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FY17 end of year MARMOT materials model development update

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ABSTRACT

This document summarizes the lower length scale progress made in FY17 in developing materials models for fuel performance modeling of oxide fuels and their implementation in the MARMOT code. The progress covers the development of a phase field recrystallization model for high-burnup structure formation in oxide fuels, a phase field model for studying stage-3 fission gas release in UO_2, a phase-field model for irradiation-enhanced densification in UO_2, a quantitative model for hydride morphology in Zirconium based cladding, and the calculation of hydrogen diffusivity under stress in various Zirconium based alloys as cladding candidates. Further development in the MARMOT code has been carried out as well to improve the multi-phase, multi-component phase field model, the representation of interfacial energy in the Kim-Kim-Suzuki model, and the description of interfacial stress. For most of the above, separated reports have been generated with detailed description. Here a brief summary is provided, with references pointing to the detailed reports or publications to guide interested readers.

1. Introduction

A critical component needed in engineering-scale fuel performance modeling is the database of materials models describing the responses of fuels and claddings in the harsh reactor environments. In traditional fuel performance modeling, empirical materials models are usually used. Most times these models are fitted to experimental data and correlated with temperature and burn-up, which by itself is insufficient to represent the evolution in chemistry and microstructure in fuels. Other factors such as fission rate, fuel type, and initial microstructure can play important roles as well. The insufficient or inaccurate physics in the empirical correlations strongly limit the performance of these models for existing fuels in scenarios that have not been included in the fitting and for new fuel types. To address this issue, a microstructure based fuel performance modeling approach is taken by the Fuels Product Line of the Nuclear Energy Advanced Modeling and Simulation (NEAMS) program, targeting mechanistic materials models based on microstructure evolution in fuels in order to improve or replace the burn-up based models.

The details of the new approach have been documented in a recent publication [1] and previous report [2, 3]. In the past few years, this approach has been applied to oxide fuels [3], primarily UO_2, and U_3Si_2 [4], which is a promising concept for accident tolerant fuels [5]. Meanwhile, a mesoscale modeling tool, the MARMOT code [6], which is based on the MOOSE framework [7], has been created and is under active development [8] to explore the fuel behavior at the mesoscale scale and to assist the development of microstructure-based mechanistic materials models, which will be used in the engineering scale fuel performance modeling code BISON [9]. Each year, a new version of MARMOT is released with an assessment report [2, 10, 11], where the capabilities of MARMOT, guidance on code development, and information on code access and user support are given.

In FY17, the effort has been focused on the development of a phase field recrystallization model for high-burnup structure formation in oxide fuels, a phase field model for studying stage-3 fission gas release, a phase-field model for irradiation-enhanced densification in UO_2, a quantitative model for hydride morphology in Zirconium based claddings, and the calculation of hydrogen diffusivity under stress in various Zirconium based alloys as cladding candidates. Separated reports have been generated to describe the progress with appropriate amount of details [12-16]. Here, a brief summary is provided to guide interested readers for the overall accomplishments to guide interested readers.

2. Accomplishment highlight

2.1 MARMOT capability improvement [11]

The present capabilities of MARMOT have been assessed in a recent report [11] along with the release of MARMOT version 1.2. To improve the quantitative predictivity of MARMOT phase field models, new capabilities have been developed and implemented including the athermal ballistic mixing for irradiation enhanced resolution, the grand potential formalism for second phase formation, the decoupling of interfacial energy from elastic strain energy [17], and a novel description of interfacial stress. These improvements are important for using MARMOT as a predictive tool to obtain quantitative results on microstructure evolution and structure-property

correlations in fuels. Meanwhile, the robustness of the code has been improved by increasing the regression test coverage to 93% of the lines of code. A procedure has been established for external users to gain access and contribute to the code, and improvements in the documentation and simplifications of the input syntax have made the use of the code more convenient for new users.

