MBM UFD Development Plan

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Veena Tikare of Sandia National Laboratories has provided essential guidance regarding the SPPARKS-MBM coupling. In addition, she contributed signficantly to an interim report [1]. Much of the description in this report of SPPARKS and of the test problems comes from the interim report. Her assistance with the coupling and with the report is gratefully acknowledged.

1 Introduction

Following irradiation in reactor, spent fuel is moved to wet storage where decay heat can be easily removed. After the decay heat is acceptably low, spent fuel is dried and placed in storage casks. Given that the U.S. does not have a spent nuclear fuel facility, used fuel will remain above ground in casks for the foreseeable future. It is desirable that the fuel rods maintain structural integrity during during handling, transportation, and eventual retrieval.

Perhaps the principal concern with respect to spent fuel rods is hydride precipitation and embrittlement. In reactor, fuel rod cladding oxidizes with an associated release of H_2. Some of this hydrogen enters into the zirconium alloy. At high temperatures, the hydrogen is primarily in solution but will precipitate as ZrH_x at lower temperatures. With significant hydrides, and depending on the hydride orientation, the ductility and fracture toughness of the cladding can change significantly.

The orientation of these hydrides has been shown to depend on the stress state in the cladding. In particular, hydrides primarily form perpendicular to the dominant stress field in the cladding. As temperatures drop following irradiation, hydrides form in a circumferential direction. During drying, temperatures are high, and the hydrogen undergoes dissolution. Following drying, lower temperatures and the changed stress state (strong hoop stress due to internal gas pressure) result in radial hydride formation. Radial hydrides are of concern because they significantly weaken cladding, including increasing susceptibility to crack formation and propagation.

Computational modeling of spent fuel is attractive due to both the expense of testing irradiated materials and the long time frames (decades) involved in storing spent fuel. The Used Fuel Disposition Campaign Gap Analysis report [2] identifies understanding the effect of hydrides as a high-priority research issue.

2 UFD Analysis Requirements

This chapter gives a brief description of the capabilities required in order to model spent fuel and assess its structural integrity.

2.1 Irradiation History

The starting point for used fuel analysis is the state of the fuel following irradiation. Modeling the irradiation history enables accurately characterizing the cladding, possibly including elastic and inelastic strains, stress, fluence, oxidation, and hydrogen content.

2.2 Wet Storage and Drying

The environment the used fuel experiences after irradiation directly impacts the formation of radial hydrides. Modeling this environment allows evaluation of a particular history as well as enabling a design loop in which optimal temperature change rates and hold periods may be determined.

2.3 Hydrogen Uptake

Hydrogen uptake occurs during irradiation through the oxidation of zirconium and water. Hydrogen is a byproduct of this reaction, some of which enters the cladding. Modeling hydrogen uptake is necessary to understand the distribution of hydrogen at the end of irradiation.

2.4 Hydrogen Diffusion

Hydrogen diffuses through the cladding due to both Fickian and Soret diffusion. Modeling diffusion is of course also important in order to know the hydrogen distribution in the cladding.

2.5 Hydrogen Precipitation/Dissolution

Hydrogen in solution will precipitate once the concentration reaches the terminal solid solubility. Likewise, hydrogen will return to solution if the concentration reaches the terminal solid solubility limit for dissolution. These changes are clearly important to model.

Details concerning hydrogen uptake, diffusion, precipitation, and dissolution may be found in [3].

2.6 Hydride Orientation

Due to the fact that the strength of hydrided cladding depends on the hydride orientation, computational models must predict orientation. Perhaps the least research has occurred to date on this issue.

2.7 Hydride Damage

The purpose of the previously-named analysis requirements is to provide an accurate description of the cladding, including hydrides, to a model that will give information about the structural integrity of the cladding. The model or models chosen will take into account the original cladding strength as well as the embrittlement effect of hydrides. Possible approaches include homogenizing grain-level model results to compute macro-level material properties for use in macro-level analysis, developing an empirical threshold beyond which failure is anticipated, and developing a damage model (see, for example, [4]).

3 Current Capabilities

Current capabilities may be grouped into three areas: 1) coupling SPPARKS and MBM, 2) modeling hydrogen uptake, diffusion, precipitation, and dissolution, and 3) modeling the history of a fuel rod from beginning of life through long-term storage.

