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FLUORINATION OF A DEPLETED URANIUM-PLUTONIUM-NITRIDE FUEL WITH ELEMENTAL FLUORINE

V. A. Karelin, V. N. Brendakov, and M. V. Popadeikin UDC 621.039.5

A physical and a mathematical model have been developed to describe the physicochemical process of torch fluorination of an uranium-plutonium-nitride fuel. An algorithm for calculating the velocity, temperature, and concentration of the chemical components has been constructed which makes it possible to optimize the parameters and operating modes of a torch reactor based on mathematical simulation methods.

INTRODUCTION

The uranium-plutonium-nitride fuel that is used in inherently safe fast reactors (BREST) is one of the most promising fuels [1] and the reactors in which it is used are highly efficient and technologically advanced. However, utilization of depleted fuels is a serious problem [2].

The fluoride technology for processing uranium-containing fuels is one of the most promising technologies in modern practice [3–5]. The process in which a gas jet containing a dispersed solid phase comes in contact with a chemically active oxidizer gas flow is often used in various technologies. To make the process of torch oxidation more efficient and technologically advanced calls for relevant research. However, full-scale experiments are very expensive and involve special safety measures. To evaluate the factors that affect the stability of process control and to optimize the process mode and geometry, it is necessary to use simulation methods.

Since the processes occurring in a torch reactor are physically intricate and difficult to describe mathematically, the basic challenge is choosing approximations which would be suited to the principal physicochemical processes and offer the possibility to describe them by a completed system of equations.

The study under consideration was aimed at developing a physical and a mathematical model of a torch reactor for fluorination of uranium nitride. For doing this, it was necessary to choose physically reasonable approximations to formulate a mathematical model of the process. In this paper, the relevant differential equations and algebraic expressions are given together with boundary conditions. The equations were solved by numerical methods. Some of the results obtained are discussed.

THE MODEL OF THE PROCESS

Let us consider an apparatus shaped as a cylinder having a round hole in its top flange through which argon inert gas is fed which contains a fine powder of mixed uranium nitrides and plutonium. Elemental fluorine is supplied through an annular hole in the peripheral part of the apparatus top flange. The outlet of the reactor is made as a central hole of larger diameter in the bottom flange. To create conditions which would provide for the onset of the reaction, the top flange of the apparatus is heated to $T_f = 50°C$ and the side wall to $T_w = 300°C$.

As a heterogeneous jet of the fuel comes in contact with the oxidizer, a burning torch appears in whose flame there ocur irreversible heterogeneous exothermal reactions

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$$2UN+9F_2 \rightarrow 2UF_6+2NF_3,$$
$$2PuN+6F_2 \rightarrow 2PuF_6+N_2 \quad (1)$$

proceed whose rates are assumed to be the same.

Since the particle size is small, 50–100 μm, it can be shown that the burning mode will be kinetic and the flame thickness that would provide complete combustion of a particle should be 1–3 mm. Hence, it can be assumed that the reactions (1) proceed within a thin layer at the flame front. The energy balance in the apparatus and the composition of the final combustion products can be affected by the secondary reaction in the gaseous phase. Analysis has shown that reactions of this type have short lifetimes and the compounds resulting from these reactions are unstable. In the oxidizing medium, these compounds interact with the oxidizer to form stable substances that are used in constructing the model. Estimation of the flow conditions in the working zone of the reactor shows a high degree of turbulization of the flow. This is favored by the entrance conditions for the fuel and oxidizer and by the burning process itself. In view of the great contribution of turbulence to the heat and mass transfer, the authors did not restrict themselves to the use of an algebraic formula for the calculation of the turbulent viscosity factor, but employed a new three-parameter model of turbulence that was specially constructed for flows of this type [6]. The system of equations that describes the heat and mass transfer can be substantially simplified by using the Shvab–Zeldovich approximation according to which the effective diffusion coefficients are assumed to be the same for all components of the mixture and coincide with the effective thermal diffusivity. For this case, the Prandtl and Lewis numbers are equal to unity.

