VALIDATION OF BISON CALCULATION OF HYDROGEN DISTRIBUTION BY COMPARISON TO EXPERIMENT

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Abstract

During normal operation in nuclear reactors, the nuclear fuel cladding corrodes as a result of exposure to high temperature cooling water. 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 spots. Because high local hydride concentrations increase the risk of cladding failure, it is important to predict the local hydrogen distribution. To that end, a hydrogen transport model has been implemented in the 3D fuel performance code BISON. In this study, we present an initial attempt of using this model for benchmarking the BISON code as applied to a case of the hydrogen distribution measured in a nuclear fuel rod, which had undergone a five cycles exposure. The prediction of hydrogen distribution show good agreement with the post irradiation measurement, indicating the promise of this benchmarking method.

Introduction

In the core of a Light Water Reactor (LWR), the fuel pellets (UO_2) are contained in fuel rods. The nuclear fuel cladding tube prevents fission products from reaching the primary loop water. This tube is made of Zr-based alloys, because of their low neutron absorption cross-section (only 0.185 barns for thermal neutrons), superior resistance to high temperature corrosion and good mechanical properties [6]. However different phenomena can put the clad integrity into jeopardy. One of the main limiting factors to cladding performance is the ingress of hydrogen and consequent hydride embrittlement.

During corrosion, zirconium reacts with the oxygen present in the water. A fraction of the hydrogen generated by the corrosion reaction (i.e. the hydrogen pickup fraction) is absorbed by the cladding. Once in the cladding, the hydrogen either dissolves in solid solution in the interstitial sites of the $\alpha-$Zr matrix or precipitates in the form of zirconium hydrides (ZrH_x).During mechanical deformation of the cladding tube, those hydrides can fail at low strain thus decreasing cladding ductility [7]. Hydrogen in solid solution is mobile in the zirconium matrix and responds to concentration gradients (following Fick's Law) and temperature gradient (Soret Effect) as described in [1].

During normal operation, temperature gradients may exist in the cladding in the three directions (radial, axial and azimuthal). Radial gradients exist during operation because of the heat flux passing through the cladding. Axial temperature gradients occur not only due to the gradual heating of the coolant along the axis such that at the higher grid spans corrosion is higher but also at the inter-pellet gaps where a local decrease of temperature is observed, which causes a higher hydrogen concentration. Finally, azimuthal variations can also be observed for various reasons such as: (i) a power gradient, caused for example by a pin having a control rod on one side and a highly enriched pin on the other [2] or (ii) oxide spallation causing a differential cooling around the cladding circumference. The resulting temperature gradients may create an inhomogeneous repartition of the hydrogen in the cladding.

Because the terminal solid solubilities for hydride dissolution increases with temperature, hydrogen tends to precipitate at the cooler spots near the outer rim. Therefore, a higher concentration of hydrogen at the cladding rim is observed at high burnup (the hydride rim). The hydride rim is located at the outer edge of the cladding and shows a very high hydrogen concentration, going up to 6,000 wt.ppm, as observed by Zhang [3].

The previous discussion has illustrated the importance of a tool that can predict hydrogen transport and precipitation, and that is benchmarked to reactor data. This study is an initial attempt to perform such a benchmarking study.

Experimental Data

Gravelines' Fuel Pin Data

The data used in this article was used in a previous study by J.-H. Zhang [3] and comes from the rods irradiated at the Gravelines nuclear power and which were and subjected to post irradiation examination. The characteristics of the pin studied are shown in Table I.

The interest of studying this pin was that it has reached a high burnup (about 60 GWd/tU) and was very well characterized in terms of hydrogen content. This study also included a hot vacuum extraction analysis to obtain the overall hydrogen content.

Table I: Studied fuel pin characteristics

Clad Material	Zircaloy-4	Number of cycles	5
Fuel	UO_2	Enrichment (%^{5}U)	4.5
Z position (mm)	3250	Burnup (MWd/tU)	58 230
Outside diameter (mm)	9.49	Inside diameter (mm)	8.36
Pressure (bars)	155	Mass Flux ($\mathrm{g\,cm^{-2}\,s^{-1}}$)	314
Inlet Enthalpy ($\mathrm{J\,g^{-1}}$)	1 264	Inlet Temperature (°C)	286
Outlet Temperature (°C)	323		

Measurements

Measurements of the hydrogen content were performed using image analysis of micrographs taken from a cross-section of the fuel cladding, which had been etched to reveal the hydrides. This analysis allowed to have a radial and azimuthal experimental distribution of the hydrogen in the cladding. The experimental data consists of four radial data points in four different azimuthal zones, defined by a rotation of 90° on the tube, which makes a total of sixteen data points for this pin. After abrasion of 9 µm off the outer cladding surface, it was possible to determine that the hydrogen content in that part of the cladding was 6 000 wt.ppm, which gives one more data point.

Data from the reactor operating conditions were also given. With these parameters, it is straightforward to compute the cladding outside temperature at the given z coordinate (given in Table I.) This allowed to calculate the hydrogen distribution with BISON and to compare it to the experimental data (Table II).