2.2 High-burnup structure formation in UO_2 [12]

The high-burnup structure, HBS, forms at regions with high burnup in oxide fuels as well as metallic fuels. It features nanoscale grain sizes and high porosity with large gas bubbles [18]. As it forms, the local fuel properties change dramatically from coarse-grained fuels. Therefore, it is important to monitor the development of HBS and the consequent change in fuel properties. In FY16 and FY17, a phase field model for the formation of HBS was developed and implemented in MARMOT. The model treats the HBS formation as an irradiation-induced recrystallization, following one of the primary hypotheses proposed for HBS formation [18]. The model takes into consideration the effect of the stored energy associated with dislocations formed under irradiation. The accumulation of radiation damage, hence, increases the system free energy and triggers recrystallization. The increase in the free energy due to the formation of new grain boundaries is offset by the reduction in the free energy by creating dislocation-free grains at the expense of the deformed grains. This model can be generalized to a free energy based model to account for other possible mechanisms, such as fission products driving HBS formation. The model was first used to study the growth of recrystallized flat and circular grains. The model results were shown to agree well with theoretical predictions. The case of HBS formation in UO_2 was then investigated. It was found that a threshold dislocation density of (or equivalently a threshold burn-up of 33-40 GWd/t) is required for HBS formation at 1200K, which is in good agreement with theory and experiments. The present model is promising in providing a quantitative description of HBS formation. However, further studies are still needed to fully elucidate the mechanisms for HBS formation and the resulting property degradation. The planned studies include: 1) establishing a quantitative description of radiation damage and fission products with the purposes of elucidating an HBS formation mechanism and quantifying the driving forces, 2) constructing structure-property correlations for fuel properties in regions with HBS formation, and 3) developing materials models to inform the engineering scale fuel performance modeling tool BISON regarding the transient microstructure and properties.

2.3 Stage-3 fission gas release in UO_2 [13]

Fission gas release has potentially negative consequences for fuel performance in light water reactors. In classic fuel performance models for fission gas release, an empirical criterion is commonly used for the fission gas stored at grain boundaries to be released [19]. The importance of percolation of grain faces towards cracks or free surfaces is ignored. To gain an improved understanding of the processes leading to fission gas release, a phase-field model has been used to simulate the growth and interconnection of grain face and grain edge bubbles in UO_2 fuel. The phase-field model is based on a grand-potential functional and allows simulations of multiple grains of fuel and the bubble phase. The solid phase free energies are based on an ideal solution model, while the gas phase free energies are based on the Helmholtz free energy of a van der Waals gas. Parabolic approximations are used for the bulk free energy of each phase. A new approach is developed to include the hydrostatic pressure of the gas phase on the surrounding fuel matrix by adding a constant extra stress dependent on the hydrostatic pressure in the bubble.

Source terms are used to include the production of fission gas atoms and the net production of vacancies. This effort represents the most physical model of fission gas bubbles developed to date. The model is used to simulate the growth of grain face and grain edge bubbles in a hexagonally periodic geometry. The source strength representing the net vacancy production was varied parametrically since its value is unknown. The grain face bubbles have the expected lenticular shape, while the grain edge bubbles have an approximately triangular cross-section. Grain boundary bubbles that were near grain edges in the initial conditions grow preferentially toward the grain edges and thus become grain edge bubbles. The grain boundary coverage and trijunction coverage were calculated and plotted as a function of time. Although the grain boundary coverage increases rapidly at very early times, soon after, trijunction coverage begins to grow more rapidly than grain boundary coverage. At the end of the simulation time, for the higher vacancy source terms, the grain edges are completely percolated.

Ultimately, the objective of this work is to inform the fission gas release model used in BISON. Currently, BISON tracks grain boundary coverage, and allows all gas within a local volume element to be released when grain boundary coverage exceeds 0.5, which is an empirical criterion commonly used for grain boundary saturation. However, to release the gas stored at a saturated grain boundary to be released, it has to be connected to either the fuel surface or open cracks through a percolation path. This path is likely established via saturated trijunctions (or grain boundaries) connected with each other. The phase-field model developed in this work provides a means to explore if such a percolation path, which required trijunction saturation, is established before grain boundary saturation. Note that due to the 1D geometry of a trijunction, the critical bubble coverage of 1 is needed for saturation, in contrast to 0.5 for grain boundaries. In the current simulations, the trijunction coverage reaches 1 after grain boundary coverage reaches 0.5, meaning that no significant gas release, or trijunction percolation will occur from grain edges before release from grain boundaries. However, due to lack of data, the phase-field simulations in this work did not employ initial conditions with a physically justifiable number of grain edge bubbles. In future work, a physics-based estimate of the number of grain edge bubbles in the initial conditions will be developed. This will allow definitive conclusions to be made about the assumptions in the current fission gas release model.

2.4 UO_2 fuel densification [14]

Oxide fuels used in current commercial reactors are usually fabricated with an initial porosity measured by the sintering density. During reactor operation, pores existing within the fuel close, causing the fuel pellets to decrease in volume [20]. The volume change in turn effects the heat conduction, temperature profile, and performance of the fuel. The in-reactor densification is similar to the ceramic process of sintering, but it is sped up by fission products. This project seeks to develop a mesoscale phase-field model for densification in MARMOT. To begin, a simplified model of sintering without irradiation effects is developed. From the initial model, it is determined that we can simulate sintering without using the rigid-body motion model. Rather, the standard phase-field equations should be sufficient to capture sintering. In the future, this model will be extended to include irradiation effects. Method to accurately determine simulation densities in both 2D and 3D will be developed. Finally, an engineering scale model will be developed and implemented in the NEAMS fuel performance code BISON [9].

