HEAT TRANSFER DURING EVAPORATION OF CESIUM FROM GRAPHITE SURFACE IN AN ARGON ENVIRONMENT

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Abstract. The article focuses on discussion of problem of graphite radioactive waste formation and accumulation. It is shown that irradiated nuclear graphite being inalienable part of uranium-graphite reactor may contain fission and activation products. Much attention is given to the process of formation of radioactive cesium on the graphite element surface. It is described a process of plasma decontamination of irradiated graphite in inert argon atmosphere. Quasi-one mathematical model is offered, it describes heat transfer process in graphite-cesium-argon system. Article shows results of calculation of temperature field inside the unit cell. Authors determined the factors which influence on temperature change.

1 Introduction

By now all of the industrial uranium-graphite reactors (IUGR) are stopped in Russia. Upon that major part of them is either on the stage of shut down preparation or on the stage of shut down. Exception is IUGR EI-2, which was replaced on special radioactive waste parking site. However it is necessary to conduct operations connected with graphite stack disassembling and further radioactive waste burial for reactors of types of RBMK, EGP, AMB, AM (in Russia) reactors of types of Magnox, UNGG, AGR, HTR (not in Russia).

One of the most common radionuclides present in irradiated structural elements is ^{137}Cs. Thus, the specific activity of the nuclide in some samples of radiation sources can reach 3.2 TBq/kg [1]. In this case the radioactive cesium contamination observed in the materials that were in contact with nuclear fuel, since it is the product nuclear fission.

At the present time industry organizations offer different ways of processing radioactive waste (RW): electrooxidation (anodizing) in an oxygen-containing electrolyte [2], combustion in thermal oxidizer environment and pyrolysis [3], plasma treatment [4], thermochemical treatment [5], melting [6], vitrification [7]. Most of the known methods lead to increased volumes of waste, produced as by-product gases of reaction, which requires additional trapping means (bubblers, scrubbers or filters).

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In the case of using an inert gas as a transfer medium by thermal deactivation of radioactive waste the amount of secondary waste is reduced. With increasing gas temperature the rate of nuclides evaporation from the surface of materials is increased, which leads to increased productivity and the recovery rate of radionuclides. At the same time there is a transition from surface to volume processes. This is especially true during deactivation of graphite structural elements, the porosity of which is about 23 % [8]. Therefore, for the thermal deactivation of RW can be used argon low-temperature plasma.

In this case, simulations of heat transfer process in a high temperature argon flow for selecting of optimal regimes of graphite cleaning process by ^{137}Cs.

2 The methodology and calculations

Irradiated graphite constructional elements usually have surface contamination by radioactive cesium [9]. Heating of graphite-cesium surface by gas flow leads to a metal sublimation. Computational cell scheme is in the figure 1. There δ_{Cs} is layer thickness of metal cesium, δ_C is layer thickness of graphite surface. High temperature plasma flow consisting of noble gas (argon) is oriented along the graphite-cesium surface. The gas velocity at the entrance to computational cell with height of R above cesium surface is u_0. It must be noted that transversal and longitudinal dimensions of cell are much bigger than thickness of cesium layer δ_{Cs}. Graphite surface heats by high temperature flow contact and by bottom wall-graphite surface heat transfer. Authors used thermodynamic parameters of cesium as a mixture of its natural occurring isotopes.

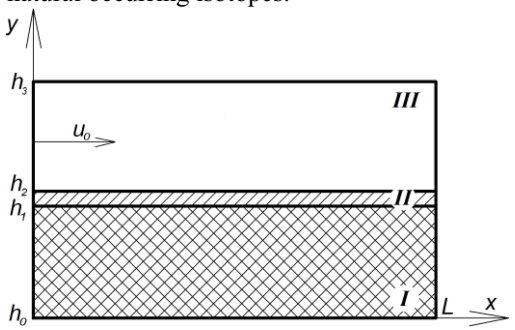

Fig. 1. Computational cell scheme: I – graphite, II – cesium, III – high temperature flow.