3.1 Coupling SPPARKS and MBM

The Used Fuel Disposition (UFD) program has initiated a project to develop a hydride formation modeling tool using a hybrid Potts-phase field approach. The Potts model is incorporated in the SPPARKS code from Sandia National Laboratories. The phase field model is provided through MARMOT from Idaho National Laboratory. MARMOT is based on MOOSE, as is BISON. MOOSE, BISON, and MARMOT are sometimes referred to as MBM.

3.1.1 SPPARKS and the Potts model [1]

SPPARKS, an acronym for stochastic parallel particle kinetic simulator, is a Sandia-developed open-source code that can simulate many microstructural evolution processes. SPPARKS is a kinetic Monte Carlo (KMC) code that has algorithms for both rejection-free KMC and rejection KMC, which is sometimes called Metropolis Monte Carlo [5].

SPPARKS is distributed as an open source code under a GNU-like license. It is highly versatile, supports a number of different applications, can be extended to add new functionalities and is able to run in serial or in parallel. The parallel version uses the message passing interface (MPI) to perform concurrent computations over all processors while minimizing communication overhead between processors. To accomplish this, the space of 3D particles (domain) is partitioned between the processors, and Monte Carlo dynamics is calculated concurrently in each processor. Note that 1-D and 2-D simulation domains are similarly partitioned by SPPARKS. Information about border sites whose neighborhood could belong to other processors is kept locally in ghost sites. While each processor can change the state or spin of its owned sites, it cannot change the state or spin of the ghost sites; it can only accesses the information contained at the ghost sites. Communication between processors is used to update the state of the ghost sites. Communication is also required to collect some of the statistical information that can be computed while simulating the evolution of the system at hand.

3.1.2 MOOSE-BISON-MARMOT (MBM)

Idaho National Laboratory is developing a modern computational simulation framework called MOOSE. MOOSE is based on the finite element method, runs on a single CPU or on a massively parallel computer, and leverages software packages that provide, among other things, advanced

numerical solvers. MOOSE itself is not built to solve any particular physical problem but provides interfaces that allow applications to be built with relative ease. These applications are tailored for a particular field of interest. Two examples are MARMOT, a meso-scale modeling application, and BISON, a nuclear fuel performance analysis tool.

Since 2008, Idaho National Laboratory (INL) has been developing a next-generation nuclear fuel modeling capability known as BISON. BISON is a massively parallel, finite element-based software product that solves the coupled nonlinear partial differential equations associated with nuclear fuel performance. BISON supports the use of one-, two-, and three-dimensional meshes and uses implicit time integration, important for the widely varied time scales in nuclear fuel simulation. An object-oriented architecture is employed which minimizes the programming required to add new material and behavior models.

BISON is written in a general way such that it may be applied to a variety of fuels and geometries. Though primarily used for LWR fuel analysis, BISON has been used to analyze TRISO-coated particle fuel and metal fuel in rod and plate form, design and interpret irradiation experiments, and investigate novel fuel concepts.

MARMOT is a multiphysics mesoscale code focused on modeling irradiation-induced microstructure evolution in reactor materials. It is based on the MOOSE framework and uses the basic phase field classes that are built into the phase field module in MOOSE (MOOSE-PF). The phase field method is used to model the microstructure evolution and it is coupled to finite deformation mechanics and heat conduction to capture the multiphysics nature of the microstructure evolution. The impact of the evolution on the material properties is quantified using advanced homogenization approaches to take the local material properties at the mesoscale and determine a single effective property over the entire mesoscale domain. The phase field method is used in MARMOT due to its flexibility in modeling many coupled categories of microstructure evolution. In the phase field model, microstructural features are represented by continuous variables, which are evolved to minimize a function describing the free energy of the system. In MARMOT, the phase field model has been directly applied to model radiation damage in UO2 and zircaloy cladding. The phase field model is coupled to linear elasticity to model the impact of stress on the fuel behavior, and to finite strain mechanics to model the impact of deformation in zirconium alloys. The phase field models are also regularly coupled to heat conduction, where the local thermal conductivity varies locally across the microstructure.

3.1.3 Verification and Code Comparison

Verification is the process of demonstrating that the implementation of a mathematical model in computer code is correct. This is done through tests of the software. Tests are developed with well-defined inputs such that the expected output can be determined independently of the software. If the software computes the expected output for a given test, that correct result is evidence that the model has been implemented correctly. Tests with analytic solutions are good verification tests.

Code comparisons may add value if an analytic solution to a given problem is not known.

This section outlines a set of verification tests and code comparison tests for the coupled SPPARKS/MBM tool. These tests are found in elk/tests/spparks/ and marmot/tests/TwoComp/ within the MBM repository.

