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Simulations of coupled heat transport, oxygen diffusion, and thermal expansion in UO_2 nuclear fuel elements

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ABSTRACT

We study the coupled thermal transport, oxygen diffusion, and thermal expansion of a typical nuclear fuel element consisting of UO_{2+x} fuel and stainless-steel cladding. Models of thermal, mechanical and chemical properties of the materials are used in a series of finite-element simulations to study the effect of the coupled phenomena on the temperature profile, oxygen distribution and radial deformation of the fuel element. The simulations include steady-state and time-dependent regimes in a variety of initial- and boundary value conditions that include sudden changes in the power density, variable oxygen content in the atmosphere, and variable temperature of the coolant. The study reveals the difference in the characteristic times associated with these phenomena and the importance of performing coupled simulations.

1. Introduction

Understanding the evolving properties of nuclear fuels and predicting the behavior of nuclear fuel elements (ceramic fuel pellets or metallic fuel rods) are major challenges for fuel manufacturing, performance, and storage. For example, sintering of ceramic fuel pellets requires a strict control of composition, thermal treatment, pressure, and atmosphere [1]. Similar issues impact the casting of metallic fuel rods [2] and the manufacturing of pebble fuels. The large number of control parameters and the uncertainty associated with them leads often to challenging problems that can be addressed by a close integration of experimental, theoretical, and computational work [3].

Once in the reactor, the fuels and structural materials (pressure vessels, pipes, ducts, etc.) are subjected to severe radiation environments that change their thermo-mechanical properties [4]. For instance, ceramic fuels exhibit radial and angular cracks and the severity of such structural damages increases with burn-up [5]. Among the main causes responsible for the deterioration of structural damage leading to a decrease in thermal conductivity are fission product migration and gas bubbles accumulation.

When multi-component systems experience significant temperature gradients, the constituents are thermally driven apart by Fickian diffusion mechanisms. Simultaneously, gradients in the concentration of constituent components build up and drive the diffusion through a distinct mechanism known as the Soret effect [6,7]. As a result, the microstructural properties evolve with position and time, sometimes producing severe changes in measurable macroscopic properties, such as thermal conductivity [8,9], or thermal expansion coefficients. This intricate scenario was confirmed by recent simulations of heat transport coupled with oxygen diffusion in typical UO_2 fuel elements [10,11] as well as in CANDU reactors [12–14].

Recent reviews of fuel performance codes [15,16] demonstrate that extrapolating materials properties to high burn-up values is a challenging task. Predicting the thermal, mechanical and chemical phenomena in the fuel element is even more challenging. In this study we demonstrate that coupling heat transport, oxygen diffusion, and thermal expansion in a fuel pellet can provide insight into the main mechanisms that cause fuel damage. This is a complex computational task that involves a detailed knowledge of UO_2 thermochemistry [4,17,18].

2. Computational set up

We focus on studying a simple schematic model aimed at elucidating general aspects of fuel behavior linked to the transport of the oxygen in the fuel element. The simulation domain consists of a cylindrical UO_{2+x} fuel pellet and steel cladding separated by a helium gap, as depicted in Fig. 1. In this “typical” fuel element, the pellet radius is $R_{\text{fuel}} = 4.3$ mm, the helium gap width is 0.03 mm, and the cladding thickness is 0.5 mm. In this fuel element we solve for the coupled equations describing thermal expansion

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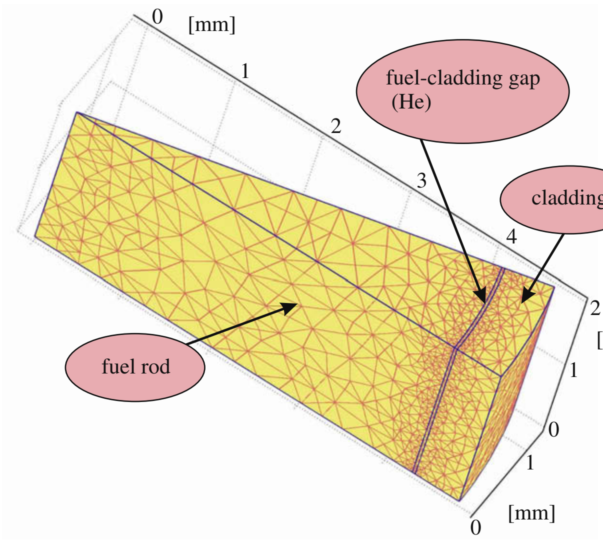

Fig. 1. Geometry of the fuel element (urania pellet, helium gap and steel cladding) for the simulations presented in this paper. While our simulation capability is ready to handle multidimensional effects, the simulations presented in this paper are essentially one-dimensional simulations.

of the fuel pellet and steel cladding, heat transport, and oxygen diffusion. The domain shown in Fig. 1, is azimuthally symmetric, and therefore all simulations reported here are effectively one-dimensional (all gradients occur in the radial direction only). Nevertheless, we set up the simulations in a three-dimensional domain as a preview of future studies which will include multidimensional effects. Assuming that the heat generated by fission reactions in the fuel is uniformly distributed, a constant, volumetric source term, Q, is added to the heat transfer equation:

$$\rho C_p \frac{\partial T}{\partial t} = \nabla \cdot (k \cdot \nabla T) + Q, \tag{1}$$

where ρ, C_P and k are the density, specific heat at constant pressure and thermal conductivity, respectively. The models of the thermal conductivity, $k(T, x)$, and the heat capacity, $C_P(T, x)$, include dependence on the deviation from stoichiometry, x, in UO_{2+x}.

