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Thermophysical characteristics of radioactive graphite – water vapor system

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Abstract. The article considers thermophysical characteristics of radioactive graphite – water vapor system in temperature range 373-3273K. The research was made by thermodynamic modeling method using TERRA software. We determined 4 temperature intervals in which changes of thermophysical characteristics of radioactive graphite – water vapor system occur.

Most of atomic power plants operate according to the scheme where low-enriched uranium is served as a fuel for thermal reactors, graphite is used as a moderator material, and water is used as a reactor coolant. [1].

In Russia and other countries in the world there is a problem of handling with radioactive graphite of uranium-graphite reactive cores after their taking out of service [2].

Radionuclides (^{137}Cs, ^{90}Sr, ^{154}Eu and others) are formed in graphite due to the leak of reactor coolant and transfer of fuel segments.

There are several methods of treatment of radioactive graphite: "Conservation", "Disposal", "Elimination" [3]. First two methods require the construction of deep storages and large financial costs. Method "Elimination" is more effective as the amount of irradiated graphite falls by 90 times [4].

Elimination of irradiated graphite can be performed by the following methods: oxidation by salts, oxidation in molten alkali and carbonates, liquid-phase oxidation in acids, bonding of graphite with metals, oxidation by air, oxygen, carbon dioxide, water vapor [3-6].

The purpose of this research is to study thermophysical characteristics of radioactive graphite – water vapor system in temperature range 373-3273 K.

The tasks of the study are carrying out the experiment with investigated system by thermodynamic modeling method, analysis of thermophysical characteristics (V – specific volume (m^3/kg), S – entropy (kJ/(kg K)), I – total enthalpy (kJ/kg), U – total internal energy (kJ/kg), M – number of moles (mole/kg), Z – mass fraction of condensed phases).

Full-scale experiments do not always give reliable information at high temperatures due to their complexity and measurements errors. So, calculations were made by

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thermodynamic modeling using TERRA software which is used for studying systems with complex chemical composition in high temperature conditions.

The software is used to calculate phase composition, thermodynamic and transport properties of arbitrary systems.

The calculation of phase composition and characteristics of equilibrium was made according to reference database for properties of individual substances. The basis of information in the database are thermodynamic, thermophysical and thermochemical properties of individual substances systemized in the Institute of High Temperatures of the Russian Academy of Sciences (database INVATERMO), by national US Bureau of Standards, calculated in Moscow State Technical University using molecular calorimetric and spectrochemical data, published in reference books [7, 8].

Thermodynamic modeling with the use of TERRA software is successfully used in thermal physics [5, 6].

Thermodynamical modeling was carried out at initial pressure of one atmosphere, at initial temperature 373 K and at final temperature 3273 K. Temperature measurement interval – 100 K. Phase composition: gaseous phase – 75% (H_2O – 100% mass), condensed phase – 25% (C ~ 99.98% mass, U ~1.15 10^{-2}% mass, Cl ~1.88 10^{-3}% mass, Ca ~2.69 10^{-4}% mass, Pu ~7.19 10^{-5}% mass, Be ~1.19 10^{-5}% mass, Ni ~7.99 10^{-6}% mass, Cs ~3.99 10^{-6}% mass, Sr ~9.99 10^{-6}% mass, Am ~9.99 10^{-6}% mass, Eu ~7.99 10^{-6}% mass.

Figure 1a shows dependence of specific volume on temperature. In temperature range from 373 to 573 K specific volume of the system increases from 1.29 to 2.01 m^3/kg. At temperature 573 K the bend point is observed and in temperature range from 573 to 973 K specific volume increases from 2.01 to 4.98 m^3/kg. At temperature 973 K the bend point is observed and in temperature range 973-2373 K specific volume increases from 4.98 to 12.38 m^3/kg. At temperature 2373 K the bend is observed and in temperature range from 2373 to 3273 K specific volume increases from 12.38 to 19.7 m^3/kg.

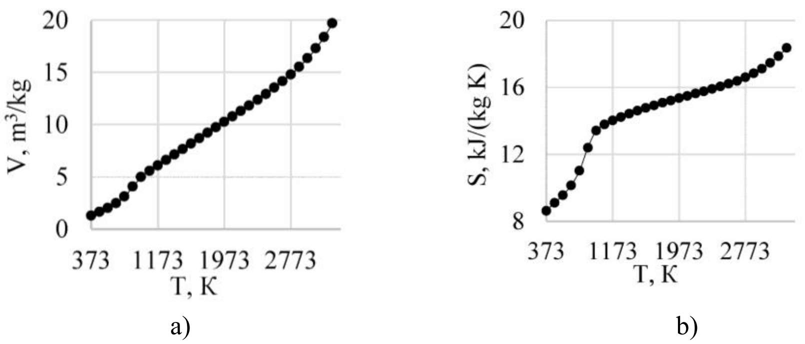

Fig. 1. Dependences of thermophysical characteristics of radioactive graphite – water vapor system on temperature: a) dependence of specific volume on temperature, b) dependence of entropy on temperature.