Thus, a mathematical model to be constructed should describe the motion of a gas mixture and a fine powder of uranium nitrides and plutonium that interact in the working zone, taking into account heterogeneous exothermal reactions and heat and mass transfer processes. In view of the high degree of dispersion of the solid fuel, we restrict our description of the burning torch to the flame front. To obtain a completed system of differential equations and boundary conditions which would allow one to calculate the velocity, temperature, and concentration fields in a torch reactor, we use additional constraints on the Prandtl and Lewis numbers as a first approximation of the model.

To describe the field of averaged velocities for a stationary turbulent axisymmetric gas flow in the working chamber of a torch reactor, we use the Navier–Stokes equation. The velocity field is written in terms of the stream function and vorticity in cylindrical coordinates [7, 8]:

$$\frac{\partial}{\partial r}(\rho u_r \omega)+\frac{\partial}{\partial z}(\rho u_z \omega)=\frac{1}{\mathrm{Re}}\left\{\frac{\partial^2}{\partial z^2}(\mu\omega)+\frac{1}{r^2}\frac{\partial}{\partial r}\left[r^3\frac{\partial}{\partial r}\left(\frac{\mu\omega}{r}\right)\right]\right\}$$
$$+\frac{1}{2}\left[\frac{\partial}{\partial z}\left(u_r^2+u_z^2\right)\ \frac{\partial\rho}{\partial r}-\frac{\partial}{\partial r}\left(u_r^2+u_z^2\right)\ \frac{\partial\rho}{\partial z}\right],$$
$$\frac{\partial}{\partial z}\left(\frac{1}{\rho r}\frac{\partial\psi}{\partial z}\right)+\frac{\partial}{\partial r}\left(\frac{1}{\rho r}\frac{\partial\psi}{\partial r}\right)=-\omega, \quad (2)$$
$$u_r=-\frac{1}{\rho r}\frac{\partial\psi}{\partial z}\ ,\ u_z=\frac{1}{\rho r}\frac{\partial\psi}{\partial r}\ ,\ \omega=\frac{\partial u_r}{\partial z}-\frac{\partial u_z}{\partial r}.$$

This equation relates the velocity vector components, the mixture density, the stream function, and the vorticity. To calculate the turbulent viscosity factor μ, we use the turbulence model described in [6].

Taking into account that the fine powder of uranium nitride and plutonium in the fuel mixture is in the 1000:1 proportion, we may use, without sacrifice of precision, only the first reaction of (1). We restrict our consideration to a reacting mixture consisting of the five chemical components

subscript	i	1	2	3	4	5
component	B_i	UN	Ar	F_2	UF_6	NF_3

(3)

that are involved in the first reaction of (1). In symbolic form, this reaction can be represented as

$$m_{11}B_1 + m_{13}B_3 \rightarrow n_{14}B_4 + n_{15}B_5\,, \tag{4}$$

where m and n are stoichiometric coefficients; the first and the second subscript denote the reaction number and the chemical component, respectively.

The equation of transfer of deceleration enthalpy for a mixture can be written in the form

$$\frac{\partial}{\partial z}\left(h\frac{\partial \psi}{\partial r}\right) - \frac{\partial}{\partial r}\left(h\frac{\partial \psi}{\partial z}\right) = \left[\frac{\partial}{\partial z}\left(r\mu\frac{\partial h}{\partial z}\right) + \frac{\partial}{\partial r}\left(r\mu\frac{\partial h}{\partial r}\right)\right]. \tag{5}$$

This quantity is related with the temperature T and mass fractions Y_i of the chemical components (3) at every point of the calculation region by the caloric equation of state for the mixture:

$$\begin{aligned} h &= \sum_{1=1}^{5} h_i(T)Y_i\,, \\ h_i(T) &= h_i^0 + \int_0^T c_{pi}(T)dT\,, \end{aligned} \tag{6}$$

where h_i is the specific enthalpy for the ith component and h_i^0 is its standard formation heat.