To simulate the heat transfer process we solved unsteady heat transfer equation in the Cartesian coordinate system. The direction of the x axis coincides with the direction of velocity vector u of high temperature gas flow.

$$\begin{cases} c_1\rho_1 \dfrac{\partial T}{\partial t} = \dfrac{\partial}{\partial y}\left(\lambda_1 \dfrac{\partial T}{\partial y}\right), \ h_2 < y < h_1 \\ c_2\rho_2 \dfrac{\partial T}{\partial t} = \dfrac{\partial}{\partial y}\left(\lambda_2 \dfrac{\partial T}{\partial y}\right), \ h_1 < y < h_0 \end{cases}, \tag{1}$$

where c_1, ρ_1, λ_1 – heat capacity, density and thermal conductivity of the metal cesium; c_2, ρ_2, λ_2 – heat capacity, density and thermal conductivity of graphite; h_0 – coordinate corresponding to the bottom surface of deactivating graphite; h_1 – the distance from the bottom to the top surface of graphite; h_2 – distance from the top surface to the surface of graphite in contact high temperature argon flow with cesium.

It was thought that the thermal and gas dynamic properties of argon dependent by temperature, but properties of graphite and cesium remain constant. On this basis the parameters of the plasma forming gas were chosen as follows:

$$u_{Ar} = u_0(\frac{T_{Ar}}{298}), \tag{2}$$

$$\nu_{Ar} = 1.37 \cdot 10^{-5}(\frac{T_{Ar}}{298})^{1.75}, \tag{3}$$

$$\lambda_{Ar} = 1.54 \cdot 10^{-3}(\frac{T_{Ar}^{1.5}}{T_{Ar}+160}), \tag{4}$$

$$\alpha_{Ar} = 2.04^{-5}(\frac{T_{Ar}}{298})^{1.75}, \tag{5}$$

where ν_{Ar}, λ_{Ar}, α_{Ar} – kinematic viscosity, thermal conductivity, thermal diffusivity of argon at a temperature T_{Ar}, u_{Ar} – gas flow rate at the temperature $T_{Ar.}$

Heat balance equation on the surface of the metal cesium sublimation for the problem:

$$-\lambda_1 \left.\frac{\partial T}{\partial y}\right|_{y=h_2(t)} = a(T_{Ar} - T_s), \tag{6}$$

where a – heat transfer coefficient under convective heat transfer of a gas flow with a surface layer of metallic cesium, T_s,– cesium sublimation temperature.

From the equation 6 shows that the process of heat transfer at the boundary cesium-argon will be determined by the gas flow regime, since the amount of heat transfer coefficient a is uniquely associated with a Nusselt numbers and Reynolds numbers.

When solving equation 1 it was considered that the change in temperature between the layer of metallic cesium and graphite surface occurs by heat exchange, which characterized by a_1 at a constant coefficient of thermal conductivity of graphite. Changing of temperature of the external wall of graphite element is described by the equation 7:

$$-\lambda_2 \left.\frac{\partial T}{\partial y}\right|_{y=h_0} = a_2(T_{h_1} - T_w), \tag{7}$$

where a_2 – thermal conductivity of graphite, T_{h1} – the temperature on the boundary of cesium-graphite, T_w – temperature of the bottom surface of the graphite element.

We thought that at the initial time the temperature of the selected cells was the same in all areas and was T_0.

The numerical values of the parameters in the equation 2–7 take the following: c_1 = 273 J/(kg·K), c_2 = 710 J/(kg·K), ρ_1 = 1873 kg/m^3, ρ_2 = 1650 kg/m^3, λ_1 = 10 W/m·K, λ_2 = 150 W/m·K, μ = 137 g/mol, T_s, = 961 K, , $T_0 = T_w$ = 25 ^{o}C, u_0 = 10 m/s, R = 30 mm.

The initial data for modeling cesium evaporation process with a graphite surface was chosen to the initial temperature of the plasma gas T_{Ar} equal to 600 ^{o}C, 1000 ^{o}C, 1500 ^{o}C, 2000 ^{o}C. It was considered that the thickness of the graphite layer δ_C = 20 mm, and cesium layer δ_{Cs} was not more than 2 microns. The results of calculation of temperature changes on the surface of the cesium film shown in figure 2. The figure shows that the maximum temperature in the surface layer is lower than the initial temperature of the gas flow due to convective heat transfer between the argon and cesium as well as the heat transfer between the cesium and graphite. For example, cesium film temperature reaches 450 ^{o}C temperature with an initial weight average gas temperature plasma 600 ^{o}C, and at 2000 ^{o}C not exceeding 1950 ^{o}C. This is due to the fact that the lower boundary of the graphite sample maintained

at a constant temperature equal to 25 °C. The time of heating the thin film of cesium to gas flow temperature does not exceed 8 minutes.

