gas environment is typical for Russian graphite brands. Chlorine and fluorine or their compounds are used as the gas. Some graphite is purified using Freon-12 (difluorodichloromethane, CCl_2F_2), sulfur hexafluoride (SF_6) and Freon-10 (carbon tetrachloride, CCl_4) [19]. Thus, the cleaning of graphite blanks leads, on the one hand, to the removal of most microimpurities, and, on the other hand, to the accumulation of chlorine and sulfur compounds. Table 4 shows generalized data on the content of microimpurities leading to ^{36}Cl radionuclide formation in unirradiated graphite elements.

As can be seen from Table 4, the content of microimpurities leading to ^{36}Cl-radionuclide formation is different for different reactor-graphite brands. According to some reports, the spread exceeds 100 times.

Thus, graphite produced in the USA is the most contaminated, while the purest ones are those produced in the UK and Russia. This is due to the fact that American graphite was historically first produced, and the technology for its purification has still not yet been developed. French graphite (P3AN, 3JHA2N) has a low contamination, which is explained by the use of specialized chemically active gases in the process of its purification upon production.

DETERMINING THE CHEMICAL FORM OF ^{36}Cl

The chemical form of the ^{36}Cl radionuclide in the structure of graphite RW will depend on the channel of its formation. All sources of formation of this radionuclide can be divided into two groups: sources accumulated directly during the production of nuclear graphite and after (up to the moment of unloading irradiated graphite from the reactor). The formation and accumulation of microimpurities during the production of graphite occurs in the preparation of coke and pitch, as well as in the process of high-temperature cleaning and graphitization. After the manufacture of graphite, the adsorption of various chlorine-containing and sulfur-containing compounds from the surrounding environment can occur on its surface (for example, HCl, HOCl, NaCl, oxychlorides, SO_2, H_2S, $(CH_3)_2S$, SO_4^{2-}). Due to incomplete purification of the purge gas, its gaseous chlorides can also be adsorbed during nuclear-reactor operation. Foreign graphite elements are characterized by their transportation to the place of use in a container of polyvinyl chloride ($[-CH_2-CHCl-]_n$), which also leads to contamination of the surface of unirradiated graphite.

COMPOSITION OF REACTION PRODUCTS DURING GRAPHITIZATION AND IN THE PROCESS OF REMOVING MICROIMPURITIES

In order to determine the possible chemical compounds of the ^{36}Cl radionuclide, we carried out thermodynamic simulation of the equilibrium composition of reaction products during graphitization and in the process of removing microimpurities. Achieving the maximum of the characteristic function is a criterion for the system to reach the state of thermodynamic equilibrium. For an isolated system, this function is entropy, and the maximum value of entropy is a criterion for achieving the equilibrium state [23, 24]. For modeling, the specialized program TERRA was used [25]. The phase composition and characteristics of the equilibrium composition were simulated using the reference database [26]. The initial impurity content in unirradiated graphite was chosen according to the data presented in [20]. Calculation was carried out in the graphite-gas system, where Cl_2, CCl_2F_2, SF_6 and CCl_4 were considered as the gas. Figure 2 shows the results.

From Fig. 2 we see that during graphite-billet purification, using chlorine-containing gases (Figs. 2a, 2b, 2d), gaseous Cl_2 is accumulated, penetrates graphite through a developed system of pores and is adsorbed on the surface. When using Cl_2 and CCl_4 gases, most of the reaction products, which are able to form the ^{36}Cl radionuclide during the irradiation process, are

