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Journal of Alloys and Compounds 444–445 (2007) 415–423

Journal of ALLOYS AND COMPOUNDS

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Models and simulations of nuclear fuel materials properties

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Received 1 September 2006; received in revised form 17 January 2007; accepted 19 January 2007
Available online 24 January 2007

Abstract

To address the complexity of the phenomena that occur in a nuclear fuel element, a multi-scale method was developed. The method incorporates theory-based atomistic and continuum models into finite element simulations to predict heat transport phenomena. By relating micro and nano-scale models to the macroscopic equilibrium and non-equilibrium simulations, the predictive character of the method is improved. The multi-scale approach was applied to calculations of point defect concentration, helium bubbles formation, oxygen diffusivity, and simulations of heat and mass transport in UO_{2+x}.

Keywords: Actinide alloys and compounds; Nuclear reactor materials; Oxide materials; Microstructure; Point defects; Thermodynamic properties; Thermochemistry; Computer simulations; Molecular dynamics Simulations; Thermodynamic modeling

1. Introduction

Nuclear fuel materials (alloys and ceramics) are complex and often exposed to severe irradiation environments during manufacturing, operation, and storage. Most commercial fuel performance codes incorporate models of materials properties that are based on empirical correlations. The applicability of such models is limited to a regime where experimental data is available and extrapolation outside this regime may lead to misleading results.

An example of integration of various time and length scales is the prediction of the Pu–Ga phase diagram, in the low Ga content region [1]. Based on a method for determining phase stability in binary systems [2], the equilibrium phase diagram of Pu–Ga was calculated using limited information about the components. Electronic structure calculations provided input for the modified embedded atom method (MEAM). Molecular dynamics calculations using the MEAM many-body inter-atomic potential led to predictions of the free energy of all phases in the low temperature, low Ga content region of the Pu–Ga system [1]. The existence of a eutectoid point that involves monoclinic Pu, face centered cubic (fcc) Pu, and the Pu_3Ga compound was predicted confirming the metastable character of the fcc Pu–Ga solution.

In this work we propose a multi-scale and multi-physics approach to develop a better understanding of heat transfer and oxygen diffusion in urania (UO_2), a widely used nuclear fuel material. The method requires a theoretical framework to allow for prediction of phenomena such as phase stability, heat transfer, species diffusion, and fission products retention. As a first step, several methods are used at specific time and length scales to determine properties of UO_2.

Fig. 1 shows the time and length scales that are involved in the simulations and the main theoretical methods that are currently used at Los Alamos National Laboratory to develop the models. The multi-scale method relies on enhanced contributions from the electronic structure theory and atomistic calculations. Electronic structure calculations are used to derive the parameters of the inter-atomic potentials. The potentials are validated against a minimal set of thermo-mechanical data. The use of molecular dynamics allows for incorporating vibrational components of thermodynamic properties. Using atomistic methods, the free energy of point defects is determined and the concentration of various types of defects calculated. That serves as input for developing models of non-stoichiometry, thermal conductivity, and oxygen diffusivity. This information is then provided to continuum level simulations of transport phenomena such as

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doi:10.1016/j.jallcom.2007.01.102

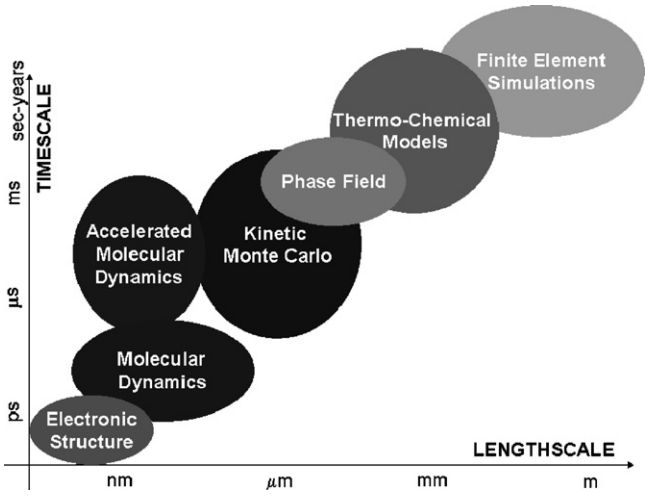

Fig. 1. Length and time scales involved in simulating phenomena relevant for nuclear materials as covered by computational methods.

heat conduction and oxygen diffusion. In this method, the information is transfer via key material properties such as density, formation and migration free energy, thermal conductivity, and species diffusivity.

At this time we are not able to fully interconnect the components and more work is necessary before producing reliable simulations. In this paper, we present preliminary steps towards integrating various length and times scales into a self-consistent simulation of transport phenomena in a nuclear fuel element. To reach the level of precision required by the multi-scale method, the properties are determined with high accuracy. However, the link between scales is imperfect and sometimes missing. Future studies will be dedicated to improving the transfer of information and determining the uncertainty associated with the models and simulations.

2. Results and discussion

2.1. Finite element simulations of heat transport and species diffusion

The temperature and oxygen distribution in UO_2 nuclear fuels are influenced by several phenomena such as neutron flux gradients that produce non-uniform fission density, volumetric heat rate and fission product yields, density changes including swelling, radial cracks, and porosity formation, and microstructural changes such as recrystallization and grain growth. In this work we are focusing on the coupled heat transport and oxygen diffusion phenomena, as a first step towards a more complex study. In a typical fuel element, cylindrical UO_2 fuel pellets are stacked in a metal alloy cladding and the gap between the cladding and the fuel pellet is filled with helium. The fission reaction generates heat that is diffused mostly outwards in the radial direction. Due to the Soret effect, the gradients in temperature induce oxygen diffusion which in turn triggers the conventional Fickian diffusion. The counterbalancing effects of the Soret and Fickian fluxes are responsible for a variation of

Table 1
Summary of materials properties (temperature in K)

Property	Dependence on temperature $T(K)$ and stoichiometry x		Units	Source
$F(UO_2)$	$F(T,x) = \dfrac{2+x}{2(1-3x)(1-2x)}$			[5]
$Q^*(UO_2)$	$Q^*(x) = -1380.8 - 134435.5\exp(-x/0.0261)$		J mol^{-1}	[6]
$\rho\ (UO_2)$	$10960(a+bT+cT^2+dT^3)^{-3}$,	$a = 0.997, b = 9.082\times10^{-6}$ $c = -2.705\times10^{-10}, d = 4.391\times10^{-13}$ } $T < 923$ K, $a = 0.997, b = 1.179\times10^{-5}$ $c = -2.429\times10^{-9}, d = 1.219\times10^{-12}$ } $T > 923$ K	kg m^{-3}	[6]
$k(UO_2)$	$k(T,x) = \lambda0(T)\dfrac{\arctan[\theta(T,x)]}{\theta(T,x)} + 5.95\times10^{-11}T^3$ $\lambda0(T) = [3.24\times10^{-2} + 2.51\times10^{-4}T]^{-1}$ $\theta(T,x) = 3.67\exp(-4.73\times10^{-4}T)\sqrt{2x\lambda0(T)}$		W m^{-1} K^{-1}	[7]