Research Article

Computer simulation of thermal processes involving Sr and Ca radionuclides in the process of heating radioactive graphite in an air atmosphere[*]

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Abstract

The paper presents the results from a thermodynamic analysis of the behavior of Sr and Ca radionuclides in the process of heating radioactive graphite in an air atmosphere. The TERRA software package was used for the thermodynamic analysis in a temperature range of 300 to 3600 K to determine the possible composition of the ionized, gaseous and condensed phases. It has been found that strontium is in the form of condensed $SrCl_{2(c)}$ and gaseous $SrCl_2$ in a temperature range of 300 to 1600 K, and in the form of gaseous $SrCl_2$, SrO, $SrCl$ and Sr and ionized $SrCl^+$, Sr^+ and SrO^+ when the temperature is increased from 1600 to 3600 K. Calcium is in the form of condensed $CaCl_{2(c)}$, $CaUO_{4(c)}$, $CaO_{(c)}$ and gaseous $CaCl_2$ in the temperature interval between 300 and 2100 K, and in the form of gaseous Ca, $CaCl$ and CaO and ionized Ca^+, CaO^+ and $CaCl^+$ when the temperature is increased from 2100 to 3600 K. The paper determines the key reactions within individual phases and among condensed, gaseous and ionized phases. The equilibrium constants of their reactions have been calculated. Based on the results obtained, dependence plots are presented for the Sr and Ca radionuclide distribution by phases.

Keywords

Introduction

Presently, nuclear power is one of the major sources of electricity generation worldwide. Altogether, the Russian Federation operates 11 nuclear power plants (NPP) of the total installed capacity in excess of 29.5 GW with 37 serviceable power units of which 8 are of the RBMK series (large graphite-moderated water-cooled pressure-tube boiling reactors) (Rosatom State Atomic Energy Corporation 2023; The IAEA Database on Nuclear Power Reactors 2023). The RBMK-1000 reactor core and reflector contain 1850 tons of nuclear pure graphite.

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Therefore, decommissioning of such reactor requires to be followed by reliable procedures for handling tens of thousands of tons of irradiated graphite. The uranium-graphite reactor decommissioning process represents a set of complex tasks involving the selection of appropriate ways and techniques for radioactive graphite handling (Skachek 2007; Safety of Nuclear Power Plant Design 2016; Velkin 2021).

One of the ways to reduce the radioactivity of graphite is its high-temperature treatment in different environments. Under the action of temperature, a part of the radionuclides passes into a gaseous state and is removed from the system (Skachek 2007).

Oxidation of radioactive graphite with gaseous and gasifying agents, such as air, oxygen, carbon dioxide and their mixtures, is tested in laboratory conditions. Computer-aided simulation of the reactor graphite reprocessing via heating in an atmosphere of air allows one to evaluate in advance the behavior of radioactive elements.

Thermodynamic simulation of the equilibrium high-temperature behavior of the system under investigation makes it possible to obtain information on the aspects of a probable beyond-design-basis accident. The results obtained allow evaluating some of the capabilities for reprocessing radioactive graphite from a nuclear power reactor series.

The purpose of the study is to determine the equilibrium gas phase composition in the course of the radioactive graphite interaction with air in a broad temperature range.

The study aims to simulate thermodynamically the system under consideration and determine the equilibrium constants for the key reactions taking place in the process of heating radioactive graphite containing Sr and Ca radionuclides.

Calculation methodology

The behavior of Sr and Ca the nuclear reactor graphite contains was investigated via thermodynamic simulation using the TERRA computer code and the IVTANTERMO database expanded at the expense of the HSC5 database information (Moiseev et al. 2002; Belov and Trusov 2013).

The calculation of the TERRA equilibrium composition is based on the entropy maximum principle which is just in accordance with the second law of thermodynamics for any equilibrium system irrespective of the way by which the system has reached equilibrium (Sinyarev et al. 1982).

$$S = \sum_{i=1}^{k} S_i^{(p_i)} \cdot n_i + \sum_{l=1}^{L} S_l \cdot n_l = \sum_{i=1}^{k} \left(S_i^0 - R_0 \ln \frac{R_0 T n_i}{v} \right) \cdot n_i + \sum_{l=1}^{L} S_l^0 \cdot n_l \quad (1)$$

where $S_i^{(p_i)}$ is the entropy of the ith gas phase component at a partial pressure of $p_i = (R_o T n_i)/v$; S_l is the entropy of the condensed phase, l, depending only on temperature; v is the specific volume of the entire system; and S_i^0 is the standard entropy of the ith gas phase component at temperature T and at a pressure equal to 1 physical atmosphere.

