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# Nuclear Energy Advanced Modeling and Simulation (NEAMS) Accident Tolerant Fuels High Impact Problem: Coordinate Multiscale FeCrAl Modeling

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# Introduction

Since the events at the Fukushima-Daiichi nuclear power plant in March 2011 significant research has unfolded at national laboratories, universities and other institutions into alternative materials that have potential enhanced accident tolerance when compared to traditional $UO_2$ fuel zircaloy clad fuel rods. One of the potential replacement claddings are iron-chromium-alunimum (FeCrAl) alloys due to their increased oxidation resistance [1–4] and higher strength [1, 2]. While the oxidation characteristics of FeCrAl are a benefit for accident tolerance, the thermal neutron absorption cross section of FeCrAl is about ten times that of Zircaloy. This neutronic penalty necessitates thinner cladding. This allows for slightly larger pellets to give the same cold gap width in the rod. However, the slight increase in pellet diameter is not sufficient to compensate for the neutronic penalty and enriching the fuel beyond the current 5% limit appears to be necessary [5]. Current estimates indicate that this neutronic penalty will impose an increase in fuel cost of 15-35% [1, 2]. In addition to the neutronic disadvantage, it is anticipated that tritium release to the coolant will be larger because the permeability of hydrogen in FeCrAl is about 100 times higher than in Zircaloy [6]. Also, radiation-induced hardening and embrittlement of FeCrAl need to be fully characterized experimentally [7]. Due to the aggressive development schedule for inserting some of the potential materials into lead test assemblies or rods by 2022 [8] multiscale multiphysics modeling approaches have been used to provide insight into these the use of FeCrAl as a cladding material.

The purpose of this letter report is to highlight the multiscale modeling effort for iron-chromium-alunimum (FeCrAl) cladding alloys as part of the Nuclear Energy Advanced Modeling and Simulation (NEAMS) program through its Accident Tolerant Fuel (ATF) High Impact Problem (HIP). The approach taken throughout the HIP is to utilize lower length scale approaches (e.g., density functional theory, cluster dynamics, rate theory, phase field, and Visco-Plastic-Self-Consistent (VPSC)) to develop more physically informed models at the engineering scale for use in the BISON [9] fuel performance code.

# The Multiscale Approach for Predicting FeCrAl Behavior

The majority of lower length scale work has been focused on understanding the creep behavior, irradiation effects, and burst behavior under normal operating and accident conditions. The lower length scale model to be incorporated into the BISON code is the VPSC model under development at Los Alamos National Laboratory (LANL). VPSC in and of itself is a separate computer code which simulates the plastic deformation of polycrystalline aggregates [10]. The code can be applied to materials of various lattice structures including hexagonal (e.g., Zircaloy) and cubic (FeCrAl). The algorithm utilizes constitutive laws to connect the lower length scale grain and dislocation behaviors to the macroscale stress behavior. VPSC is able to provide mesoscale informed predictions of macroscopic parameters such as creep, hardening, and burst (failure).

Before coupling of the VPSC to large scale BISON simulations, work by other collaborators at Idaho National Laboratory (INL), LANL, and the University of Tennessee Knoxville is ongoing to inform the constitutive laws used by VPSC for irradiation hardening and creep. This work includes the following:

Mesoscale modeling efforts focused on using a cluster dynamics based reaction diffusion model of radiation induced defect cluster evolution in Fe-Cr-Al alloys under prototypic neutron irradiation conditions are able to predict the evolution of prismatic dislocation loops with Burgers vectors of a<100> and a/2<111>, as well as the next flux of point defects to network dislocations that induce climb. The modeling predictions qualitatively capture the observed neutron irradiation trends with respect to irradiation temperature, but quantitatively the model predicts too many, too small <a/2>111 loops, and too few, but too large a<100> loops. One possibility for this discrepancy may be related to a change in elastic displacement fields surrounding the essentially immobile, larger a<100> loops, which changes with time as a result of the migration of interstitial atoms to the loop periphery that shields the surrounding stress fields and effectively reduces the dislocation bias. Preliminary modeling with time dependent bias of the dislocation loops shifts the quantitative predictions closer in comparison to the experimental results, and future efforts will encompass atomistic modeling to provide a self-consistent foundation for the cluster dynamics modeling. Ultimately, the cluster dynamics model will inform the self-consistent viscoplastic constitutive modeling of irradiation hardening and creep of FeCrAl accident tolerant fuel cladding.

