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The behaviour of Eu, Pu, Am radionuclide at burning radioactive graphite in an oxygen atmosphere. Computer experiments

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Abstract. Be means of the method of computer thermodynamic simulation we studied the behaviour of the europium, plutonium and americium from the combustion of radioactive graphite in oxygen. Europe is in the form of condensed EuOCl, Eu_2O_3 and vapour EuO. Pluto is in the form of condensed vapour PuO_2 and PuO_2. Americium is a condensed AmO_2, Am_2O_3 and vapour Am. The basic reactions occurring compounds with europium, plutonium and americium. Equilibrium constants of the reactions have been determined.

Introduction

In connection with the completion of the operation and as a result of accidents and man-made emergencies, there are a large amount of radioactive graphite waste. Experts estimate that the total number of irradiated graphite in the world is about 250 thousand tonnes. Exactly reactor graphite is the most of the accumulated solid waste [1, 2].

In the Russian Federation there are about seventeen units. The weight of the graphite stack one reactor, depending on its type, is 1.5–2.5 thousand tonnes. The total mass of irradiated graphite in Russia is approximately equal to 60 thousand tonnes [3].

Technologies for waste disposal of nuclear energy, in the majority, based on the principle of isolation of radioactive graphite from the environment. Considered the most appropriate construction of underground storage in solid granite or basalt rocks. Such a method, by far, the safest, cheapest, and is used by many countries in the fifties of the twentieth century. However, this method also has a number of disadvantages, the main of which is the fact that the area for disposal not unlimited, and this disposal method does not require a reduction of waste [4].

Become a promising high-temperature treatment of radioactive waste. Today, there are several ways to burn radioactive graphite, oxidation air, oxygen, oxidation in molten alkali chlorides, carbonates [2, 5, 6]. The criterion for the successful development of the combustion process is acceptable emissions of radioactive elements to preserve the environment [7].

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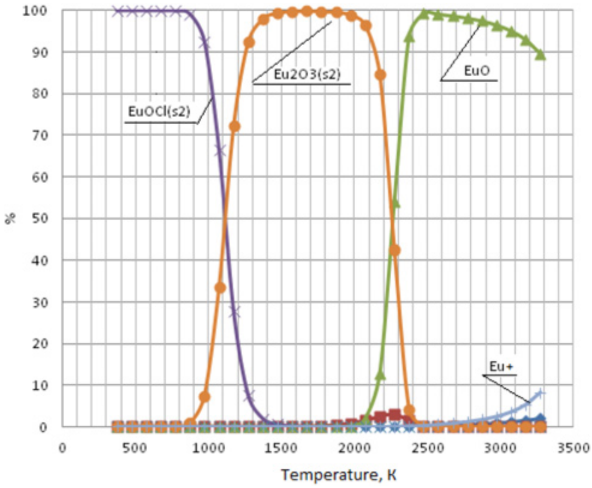

Figure 1. The distribution of Eu in the incineration of radioactive graphite.

Radioactive elements cannot be incinerated. Either they are part of a non-combustible waste, or evaporate, depending on their volatility. The gaseous components condense on the larger particles in the flue gases are removed scrub system [7].

Objective

The main objective of this study is the behaviour of some radioactive elements in combustion of the reactor graphite under oxygen.

Computer experiment

The behaviour of radioactive elements is not different from that of their non-radioactive isotopes. Studies conducted by thermodynamic modelling.

Thermodynamic modelling is a thermodynamic analysis of the equilibrium state of the system as a whole (complete thermodynamic analysis) [8, 9]. Calculation methods are developed based on variation principles of thermodynamics.

One of the most advanced and efficient programs that implement such thermodynamic calculations is a software package TERRA, which is a step further development of the software package ASTRA [10, 11].

Calculations of the equilibrium phases and characteristics are carried out using the reference database on the properties of individual substances [12, 13].

Results and discussion

Fused EuOCl is predominant compound in the temperature range from 375 to 800 K, then the quantity begins to decrease and reaches a value of 0 mol. % At 1500 K. The increase in the number of condensed Eu_2O_3 observed in the temperature range 900–1300 K, and the reduction 1800–2500 K. The growth of gaseous EuO happening to 2000 K at 2500 K. On further heating to 3300 K, the reduction in the amount of vapour EuO (with 99 mol.% to 89 mol.%) with a simultaneous increase in the amount of ions Eu + (8 mol.%) and vaporous Eu (2 mol.%).