Thermodynamic simulation consists in analyzing thermodynamically the equilibrium state of systems as the whole (complete thermodynamic analysis). The theoretical fundamentals for thermodynamic simulation are set forth in Barbin et al. 2017, 2019, 2022. The initial composition of the radioactive graphite system in air is presented in Table 1. The chemical compound forms of Sr and Ca radionuclides required for thermodynamic simulation are presented in Table 2 (Bazhenov et al. 1990; Shidlovsky et al. 2010).

Table 1. Initial composition of a radioactive graphite system in air

Phase	Phase composition	Content, mass %
Gas (89.44%)	O_2	21.42743167
	N_2	78.57256833
Condensed (10.56%)	C	99.98612976
	U	$1.15 \cdot 10^{-2}$
	Cl	$1.89 \cdot 10^{-3}$
	Ca	$2.70 \cdot 10^{-4}$
	Pu	$7.27 \cdot 10^{-5}$
	Be	$1.20 \cdot 10^{-5}$
	Ni	$8.19 \cdot 10^{-6}$
	Cs	$3.36 \cdot 10^{-6}$
	Am	$9.27 \cdot 10^{-6}$
	Sr	$1.10 \cdot 10^{-6}$
	Eu	$1.15 \cdot 10^{-6}$

Results and discussion

The distribution of strontium by equilibrium phases in the process of radioactive graphite heating in air is presented in Fig. 1.

As temperatures increase from 1000 to 1600 K, condensed strontium chloride passes into gaseous strontium chloride in accordance with reaction (1) (Table 3). In a temperature interval of 1700 to 2100 K, gaseous strontium chloride decomposes reversibly into ionized chlorous strontium and gaseous chlorine according to reaction (2).
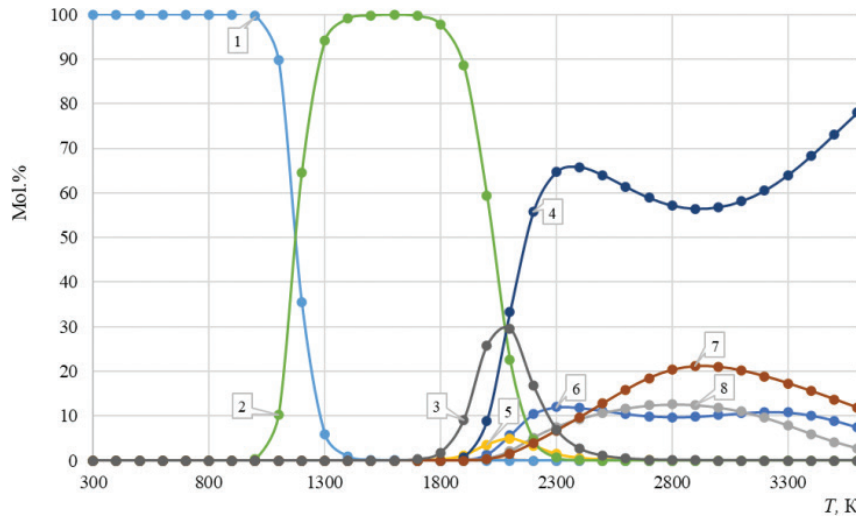
Table 2. Expected forms of radionuclide existence

Radionuclide in graphite	Type of compound in equilibrium system
[90]Sr	Sr, $SrO_{(c)}$, SrO, SrCl, $SrCl_{2(c)}$, $Sr_3N_{2(c)}$, $SrN_2O_{6(c)}$, $SrO_{(c)}$, $SrC_{2(c)}$, $SrCO_{3(c)}$, $SrCl_2$, Sr^+, SrO^+, $SrCl^+$
[41]Ca	Ca, $CaO_{(c)}$, CaO, CaCl, $CaCl_{2(c)}$, $CaCl_2$, $CaO_{2(c)}$, $CaOCl_{2(c)}$, $CaC_{2(c)}$, $CaO_2H_{2(c)}$, $Ca_3N_{2(c)}$, $CaN_2O_{6(c)}$, $CaUO_{4(c)}$, $CaCO_{3(c)}$, Ca^+, CaO^+, $CaCl^+$

Figure 1. Distribution of strontium by equilibrium phases in the process of radioactive graphite heating in air: $1 - SrCl_{2(c)}$; $2 - SrCl_2$; $3 - SrCl^+$; $4 - Sr^+$; $5 - SrCl$; $6 - Sr$; $7 - SrO^+$; $8 - SrO$.