Eq. (1) is coupled with the equation describing the diffusion of oxygen in the fuel pellet:

$$\frac{n}{2}\frac{\partial x}{\partial t} = -\nabla \cdot J, \tag{2}$$

Here, n is the concentration of oxygen sites and the oxygen flux J is given by [19,20]

$$J = -\frac{n}{2} D(T,x) \cdot \left[\nabla x + \frac{x}{F(T,x)} \frac{Q}{RT^2} \nabla T \right], \tag{3}$$

where $D(T, x)$ is the oxygen diffusivity in the fuel, $F(T, x)$ is the thermodynamic factor, Q^* is the heat of transport of oxygen, and R is the universal gas constant. In the parenthesis of Eq. (3), the first term is associated with the conventional Fickian diffusion, whereas the second term represents the Soret effect which accounts for the oxygen diffusion driven by the temperature gradient [6,7].

The predictive power of these simulations depends on the accuracy of solving self-consistently the system of coupled Eqs. (1)–(3) for temperature, non-stoichiometry, and thermal expansion profiles, and the uncertainty associated with the models of UO_{2+x} properties (i.e. the specific heat $C_P(T, x)$, the thermodynamic factor of oxygen $F(T, x)$, the thermal expansion coefficient $k(T, x)$ and the oxygen diffusivity in urania, $D(T, x)$. Developing models of these properties as functions of temperature and oxygen content, proves to be a challenging task, because in a nuclear reactor environment strong irradiation induces complex defect species, microstructural changes, and the properties evolve with time.

In this work, the properties of the materials in the fuel element were obtained from previously published correlations or from analysis of previously published data, and are summarized in Table 1. A few comments regarding our choice of parameters for UO_{2+x} are in order: (i) First, we note that the expression for the heat of transport of oxygen, Q^*, was obtained by fitting experimental data from Ref. [19] for mixed fuels instead of UO_{2+x}. However, in the absence of data for UO_{2+x}, this should be acceptable because Q^* depends only weekly on the plutonium content, as seen from Fig. 1in Ref. [19]. (ii) Second, we note that the temperature-dependence of the density and thermal expansion of UO_{2+x} in Table 1 do not include an explicit composition dependence [21,22] in accordance with the work of Martin [22], which suggests that there is little difference for these properties between stoichiometric and hyper-stoichiometric fuel. (iii) and, finally, we note that in contrast with our previous work [10] where the specific heat of UO_2 was calculated in the temperature domain 600–1600 K based on a linear interpolation of the experimental data evaluated in SGTE and JANAF tables, i.e.

$$C_p(UO_2) = 264.256 + 0.047T \ [\mathrm{J/(kg\ K)}], \tag{4}$$

in this work we employ the composition-dependent model put forward in Ref. [12]. A comparison of the two models for the temperature and stoichiometry domains of interest here is depicted in Fig. 2. We emphasize that the results of all transient simulations presented next are insensitive to the choice of heat capacity model for UO_{2+x}, and therefore we conclude that the stoichiometry effects of the UO_{2+x} heat capacity are not important and may be neglected, at least as our simulation scenarios are concerned.

With respect to the steel cladding material properties, in this paper we use a 316 grade stainless-steel cladding material instead of the 347 grade stainless-steel cladding used in Ref. [10]. This change was made for convenience, based on our ability to compile a consistent set of material properties for a cladding made of 316 grade austenitic stainless-steel based on the data readily available in the literature.

It is important to note that although both urania and plutonia adopt the same fluorite structure, plutonium oxide tends to be hypo-stoichiometric PuO_{2-x}, while uranium oxides are most often hyper-stoichiometric (UO_{2+x}). More remarkably, urania exhibits a negative heat of transport of oxygen [30]. Therefore, in urania the oxygen ions will migrate from the regions of low temperature to the regions of high temperature, leading to an increase in oxygen concentration close to the center of the fuel pellet. This effect is clearly seen from Eq. (3) which shows that the oxygen flux vanishes for steady-state conditions. Under this constraint, in materials with negative heat of transport, Q^*, the gradient of oxygen concentration follows the temperature gradient. This is the opposite of what occurs in a material with positive Q^*, such as hypo-stoichiometric PuO_{2-x} [31], in which the oxygen atoms migrate from the hot to the cold regions.

3. Results and discussions

In the following, we report results for both steady-state and time-dependent simulations corresponding to the fuel element described in Fig. 1. Solutions of the structural mechanics problem in the fuel and cladding are obtained self-consistently by solving for the temperature (T) distribution within the fuel, helium gap and steel cladding, and by solving for the oxygen non-stoichiometry (x) profile within the fuel. Symmetrical boundary conditions are used for solid mechanics, T, and x along the straight edges of the fuel element (see Fig. 1). The fuel pellet and steel cladding are al-