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Oxygen Behavior and Diffusion Model in LWR Fuel Using the THERMOCHIMICA Module in Moose/Bison

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Materials Science and Technology Division

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Executive Summary

A comprehensive thermochemical database has been developed for representing irradiated fuel with fission products. Calculations of oxygen potential as a function of burnup were performed using the database, and when clad oxidation is considered the results appear to agree well with the limited available measurements. The thermochemical equilibrium solver software library THERMOCHIMICA was used for modeling oxygen transport in UO_2 fuel. Similar to recent publications, we assume that the hyper-stoichiometric UO_{2+x} systems are pseudo-binary systems that can be described as dilute solutions of oxygen interstitials in the oxygen sub-lattice. The model is coupled with thermal diffusion and accounts for oxygen diffusion due to temperature gradients. The thermal and transport material properties are functions of temperature and non-stoichiometry of the fuel. Another implementation of oxygen transport is under development that is more applicable to heterogeneous composition of irradiated fuel and is based on chemical potentials, and not simply the concentrations. This implementation involves original scientific development and will be a subject of a planned journal publication.

Introduction

Irradiated LWR UO_2 fuel generates up to sixty transuranic and fission product elements resulting in a complex chemical system. To represent that system a thermodynamic database is under development that can be used with equilibrium solvers to determine the local chemical state, including vapor pressure and oxygen potential. To date the database contains assessed representations of U-Pu-O, U-La-O, U-Gd-O, U-Ce-O because plutonium and the lanthanoids are high concentration fission products that readily dissolve in UO_2 and have a strong influence on behavior. Thus these together can accurately predict behavior within the combined systems in fuel. Other important fission products are also included in the database, along with secondary phases, only a subset of which has been assessed. Model approximations in the form of ideal solutions in the fluorite phase are being used for the remainder of the systems, and these can provide reasonable results pending completion of the remaining assessments of important fission products.

THERMOCHIMICA [1] is a software library developed for calculation of thermochemical equilibrium at a prescribed composition, temperature and pressure. The equilibrium solver determines a unique combination of phases and their compositions at thermochemical equilibrium. The computational algorithms and software were developed at the Royal Military College of Canada and at the Oak Ridge National Laboratory and have been released in the public domain. THERMOCHIMICA routines were developed for incorporation of equilibrium thermo-chemical calculations into other simulation codes, with emphasis on nuclear fuel problems. Considerable research efforts have been undertaken to achieve high computational performance, primarily through advanced algorithm development, such as improved estimation techniques and domain tailored non-linear solvers [2, 3].

THERMOCHIMICA was incorporated into the MOOSE (Multiphysics Object Oriented Simulation Environment) [4] code that was developed by the Idaho National Laboratory. MOOSE is a computational framework for development of simulation tools for solving coupled, nonlinear partial differential equations (PDEs). BISON [5] is a finite element method (FEM) code built within MOOSE for simulation of coupled multi-physics phenomena in nuclear fuel elements and nuclear reactor engineering. The physical phenomena at the continuum scale are modeled by conservation laws expressed by PDEs and the coupling of the physics models is then formulated and modeled by systems of coupled PDEs. A detailed description of the implementation of THERMOCHIMICA can be found in a technical report [6].

THERMOCHIMICA Implementation for Nuclear Fuel

THERMOCHIMICA software consists of the library of material thermodynamic properties, a select set of thermodynamic models, and a numerical solver to minimize global Gibbs energy while respecting the Gibbs Phase Rule and conservation of mass. THERMOCHIMICA utilizes input parameters and a specific model framework to predict a unique combination of species and phases that are stable at thermodynamic equilibrium and then outputs material properties and boundary conditions. Specifically, the software uses input parameters such as temperature, pressure, and species mass together with a thermodynamic database such as that described above, to calculate species mole fraction, chemical potentials, thermochemical activities, moles of phases present, enthalpy, heat capacity, and Gibbs energy.