Table 3. Key reactions and their respective equilibrium constants

№	Reaction	ΔT, К	A	B	ΔA	ΔB
1	2	3	4	5	6	7
1	$SrCl_{2(c)} = SrCl_2$	1000–1600	16.56	-35451	0.311	389.04
2	$SrCl_2 = SrCl^+ + Cl + e^-$	1700–2100	14.13	-57112	0.659	1242.22
3	$SrCl_2 = Sr^+ + 2Cl + e^-$	1900–2400	24.45	-106941	0.236	502.71
4	$SrCl_2 = Sr + 2Cl$	1900–2400	23.47	-108601	0.013	29.16
5	$SrCl_2 = SrCl + Cl$	1800–2100	-11.92	-3161.6	0.281	574.25
6	$SrCl_2 + CO_2 = SrO^+ + CO + 2Cl + e^-$	2000–2300	30.81	-125548	0.333	714.49
7	$SrCl_2 + CO_2 = SrO^+ CO + 2Cl + e^-$	2000–2300	29.03	-121148	0.024	52.24
8	$SrCl^+ = Sr^+ + Cl$	2100–2300	12.32	-53851	0.011	23.41
9	$SrCl^+ = Sr + Cl - e^-$	2100–2300	11.53	-55866	0.574	1260.91
10	$SrCl^+ + CO_2 = SrO^+ + Cl + CO$	2100–2600	19.21	-73595	0.003	8.78
11	$SrCl^+ + CO_2 = SrO + Cl + CO - e^-$	2100–2600	16.13	-66319	0.221	516.71
12	$SrCl = Sr+ + Cl$	2100–2400	11.93	-49340	0.438	983.81
13	$SrCl = Sr + Cl$	2100–2400	10.59	-50175	0.0093	20.83
14	$SrCl + CO_2 = SrO^+ + CO + Cl + e^-$	2100–2500	19.15	-69803	0.318	727.97
15	$SrCl + CO_2 = SrO + CO + Cl$	2100–2500	16.22	-62860	0.019	44.88
16	$Sr^+ + CO_2 = SrO^+ + CO$	2400–2900	6.78	-19496	0.011	26.51
17	$Sr^+ + CO_2 = SrO + CO - e^-$	2400–2900	3.83	-12557	0.146	386.69
18	$Sr + CO_2 = SrO^+ + CO + e^-$	2300–2900	8.84	-20328	0.118	305.42
19	$Sr + CO_2 = SrO + CO$	2300–2900	5.74	-12976	0.018	48.51
20	$SrO^+ = Sr^+ + O$	2900–3600	11.29	-44572	0.005	17.95
21	$SrO^+ = Sr + O + e^-$	2900–3300	9.36	-43979	0.345	1068.25
22	$SrO = Sr^+ + O + e^-$	2900–3600	16.24	-57842	0.705	2276.03
23	$SrO = Sr + O$	2900–3300	11.94	-49980	0.024	75.35
24	$Sr = Sr^+ + e^-$	3300–3600	8.53	-22421	0.793	2733.27
25	$CaCl_{2(c)} = CaCl_2$	1000–1500	14.99	-31816	0.281	340.41
26	$CaCl_2 + UO_{2(c)} + 2CO_2 = CaUO_{4(c)} + 2Cl + 2CO$	1500–1800	13.35	-48601	0.033	55.08
27	$CaCl_2 + CO_2 = CaO_{(c)} + 2Cl + CO$	1500–1800	11.35	-47684	0.033	55.18
28	$CaUO_{4(c)} + Cl = CaCl + UO_3 + O$	1800–2100	40.29	-139686	0.016	32.61
29	$CaO_{(c)} + 2Cl = CaCl_2 + O$	1800–2000	6.51	-15980	0.057	109.26
30	$CaCl_2 = Ca + 2Cl$	1900–2400	24.81	-109604	0.005	11.72
31	$CaCl_2 = Ca^+ + 2Cl + e^-$	1900–2400	25.79	-112786	0.232	495.71
32	$CaCl_2 = CaCl + Cl$	1900–2100	14.06	-60059	0.011	23.82
33	$CaCl_2 + CO_2 = CaO_{(c)} + 2Cl + CO$	1900–2100	11.68	-48293	0.004	9.31769
34	$CaCl_2 = CaCl^+ + Cl + e^-$	1900–2100	13.36	-62200	0.664	1325.78
35	$CaCl = Ca + Cl$	2100–2300	10.78	-49615	0.011	22.31
36	$CaCl = Ca^+ + Cl + e^-$	2100–2600	12.65	-54812	0.247	577.98
37	$CaCl + CO_2 = CaO + CO + Cl$	2100–2600	16.45	-67409	0.025	59.52
38	$CaCl + CO_2 = CaO^+ + CO + Cl + e^-$	2100–2600	19.33	-76763	0.239	558.21
39	$CaCl^+ = Ca + Cl - e^-$	2100–2300	11.81	-48091	0.575	1263.72