A crystal plasticity based constitutive model was developed in order to simultaneously predict the mechanical response and microstructure evolution of FeCrAl during loading at temperature (e.g. monotonic., biaxial, creep, relaxation etc.). The constitutive relation was further developed with the intent to be able to capture the major effects of chemistry on mechanical properties and on the activation of dissipative processes (dislocation glide and its relationship with solute diffusion, dislocation climb and its relationship with vacancy diffusion). Further, developments will aim at accurately predicting the evolution of damage; the model proposed already introduces a quantification of type III stresses, due to the stress field inherent to the presence of dislocations, and of their evolution during plasticity. The constitutive law was successfully fitted against experimental data from ORNL. The model was further introduced in Vulture. Vulture was used in combination with the MOOSE package in order to predict the performance of FeCrAl based clad during Loss of Coolant Accident Scenario. Further, the predicted performance (i.e. dimensional changes as a function of temperature ramp, time etc.) were compared to those obtained in the case of Zircaloy-4 clad such as to obtain a prediction of the safety gained from the use of FeCrAl. As the current constitutive models are not sensitive to irradiation history, a series of lower length scale simulations, based on the use of Discrete Dislocation Dynamics were performed. The intent is to quantify the role of Cr precipitate formation, resulting from neutron bombardment, on the resistance to dislocation glide.

An atomic kinetic Monte-Carlo (AKMC) model for ternary Fe-Cr-Al alloys has been developed to simulate Cr precipitation, which may occur in high Cr alloys ($>$12% Cr) during thermal aging and under irradiation. This model describes the interaction between Fe, Cr and Al atoms using pairwise bonds. The bond energies are fitted to DFT results on the diffusion barriers with various compositions. Concentration dependency is introduced for Fe-Cr bonds by fitting to the Cr-Cr interaction within the 5th nearest neighbor distance. The model can predict the short range ordering of Cr solid solution when the Cr concentration is low, and the precipitation of $\alpha$' phase when the Cr concentration is high. The addition of Al is found to delay $\alpha$' phase precipitation kinetics, because of the strong binding between Al atoms and vacancies. Effectively, vacancies are trapped around Al atoms, reducing the diffusion of Cr atoms. The Al atoms have a stronger tendency to be mixed with Fe than with Cr atoms. As a result, the Al concentration in the $\alpha$' phase is much lower than that in the Fe matrix. The precipitation information obtained from the AKMC simulations will be used to access the possible hardening and embrittlement in FeCrAl alloys. This can be done by coupling the AKMC model with either continuum scale theories or with the crystal plasticity model.

As the constitutive laws are finalized, a meeting was held in mid June between the current LANL VPSC developers and select BISON developers from the INL to coordinate efforts of utilizing the VPSC code in large scale BISON simulations of FeCrAl cladding. Currently, based upon the meeting discussions, the VPSC code in Vulture can be coupled to BISON. However, convergence is extremely slow. Two paths forward have been identified to improve the predictive capabilities of BISON predictions of FeCrAl cladding using VPSC. First, improve robustness by examining other alternatives to solvers available in PETSc that work well with both Vulture and BISON. Unfortunately due to the need to account for mechanical contact in BISON simulations, the amount of robust solvers available may be limited. Secondly, reduced order models (ROMs) that represent the constitutive laws available in the VPSC code could be added to BISON for increased computational efficiency. The purpose of ROMs is to replace the complex mathematical models for the given process by a much simpler mathematical representation that still captures the certain aspects of the process. This is done by running many standalone VPSC calculations to develop or train the reduced order representation of a particular constitutive law. As an example, a computationally intensive creep constitutive law may be able to be reduced to a higher order polynomial expression. From a computational perspective, polynomials are extremely efficient to evaluate and would increase the speed at which a coupled BISON/VPSC model could be completed. However, depending on the case, the number of simulations required to develop the ROM may be prohibitive. As these ROMs become available they will be incorporated directly into BISON and bypass the need of coupling to Vulture entirely.

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