SHAPE CHANGE OF PROCESS PORES IN KERNELS OF HIGH-TEMPERATURE URANIUM DIOXIDE BASED FUEL ELEMENTS

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A computational method and results for calculating the shape change of an initially spherical vacuum pore with migration in uranium dioxide fuel during operation of a high-temperature fuel element are presented. The conditions for a spherical fuel element to be elongated and flattened in the direction of the temperature gradient, i.e., its transformation into prolate and lenticular, are determined. It is shown that the sign of the second derivative of the saturated vapor pressure of uranium dioxide with respect to the radius of the fuel kernel is decisive. It is established that the region of formation of the prolate pores is located at the periphery of the cross section of a fuel kernel and that of lenticular pores at the center. As the thermal neutron fraction in the reactor spectrum increases, the formation region becomes narrower for prolate pores and wider for lenticular pores.

The initial process porosity of uranium dioxide strongly affects the evolution of the structure, strength properties, and radiation characteristics. For this reason, the shape change of the pores as they migrate in a temperature gradient field is of great interest [1–4]. Large pores which initially are approximately the same size as a grain become prolate as they migrate in the direction of a temperature gradient right up to the formation of cylindrical channels. The observed migration of lenticular pores forms a columnar structure of uranium dioxide grains. The sources of such pores are rounded microcracks in uranium dioxide, which arise in the starting–stopping regimes of a reactor [3]. Lenticular pores can also form during migration of grain boundaries oriented perpendicular to the direction of a temperature gradient with pores arranged on the boundaries and acquiring the shape of a lens as the curvature of the grain boundary increases as it migrates together with the pore [5].

As a supplement to the indicated mechanism, the present work examines the effect of the character of the radial temperature distribution and temperature gradient in a high-temperature fuel element on the shape change of the technological pores distributed in the volume of a fuel pellet. A ventilated fuel element of a thermionic converter reactor with single-element electricity-generating channels based on uranium dioxide with mostly open porosity with ten-micron pores stabilized to thermal sintering is examined [6].

Computational estimates show that vacuum pores with the indicated sizes at the operating temperature migrate as a result of the transfer of uranium dioxide vapors in the pore volume in the free-molecular regime. In this case, the migration rate and pore shape are most sensitive to the local values of the temperature and the temperature gradient in the fuel-element kernel which vary in the migration process. The effect of these factors on the porosity of a uranium dioxide kernel was studied in the present work.

Mass transfer in a vacuum spherical pore with an arbitrary temperature distribution on its surface was investigated in [7]. The intensity of the mass $M(r)$ migration from the wall of a pore is determined by the expression

$$M(r) = \beta \sqrt{\frac{m}{2\pi k}} \left( \frac{p(r)}{\sqrt{T(r)}} - \frac{1}{4}\int r \frac{p(r)}{\sqrt{T(r)}} \, dr \right),$$

(1)
where \( \mathbf{r} \) and \( \mathbf{r}_1 \) is the radius vector from the origin of the spherical coordinates, which is located at the center of the spherical pore, to a point on the surface of a pore; \( \beta \) is the coefficient of evaporation and condensation of the matrix material (in our case uranium dioxide); \( m \) is the mass of a uranium dioxide molecule; \( k \) is Boltzmann’s constant; \( T(\mathbf{r}) \) is the temperature distribution over the surface \( S \) of a pore; \( p(\mathbf{r}) \) is the equilibrium vapor pressure of uranium dioxide at temperature \( T(\mathbf{r}) \); and \( R \) is the radius of a pore.

According to Eq. (1), the pressure and temperature are integrated over the surface of a pore. In the case at hand, the azimuthal variation of the temperature and pressure of the vapor in a plane perpendicular to the temperature gradient can be neglected because of the smallness of the pores. In addition, in a spherical pore \( |\mathbf{r}| = |\mathbf{r}_1| = R \). For this reason, the temperature and pressure in spherical coordinates are a function of only the polar angle \( \theta \). Expression (1) for the intensity of the mass migration in this case assumes the form

\[
M(\theta) = \beta \sqrt{\frac{m}{2\pi k}} \left( \frac{p(\theta)}{\sqrt{T(\theta)}} \right) - \frac{1}{2} \int_0^{\pi} \frac{p(\theta_1)}{\sqrt{T(\theta_1)}} \sin \theta_1 d\theta_1 .
\]  

(2)