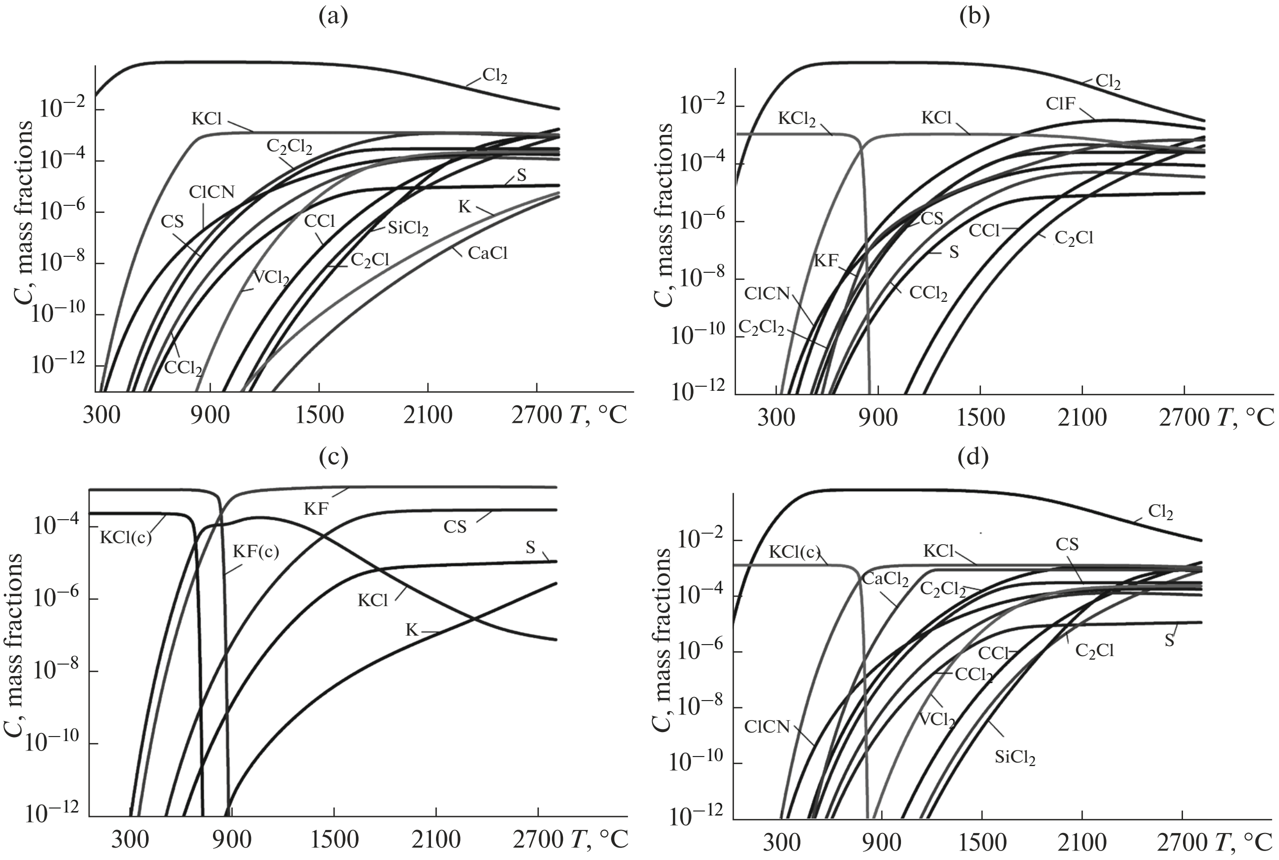

Fig. 2. Thermodynamic modeling of the equilibrium composition of reaction products during unirradiated graphite purification with various gases: (a) Cl_2, (b) CCl_2F_2, (c) SF_6, (d) CCl_4.

presented in the form of chlorides: C_2Cl_2, CCl, CCl_2, ClCN, KCl, $SiCl_2$, VCl_2, $CaCl_2$. To a lesser extent, sulfur compounds are formed, which, as a rule, are present in the form of carbon disulfide. When using difluorodichloromethane as a cleaning gas, chlorination and fluorination of the graphite surface occur simultaneously. Almost all of the existing impurities interact with fluorine and are removed from the surface being cleaned. However, the formation of ClF and KF compounds is possible, which are subsequently irradiated with neutrons.

The most complete cleaning of graphite blanks occurs when sulfur hexafluoride SF_6 is used as the heat carrier. This is due to the fact that fluorine interacts with all microimpurities and forms gaseous reaction products, and the lack of chlorine eliminates the appearance of chlorides in the graphite structure. In this case, the amount of sulfates and sulfides formed is insignificant. After treatment with sulfur hexafluoride, the main compounds found in unirradiated graphite are CS_2, K, KF, and KCl.

The calculation results obtained are valid only for the processes of graphitization and purification. When removing a blank from furnaces and chambers, interaction of the compounds contained in the graphite blank with air, atmospheric impurities and water vapor can occur.

