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The kinetics of fission products release from microfuel taking into account the trapped fraction and limited solubility effects

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ABSTRACT

In this paper the effect of the oxygen getter on fission products release from the coated particle was studied by the "FP Kinetics" code. Trapped fraction and limited solubility effects taken into consideration. It was shown that these effects have a significant impact on the concentration profile and integral release of fission products.

1. Introduction

Possibilities of making coated fuel particles with kernels of plutonium oxide, uranium oxide, MOX fuel, thorium-containing fuel and other kinds of fuel are being explored. TRISO-coated particles are spherical fuel kernels, which coated a carbon buffer layer and layers of pyrolytic carbon and silicon carbide. Development of new types of coated fuel particles for high-temperature gas cooled reactors, which will ensure high levels of fuel burnup, call for essential advancement of our knowledge. In many instances a judgment on operational reliability of coated particles (CP) cannot be made on the basis of the available experience. Of course, it is advisable to do direct experiments to acquire necessary information, but they are challenging and costly. In view of this, evolution of prediction methods based on more in-depth physical and physico-chemical models of fuel behavior at high burnup and development of related computer codes become particularly important. One of the key aspects of the problem is the study of changes in the CP morphology, composition and structure during almost complete transformation of the kernels. Another important problem is release of fission products both from damaged and intact coated particles. Calculations of coated fuel particles thermodynamics and thermomechanics play a critical role in its solution. However, at present there are no computer codes that would reliably calculate thermodynamics of such complex systems as high burnup nuclear fuel. The existing models and codes were tested on rather simple systems containing just a few chemically reacting components. Therefore, performance of test experiments is one of the most vital tasks in the effort to develop new types of coated fuel particles for high burnup systems.

2. Diluted kernels

It is well known that achievement of high levels of burnup in HTGR coated particles is a rather complicated technical problem. This is connected, first of all, with a considerable internal pressure buildup due to formation of carbon oxides and gaseous fission products with increasing burnup. Various solutions allowing an essential improvement in operational reliability of TRISO coated particles have been analyzed in recent years. First, this is the use of diluted kernels. Dilution of kernels allows, on the one hand, significantly reducing the amount of fission products produced, and on the other hand, increasing the free volume where gases are accumulated. Zirconium dioxide can be used as diluting material. Numerous studies of yttrium-stabilized ZrO_2 (Y-FSZ) as an inert matrix candidate for plutonium-based coated particles produced good results on radiation resistance, low irradiation-induced swelling within the required temperature range, phase stability and compatibility with various nuclear fuel fission products (Damen et al., 2003; Rounet, 1971; Degueldre et al., 1995; Arima et al., 2003). Second, an oxygen getter that drastically reduces the CO concentration during the burnup process is a required element of plutonium-based coated particles under consideration. To date, detailed computational and theoretical analyses of thermodynamics of CP with diluted kernels and oxygen getters based on SiC and ZrC during the burnup process have been carried out (Rusinkevich and Ivanov, 2013).

3. Oxygen getters

The employment of an oxygen getter has an essential beneficial effect as it reduces pressure in CP due to reduction of the carbon oxide pressure, which is especially important at high levels of burnup and temperature. However, its use leads to a significant

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increase of the metal component in the CP, especially for such metals as Cs, Rb, Zr, Sr, Ba, Y, La, Ce, Pu, Mo and Cd, which may result in an increased release of these components from the CP. At temperatures up to ~1500 °C, mass fractions of components that can be in a liquid state in CP with an outer SiC oxygen getter are only slightly higher than in CP with a ZrC oxygen getter. But at higher temperatures a significant buildup of the liquid phase due to formation of FP silicates and silicides is observed. One of distinct advantages of using an outer SiC getter over a ZrC one is the almost complete lack of interaction between the components of a kernel when a SiC coating of the CP is used. The reason is that one of the main factors responsible for degradation of the CP load-bearing layer is corrosion interaction between the SiC layer and kernel components.

The question is when there will be no corrosion interaction. It will be weak if the SiC layer is tightly covered with dense pyrolytic carbon. However, as fast neutron fluence increases, a significant anisotropic shrinkage of PyC takes place, and it cracks. The probability of failure of the inner PyC layer goes up, reaching the value of 5×10^{-5} at the fast neutron fluence Wc of $\sim 1 \times 10^{25}\ \mathrm{m}^{-2}$, which is several times less than the limit value of $5{\cdot}10^{25}\ \mathrm{m}^{-2}$ (Kaae, 1977; NERI, 2004; Koss et al., 1973; Kaae, 1973).

If, however, the fuel contains a SiC oxygen getter, the interior of the CP consisting of the kernel, fission products and buffer layer will be in a state close to thermodynamic equilibrium with SiC. Therefore, damages produced in the dense pyrolytic carbon layer shall not result in significant corrosion of exposed parts of the SiC load-bearing layer.