To simplify the problem of mass transfer, we consider the mass fractions Y_a of the chemical elements that constitute the mixture. Taking into account that the Lewis number for mass fractions of chemical elements is commonly taken equal to unity, we obtain differential equations similar to (5):

$$\begin{aligned} &\frac{\partial}{\partial z}\left(Y_a\frac{\partial \psi}{\partial r}\right) - \frac{\partial}{\partial r}\left(Y_a\frac{\partial \psi}{\partial z}\right) = \frac{\partial}{\partial z}\left(r\mu\frac{\partial Y_a}{\partial z}\right) + \frac{\partial}{\partial r}\left(r\mu\frac{\partial Y_a}{\partial r}\right), \\ &a\text{-subscript}\quad 1 \quad 2 \quad 3 \quad 4 \\ &\text{element} \qquad \text{U} \quad \text{Ar} \quad \text{N} \quad \text{F}. \end{aligned} \tag{7}$$

The mass fractions of the elements and components are related as

$$Y_a = \sum_{i=1}^{5} \mu_{ai} Y_i \;\;,\;\; a = 1, ..., 4\,, \tag{8}$$

where μ_{ai} is the mass fraction of the ath element in the ith component of the mixture. However, these four equations are lacking for finding the five unknown Y_i.

Let us write down expressions for the mole fractions of the mixture components, x_i, in terms of the mass fractions Y_i of the components, their molecular weights W_i, and the mean molecular weight of the mixture, W:

$$x_i = \frac{W}{W_i}Y_i\,, \quad W = \left(\sum_{i=1}^{8}\frac{Y_i}{W_i}\right)^{-1}.$$

Assuming that the irreversible reaction of fluorination (1) proceeds within a thin layer, we approximate the surface of the flame front by an equation of the form

$$\varphi(r, z) = 0\,,$$

where the function φ is determined from the condition that at the flame front the fuel and the oxidizer are mixed in a stoichiometric proportion. Proceeding from the representation of the chemical reaction in the form of (4), we may write the relation

$$\varphi = \frac{m_{13}}{m_{11}} x_1 - x_3 = 4.5 x_{\mathrm{UN}} - x_{\mathrm{F}_2} ,$$

such that the fuel and the oxidizer content cannot be equal to zero simultaneously at the same point of the calculation region. There is no fuel behind the front and no oxidizer at the front. Hence, we may write

$$\begin{aligned} 4.5 x_{\mathrm{UN}} &= 0 \quad \text{for} \quad \varphi \le 0, \\ x_{\mathrm{F}_2} &= 0 \quad \text{for} \quad \varphi \ge 0. \end{aligned} \tag{9}$$

Thus, relations (8) and (9) constitute a system of five equations for finding Y_i, $i = 1, \ldots, 5$, at every point of the calculation region for known values of φ, Y_a, and T.

The system of equations involved in the mathematical model is complete with the equation of state for the mixture

$$p = \frac{\rho R T}{W} \sum_{i=2}^{5} x_i ,$$

in which the summation refers to only gaseous components since the volumetric content of the solid phase is as small as of the order of 10^{-4}. In view of the low velocities, we assume that the pressure is the same throughout the apparatus volume.

CALCULATION RESULTS AND DISCUSSION

The system of equations involved in the mathematical model developed was solved numerically by the finite difference method. The calculation region is shown schematically in Fig. 1; it possesses axial symmetry. The principal geometric parameters used in the calculations are indicated. The apparatus diameter is taken as a length scale. The calculations were performed for a half of the apparatus. The finite-difference analogs of the differential equations (2), (5), and (7) were written for a uniform network. For this purpose, a matrix with 21×801 nodes was used. The system of algebraic equations obtained by discretization was solved by the sweep method with fixation in time. At every step of the iteration process, the system of equations for the determination of the temperature and concentration fields was solved for the mixture components. For doing this, Eq. (8), in view of the conservation of atoms of each species, was rearranged to the form

$$\sum_{i=2}^{5} k_{ai} x_i = (k_{a1} x_1 + k_{a2} x_2)\varphi + k_{a3} x_3 (1 - \varphi) ,$$

where

$$k_{ai} = \frac{W_i}{W_a} \mu_{ai} .$$

This is the number of atoms with an atomic weight W_a in a molecule of the ith species.