REACTION-PRODUCT COMPOSITION DURING BILLET EXTRACTION FROM FURNACES AND CHAMBERS

Consider the main chemical reactions that describe such processes:

$$ClCN + H_2O \rightarrow HNCO + HCl, \quad (1)$$

$$HNCO + H_2O \rightarrow NH_3 + CO_2, \quad (2)$$

$$ClCN + NH_3 \rightarrow H_2N\text{-}CN + HCl, \quad (3)$$

$$4ClF + 2H_2O \rightarrow 2Cl_2 + O_2 + 4HF, \quad (4)$$

$$ClF + H_2 \rightarrow HCl + HF, \quad (5)$$

$$CS_2 + 3O_2 \rightarrow CO_2 + 2SO_2, \quad (6)$$

$$CS_2 + 2H_2O \rightarrow CO_2 + 2H_2S, \quad (7)$$

Reactions (1)–(2) reflect the process of the hydrolysis of cyanogen chloride with water vapor to form isocyanic acid, which, in turn, hydrolyzes to form ammonia and carbon dioxide. The resulting ammonia can also interact with ClCN, leading to HCl formation. Chlorine fluoride, interacting with water vapor and hydrogen, can form molecular chlorine Cl_2 by reaction (4) and hydrogen chloride HCl by reaction (5). Carbon disulfide CS_2 is able to oxidize in air to SO_2 via reaction (6). When CS_2 interacts with water, the formation of hydrogen sulfide H_2S occurs according to reaction (7). However, the latter reaction proceeds at temperatures above 150°C, which is achieved only during nuclear-reactor operation and in the case of process-channel depressurization [27].

EXPERIMENTAL DETERMINATION OF THE CHEMICAL FORM OF THE ^{36}Cl RADIONUCLIDE IN GRAPHITE

The results of determining the chemical form of the ^{36}Cl radionuclide by X-ray photoelectron spectroscopy of the surface of irradiated nuclear graphite are presented in [28]. Chlorine was shown to have a covalent bond with carbon on the studied surface. In this case, the sp^2 and sp^3 hybridization of carbon atoms occurs. The considered radionuclide is most likely located in the Cl_2 molecule or bound to C–Cl carbon. On the other hand, mass spectrometry studies show the presence of the HCl compound [29]. Based on this, the possible reactions between carbon and chlorine are [30]:

—reaction of hydrogen substitution by chlorine:

$$-C-H + Cl_2 \rightarrow -C-Cl + HCl; \quad (8)$$

—reaction of joining to conjugated double bonds:

$$C{=}C + Cl_2 = ClC-CCl, \quad (9)$$

—dehydrogenation reaction:

$$HC-CH + Cl_2 = C{=}C + 2HCl. \quad (10)$$

It was shown in [31] that reaction (8) is multistage and includes the chemisorption of gaseous Cl_2, the diffusion of compounds onto the graphite surface (including the pore surface) and HCl formation. In this case, the reaction of hydrogen substitution with chlorine is irreversible, and chlorine itself is not removed by heating or chemical purification [32]. During graphite treatment at a temperature of 500°C, an irreversible process of formation of the C–Cl covalent bond occurs, as a result of which chlorine occupies the position between the graphite layers.

In addition, it was experimentally determined that the ^{36}Cl radionuclide (to a lesser extent) may be present in the following compounds: $NaClO_4$, $NaClO_3$, $NaClO_2$, NaCl, $[-CH_2-CHCl-]_n$, HClO, CH_3Cl, $ClONO_2$, HClO, $HClO_2$, $HClO_3$, ClO, ClO_2, ClO_3 [33, 34].

CONCLUSIONS

The considered channels for ^{36}Cl-radionuclide formation explain its localization in irradiated graphite. This is manifested in an absence of regularities in its distribution in graphite volumes with characteristic dimensions of at least ≈1–2 mm (sample thickness), as well as in volumes with characteristic dimensions of tens of cm (block size). This is likely one explanation of the Nugget effect.

The experimental results presented here are in qualitative agreement with the results of the thermodynamic modeling of the equilibrium composition. Therefore, the maximum amount of impurities from which ^{36}Cl is formed when using graphite in the nuclear reactor, is accumulated in the process of graphitization, purification in a medium of chemically active gases and when transporting prefabricated construction elements. In this case, the compounds in which the considered radionuclide can exist are most likely HCl, KCl and, to a lesser extent, the following: $NaClO_4$, $NaClO_3$, $NaClO_2$, NaCl, $[-CH_2-CHCl-]_n$, HClO, CH_3Cl, $ClONO_2$, HClO, $HClO_2$, $HClO_3$, ClO, ClO_2, ClO_3.

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