Table II: *Hydrogen experimental distribution determined by Image Analysis, quantities are given in wt.ppm*

Radial position µm	left	top	right	bottom	Average [H] per layer
499	685	713	726	745	717 ± 80
356	430	469	537	490	482 ± 80
214	195	142	244	296	219 ± 80
71	206	173	169	129	169 ± 80
Average	379	374	419	415	392 ± 40

The hydrogen distribution measurements made by J.-H. Zhang [3] have an accuracy of ± 80 wt.ppm, which creates a significant difference in overall hydrogen content: 392 wt.ppm is found by image analysis when 541 wt.ppm is found by hot vacuum extraction (HVE). A difference between Image analysis and the HVE measurements should be observed because the hydrogen content in this sample is very high and the software see overlapping hydrides and therefore calculates a hydrogen concentration below the real one. To address this issue, it is assumed that the missing hydrogen content is not seen in the first layer (position 499 µm). Therefore, since the hydrogen content found by HVE is 541 wt. ppm. and since Zhang found a hydrogen rim of 6000 wt. ppm. on a 9 µm thickness, it is possible to calculate the hydrogen content that is actually present in this layer. The value found is 861 wt.ppm.

BISON Simulations And Results

Assumptions And Introduction To The Subject

A few parameters are missing from the data set used in this work. For example, the environmental conditions or cladding outside temperature distribution were not given, and it was not possible to retrieve the position of the fuel pin in the assembly, and thus, it was difficult to determine the temperature gradient experienced by this pin. As a first approach it was decided not to implement an azimuthal temperature gradient, and ton only focus on the radial hydrogen distribution: the experimental data was averaged over the azimuth (average per layer) which thus gave an average radial distribution over the whole azimuth.

In the study, no data of the neighboring fuel pins was provided, it was hence assumed that those pins do not influence the pin studied other than providing neutrons and heating the coolant. Furthermore, it has been considered that the influence of the stress gradient in the cladding on hydrogen migration is negligible compared to the influence of a temperature gradient by the Soret effect and a concentration gradient described by Fick's law, as shown by Puls in 2002 [9].

Problem Description, And Input Parameters

The hydrogen concentration in solid solution gradually increases in the cladding during reactor exposure due to hydrogen pickup until reaching the terminal solid solubility for precipitation ($\mathrm{TSS_p}$) in colder spots. This leads to a complex picture of hydrogen migration, hydride dissolution and precipitation, which can be predicted by the hydrogen model in BISON. In this study, the operating conditions used are those used in the calculations made by J.-H. Zhang [3] based on the data given in Table I.

For Zircaloy-4, it has been reported that 10-20% of the hydrogen produced during the corrosion reaction is picked up by the cladding [5]. The value of 15% has been chosen here as an average value while noting that recent studies have shown the hydrogen pickup fraction to be variable over the exposure time. A reactor shutdown schedule needs to be included in the simulation since the observations were made at room temperature, and most hydrides precipitate during shutdown and the specific cooling rate should affect the hydride distribution seen at low temperature [1].

It is also considered that after the beginning of the shutdown, the fuel pin delivers 7% of the power it was delivering during its cycle at Hot Full Power, and slowly decreases following the equation (1):

$$\frac{P}{P_0} = 0.066\left((t-t_0)^{-0.2} - t^{-0.2}\right) \tag{1}$$

Where P_0 is the reactor power before shutdown, t the time elapsed since reactor startup, t_0 the time of reactor shutdown and P the power at the considered time t as shown by A.R. Knief [8]. During the same time, the bulk temperature slowly cools down by steps, according to the reactor shutdown procedure given by J.-H. Zhang [3]. The power distribution and average clad outside temperature distribution are shown in figure 1 while figure 2 shows the outside clad temperature of the pin as a function of time during shutdown. With these input parameters it is possible to calculate the hydrogen content absorbed by the cladding and its distribution within the cladding, and compare it to the experimental data, as shown in figure 2 (right graph).

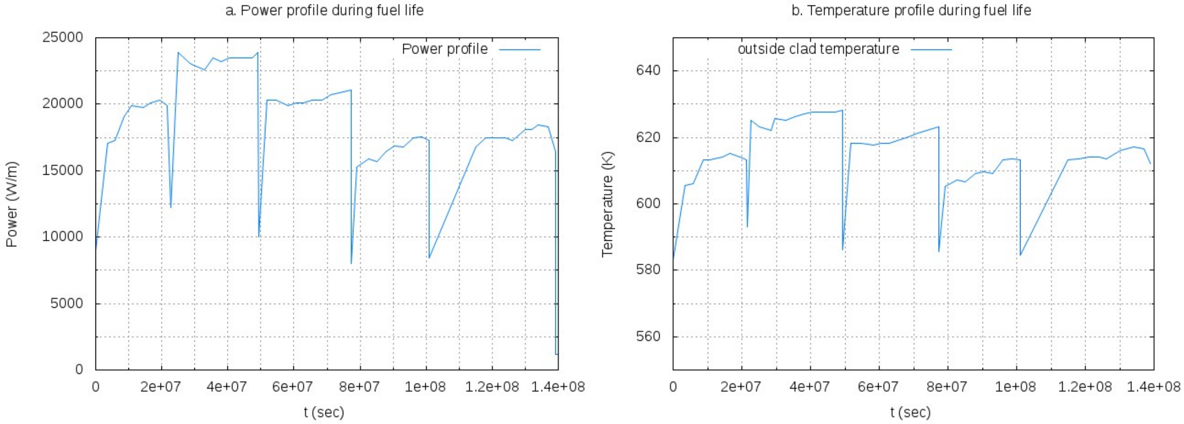

Figure 1: *linear power at the axial location (a.) and outside clad Temperature (b.) variation of the pin 1079 with respect to time*