THERMOCHIMICA solves for thermodynamic equilibrium and does not take chemical kinetic information into consideration. It is implicit to the method that the chemical reactions are controlled by the thermodynamics and not the kinetics. In reality, chemical equilibrium is not achieved instantaneously, but chemical kinetics is not very significant for sufficiently high temperatures, long time periods, and when atoms of the various chemical elements are randomly mixed in the system. In nuclear fuels under normal operating conditions, chemical equilibrium is achieved quickly due to the high temperatures, and the chemical reactions occur over long time periods between refueling. Also, due to the nature of fission, the atoms of the various elements representing the transuranics and fission products are randomly mixed in irradiated nuclear fuel. This implicit assumption of local thermodynamic equilibrium must be taken into account in model development and in the solution procedures in the code. The current implementation of chemically reactive transport for THERMOCHIMICA in MOOSE is based on a quasi-rate formulation in order to be compatible with the MOOSE framework. Therefore, at each time step thermodynamic equilibrium of the system is determined. The quasi-rate is not related to kinetics, it is merely the mechanism to implement the thermodynamics solver into the rate formulations of MOOSE. The time integration is first order in time, and further enhancements are under development in MOOSE to minimize the computational cost of chemical equilibrium calculations and transport of multiple species.

Modeling of Oxygen Transport in LWR Nuclear Fuel Material

Modeling of species diffusion in multicomponent systems is a challenging and still an open science problem [7, 8]. For simple systems, a Fick's law [7] approximation is commonly used that assumes that the concentration gradients are the driving forces of diffusion. The recent papers on multiphysics modeling of oxygen transport in hyper-stoichiometric LWR fuel [9-12] assume that they are pseudo-binary systems that can be described as dilute solutions of oxygen interstitials in the oxygen sublattice. The atomic fractions of oxygen interstitials, c, are then related to the deviation from stoichiometry in the fluorite phase as:

$$x = \frac{O}{M} - 2$$

where O/M denotes the oxygen to metal ratio of the fluorite phase. Assuming that c corresponds to x, the problem is then cast in the form of standard Fickian diffusion with x as the primary transporting variable. The assumption of a binary system is adequate for fresh fuel, but for complex multicomponent systems, such as the irradiated LWR nuclear fuels, a more fitting form of the driving force for species transport is the gradient of their chemical potential [13-15]. Oxygen potential measurements have been made for high burnup LWR UO_2 fuel [16, 17], and examples of computed values are seen in this report, but they have not yet been extensively used in transport models. On the other hand, formulating a diffusion model using the chemical potential increases its mathematical complexity. Given the multitude and complexity of the operating phenomena in the reactor environment, existing models [18] use LeClaire's approach [19] of using Fick's law with the diffusion coefficient fitted as an empirical function of concentration. Implementation of THERMOCHIMICA into the MOOSE framework will allow for easier use of chemical potentials as driving forces in multicomponent systems. The current THERMOCHIMICA implementation for modeling of oxygen transport, however, employs the Fickian, O/M approach to demonstrate the capability. In depth modeling of oxygen transport in irradiated LWR is the subject of the ongoing project.

Thermochemical Database for LWR Nuclear Fuel

A global LWR nuclear fuel database has been under development at ORNL. It is currently based on the original work of Gueneau, et al. [20] who developed a thermochemical model for the fluorite structure urania phase with solutes, and in their work plutonium was considered as a solute. The model utilizes the compound energy formalism (CEF), which in this case allows for consideration of cation solutes and oxygen vacancies and interstitials.

The urania-plutonia CEF model has been expanded to include all the relevant rare earth fission products as well as yttrium, zirconium, molybdenum, and tellurium. Given the fact that fission product concentrations will be relatively dilute, a good assumption was that interactions would occur only between uranium and the respective other cations in the structure. Thus thermochemical models of urania with the fission products integrated into a single CEF model for the fluorite phase would be sufficiently accurate. To date, assessed models for urania with plutonium, lanthanum, cerium, gadolinium (as a burnable poison) have been included in the urania representation. The remaining cations are so far being treated as ideal solution components, which will be adequate for the demonstration of fuel behavior in the current work.

Secondary phases will from in the urania fuel, although most only at relatively high burnups [21]. Phases that form at lower burnups include the white or noble metal phase. These are alloys of Rh, Ru, Mo, Pd, and Tc which can be present in the bcc, fcc, or hcp structures. Thermochemical models for these phases have been adopted from the work of Kaye, et al. [22].