№	Reaction	ΔT, К	A	B	ΔA	ΔB
1	2	3	4	5	6	7
40	$CaCl^+ = Ca^+ + Cl$	2100–2300	12.59	-50920	0.011	22.91
41	$CaCl^+ + CO_2 = CaO + Cl + CO + e^-$	2100–2400	16.85	-64527	0.405	909.23
42	$CaCl^+ + CO_2 = CaO^+ + Cl + CO$	2100–2400	19.31	-72925	0.001	4.12
43	$Ca = Ca^+ + e^-$	2300–2700	2.33	-6336.2	0.046	115
44	$Ca + CO_2 = CaO + CO$	2300–2900	5.79	-18085	0.023	60.19
45	$Ca + CO_2 = CaO^+ + CO + e^-$	2300–3000	8.52	-27082	0.131	343.56
46	$CaO = Ca^+ + O + e^-$	2900–3600	16.22	-57656	0.708	2285.16
47	$CaO^+ = Ca^+ + O$	3000–3600	11.47	-42332	0.005	18.9981

In the temperature region between 1900 and 2400 K, thermal dissociation of gaseous strontium chloride takes place by reactions (3)–(4). In the temperature range between 1800 to 2100 K, gaseous strontium chloride passes into gaseous chlorous strontium and gaseous chlorine in accordance with reaction (5). In the temperature region between 2000 and 2300 K, gaseous strontium chloride interacts with carbon dioxide and passes into ionized strontium oxide, carbon monoxide and gaseous chlorine according to reaction (6). In the same temperature interval, based on reaction (7), gaseous strontium chloride reacts with carbon dioxide and passes into gaseous strontium oxide, carbon monoxide and gaseous chlorine. At a temperature of 2100 to 2300 K, thermal dissociation of ionized chlorous strontium takes place in accordance with reactions (8)–(9). In a temperature range of 2100 to 2600 K, the interaction of ionized chlorous strontium with carbon dioxide results in ionized strontium oxide, gaseous chlorine and carbon monoxide formed (reaction (10)). In the same temperature region, ionized chlorous strontium starts to interact with carbon dioxide, this resulting in gaseous strontium oxide, gaseous chlorine and carbon monoxide formed (reaction (11)). In the temperature region between 2100 and 2400 K, thermal dissociation of gaseous chlorous strontium takes place according to reactions (12)–(13). As temperatures increase from 2100 to 2500 K, according to reaction (14), gaseous chlorous strontium interacts with carbon dioxide and forms ionized strontium oxide, carbon monoxide and gaseous chlorine. In the same temperature interval, as gaseous chlorous strontium interacts with carbon dioxide,

