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Three-dimensional multi-physics modeling of hydrogen and hydride distribution in zirconium alloy cladding

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ABSTRACT

Localized phenomena within the reactor core, specifically those associated with the nuclear fuel, require high-fidelity simulations to enable accurate physics predictions. One example is the zirconium cladding, which absorbs hydrogen from the light water coolant during normal reactor operation. Absorbed in the cladding, this hydrogen is in solid solution and its distribution is sensitive to temperature and concentration gradients. At high enough concentrations, the hydrogen will precipitate as a hydride. Thus, the hydrogen distribution as a hydride precipitate in cladding has been identified as an important safety concern, and a possible ersatz for validating reactor simulation code temperature models. This study reports development efforts of using high-fidelity multi-physics codes to model temperature, hydrogen, and hydride distributions in three dimensions under realistic operating conditions. The Consortium for the Advanced Simulation of Light Water Reactors multi-physics code, Tiamat, is used to model selected sub-assemblies. Then, a single fuel pin is selected from the sub-assembly and modeled as a three-dimensional BISON problem. The outer cladding temperatures from the Tiamat calculation are used as boundary conditions for the BISON problem in order to obtain hydrogen and hydride distributions. Areas of interest for hydride precipitation include locations along the fuel rod experiencing highest temperatures with significant spatial variation, particularly in the vicinity of the spacer grids and mixing vanes.

1. Introduction

Zirconium is a refractory metal having excellent corrosion resistance, low thermal neutron cross section, and good heat resistance. Zirconium alloys have higher thermal properties than other refractory alloys and high thermal conductivity than stainless steel alloys. High corrosion resistance of these alloys is due to the protection of a metal-oxide interface by a tightly adhered protective oxide film. Standard fabrication techniques are used to manufacture the alloys. Because of these properties zirconium alloys are utilized as cladding material for nuclear fuel. One of these alloys used for fuel cladding material in Pressurized Water Reactors (PWRs) is Zircaloy-4 (Alloy Zr4) (UNS R60804), which contains in its chemical composition 97.56–98.27% zirconium (Zr).

Nuclear fuel undergoes various changes during reactor operation including cladding oxidation and hydride precipitation. At high temperatures, the zirconium alloy cladding oxidizes and leads to hydrogen production in the following process:

$$Zr + 2H_2O \rightarrow ZrO_2 + 2H_2.$$

During this process, hydrogen can enter the zirconium-alloy of the fuel cladding, and under proper conditions, precipitate as brittle hydride platelets which can severely impact cladding ductility and fracture toughness. Hydrogen tends to migrate to and precipitate at colder locations. Because high local hydride concentrations increase the risk of cladding failure, it is important to predict the local hydrogen distribution. The absorbed hydrogen redistributes within the cladding based on temperature, concentration, and stress gradients. Hydrogen migrates down temperature gradients and, at a high enough concentration, it precipitates as a hydride, which may embrittle the cladding. Due to the sensitivity to temperature gradients, hydride distribution in cladding can be used as an ersatz for temperature model validation (Courty, 2013). The new US Nuclear Regulatory Commission (NRC) requirements on hydrogen level in fuel cladding are described in 10 CFR 50.46c - Acceptance criteria for emergency core cooling systems for light-water nuclear power reactors (Federal Register, 2015) and serve as motivation for this study.

Hydrogen transport depends on different phenomena. Even though migration can only occur when the hydrogen is in solid solution, the cladding temperatures during operating condition allow a portion of

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hydrogen to be in solid solution. Therefore, as hydrogen is picked up during the corrosion reaction between the cladding and the coolant, it can migrate following Fick's law and the Soret effect. Another phenomenon that affects hydrogen transport is stress gradients; however, previous studies showed that it is usually negligible compared to Fick's law and the Soret effect (Couet et al., 2014). Once the local hydrogen content reaches the terminal solid solubility for precipitation, the hydrogen will start precipitating as a zirconium hydride phase. With hydrogen diffusing in the cladding, it might at certain locations exceed its terminal solid solubility of precipitation (TSSp) (Kearns, 1972). In that case, some of the hydrogen might precipitate as zirconium hydride. The TSSp is modeled by the following equation:

$$TSSp = 138746e^{-\frac{4145.7}{T}}.$$

The precipitated hydride can dissolve back to the solution if the concentration of hydrogen in the solid solution drops to a lower limit known as the terminal solid solubility of dissolution (TSSd):

$$TSSd = 106446.7e^{-\frac{4328.67}{T}}.$$

The effect of both factors as a function of temperature can be seen in Fig. 1 (Mankosa, 2015). It is shown in Fig. 1 that there is a difference between solubility of hydrogen in case of dissolution and precipitation. This hysteresis effect is because the precipitation occurs by a nucleation and growth process. It is necessary to create an initial nucleus for the precipitate to grow.

The dynamics of hydrogen in zirconium alloy cladding has been implemented in the BISON fuel performance code (Courty et al., 2014). The code is able to model hydrogen production, hydrogen absorption, hydrogen redistribution, hydride precipitation, and hydride dissolution in the cladding as a function of concentration and temperature. A first attempt to benchmark this model has been made by comparing the results given by BISON to the hydrogen distribution measured in a nuclear fuel rod, which had undergone an exposure of five cycles (Lacroix, 2016). This was feasible because of the very detailed information about that reactor and fuel pin were available, and a very good agreement was found between the calculations and what was observed experimentally. Based on this validation studies the hydrogen model, implemented in BISON, was revised in the part related to the initial hydride precipitation kinetics. A closer agreement with the experimental data, described above, was obtained with the revised model.