A model for the perovskite phase formed from fission products and seen in high burnup oxide fuel was generated as a solid solution of BaO, SrO, Cs_2O, ZrO_2, MoO_2, RbO_2, and actinide dioxides. This model remains unoptimized, but the phase does appear in calculations of fuel at high burnup and is seen as adequately representative.

Other secondary oxide phases are observed at high burnups in fuel and are represented in the database. These include (Ba,Si)O, $(Rb,Cs)_2ZrO_3$, $(Sr,Ba)ZrO_3$, $(Rb,Cs)_2UO_4$, $(Sr,Ba)UO_4$, and$(Rb,Cs)MoO_4$.

ORIGEN Calculations

A series of calculations were performed using ORIGEN to obtain the average composition of PWR fuel undergoing burnup to 102 GWd/t over 5.75 years. A 14×14 fuel assembly lattice with 4.5–5% enriched uranium was used for depletion calculations; this configuration will be improved in future work but should provide reasonable estimates of the burnup-dependent elemental fuel composition for now. The initial O/U ratio was 2.

Thermochemical Modeling of Burnup

The elemental compositions provided from the burnup calculations performed in ORIGEN were used as input to thermochemical equilibrium calculations. The FactSage [23] software package was used for computing the equilibrium state utilizing the database described above. The equilibrium oxygen potential was determined, as defined by

$$= RT \ln (p_{O2})$$

where R is the ideal gas law constant, T is temperature, and p_{O2} is the oxygen partial pressure. For these calculations the total pressure was 1 bar and temperature was 1023K. The conditions were selected to match those of reported oxygen potential measurements on irradiated fuel samples to allow direct comparison.

The computed oxygen potentials together with reported measurements are shown in Fig. 1. The blue curve containing relatively more positive oxygen potential values are the result of the direct calculation of oxygen potential based on the composition of fuel at the specific burnup values. The results can be seen to be generally more positive than the reported measurements of Adamson, et al. [24], Une, et al. [25], Matzke [17], and Walker, et al. [26]. The calculations, however, do not take into account the observed oxidation of the inner zirconium clad surface and therefore the resultant loss of oxygen from the fuel. In order to represent the oxidation of the clad and therefore to better fit the measured fuel oxidation potentials, the equivalent of the amount of oxygen that would be lost to ZrO_2 was removed from the calculations. The surface area of clad directly adjacent to a fuel pellet was assumed to oxidize, and the depth of the oxide layer was assumed to increase from ~1 µm at 12.5 GWd/t to a steady state 6 µm at > 50GWd/t to fit the observed oxygen potential values. The internal cladding oxide layer thickness also agrees well with the typical observation of about a 6 µm layer reported by Walker, et al. [26]. The orange, more negative oxygen potential curve in Figure 1 is seen to well represent the limited reported measurements. Changes in slope and points of inflection are generally caused by the changes in phase assemblages computed with increasing fission product content.