Do-nothing test

The foundational verification exercise is to execute a SPPARKS model from within MBM and check that the results match those computed by a stand-alone SPPARKS run.

Check transfer to MARMOT

Another worthwhile test is one in which the computed values from SPPARKS are transferred to and written out by MARMOT. If done correctly, the output from MARMOT should match that the native output from SPPARKS.

Check transfer to SPPARKS

A similar test is on in which data from MARMOT is sent to SPPARKS. The data in SPPARKS should match that in MARMOT.

The following verification tests are based on examples from [6].

Equilibrium of binary, two-phase interface

Characterize the equilibrium configuration of a binary, two-phase interface and compare to the SPPARKS solution. Allow an α-grain shaped like a rectangle or cube (in 3D) to come to equilibrium with the same-shaped β-grain so that they share a flat interface. The starting composition for both grains is the equilibrium composition of that phase. When the composition across the interface no longer changes, characterize the interface structure and composition change. See Section 3.1 in the paper.

Diffusion kinetics of binary, two-phase diffusion couple

Characterize the diffusion kinetics of a binary, two-phase diffusion couple and compare to SPPARKS solution and to analytical solution. Form a diffusion couple of α-grain shaped like a rectangle or cube (in 3D) with the same-shaped β-grain so that they share a flat interface. The composition of both is to be off-set from equilibrium so that the total volume of the two phases does not change. (This is done by choosing the compositions of the two phase, so that the lengths from the overall composition to the free energy of the two phases does not change as the composition of the two phases change.) Allow the two components to diffuse across the interface and compare the diffusion kinetic results to the analytic solution for this configuration. See Section 3.2 in the paper.

3.2 Modeling Hydrogen

Modeling hydrogen in Ziracloy is the subject of [3]. Many details of BISON's hydrogen modeling capability may be found there. Further work has occurred this year to revamp the hydride modeling in BISON, focusing on improving the quality of the implementation and removing issues with time stepping. As part of this work, the small time step restriction imposed by the original hydride model was removed. The upgrade enables large, multidimensional simulations

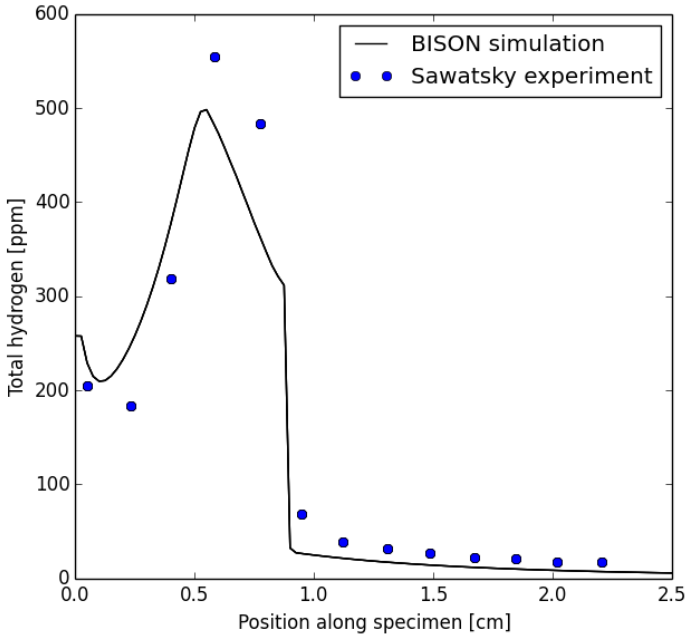

Figure 3.1: Validation of BISON hydride model against Sawatzky's 1D hydride redistribution experiment [7].

to be run efficiently while still capturing hydrogen pickup, diffusion, precipitation, and dissolution.

In addition to verification cases, the hydride model has recently been validated against [7]. In this experiment, an initially uniform hydrogen distribution is redistributed in one dimension using a thermal gradient so that some hydrogen forms zirconium hydride at one end of the domain. Modeling this in BISON tests the Fickian diffusion, Soret diffusion, precipitation, and dissolution kernels (i.e. everything except for hydrogen pickup from the cladding). As shown in Figure 3.1, BISON predicts the major features of the experiment.

We have also begun work to simulate UFD scenarios that include hydride formation, beginning with high-burnup irradiation, continuing with cooling in the spent pool, and finishing with drying and long-term storage in casks. The model currently includes separate thermo-mechanical and precipitation models, which eases time stepping restrictions that the fully-coupled case would impose.

