



# Computer modeling of thermal processes involving calcium, strontium and cesium during radioactive graphite heating in the carbon dioxide atmosphere

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Available online 26 May 2017

## Abstract

The design of nuclear reactors did not provide for decommissioning solutions, and there were no safe technologies to handle irradiated reactor graphite. Decommissioning of uranium–graphite reactors is a combination of complex tasks involving the selection of appropriate methods and techniques for the radioactive graphite handling. Computer modeling of the reactor graphite reprocessing by heating in a carbon dioxide environment makes it possible to determine the behavior of radioactive elements. The behavior of Ca, Sr and Cs during radioactive graphite heating in the carbon dioxide atmosphere was studied through computer modeling. It has been found that calcium is present as vapors of Ca, CaO, CaCl and CaCl<sub>2</sub>, as Ca<sup>+</sup> and CaO<sup>+</sup> ions, and as condensed forms of CaCO<sub>3</sub>, CaCl<sub>2</sub> and CaO. Strontium is present as vapors of Sr, SrO, SrCl and SrCl<sub>2</sub>, as Sr<sup>+</sup> and SrO<sup>+</sup> ions, and as condensed forms of SrCl<sub>2</sub>, SrCO<sub>3</sub> and SrO. Cesium is present as vapors of Cs and CsCl, as Cs<sup>+</sup> ions, and as a condensed phase of CsCl<sub>2</sub>. Basic reactions have been identified and their respective equilibrium constants have been determined. The data obtained has shown that formation of calcium, strontium and cesium chloride vapors takes place at temperatures of 573 to 973 K. A temperature increase to 1373 K leads to thermal ionization of cesium chloride and to formation of ionized cesium. As the temperature increases to 2273 K, thermal ionization of strontium and cesium is observed and ionized calcium and strontium form.

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**Keywords:** Thermodynamic modeling; Thermal processes; Equilibrium constant; Radionuclides; Radioactive graphite; Heating; Carbon dioxide.

## Introduction

Nuclear power plants are one of the major sources of electricity generation. In Russia, 40% of NPPs have nuclear

reactors with graphite used as the neutron moderator and reflector. The design of nuclear reactors did not provide for decommissioning solutions, and there were no safe technologies to handle irradiated reactor graphite [1,2].

Decommissioning of uranium–graphite reactors is a combination of complex tasks involving the selection of appropriate methods and techniques for the radioactive graphite handling [3].

One of the ways to reduce the radioactivity of graphite is high-temperature treatment in different environments. Some of the radionuclides pass to a gaseous state under the action of temperature and are removed from the system [4,5].

Oxidation of radioactive graphite by gaseous gasifying agents, such as air, oxygen, carbon dioxide, steam and their respective mixtures, is tested on a laboratory scale. Computer

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Peer-review under responsibility of National Research Nuclear University MEPhI (Moscow Engineering Physics Institute).

Russian text published: *Izvestiya vuzov. Yadernaya Energetika* (ISSN 0204-3327), 2017, n.1, pp. 73–82.

Table 1  
Radionuclides in graphite.

Radionuclide	Half-life, years [17]	Compound type
<sup>41</sup> Ca	102 000	Ca(g), CaO(cond.), CaO(g), CaCl(g), CaCl <sub>2</sub> (cond.), CaCl <sub>2</sub> (g), CaCO <sub>3</sub> (cond.), CaUO <sub>4</sub> (cond.), Ca <sup>+</sup> , CaO <sup>+</sup> , CaCl <sup>+</sup>
<sup>90</sup> Sr	28.90	Sr(g), SrO(cond.), SrO(g), SrCl(g), SrCl <sub>2</sub> (cond.), SrCl <sub>2</sub> (g), SrCO <sub>3</sub> (cond.), Sr <sup>+</sup> , SrO <sup>+</sup> , SrCl <sup>+</sup>
<sup>137</sup> Cs, <sup>134</sup> Cs	30.1671 2.0652	Cs(g), CsO(g), CsCl(cond.), CsCl(g), Cs <sup>+</sup>

modeling of the reactor graphite reprocessing by heating in a carbon dioxide environment makes it possible to predict the behavior of radioactive elements.

The paper considers the behavior of calcium, strontium and cesium during the reactor graphite heating in the carbon dioxide atmosphere. The heating of graphite in the carbon dioxide atmosphere may take place during a beyond design basis accident in a graphite moderated gas cooled nuclear reactor.