Let the spherical coordinate system be arranged so that for \( \theta = 0 \) the direction of the radius vector \( \mathbf{r} \) is also the direction of motion of a pore, i.e., the temperature gradient lies in the axis of the fuel kernel, as shown in Fig. 1. For elongation of the pore in the direction of motion the migration of mass from the frontal surface of the pore must be greater than the arrival of mass on the back surface. Let

\[
A = \frac{1}{2} \int_0^{\pi} \frac{p(\theta_1)}{\sqrt{T(\theta_1)}} \sin \theta_1 d\theta_1 ,
\]

and since at \( \theta = \pi \) the expression in the parentheses in Eq. (2) is negative (mass arrives on the back surface of the pore), we obtain the condition for a pore to be elongated:

\[
\frac{p(0)}{\sqrt{T(0)}} - A > -\left( \frac{p(\pi)}{\sqrt{T(\pi)}} - A \right).
\]  

(3)

Inequality (3) can be rewritten in the form

\[
\frac{p(0)}{\sqrt{T(0)}} + \frac{p(\pi)}{\sqrt{T(\pi)}} > 2A.
\]  

(4)

In addition, when a pore is elongated in the direction of motion the mass migration from the side surface must be negative (i.e., mass must arrive):

\[
\frac{p(\pi/2)}{\sqrt{T(\pi/2)}} - A < 0.
\]  

(5)
We obtain from inequalities (4) and (5) the condition for elongation of the pore in the direction of motion in the form

\[
\frac{p(0)}{\sqrt{T(0)}} \frac{2p(\pi/2)}{\sqrt{T(\pi/2)}} + \frac{p(\pi)}{\sqrt{T(\pi)}} > 0.
\]  

(6)

The left-hand side of equality (6) is a finite-difference expression for the second derivative of the quantity \( p/\sqrt{T} \) along the radius of the fuel kernel at the location of a pore, multiplied by the squared radius of a pore.

Indeed, as one can see in Fig. 2, in a one-dimensional radial model of heat transfer the value of \( f = p/\sqrt{T} \) at the point of a pore with the coordinate \( \theta = \pi/2 \) equal to the value of this quantity at the point of the kernel with radial coordinate \( \rho: f(0) = f(\rho - \Delta \rho), f(\pi) = f(\rho + \Delta \rho) \). Since \( \Delta \rho = R \ll \rho \), the values of \( \rho - \Delta \rho \) and \( \rho + \Delta \rho \) can be regarded as three neighboring radial nodes of a grid partitioning the kernel and the finite-difference representation of the second derivative can be used:

\[
\frac{d^2 f}{dp^2} \frac{f(\rho - \Delta \rho) - 2f(\rho) + f(\rho + \Delta \rho)}{(\Delta \rho)^2} = \frac{f(0) - 2f(\pi/2) + f(\pi)}{R^2}.
\]

Thus, the inequality \( d^2(p/\sqrt{T})/dp^2 > 0 \) is equivalent to

\[
\frac{1}{R^2} \left( \frac{p(0)}{\sqrt{T(0)}} \frac{2p(\pi/2)}{\sqrt{T(\pi/2)}} + \frac{p(\pi)}{\sqrt{T(\pi)}} \right) > 0.
\]

Multiplying the last inequality by the positive quantity \( R^2 \) gives inequality (6).

Neglecting the small, as compared with the change of the equilibrium vapor pressure, change of \( T^{1/2} \) in the operating range of the fuel temperature, the condition for elongation of a spherical pore in the direction of motion at a point with the coordinate \( \rho \) along the radius of the fuel kernel can be written in the form \( d^2 p/ dp^2 > 0 \). Similar calculations show that for \( d^2 p/ dp^2 < 0 \) a pore flattens, i.e., the spherical shape becomes lenticular.

This is clearly shown in Fig. 3. If \( d^2 p/ dp^2 > 0 \), then on the long radius of the fuel kernel, i.e., on the back surface of the pore at \( \theta = \pi \) the derivative of the equilibrium vapor pressure of uranium dioxide with respect to the radius of the kernel \( dp/ dp \) is larger than on the shorter radius, i.e., on the front surface of the pore at \( \theta = 0 \). Since the derivative \( dp/ dp \) for the entire layer of fuel is negative because of the temperature increase in the direction of the axis of the fuel kernel, we have
Keeping in mind that the rate of evaporation and condensation are proportional to the modulus of the pressure gradient, the rate of evaporation on the front surface of the pore is higher than the rate of condensation on the back surface and the pore becomes elongated. Conversely, if \( \frac{d^2 p}{d \rho^2} < 0 \), then \( \left| \frac{dp}{d\rho} \right|_{\theta=\pi} > \left| \frac{dp}{d\rho} \right|_{\theta=0} \) and the pore becomes flattened.