The boundary conditions for temperature were as follows: 20°C at the inlet holes, 50°C at the top flange, and up to 300°C on the side wall; at the outlet of the apparatus, the derivative was put equal to zero. The geometric dimensions of the torch reactor were chosen to correspond to those of an actual apparatus: 6 m for the height, 0.3 m for the apparatus diameter,

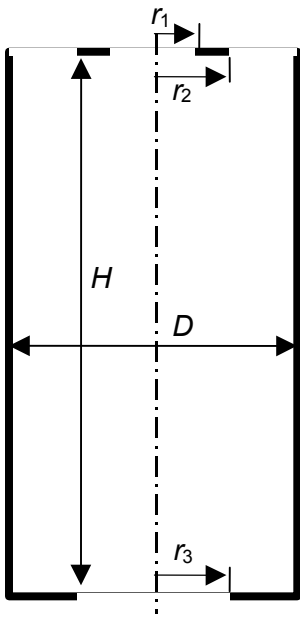

Fig. 1

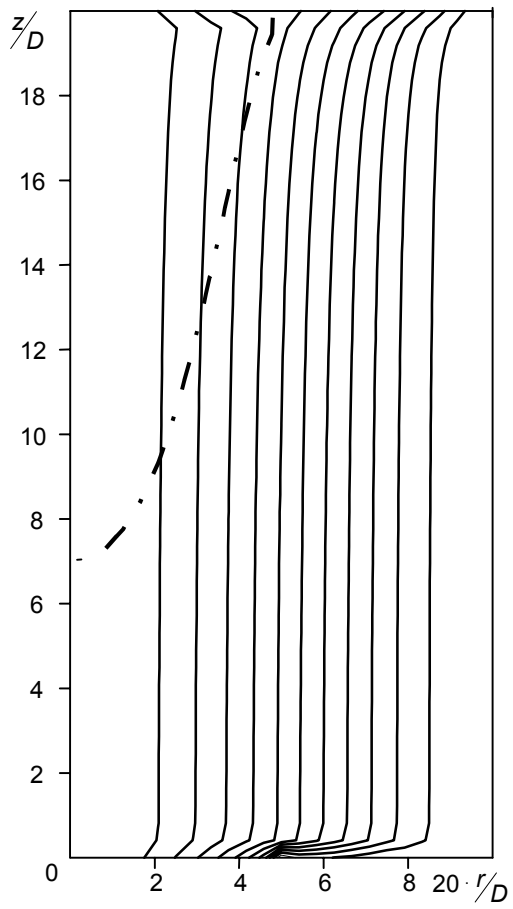

Fig. 2

Fig. 1. Geometry of the calculation region: r_1 – inlet radius for the UN fuel, r_2 – inlet radius for the F_2 oxidizer, r_3 – outlet radius for combustion products, D – apparatus diameter, and H – apparatus height.

Fig. 2. Stream lines for gases in a torch reactor: dot-and-dash line – combustion torch front, z/D – dimensionless axial coordinate, and $20 \cdot r/D$ – radial coordinate scale.

0.12 m for the inlet diameter for the fuel, and 0.15 m for the outlet diameter. In all pictures, the radial coordinate scale is increased 20 times.

Figure 2 presents the stream lines for the gaseous components in the calculation region. The dot-and-dash line indicates the flame front for the given mode of operation of the reactor. The isolines correspond to different values of the stream function with the difference between the values corresponding to neighboring lines being the same. As can be seen from this figure, the nonuniform temperature distribution between the top flange and the side walls has no effect on the flow pattern. The stagnation zone at the side surface is rather small, increasing toward the bottom flange of the apparatus. The outlet condition for the flow at the central hole results in a considerable gradient of the radial component of the velocity vector near the bottom flange.

