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John Wagner
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Dear Dr. Wagner,

Per your request, this is to inform you of the activities in the SNL UFD Work Package: FT-12SN081005 and the two related ones, INL UFD Work Package FT-12IN081001 and SNL NEAMS Work Package MS-12SN060507.

SNL UFD Work Package: **Modeling Hydride Reorientation in Cladding**
This UFD project is focused on the simulation of hydride precipitation and reorientation during long-term dry storage at the mesoscale. In FY12, this project will develop a framework for hydride precipitation and growth in realistic microstructures. This framework will allow incorporation of materials thermodynamics, physics and chemistry as they become available. The objective of this work is to develop predictive capabilities for hydride formation during long-term dry storage. The results will inform engineering scale, mechanics models as we believe that the hydride structure is one of the key features controlling reliability of the clad. This information from the mesoscale will enable true predictive capabilities in the continuum simulations of fuel assemblies.

The mesoscale, microstructural model will be a hybrid Potts-phase field model. It is well suited for simulation of diffusional phase transformations. The hydride precipitation and growth is thought to be a diffusional phase transformation. The precipitation and growth of the hydride occurs as the temperature decreases and solubility of hydrogen decreases. The model assumes that the hydrogen diffuses to regions where the hydride is thought to precipitate preferentially. These regions are grain boundaries and other defects such as dislocation loops. Once the hydride nucleates in these preferred regions, the growth is assumed to be diffusion controlled.

The input parameters for the hydride formation and growth model are the underlying cladding microstructure and composition, especially hydrogen content, and its form and location; free energies of the phases present; the mobility of the different components; temperature and stress profile. Based on these, a microstructure-based prediction of hydride formation will be simulated. Again as mentioned before, the framework for such a model will be developed in FY12. Adding detailed materials behavior and functionality will be done in future years.

The first attempt to simulate hydride precipitation only at grain boundaries is shown in Fig. 3. In this simulation, we assume that hydrides precipitate only at grain boundaries, not any other defect. At lower temperatures more hydride is formed (shown in darker red) with the matrix phase depleted of hydrogen (shown in dark blue). As temperature increases, there is less precipitation at the grain boundaries and more hydrogen remains dissolved in the grains (shown in light blue).

We are working closely with two other projects described below:

INL UFD Work Package **Zr-Alloy Thermodynamic Calculations**
Michael Glazoff of INL is an expert in computational thermodynamics, diffusion modeling and morphological analysis used for analysis of microstructures. He will be calculating phase equilibrium diagrams and free energy curves for the phases of interest for the standard Zr-alloy cladding compositions using Thermo-Calc. The generated free energy curves and their second derivatives (driving forces) will serve at the thermodynamic input to the hybrid model for hydride formation. Accurate thermodynamic data is essential for accurate prediction of hydride formation. An example of a binary Zr – H phase diagram computed using Thermo Calc is presented in Figure 1. It can be seen that it corresponds well to the existing experimental data shown in Fig. 2[1].