In summary, one of the factors that might affect the integrity of the fuel cladding is the formation of hydride precipitates. The high velocity of the water used for cooling of Light Water Reactors (LWRs) is a highly corrosive environment to the zirconium cladding. This corrosive action involves the oxidization of the cladding accompanied by the release of hydrogen. A fraction of this released hydrogen can be absorbed by the cladding to form a solid solution of hydrogen. The hydrogen tends to diffuse from high concentration regions to lower ones (Fick's law) and from higher temperature regions to lower temperature regions (Soret effect). At certain concentrations and temperatures, the hydrogen may form localized hydride precipitates that might embrittle the cladding locally. This makes the cladding vulnerable to cracking and its integrity might be compromised. At a specific axial height and azimuthal angle, the local hydride concentration might be very high forming a weak point in the cladding. Therefore, a three-dimensional (3D model) is needed to be able to quantify this local effect.

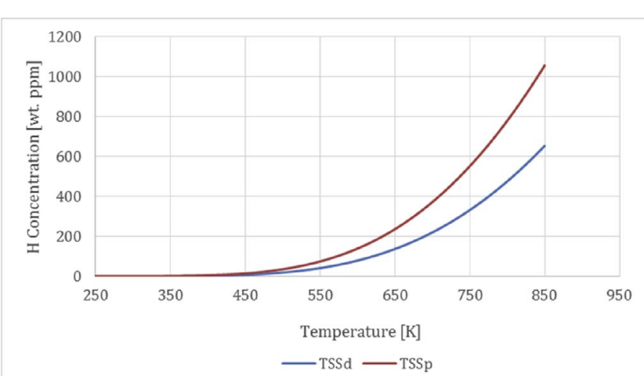

Fig. 1. TSSp and TSSd concentrations as a function of temperature (Mankosa, 2015).

The purpose of this study is to examine the 3D hydrogen and hydride distribution in a fuel pin in the complex flow region around a spacer grid using high-fidelity multi-physics simulations. Several codes are used to achieve this, including the Tiamat multi-physics framework, and standalone BISON fuel performance code with implemented and validated revised hydrogen model. The following section describes each simulation tool in the context of this investigation.

2. Multi-physics simulation framework

BISON is a 3D finite element fuel performance code developed by the Idaho National Laboratory (INL), which is built within the MOOSE (Multiphysics Object Oriented Simulation Environment) framework. BISON predicts temperature distributions, fission product swelling, densification, thermal and irradiation creep, mechanical properties, fission gas production, and distribution of hydrogen and hydrides in cladding (Hales et al., 2015).

Tiamat, a multi-physics code coupling that was developed within the Consortium for Advanced Simulation of Light Water Reactors (CASL) program, is used to model various sub-assemblies for this project. Tiamat couples a thermal hydraulics subchannel code, a neutronics transport code, and a fuel performance code (Pawlowski et al., 2015). This multi-physics coupling allows feedback from each code in order to calculate an accurate temperature distribution, which is required for hydrogen prediction. The thermal hydraulics portion of the coupling is the Coolant Boiling in Rod Arrays – Two Fluid (CTF) which is a Reactor Dynamics and Fuel Modeling Group (RDFMG) maintained subchannel code. CTF is currently being utilized in the Department of Energy (DOE) CASL project for high-fidelity core thermal-hydraulic calculations (Avramova, 2016). The neutronics solver is a University of Michigan code called Michigan Parallel Characteristics based Transport (MPACT). MPACT uses a two-dimensional (2D) method of characteristics (MOC) approach to solve the neutron flux in the radial plane and a one-dimensional (1D) diffusion method to solve in the axial direction (Kochunas et al., 2014). The third leg of Tiamat is BISON, which is utilized for temperature distributions and fuel parameters. The Tiamat code passes fluid temperature and density from CTF to MPACT. Fuel rod temperature is passed from BISON to MPACT, which passes back power. CTF passes cladding temperature to BISON, which gives heat flux to CTF in return. A basic summary of the coupling is shown in Fig. 2.

3. Modeling methodology

The modeling approach used in this research could be summarized as follows. The CASL coupled multi-physics code, Tiamat, was used to model a 3D sub-assembly with spacer grids. From this Tiamat full length 4 × 4 sub-assembly model an area of interest near a spacer grid was selected and used to create a section of single fuel pin (node) 3D BISON model. Three-dimensional BISON calculation of the selected node was performed using Tiamat boundary conditions to obtain three-dimensional hydrogen and hydride distributions at areas around spacer grids.

Tiamat was first used to model a 16-pin sub-assembly, in order to achieve detailed power distributions (from MPACT) and sub-channel flow parameters (from CTF) with feedback between codes and from the fuel performance code BISON. The coupling simulated a full-length 3D, 4 × 4 sub-assembly including fuel, plenum, and spacer grids. The problem specifications can be seen in Table 1. The sub-assembly is modeled with varying enrichment and a guide tube in order to create an