gaseous strontium oxide, carbon monoxide and gaseous chlorine form (see reaction (15). In the temperature region between 2400 and 2900 K, ionized strontium reacts with carbon dioxide and passes into ionized strontium oxide and carbon monoxide in the first case, and into gaseous strontium oxide and carbon monoxide in the second case (in accordance with reactions (16)–(17). In the temperature region between 2300 and 2900 K, gaseous strontium reacts with carbon dioxide and transforms into ionized strontium oxide and carbon monoxide in the first case (based on reaction (18), and into gaseous strontium oxide and carbon monoxide in the second case (according to reaction (19). As temperatures increase from 2900 to 3600 K, ionized strontium oxide passes into ionized strontium and oxygen according to reaction (20). In the temperature range between 2900 and 3300 K, based on reaction (21), ionized strontium oxide transforms into gaseous strontium and oxygen. In the same temperature range, thermal dissociation of gaseous strontium oxide takes place by reaction (22). In the temperature interval between 2900 and 3600 K, in accordance with reaction (23), gaseous strontium oxide decomposes reversibly into ionized strontium and oxygen. In the temperature range between 3300 and 3600 K, gaseous strontium passes into ionized strontium according to reaction (24).

The distribution of calcium by equilibrium phases in the process of radioactive graphite heating in an atmosphere of air is shown in Fig. 2. In the temperature interval between 1000 and 1500 K, according to reaction (25), condensed calcium chloride passes into gaseous calcium chloride.
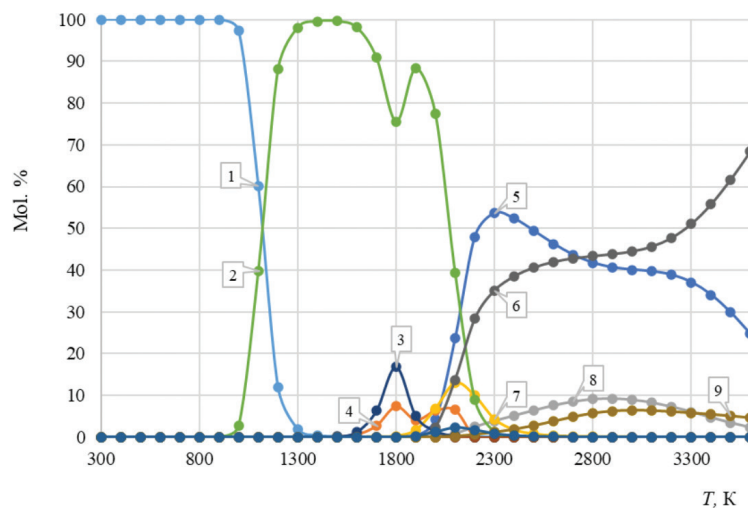

Figure 2. Distribution of calcium by equilibrium phases in the process of radioactive graphite heating in air: 1 – $CaCl_{2(c)}$; 2 – $CaCl_2$; 3 – $CaUO_{4(c)}$; 4 – $CaO_{(c)}$; 5 – Ca; 6 – Ca^+; 7 – CaCl; 8 – CaO; 9 – CaO^+; 10 – $CaCl^+$.

In the temperature region between 1500 and 1800 K, gaseous calcium chloride interacts with carbon dioxide with this resulting in condensed calcium oxide, gaseous chlorine and carbon monoxide formed by reaction (26). At a temperature of 1500 to 1800 K, in accordance with reaction (27), gaseous calcium chloride reacts with condensed uranium oxide (IV) and carbon dioxide, this resulting in condensed calcium uranate, gaseous chlorine and carbon monoxide formed. In a temperature interval of 1800 and 2100 K, based on reaction (28), condensed calcium uranate starts to react with gaseous chlorine with this resulting in gaseous chlorous calcium, uranium oxide (VI) and oxygen formed. In the temperature range between 1800 and 2000 K, according to reaction (29), condensed calcium oxide reacts with gaseous chlorine and passes into gaseous calcium chloride with oxygen formed. In a temperature interval of 1900 and 2400 K, thermal dissociation of gaseous calcium chloride takes place according to reactions (30)–(31).

In the temperature interval between 1900 and 2100 K, gaseous calcium chloride transforms into gaseous chlorous calcium and gaseous chlorine by reaction (32). In the same temperature range, based on reaction (33), gaseous calcium chloride interacts with carbon dioxide with condensed calcium oxide, gaseous chlorine and carbon monoxide formed. In the same temperature region, thermal dissociation of gaseous calcium chloride takes place according to reaction (34). As temperatures increase from 2100 to 2300 K, in accordance with reaction (35), gaseous chlorous calcium decomposes reversibly into gaseous calcium and chlorine. As the temperature increases from 2100 to 2600 K, gaseous chlorous calcium decomposes reversibly as well into ionized calcium and gaseous chlorine by reaction (36).