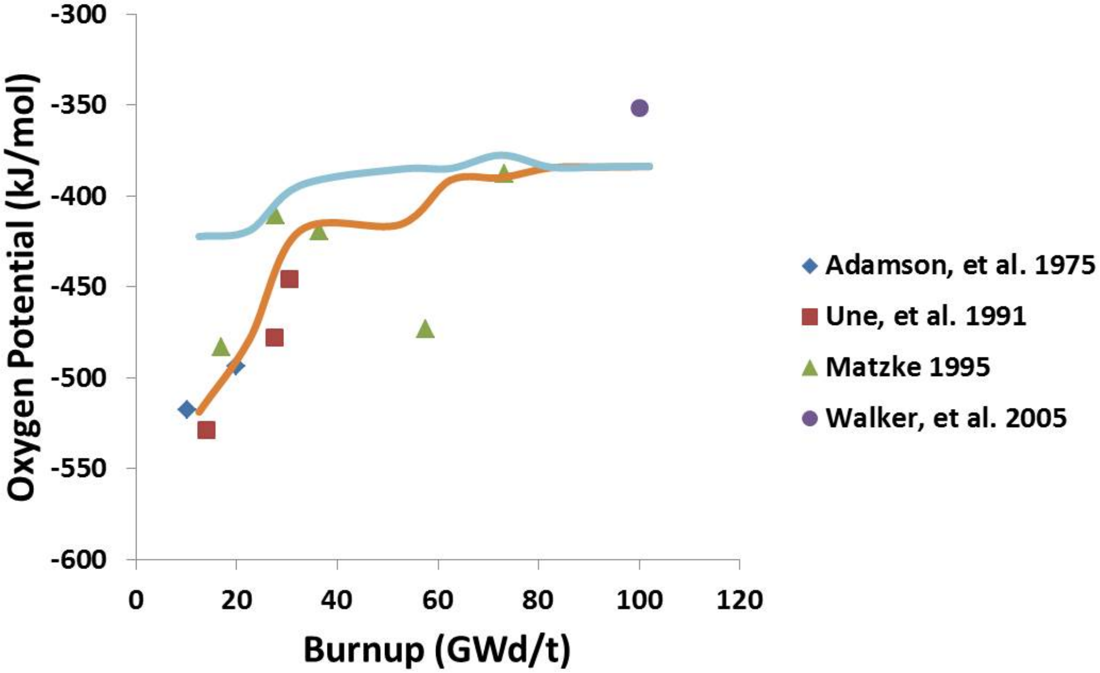

Figure 1. Computed oxygen potentials (lines) as a function of burnup for PWR fuel compared to reported measurements (points) [17, 24-26]. The blue curve represents direct equilibrium calculations from the ORIGEN results, whereas the orange curve includes oxygen loss due to clad oxidation.

THERMOCHIMICA Implementation of Oxygen Transport Model for Irradiated Fuel

The current functionality for modeling oxygen diffusion is based on the deviation of stoichiometry of the UO_2 solid solution phase. The calculation of the deviation is based on the O/M ratio of the UO_2 solid solution phase. The assumption is that a three sublattice CEF model is used to represent this phase. The first sublattice represents the cations on their normal sites, the second sublattice represents oxygen anions on their normal sites (or vacancies) and the third sublattice represents interstitial oxygen anions or vacancies. The calling format of the FORTRAN 90 subroutine is:

```
subroutine CompOtoMRatio(cPhaseIn, dOtoMRatio, INFO)

!> \param[in]  cPhaseIn   A character string representing the
name
!!                        of the phase in question.
!> \param[out] dOtoMRatio A double real scalar representing the
oxygen
!!                        to metal ratio.
!> \param[out] INFO       An integer scalar indicating
!!                        successful exit (0)
!!                        or an error (/= 0).
```

The developed subroutine was used to calculate the O/M ratio of the UO_2 solid solution phase for different stages of burnup using the composition obtained from ORIGEN simulations described above. The results are shown in Figure 2. The temperature and the pressure used in the calculations were 2000K and 1bar, respectively.