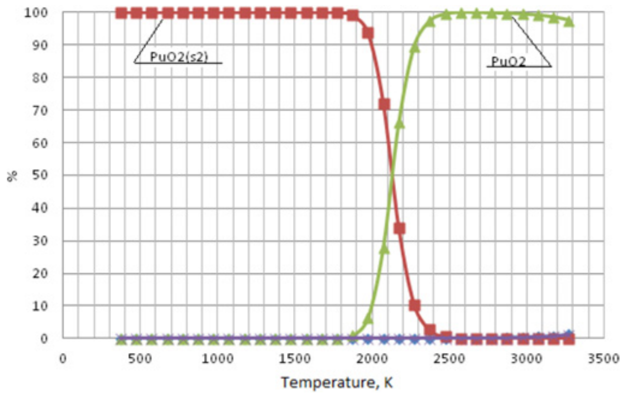

Figure 2. The distribution of Pu in the incineration of radioactive graphite.

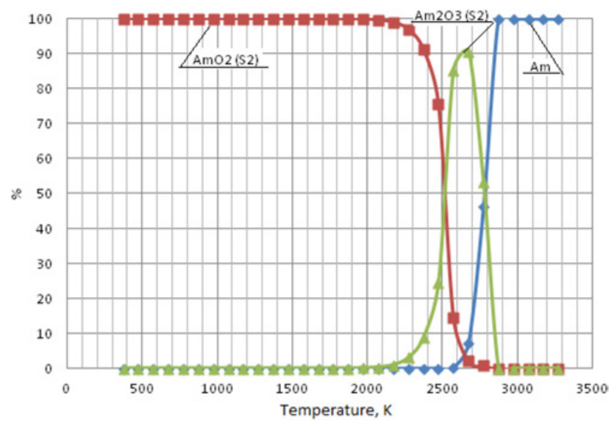

Figure 3. The distribution of Am in the incineration of radioactive graphite.

Plutonium is distributed in the following way: to a temperature of 1800 K prevails in the fused PuO_2 (98–99 mol.%). With increasing temperature, the content of the condensed PuO_2 begins to decrease, at the same time increasing the content of gaseous PuO_2. At a temperature 2400 K PuO_2 vapor content is $\approx$ 99 mol. %, The content of the condensed PuO_2 reduced almost to 0 mole. %.

At temperatures from 375 to 2300 K in the system prevails fused AmO_2 (99 mol.%). Increasing the temperature to 2700 K causes a rise in the content of the condensed Am_2O_3 to 90 mol. %, As well as vapor Am to 9 mol. % And a decrease in the amount of condensed AmO_2 to 0 mole. %. At 3300 K decreases the amount of condensed Am2O3 to 0 mole. % and amount of vapor Am increases to 100 mol. %.

As a result of thermodynamic modeling revealed that the combustion of radioactive graphite in an atmosphere of O_2, americium, plutonium and europium are present in the form of oxygen compounds. Increasing the temperature above 2000 K lead to evaporation of these compounds.

In accordance with the above set of phase transitions and guided by numerical simulation results, the minimum set of basic reaction (equilibrium) within the individual phases and at the interface is written below.

$$2\,Eu_2O_3(S) = 4\,EuO + O_2 \tag{1}$$

$$4\,EuOCl(S) + O_2 = 2\,Eu_2O_3(S) + 2\,Cl_2 \tag{2}$$

Table 1. The coefficients in equation constants reaction.

Reaction's number	**Temperature range ΔT_i, K**	**A**	**B**	**The magnitude approximation R^2**
1	1873–2373	-348975	87,165	1
2	873–1473	51740	-47,463	0,9842
3	1973–2473	-352836	89,027	0,9997
4	1873–2473	-68474	18,95	0,9959
5	2173–2773	-100978	50,264	0,991
6	2673–2873	-388183	52,927	0,9601

$$2\,Eu_2O_3(S) = 4\,EuO + O_2 \tag{3}$$

$$PuO_2(S) = PuO_2 \tag{4}$$

$$4\,AmO_2(S) = 2\,Am_2O_3(S) + O_2 \tag{5}$$

$$2\,Am_2O_3 = 4\,Am + 3\,O_2. \tag{6}$$

The dependence of the equilibrium constants of reactions (1)–(6) of the temperature is described by the equation of the form:

$$\ln k_i = A + \frac{B}{T}\cdot \tag{7}$$

The numerical values of the coefficients A and B of Eq. (7) shown in Table 1.

The values of the equilibrium constants indicate that the reaction shifted towards the formation of products.

1. Findings

The combustion of radioactive graphite oxygen europium is in the form of condensed EuOCl (to a temperature of 1200 K), the condensed Eu_2O_3 (in the temperature range 1200–2200 C) and vapor EuO (at temperatures above 2200 K).

The combustion of radioactive graphite oxygen plutonium is in the form of condensed PuO_2 (to a temperature of 2100 K) and vapor PuO_2 (above 2100 K).

The combustion of radioactive graphite oxygen americium is in the form of condensed AmO_2 (to a temperature of 2500 K), the condensed Am_2O_3 (in the temperature range 2500–2900 C) and vapor Am (at temperatures above 2900 K).

The findings should be considered when disposing of radioactive graphite by incineration.

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