Thermal gradients in cladding lead to higher hydrogen concentrations at the outer cladding radius and in the vicinity of pellet-pellet interfaces. This effect can be seen in Figure 3.2.

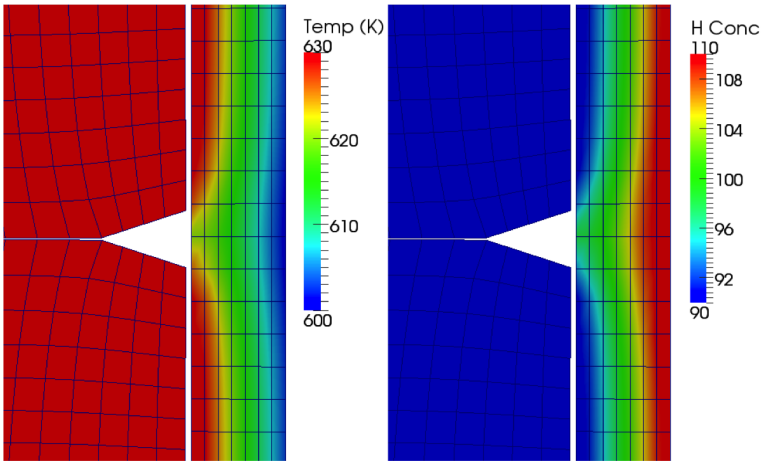

Figure 3.2: Temperature and hydrogen concentration in cladding near a pellet-pellet interface.

3.3 Model Fuel Rod Life

BISON is well-suited to model nuclear fuel throughout its lifetime. Recent additions to BISON include a decay heat capability, necessary for modeling fuel rod life after irradiation. Figure 3.3 shows the temperature history of a demonstration rodlet throughout irradiation, time in the spent fuel pool, drying, and dry storage. Figure 3.4 shows the stress and strain history for the same rodlet over the same time.

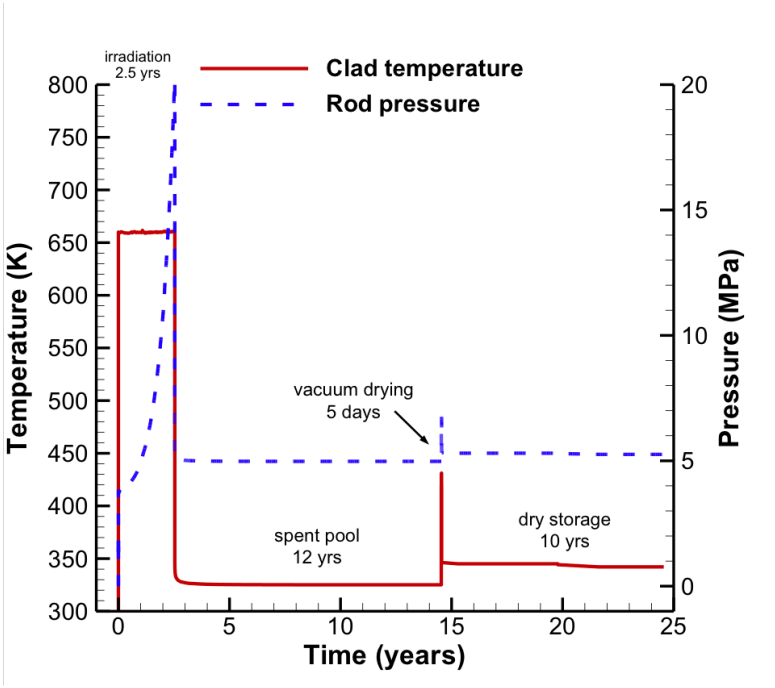

Figure 3.3: Temperature of a rodlet throughout its life.