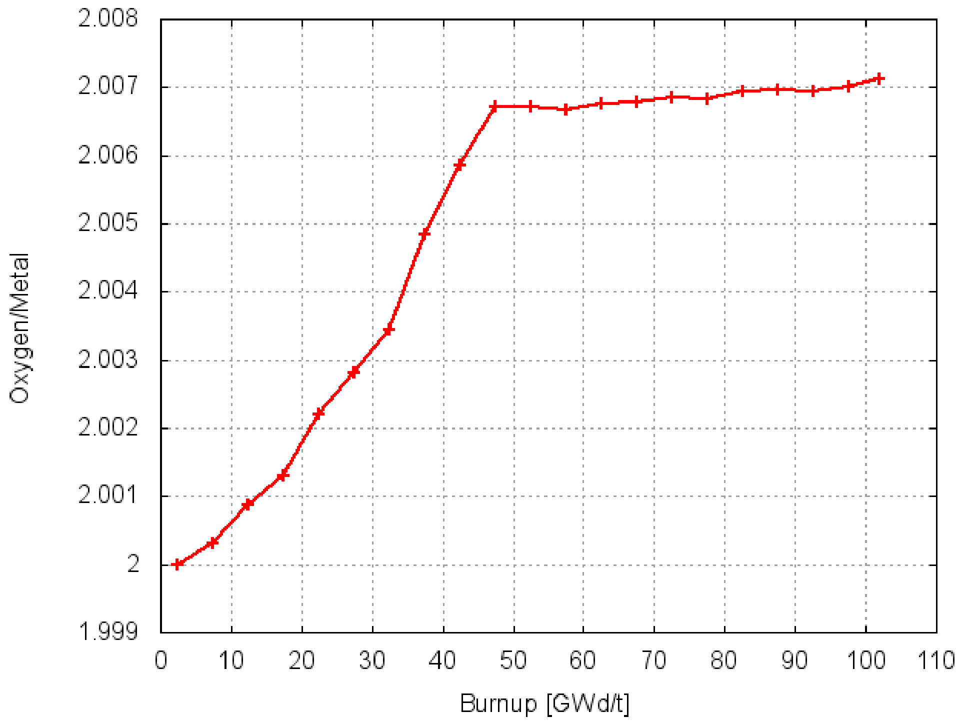

Figure 2. O/M ratio determined by CompOtoMRatio subroutine for the fuel compositions using ORIGEN simulations

Thermal and Oxygen Transport Models Implementation in BISON

A coupled thermal and transport model based on references [9-11] was developed. Various existing implementations for thermal and transport properties in MOOSE and BISON were utilized and extended. The model uses Fick’s law for thermal diffusion and combination of Fick and Soret diffusion for species transport. The module for calculating initial condition for deviation from stoichiometry in the UO_2 solid solution phase was developed using THERMOCHIMICA and its use is illustrated in the snippet of the input file of Figure 3.

```
[ICs]
  [./mat_1]
    block = 1
    type = UO2PXOxygenIc
    variable = oxygen
    temp = temp
    pressure = 1.0
    U = 3.6971E+03
    O = 8.4030E+03
    Pu = 5.7050E+01
    Ba = 3.5550E+01
    Ce = 5.4720E+01
    Cs = 5.7380E+01
    I = 5.3120E+00
    La = 2.5200E+01
    Mo = 1.0080E+02
    Nd = 7.8800E+01
    Pd = 6.7630E+01
    Pr = 2.1840E+01
    Rb = 9.3000E+00
    Rh = 7.5310E+00
    Ru = 8.9420E+01
    Sr = 2.1870E+01
    Tc = 1.8530E+01
    Te = 1.2290E+01
    Xe = 1.3150E+02
    Y = 1.1550E+01
    Zr = 9.8930E+01
  [../]
[]
```

Figure 3. Input example for defining the initial composition. The information is used for calculating thermodynamic equilibrium, oxygen/metal ratio. and deviation for stoichiometry. Pressure is in atmospheres.

In the above example, temperature is a coupled field denoted with `temp` whereas the variable `oxygen` denotes deviation from stoichiometry in the UO_2 solid solution phase. The elemental composition used corresponds to the highest burnup in Figure 2. For a simple test cube geometry and uniform temperature and pressure as above, an initial condition calculation of the deviation from stoichiometry is shown in Figure 4. The calculated value corresponds to x in UO_{2+x} as plotted in Figure 2.