In the temperature interval between 2100 to 2600 K, according to reactions (37)–(38), gaseous chlorous calcium interacts with carbon dioxide and passes into gaseous calcium oxide, carbon monoxide and gaseous chlorine in the first case, and into ionized calcium oxide, carbon monoxide and gaseous chlorine in the second case. In the temperature region between 2100 and 2300 K, ionized chlorous calcium decomposes reversibly into gaseous calcium and chlorine in the first case (based on reaction (39)), and into ionized calcium and gaseous chlorine in the second case (by reaction (40)).

At a temperature of 2100 to 2400 K, in accordance with reactions (41)–(42), ionized chlorous calcium interacts with carbon dioxide and passes into gaseous calcium oxide, gaseous chlorine and carbon monoxide in the first case, and into ionized calcium oxide, gaseous chlorine and carbon monoxide in the second case. As the temperature increases from 2300 to 2700 K, gaseous calcium transforms into ionized calcium by reaction (43). In the temperature range between 2300 to 2900 K, according to reaction (44), the gaseous calcium interaction with carbon dioxide results in gaseous calcium oxide and carbon monoxide formed. In a temperature interval of 2300 to 3000 K, gaseous calcium starts to interact with carbon dioxide and forms ionized calcium oxide and carbon monoxide according to reaction (45). In the temperature region between 3000 and 3600 K, thermal dissociation from gaseous calcium oxide is observed in reaction (46) and that from ionized calcium oxide is observed in reaction (47).

The equilibrium constants are represented by analytical equations of the following form:

$$\ln K_i = A_i + \frac{B_i}{T} \qquad (2)$$

where A_i and B_i are the equilibrium constants presented in Table 3.

The equilibrium constants of reactions as a function of $1/T$ are shown in Figs 3 through 5. In a temperature region of 1000 to 1900 K, the reaction equilibrium constants increase as the temperature increases, this change being shown in reactions (1, 2, 25, 26–29) in Fig. 3.
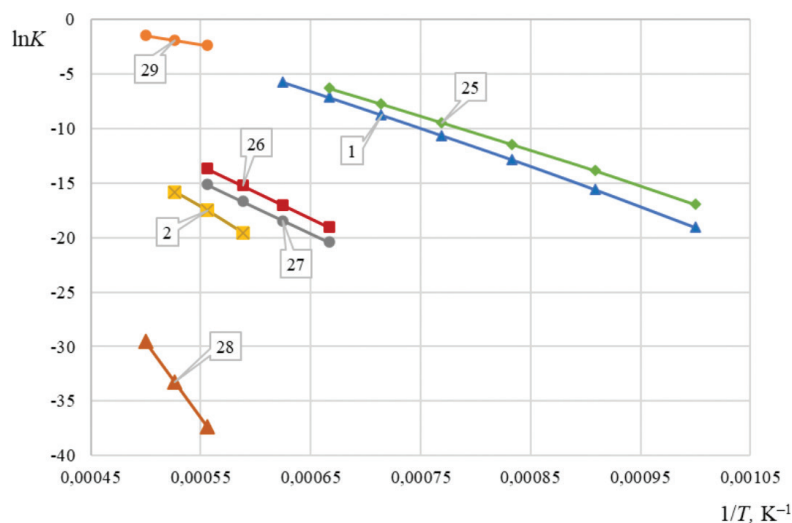

Figure 3. Equilibrium constants of reactions as a function of $1/T$ in the process of radioactive graphite heating in air at $T = 1000$–1900 K.

In a temperature interval of 1900 to 2900 K, the equilibrium constants of reactions increase as the temperature increases, this change being shown in reactions (2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12, 13, 14, 15, 16, 17, 18, 19, 29, 30, 31, 32, 33, 34, 35, 36, 37, 38, 39, 40, 41, 42, 43, 44, 45) in Fig. 4.

In the temperature range between 2200 and 3600 K, the equilibrium constants of reactions increase as the temperature increases, this change being shown in reactions (20, 21, 22, 23, 24, 46, 47) in Fig. 5.