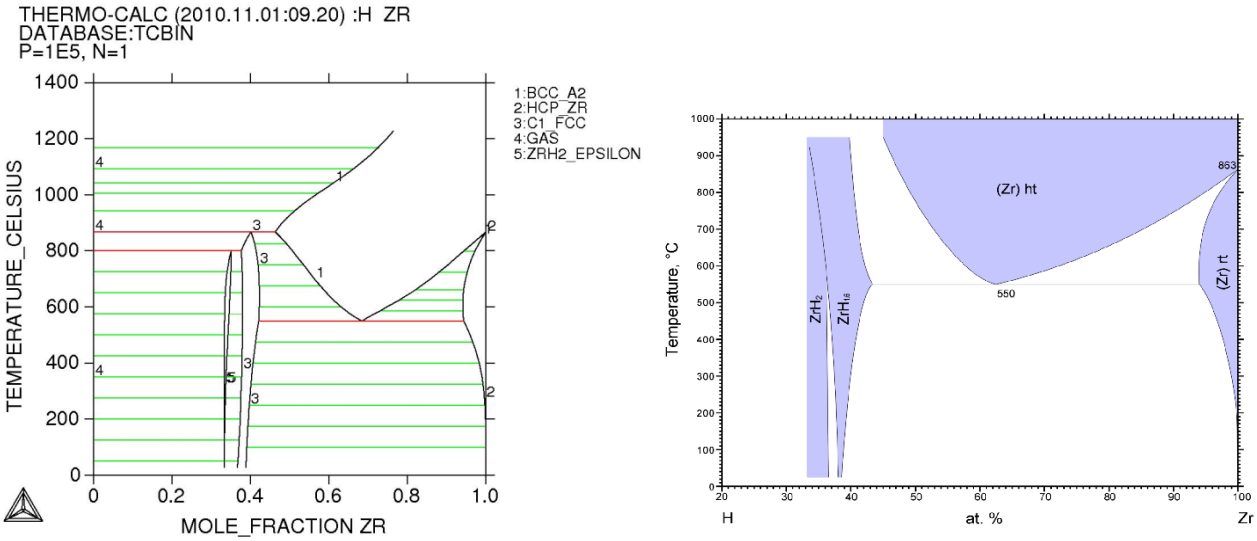

Fig. 1. The results of thermodynamic calculations using software package *Thermo-Calc*
Fig. 2. Experimental phase diagram constructed by Zuzek and Abriata[1]

However, preliminary calculations indicate that in Zircaloy-2 (composition: 1.5 wt.% Sn; 0.12 wt.% Fe; 0.10 wt.% Cr; 0.05 wt.% Ni; Zr) other phases might participate depending on the hydrogen concentration and temperature: ZrH_2 (ε-phase); $ZrH_{1.6}$ (δ-phase); Zr_4Sn; Zr_7FeSn_2; α–Zr; β–Zr; Laves phase C15; and Zr_2M (M=Fe, and Cr). This thermodynamic complexity clearly indicates that the real Zr cladding alloys and their thermodynamic properties should be studied, not just the Zr-H binary phase diagram. This project will focus on generating thermodynamic data initially for the five-component system, Zircaloy-2. In the future, other alloys will be characterized. Additionally in future years, morphological analysis of ZrH_x precipitates and their spatial distribution in the cladding microstructure

[1] Zuzek E.; Abriata J.P., National Nuclear Energy Commission, Argentina; San Martin A.; Manchester F.D., Toronto University, Toronto, Canada, in: Binary Alloy Phase diagrams, 2nd Ed., Edit. T.B.Massalsky (1990)

is proposed using the software *MorphoHawk* developed by Michael Glazoff over the last decade. This will inform the mesoscale simulations for preferred precipitation sites in the microstructure.

SNL NEAMS project, **Modeling & Simulation for Used Nuclear Fuel Very Long Term Storage.** Glen Hansen is developing a macro-scale hydride reorientation model that will be employed in the analysis of storage and transport processes to simulate cladding state and reliability at the fuel pin, fuel assembly and cask scale. The initial capabilities being developed are similar to the mixture theory model employed by EPRI (report 1015048), to allow benchmarking and comparison. It will then be extended to form a multiscale hydride model by directly linking to the meso-scale model to increase fidelity of prediction. The coupling of the meso- and macro-scale model will be done in two stages. In the first stage, the macro-scale will be decomposed into a set of representative volume elements (RVEs) that correspond to the different radial environments that exist within the cladding as shown in Fig 1. In this stage, we will pass the macro-scale cladding temperature as a function of time and spatial location to a set of meso-scale calculations to compute the meso-scale hydride morphology during the drying and storage periods, as shown in Fig. 3, below.

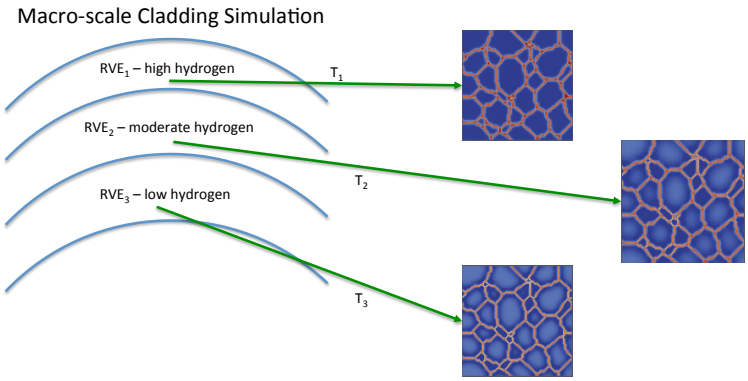

Fig. 3. Couple macro-scale temperature profile to meso-scale microstructural model