2.5 Hydrogen diffusion in Zirconium alloys [15]

The presence of hydrogen (H) can detrimentally affect the mechanical properties of many metals and alloys, including the Zirconium based alloys used for nuclear fuel claddings. To mitigate these detrimental effects a fundamental understanding of the thermodynamics and kinetics governing H pickup and hydride formation in these alloys is required. As the rate-limiting factor for hydride evolution and delayed hydride cracking, H diffusion in Zr-based alloys can be affected by alloying elements and stress, factors that have been shown to strongly affect H pickup and hydride formation in nuclear fuel claddings. To elucidate these effects, a recently developed accelerated kinetic Monte Carlo method [21] is used to quantify H diffusivities in these alloys under mechanical loading. Accurate predictions of H diffusivities have been obtained from the simulations, with analytical models derived for the usage in engineering scale modeling [22]. It is found that H diffusivity depends only weakly on composition, with negligible effect at high temperatures. In contrast, stress strongly affects H diffusivity. This effect needs to be considered for studying hydride formation and delayed hydride cracking. We note that during operation, other factors may also affect H diffusion strongly including the production of radiation damage and the incorporation of oxygen due to oxidation. These factors will be investigated in the future.

2.6 Hydride morphology in Zirconium alloys [16]

Zirconium alloys are current choices for the cladding of UO_2 fuel in Light Water Reactors for their low neutron absorption cross section, good thermal conductivity and good resistance to corrosion. However, as claddings directly face high temperature coolant water, oxidation occurs and produces H, which infiltrates into the cladding matrix. Due to the low solubility of H in α-Zr, precipitation of H into hydride phases takes place. These hydride phases, primarily δ-$ZrH_{1.66}$, are brittle, leading to embrittlement of the cladding. More importantly, the hydride phases may cause delayed hydride cracking during used fuel storage. Therefore, it is important to understand hydride formation and evolution, and the consequent degradation in cladding fracture toughness. For this purpose, a multi-phase field model and a grand potential model coupled with elasticity have been developed in MOOSE, for simulations of hydride morphology. These models have been verified by comparing the results to analytical solutions in simple cases, and were then applied to the α-Zr/δ-$ZrH_{1.66}$ system. Different elastic models were implemented, and this study has confirmed the fact that the Khachaturian mechanical property interpolation (KHS) elastic model provides more accurate results than the Voigt-Taylor elastic energy interpolation (VTS) model. The inherent decoupling of the bulk free energy from the interfacial energy in the grand potential model while retaining a low computational complexity is a distinct advantage over other phase field models. The interface thicknesses can be defined larger than in other models, allowing the use of coarser meshes in simulations and leading to lower computational cost and faster results. This could allow for observing clear orientation of the hydride along the basal plane, which was difficult before due to the computational costs of simulations when using the multi-phase field model. Future work should focus on adding plasticity to the phase field models coupled with elasticity to obtain quantitative results for the α-Zr/δ-$ZrH_{1.66}$ system that could be compared with experimental results. Simulations of hydride evolution and predictions of mechanical properties in hydrided systems are also planned.

3. Summary and discussion

In addition to the above highlighted accomplishments, progress has also been made on grain growth in UO_2 [23], center hole formation in oxide fuels for fast reactors [24], and anisotropic interfacial energies in UO_2. We note that the development of a microstructure based mechanistic fuel performance modeling approach holds a significant potential to provide a physical and predictive fuel performance modeling. On the other hand challenges intrinsic to the multiscale modeling approach adopted remain as the simulations span many time and length scales. This area is under active research and development, with impacts already emerging on selected topics. The success of this approach in oxide fuel is being extended to silicide and metallic fuels, structural materials and well beyond nuclear related materials. In the previous years, the research has been focused on oxide fuels. In FY18, effort will also be made in modeling the material behavior of metallic fuels such as U/Zr and U/Pu/Zr fuels. More specifically, we will focus on fracture, fission gas release and high burnup structure in oxide fuels, and swelling in both silicide and metallic fuels. As some mesoscale capabilities will be ready to be merged into the engineering scale in a short term, including grain growth and fracture in oxide fuels, special attention will be paid to integrate the lower length scale models into BISON, as well as performing experimental validation.

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