The isotherms of the temperature field are shown in Fig. 3. The temperature is given in Kelvin degrees; it has a maximum of 1450 K at the boundary of the flame front. The lines are drawn with an interval of 100 K. From this figure it can be seen that the fuel and oxidizer, when coming in the reactor, create a region of high temperature gradients. Downstream of the combustion torch, the temperature field is comparatively uniform. In the top corner of the apparatus

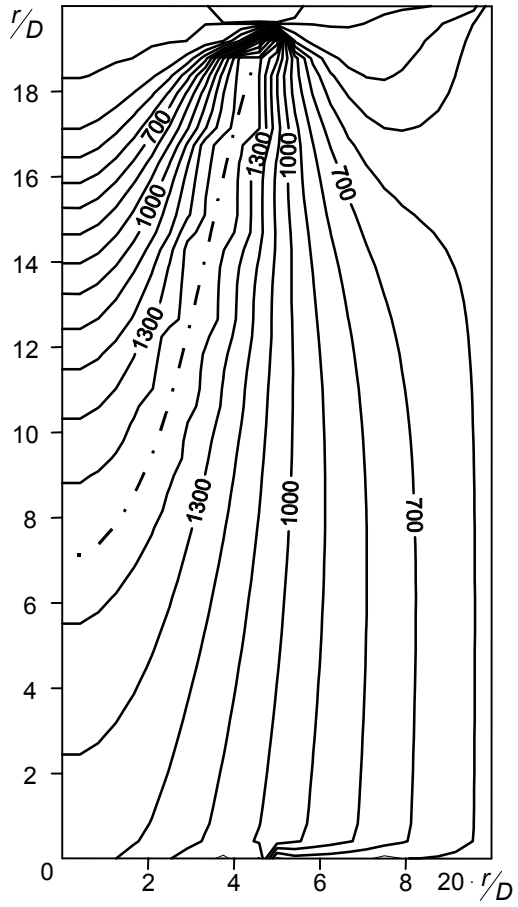

Fig. 3. Temperature distribution in the apparatus.

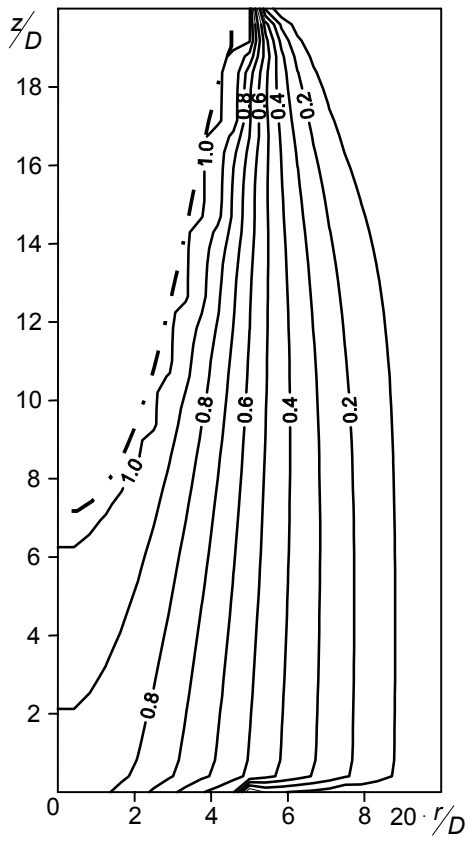

Fig. 4. UF_6 concentration distribution.

there is a low temperature region associated with the temperature of the top flange. Under certain conditions, this region may have a detrimental effect on the fluorination process.

Figure 4 presents the UF_6 concentration spatial distribution over the apparatus. The volume content of UF_6 in the mixture is given in the isolines. The decrease in concentration toward the periphery is related to that the oxidizer, F_2, is fed in excess.

CONCLUSION

The physical and the mathematical model of a plasma reactor discussed above give a simplified representation of the actual processes occurring in an apparatus of this type. The use of mathematical simulation methods makes it possible to reveal the basic trends in the behavior of various characteristics of the processes in a torch reactor and their variations on varying the operation mode and geometry of the apparatus and the fluorination mode. The mathematical model of the fluorination process can be used to estimate the basic characteristics of the processes in a reactor and to reveal the trends in the behavior of various parameters and their sensitivity on varying the geometric dimensions of the apparatus, the entrance conditions for the fuel and oxidizer, and the conditions at the outlet of the reactor. The approximations used in the model are well suited to the essence of the processes under consideration and, at the same time, substantially simplify the mathematical description of the physicochemical processes taking place in an apparatus of this type.

The algorithms for the calculation of the velocity, temperature, and concentration fields for chemical components make it possible to optimize, based on physical and mathematical simulations, the parameters and operation modes of a torch reactor.

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