Figure 1b shows dependence of entropy on temperature. In temperature range from 373 to 573 K entropy increases from 8.63 to 9.56 kJ/(kg K). At temperature 573 K the bend point is observed and in temperature range from 573 to 973 K entropy increases from 9.56 to 13.41 kJ/(kg K). At temperature 973 K the bend point is observed and in temperature range 973-2373 K entropy increases from 13.41 to 15.89 kJ/(kg K). At temperature 2373 K the bend is observed and in temperature range from 2373 to 3273 K entropy increases from 15.89 to 18.35 kJ/(kg K).

Figure 2a shows dependence of total enthalpy on temperature. In temperature range from 373 to 573 K total enthalpy increases from -9873.53 to -9432.54 kJ/kg. At temperature 573 K the bend point is observed and in temperature range from 573 to 973 K

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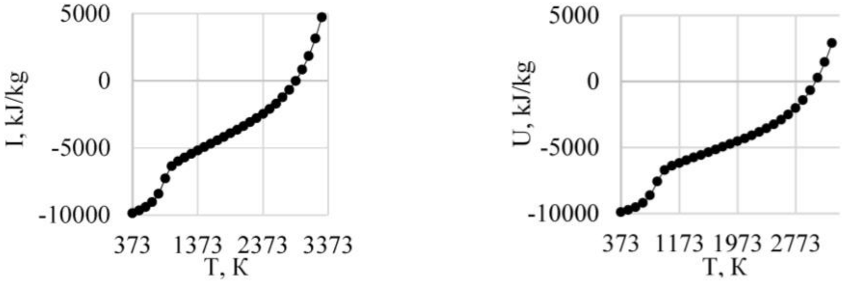
total enthalpy increases from -9432.54 to -6368.44 kJ/kg. At temperature 973 K the bend point is observed and in temperature range 973-2373 K total enthalpy increases from -6368.44 to -2482.69 kJ/kg. At temperature 2373 K the bend is observed and total enthalpy increases from -2482.69 to 4704.91 kJ/kg.

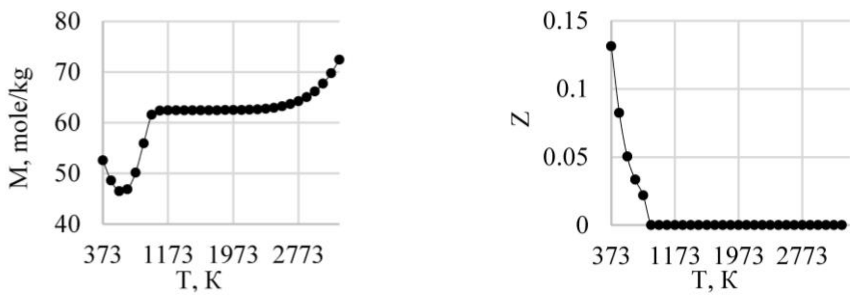


a) b)

Fig. 2. Dependences of thermophysical characteristics of radioactive graphite – water vapor system on temperature: a) dependence of total enthalpy on temperature, b) dependence of total internal energy on temperature.

Figure 2b shows dependence of total internal energy on temperature. In temperature range from 373 to 573 K total internal energy increases from -9899.44 to -9529.15 kJ/kg. At temperature 573 K the bend point is observed and in temperature range from 573 to 973 K total internal energy increases from -9529.15 to -6713.87 kJ/kg. At temperature 973 K the bend point is observed and in temperature range 973-2373 K total internal energy increases from -6713.87 to -3565.17 kJ/kg. At temperature 2373 K the bend is observed and total internal energy increases from -3565.17 to 2914.22 kJ/kg.

Figure 3a shows dependence of number of component moles on temperature. In temperature range from 373 to 573 K number of component moles in the system decreases from 52.5 to 46.4 mole/kg. At temperature 573 K the bend point is observed and in temperature range from 573 to 973 K number of component moles increases from 46.4 to 61.5 mole/kg. At temperature 973 K the bend point is observed and in temperature range 973-2373 K number of component moles doesn't change and is equal to ~ 62 mole/kg. At temperature 2373 K the bend is observed and number of component moles increases from 62.7 to 72.3 mole/kg.



a) b)

Fig. 3. Dependences of thermophysical characteristics of radioactive graphite – water vapor system on temperature: a) dependence of number of component moles on temperature, b) dependence of mass fraction of condensed phases on temperature.

Figure 3b shows dependence of mass fraction of condensed phases on temperature. In temperature range from 373 to 573 K mass fraction of condensed phases decreases from

0.13 to 0.05. At temperature 573 K the bend point is observed and in temperature range from 573 to 873 K mass fraction of condensed phases decreases from 0.05 to 0.

Condensed C gives the main contribution to thermodynamic properties of the system till its burning temperature (873 K). At temperature > 873 K thermodynamic properties are defined by gaseous phase.

According to the Figure four temperature intervals where changes of thermophysical properties of the system occur are marked.

1 temperature range from 373 to 573 K: changes are connected with the increasing of CH_4 gas content.

2 temperature range from 573 to 973 K: changes are connected with both decreasing of CH_4 gas content and burning of condensed C.

3 temperature range from 973 to 2373 K: changes have a linear behavior and are defined by gas-vapor phase.

4 temperature range > 2373 K: changes are connected with thermal decomposition of gas-vapor phase.

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