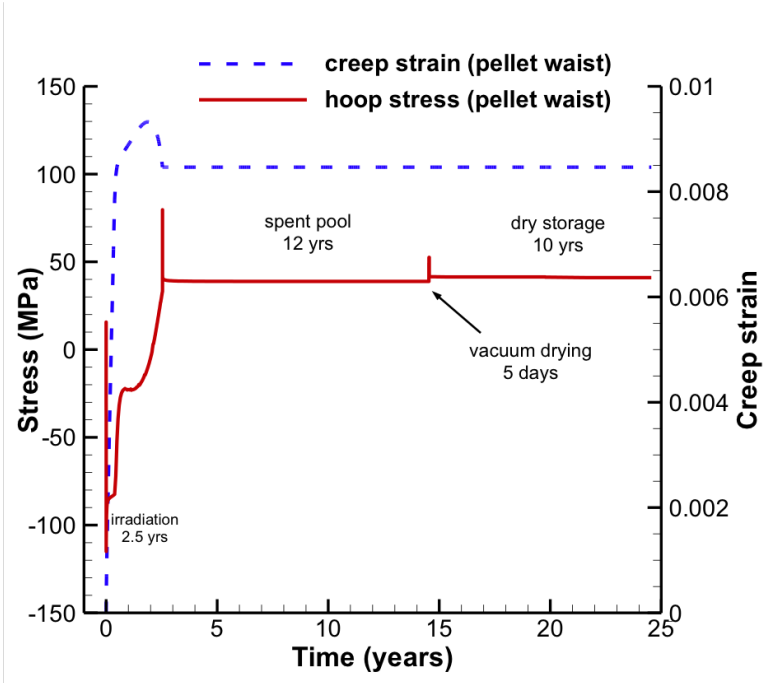

Figure 3.4: Stress and strain of a rodlet throughout its life.

4 Future Development

Future development to enable structural integrity calculations of spent fuel will encompass the following areas.

4.1 Hydride Diffusion

Much work in this area has already been completed. However, benefit would be gained by investigating ways to improve the performance of this capability. In particular, time stepping approaches and solver robustness are areas for further development.

4.2 Hydride Orientation

Knowing the orientation of hydrides is critical in calculating hydride embrittlement. Understanding hydride formation is particularly important following drying when hydrogen in solution re-precipitates and is susceptible to forming in the radial direction.

At least two possibilities exist for obtaining an orientation model. The first is from the work of Veena Tikare and collaborators. If an orientation model is available in SPPARKS, perhaps it may be used in conjunction with BISON.

The second is from the ongoing work in MARMOT. Mike Tonks and others, through INL LDRD funding, are investigating hydride formation and hope to develop an understanding of hydride orientation.

If these or other research efforts are successful, it may be possible to compute hydride orientation rather than assuming a certain fraction of radial hydrides. Such a capability would be a step toward a predictive capability.

4.3 Hydride Damage

As stated previously, calculating hydride damage is the purpose for the cladding hydride work. This capability will certainly benefit from lower length scale development. The central issue is appropriately processing the grain-level information.

Three possible approaches are reviewed below.

4.3.1 Homogenization

One approach to getting an engineering-scale measure of hydrided cladding integrity is through homogenizing lower length scale properties. This approach computes macro-scale properties such as elastic moduli using lower length scale data. With the macro-scale properties in hand,

finite element analysis of the cladding will result in stress and strain predictions which may be used to assess the integrity of the cladding.

One homogenization technique has been developed in MBM, and a paper describing the approach is under review. It would be straightforward to employ this approach within MBM for thermal conductivity and elastic moduli.

4.3.2 Empirical Threshold

A second approach to assessing structural integrity would be to develop an empirical threshold. In this approach, some measure of acceptable cladding as a function of hydride concentration, hydride orientation, temperature, and possibly other parameters would be developed. With a measure of acceptable cladding available, simulation would compute the inputs to the threshold function. That is, simulation would compute hydride concentration, hydride orientation, etc., and these would be used by the empirical function to compute a measure of acceptability. If the output of the function were beyond a critical value, the cladding would be considered failed or otherwise unsound.

4.3.3 Damage Model

A third approach is to develop or implement a constitutive model for cladding that includes the effects of hydrides. This would be a damage model, a model that computes lower strength with increasing damage (hydrides). A hydride damage model would require as input things such as hydride orientation. Hydride damage models already exist [4], and some work has been done to implement such a model.

4.4 Recommendations

Which of these approaches is best is an open question. The homogenization approach is fairly well developed for elastic moduli. However, the approach has never been applied to a realistic cladding with hydrides. The empirical threshold would be simple to use but would require considerable development and would not be predictive. The damage model would also require development and would complicate the engineering-scale analysis.

Given these challenges, it may be appropriate to perform scoping studies in attempt to see which holds the most promise. A moderate effort in two or all three areas over a few months would provide for an informed decision regarding which to pursue further.

Regardless of which approach is taken, fundamental lower length scale development is required. In one form or another, these approaches need information about the hydrides, and obtaining that information through modeling necessitates further lower length scale work.

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