According to the thermodynamic probability of occurrence in this system, the reactions are arranged as a series in the downward order (29, 25, 1, 26, 27, 2, 28) in the temperature range between 952 K and 2 222 K, in the downward order (18, 19, 43, 44, 29, 16, 17, 45, 5, 34, 40, 12, 13, 35, 10, 11, 36, 2, 37, 42, 14, 15, 39, 38, 33, 32, 3, 4, 6, 7, 8, 9, 30, 31) in the temperature range between 1818 K and 3030 K, and in the downward order (24, 22, 46, 47, 20, 23, 21) in the temperature range between 2 857 K and 3 703 K.
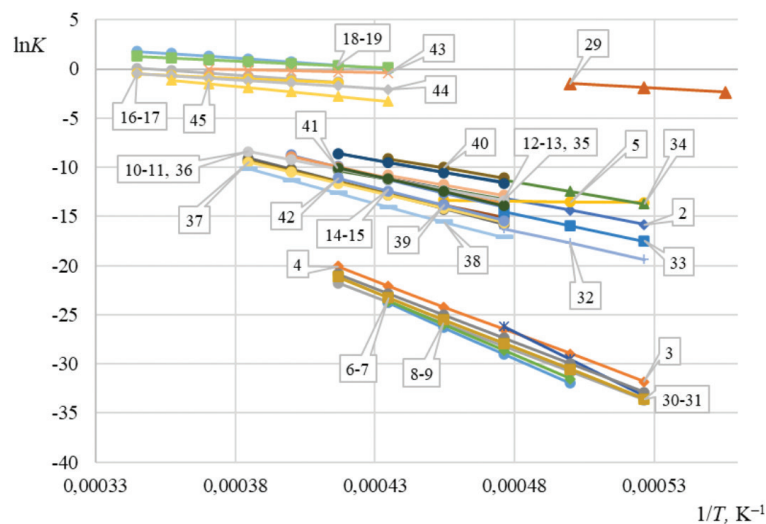

Figure 4. Equilibrium constants of reactions as a function of $1/T$ in the process of radioactive graphite heating in air at $T = 1900$–2900 K.

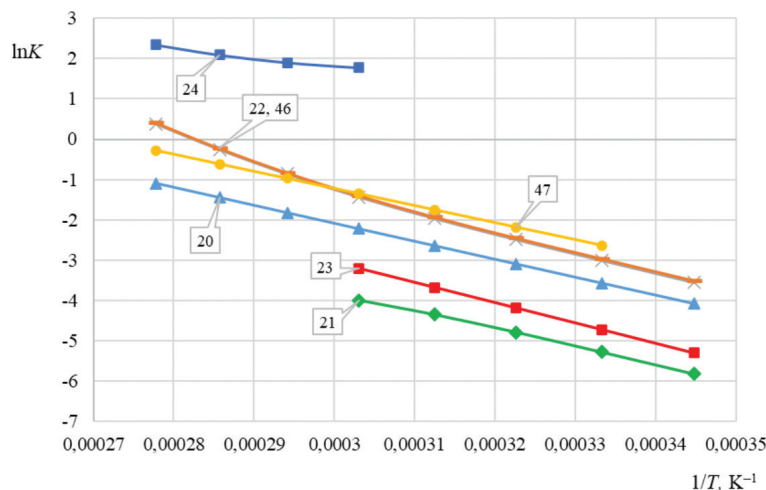**Figure 5.** Equilibrium constants of reactions as a function of $1/T$ in the process of radioactive graphite heating in air at $T = 2900$–3600 K.

Conclusions

The paper presents the results from a thermodynamic simulation of thermal processes in the course of radioactive graphite heating in an atmosphere of air. Based on the simulation results, dependence diagrams have been plotted for the Sr and Ca radionuclide distributions by phases. In accordance with the dependence diagrams for the Sr and Ca radionuclide distributions by phases, key reactions inside individual phases and on the interphase boundary

have been derived. Equilibrium constants have been calculated for these reactions. The computer-aided simulation results have shown that Sr and Ca, which are present initially in a graphite-containing system as impurities, are prone to passing into a gas phase in the process of heating in air as particular temperatures are reached. This can be explained by the graphite and condensed phase interaction with air in the explored temperature range, this leading to condensed phases becoming more unstable and their components passing into a gas phase.

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