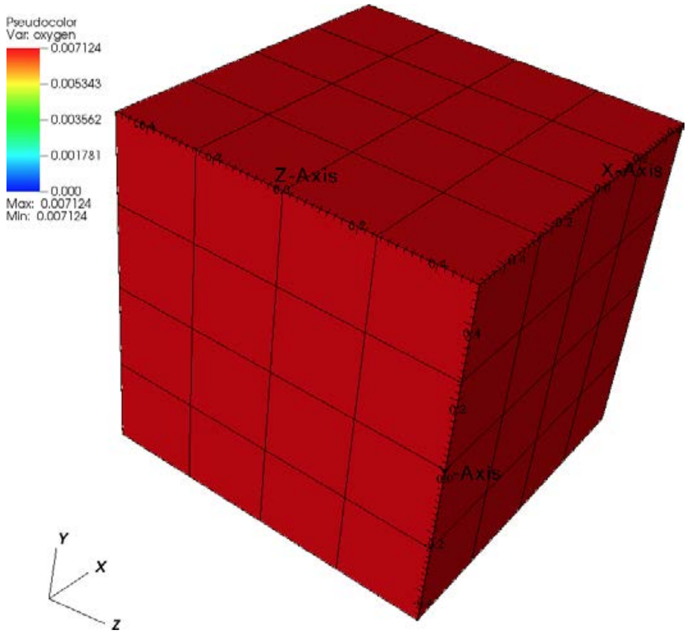

Figure 4. Initial condition calculation of the deviation from stoichiometry for the composition in Figure 3 and uniform temperature (2000K) and pressure (1 bar)

Example Problem for Oxygen Diffusion using BISON and THERMOCHIMICA Module

A simple example problem that uses thermal and oxygen transport models from the recent literature [9, 10] has been implemented in BISON. Material properties used in this model are functions of temperature and implicitly composition through the deviation of stoichiometry calculated from the O/M in the UO_2 solid solution phase. The problem is used for the illustrative purposes only and for the verification of the THERMOCHIMICA implementation. The result for the problem with imposed thermal gradient and initial composition is shown in Figure 5. The trends from the results correspond to the expected values from the composition.

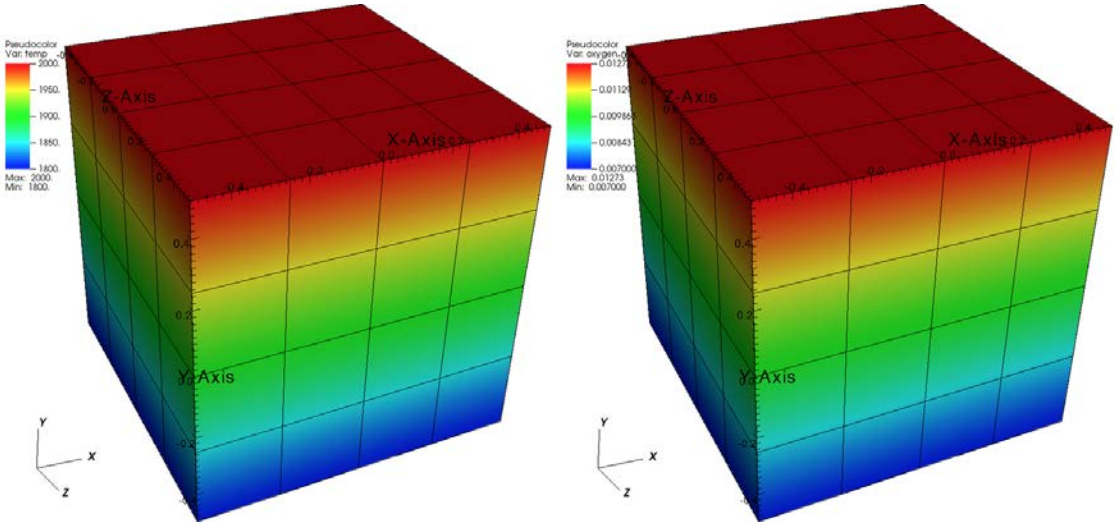

Figure 5. Temperature (left) and oxygen (right) distribution for a test problem with a temperature gradient in a cubic domain.

Summary

A thermochemical database for LWR fuel containing transuranics and fission products has been developed. The database was used in thermochemical calculations performed for average compositions representing specific burnup values. When limited clad oxidation is taken into account to adjust oxygen content, reasonable agreement is obtained between the calculations and reported measured oxygen potentials in irradiated fuel samples. An initial THERMOCHIMICA model for simulating chemical composition of irradiated LWR UO_2 fuel has been implemented in MOOSE and BISON. The model is used for the demonstration of the capabilities of the thermodynamics solver for simulation of the state of nuclear fuel. The initial implementation of the oxygen transport is based on a standard approach that does not take advantage of the thermodynamic solver information, such as chemical potential. A more rigorous model based on the chemical potential as the driving force for the diffusion is under development.

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