In stage 2 we will add stress and strain coupling from the macro- to the meso-scale, which is critical to predicting hydride formation and yield stress. Further, we will fully couple the transient meso-scale cladding strength and yield behavior back to the macro-scale calculation to calculate aged fuel response under transport and handling conditions. Uncertainty quantification is being embedded in the framework to enable uncertainty propagation across the scales spanned by the model. The stage 2 multiscale model is illustrated in Fig. 4. Note that a similar multiscale linking concept has been demonstrated for fuel performance[2].

[2] M R Tonks, G Hansen, D Gaston, C Permann, P Millett and D Wolf, "Fully-coupled engineering and mesoscale simulations of thermal conductivity in UO_2 fuel using an implicit multiscale approach," J. Phys.: Conf. Ser. **180**(1), 2009.

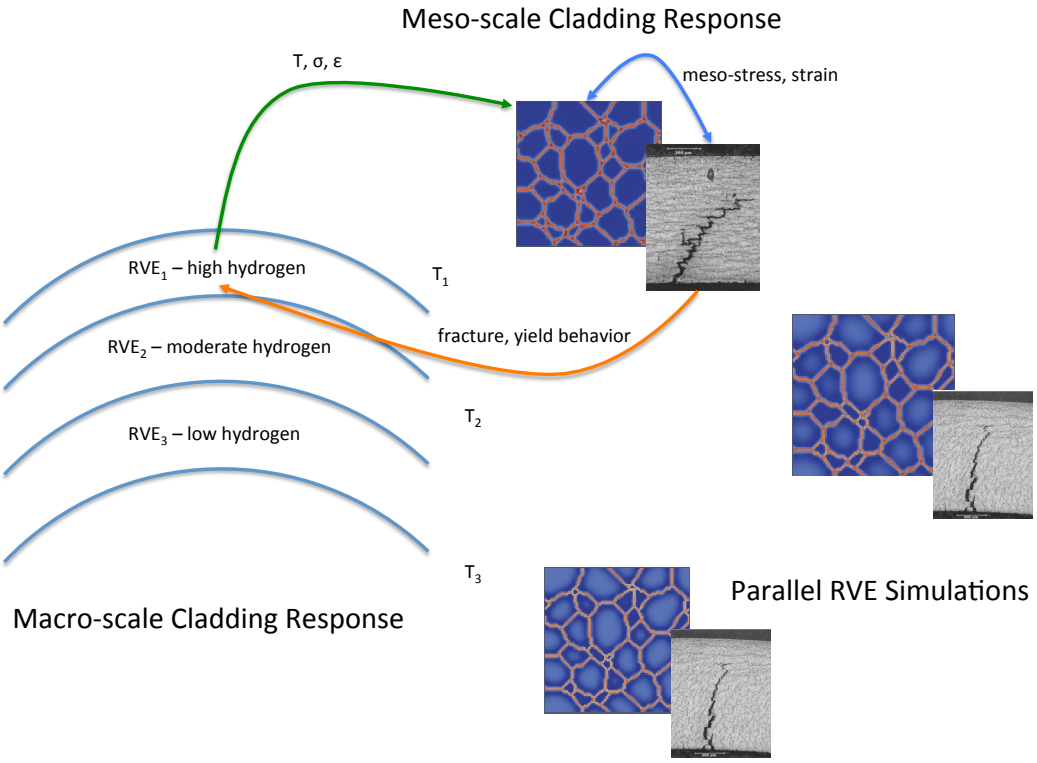

Fig. 4. Fully couple macro-scale response with the evolution of microstructure.

Please let us know, if you need more information or clarification.

Best regards,

Veena Tikare	Michael Glazoff	Glen Hansen
Computation materials science	Computational thermodynamics	Engineering mechanics
SNL	INL	SNL

cc: Ken Sorenson, SNL, MS 0747
Sandra Birk, INL