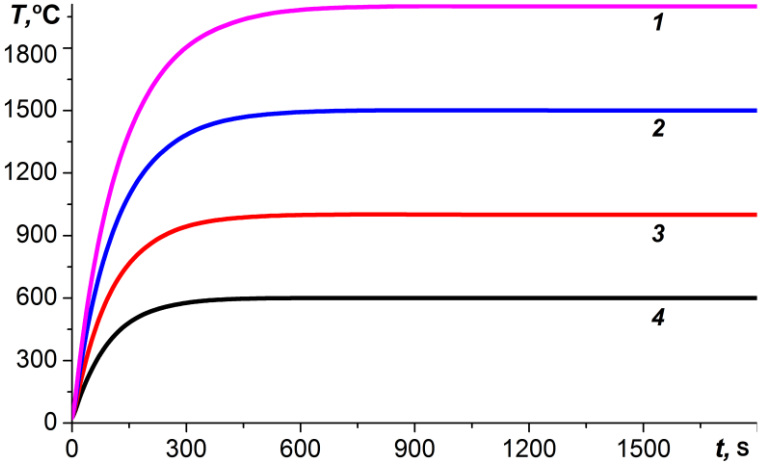

Fig. 2. Changing the cesium film temperature at different initial gas temperature of plasma: 1 – 2000 °C, 2 – 1500 °C, 3 – 1000 °C, 4 – 600 °C.

The velocity of the high temperature flow along the deactivated sample insignificantly effect on the heat transfer process (fig. 3).

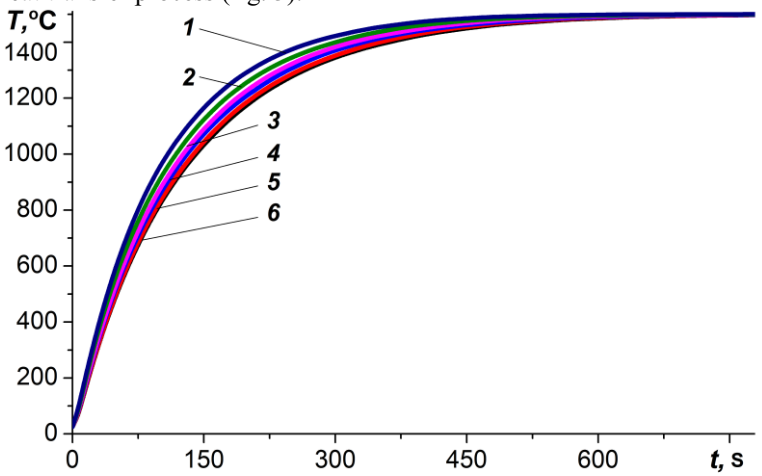

Fig. 3. Changing the cesium film temperature at different initial velocity of high temperature gas flow: 1 – 40 m/s, 2 – 20 m/s, 3 – 10 m/s, 4 – 5 m/s, 5 – 1 m/s, 6 – 0.1 m/s.

The graph shows that the release time of a thermodynamic system to a quasi-stationary mode varies from 7.5–8 minutes in the gas velocity range of 0.1–40 m/s. This is probably due to the fact that the under steady flow regime of high temperature flow does not occur significant change in the energy balance of the system. The change in the thermodynamic parameters of the system can be caused only by a change in thickness of cesium δ_{Cs}, which is negligible compared to an equivalent cell size.

3 Summary

The paper presents a mathematical model describing the heat transfer process in an argon-cesium-graphite system. The model was used to calculate the extraction rate of radioactive ^{137}Cs from the surface of graphite during plasma treatment of radioactive waste. It was shown that the significant factors influencing the degree of deactivation are weight average gas temperature of equilibrium plasma and velocity of gas flow.

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