The temperature dependence of the equilibrium vapor pressure of uranium dioxide versus the temperature has the form

\[
p(T) = p_0 \exp(-Q/\bar{R}T),
\]

where \( p_0 \) is a pre-exponential factor; \( Q = 567 \text{ kJ/mole} \) is the heat of evaporation of uranium dioxide [8]; \( \bar{R} = 8.31 \text{ J/(mole·deg)} \) is the universal gas constant.

Differentiating Eq. (7) twice with respect to \( \rho \), we obtain

\[
\frac{d^2 p}{d\rho^2} = p \frac{Q}{\bar{R}T^2} \left[ \frac{1}{T} \left( \frac{Q}{\bar{R}T} - 2 \right) \left( \frac{dT}{dp} \right)^2 + \frac{d^2 T}{dp^2} \right].
\]

In the operating temperature range of the fuel in the fuel elements of the electricity-generating channels \( Q/\bar{R}T \gg 1 \), so that the number 2 in the factor \( Q/\bar{R}T - 2 \) in Eq. (8) can be neglected.
The sign of the second derivative \( \frac{d^2 p}{d \rho^2} \) is determined by the expression in the brackets. We shall find the critical radial point \( \rho_0 \) where this expression vanishes, i.e., where the sign of the second derivative changes, assuming that the temperature distribution along the radius of the fuel kernel can be approximated by a power-law function of the form

\[ T = T_0(1 - a \rho^n). \]  

(10)

Then

\[ \frac{dT}{d\rho} = -T_0an\rho^{n-1}; \]

\[ \frac{d^2T}{d\rho^2} = -T_0an(n - 1)\rho^{n-2}. \]  

(11)

Equating the expression in brackets in Eq. (9) to zero, substituting into the equality obtained the expressions for the temperature and its derivatives from Eqs. (10) and (11), and performing simple transformations, we obtain a quadratic equation for the \( a\rho_0^n \):

\[
\frac{a\rho_0^n}{(1-a\rho_0^n)^2} = \frac{n-1}{n} \frac{1}{Q/RT}. \]  

(12)

Examination of Eqs. (9) and (12) shows that when the radial coordinate of a pore \( \rho > \rho_0 \), then \( \frac{d^2 p}{d \rho^2} > 0 \), i.e., this is the region of elongation of the pore in the direction of motion; if \( \rho < \rho_0 \), then \( \frac{d^2 p}{d \rho^2} < 0 \) and this is the region where the pore is flattened and becomes lenticular.
Since $Q/\dot{R}T >> 1$, it follows from Eq. (12) that for $ap^n \ll (1 - ap^n)^2$, so that the $ap^n$ in the denominator on the left-hand side of this equation can be neglected compared to 1. Then we obtain finally an expression for calculating the coordinates of the interface of the regions of elongation and flattening of the pores in the form

$$
\rho_0 = \left(\frac{(n-1)\dot{R}T_0}{naQ}\right)^{1/n}.
$$

The method developed was used to perform calculations for operating the electricity generating channels in fast and thermal reactors as well as in a reactor with 20% heat release due to fast neutrons, which corresponds to a reactor with zirconium hydride moderator. A fuel element of a single-element electricity generating channel is characterized by the following parameters: the outer radius of the kernel 0.865 cm, the radius of the central channel in the kernel 0.4 cm, the emitter temperature 1600°C, and the heat flux on the emitter 25 W/cm².

The least-squares temperature distribution using the (10) function along the radius of the kernel has the form $T = 1691(1 - 0.31\rho^{1.2})$ in a thermal reactor and $T = 1963(1 - 0.31\rho^{3.5})$ in a fast reactor, and $T = 1841(1 - 0.24\rho^{4.2})$ in a reactor with a zirconium hydride moderator (Fig. 4). The critical radius $\rho_0$ for these three cases is 0.81, 0.46, and 0.56 cm, respectively (Fig. 5).

**Discussion.** In a high-temperature ventilated fuel element based on uranium dioxide, conditions are created where elongated and lenticular pores are formed by mass transfer in different zones of a kernel containing large vacuum process pores. A shift of the neutron spectrum from thermal to fast increases the zone of elongated pores. Since the radial temperature differential in the kernel of a fast reactor is several hundreds of degrees higher than in a thermal reactor at the same power, the time required for the uranium dioxide structure to change in these reactors will differ considerably.

In the case of a hermetic fuel element, mass transfer of uranium dioxide inside gas-filled pores proceeds in the diffusion regime and for this reason it can no longer be described by the equations presented. For this reason, a shape change of such pores requires further quantitative analysis.

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**REFERENCES**