### Calculation procedure

The studies were based on a thermodynamic modeling method [6–10] used successfully in materials science [9–13] and physics [10–16] for investigations on inorganic substances at high temperatures.

Thermodynamic modeling involves a thermodynamic analysis of the equilibrium state of systems as the whole (a complete thermodynamic analysis). The theoretical basis for thermodynamic modeling is described in [7–10].

Table 1 presents radioactive elements contained in the reactor graphite and their chemical compounds, the thermodynamic behavior of which was taken into account in the thermodynamic modeling. The chemical properties of stable nonradioactive elements do not differ from the properties of their radioactive isotopes [6,7].

Calculations were performed using the TEPPA software designed for studies on high-temperature processes [18]. It was assumed initially in the assignment for the calculation that, with an allowance for the given carbon dioxide atmosphere, an ideal solid solution of interaction products could form in the investigated temperature interval, including the above listed oxides and salts of the radionuclides in question. It was also assumed that volatile compounds containing these radionuclides could contribute to the formation of an ideal gaseous solution of interaction products.

The numerical experiment was conducted in the carbon dioxide atmosphere at the initial pressure of one technical atmosphere. The equilibrium temperature was changed in a range of 373 to 3273 K with a step of 100 K. No phase state change time, gas exchange with the environment and the reaction rate are considered as part of the equilibrium state thermodynamic modeling procedure.

### Results and discussion

The distribution of calcium, strontium and cesium by equilibrium phases was studied as part of the computer modeling procedure.

Apart from the gas phase, as shown by the modeling results, an equilibrium system has two condensed phases: a carbon phase (graphite) and a solid oxide-salt solution of interaction products. The carbon phase exists only in a temperature range of 373 to 973 K, and the solid solution exists in a range of 373 to 2873 K. The compounds contained in the solid oxide-salt solution are indexed “cond.”

The interaction with the CO<sub>2</sub> atmosphere with formation of respective quantities of gaseous CO gas should be considered to be the cause for the carbon phase disappearance at 973 K.

The calcium phase distribution is shown in Fig. 1. In the interval between 373 and 873 K, nearly all of the calcium in the system is contained in the solid oxide-salt solution in the form of CaCO<sub>3</sub>(cond.) (~60 mol%) and CaCl<sub>2</sub>(cond.) (~40 mol%). A further temperature increase to 1473 K leads to the appearance and a growth in the content of CaO(cond.) in the solid solution, as well as of the CaCl<sub>2</sub> vapor in the gas phase. These changes in the equilibrium composition of phases match reactions 5 and 18 (Table 2). In the temperature interval between 1473 to 2073 K, the content of CaO(cond.) in the solid oxide-salt solution increases and the content of the CaCl<sub>2</sub> vapor in the gas phase decreases which may be described by reaction 23 (see Table 2). An increase in the equilibrium temperature to 2373 K leads to up to 70% of the total calcium quantity in the system passing to a gas phase in the form of the Ca vapor as the result of reaction 4. At this temperature, some 20% of the total calcium quantity remains in the solid solution in the form of CaO(cond.). The rest of the calcium (~10%) is present in the gas phase as CaO (in accordance with reaction 6). All of the calcium in the system is in the gas phase at temperatures above 2673 K. As the temperature increases to 3273 K, the calcium distribution in the gas phase is as follows: vaporous Ca (~42 mol%), vaporous ionized Ca<sup>+</sup> (~31 mol%), vaporous CaO (~18 mol%) and ionized CaO<sup>+</sup> (~9 mol%).

The distribution of strontium by phases is shown in Fig. 2. In the temperature interval between 373 and 973 K, all of the strontium in the system is contained in the solid oxide-salt solution, predominantly as SrCl<sub>2</sub>(cond.) and SrCO<sub>3</sub>(cond.). As the equilibrium temperature increases to 1073 K, the composition changes noticeably, which may be described by reaction 20 (see Table 2). A further temperature increase from 973 to 1573 K leads to the formation of vaporous SrCl<sub>2</sub> and a rapid growth in its content to 97% of the total strontium quantity in the system as the result of reaction 7. Decomposition of SrCO<sub>3</sub>(cond.) takes place in the same temperature interval in accordance with reaction 19. Decomposition of SrCl<sub>2</sub> vapors is observed and the rest of SrO(cond.) evaporates in accordance with reactions 9 and 11 in the temperature interval of 2073 to

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