The use of solutions listed above provides for high levels of CP operational reliability at burnup of up to 80% FIMA.

4. Fission products transport

Another important aspect of the problem under consideration is transport of fission products through fuel particle coating layers, which has a number of distinctive features at high burnup levels. First of all, this is a so-called “trapped fraction” effect. This effect is associated with the need to renormalize the source function for fission products produced in the course of the burnup process. The point is that chemical bonding of fission products and the effect of these processes on the diffusion transport should be taken into consideration when constructing the source function.

Another interesting and important effect is the necessity of taking into account the limited solubility of FP in fuel particle coatings. It should be noted that solubility could have an impact on FP release as a result of her limitations, and due to of jumps of concentrations at interfaces. In this article, the effect of the limited solubility has been studied. In this case at low concentrations of components, when concentrations of FP in coating materials are less than respective solubility limits, the concentration field remains continuous. However, the FP concentration in the coated particle increases with increasing burnup, and at some point the concentration of any of the components may become higher than the limit of solubility in one or another coating material. In that event the concentration of the component involved has a discontinuity. In the interior of the CP the concentration continues to grow as the fuel burns up. But at the boundary on the side of the material having low solubility the concentration cannot increase; it remains constant and equal to the limit of solubility at a given temperature. Hence consideration of the limited solubility of diffusing components in the SiC layer may introduce a significant correction to the release of radionuclides from CP, reducing it at high levels of fuel burnup.

A few years ago we made an attempt to evaluate the influence of the effects under consideration on the release of fission products from fuel particles at high burnup levels using the TRAFIC code (General Atomic). The influence of discontinuities in the concentration field on the FP Kinetics can be taken into consideration in the TRAFIC code model. Concentration jumps at the interfaces are created by setting the discontinuous boundary conditions. Boundary coefficients are entered for this purpose in the conditions of cross-linking. However, the values of these coefficients in the TRAFIC code are set initially and remain constant over time. Therefore, an adequate description of the real diffusion process with the TRAFIC code encounters considerable difficulties. To solve this problem, we developed a new code for calculation of release of fission products from coated particles – FP Kinetics – that takes into account the effects of chemical bonding and limited solubility.

5. FP Kinetics code

For the spherically symmetric problem, the diffusion equation for calculation of the field of concentrations inside the CP takes the form:

$$\frac{\partial c^{(i)}}{\partial t} = \frac{1}{r^2}\frac{\partial}{\partial r}\left(r^2 D^{(i)}\frac{\partial c^{(i)}}{\partial r}\right) + \dot{s}, \qquad \dot{s} = \frac{\mathrm{d}}{\mathrm{d}t}\left[c^*(t)\cdot(1 - F(t))\right] \tag{1}$$

where $c(r, t)$ is the nuclide concentration; $D^{(i)}$ is the coefficient of diffusion in layer i; and $\dot{s}$ is the function of the radionuclide source in the kernel. The radionuclide source function is the variation of the radionuclide concentration per unit volume of the kernel in unit time due to chemical reactions $(1 - F)$ and generation processes c^*. As demonstrated before (Belov and Ivanov, 2005), these processes proceed at much a higher rate than the diffusion transport through the CP coating layers, while allowing sufficient time for attainment of the near thermodynamically equilibrium state in the interior of the CP.

The initial and boundary conditions:

The spherical symmetry condition requires that in the central part of the kernel:

$$\left.\frac{\partial c}{\partial r}\right|_{r=0} = 0. \tag{2}$$

The boundary condition on the surface of a particle of radius R:

$$c^{(i)}(R, t) = 0, \tag{3}$$

Thus we assume that atoms arriving at the outer surface of a CP do not stay there but almost immediately get into the matrix graphite and further to the coolant. A conservative estimate of the FP release that corresponds to the maximum diffusion flow is implemented in this case.

The initial condition:

$$c^{(i)}(r, 0) = c_0^{(i)}(r). \tag{4}$$

where ${c_0}^{(i)}(r)$ is the initial nuclide concentration inside the CP.

Hence, until the concentration of the diffusing element reaches the limit value for the SiC layer, the diffusion problem coincides with the standard one. However, upon reaching the limit concentration the problem breaks down into an exterior problem and an interior one; the exterior problem becoming a problem with a constant concentration set at the inner boundary of the SiC layer.

In addition to the concentration field, the code also calculates the CP outlet flow J and the integral release of FP from the CP W.

$$J = 4\pi R^2 D^{(5)}\left(\frac{\mathrm{d}c}{\mathrm{d}r}\right)_{r=R}, \tag{5}$$

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