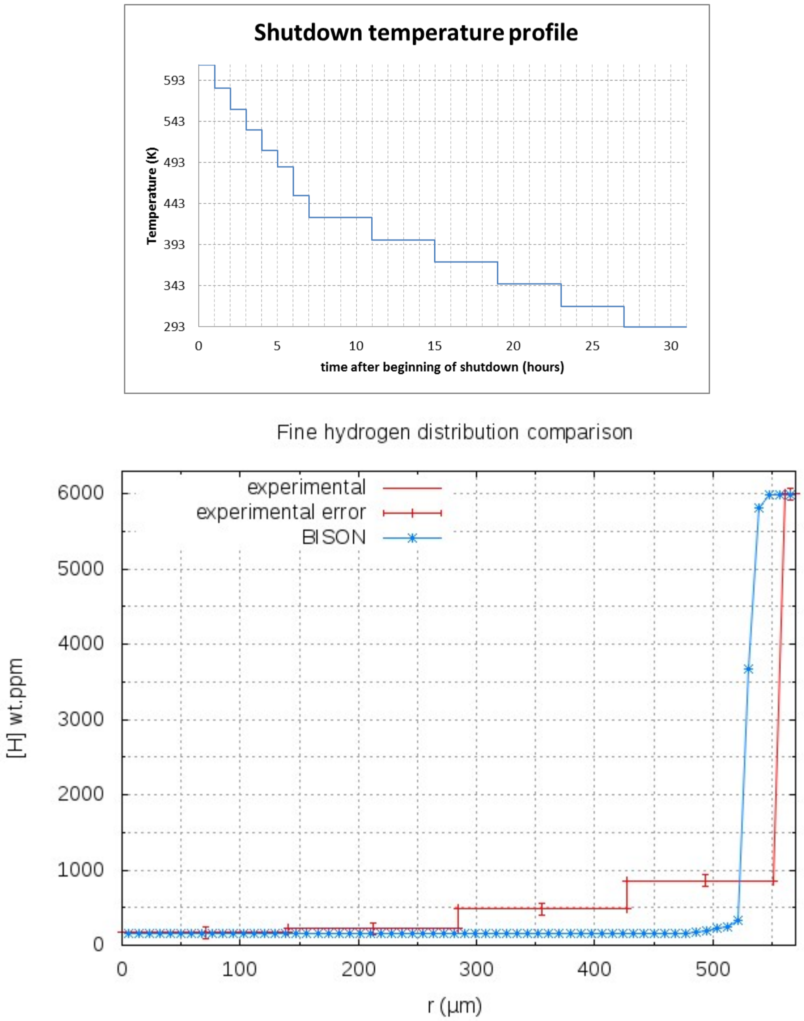

Figure 2: *Shutdown temperature profile of the pin 1079 (left) BISON caluclation results (right)*

As seen in fig. 2, the hydrogen distribution given here is not very accurate. Even though the total hydrogen content closely corresponds to the experimental measurement (542.5 versus 541), all of the hydrogen is calculated to precipitate near the metal/coolant interface. This can be explained by a numerical feature of BISON: there is no nucleation, germination and growth model of the zirconium hydrides implemented in the hydrogen model used in BISON. Therefore, the hydrogen follows the Soret effect and precipitates as soon as it enters the cladding, if the hydrogen content is higher than the $\mathrm{TSS_p}$. This suggests that a nucleation model is needed to better describe hydride precipitation, but a work around is described in the next section.

Fine Mesh Description of Hydrogen Distribution

Experimental data consisted of only five points, i.e. the regular four image analysis points and the hydrogen content in the hydride rim. In this section, results obtained using BISON are compared to the experimental data in fig. 2 and 3. For this purpose, a mesh with sixty-four radial nodes and one hundred and forty four azimuthal nodes in order to have a 8.9 µm radial precision on the hydrogen distribution and thus have a node size comparable to the rim size found experimentally.

It has been observed that in the BISON calculation, when hydrogen is initially homogeneously distributed at $t_0 = 0s$, the final distribution matches the experimental data. In order to have a simulation that both simulates the hydrogen ingress and computes its distribution, a script has been made to counteract the nucleation problem. This script runs BISON a first time on a coarse mesh to obtain the hydrogen ingress, then reads the total hydrogen content in the cladding at the end of the simulation, and then performs another BISON run with no hydrogen pickup, and with the previously obtained hydrogen content homogeneously distributed in the cladding initially. The last run is made on the fine mesh described above, to calculate the hydrogen distribution on a fine mesh.

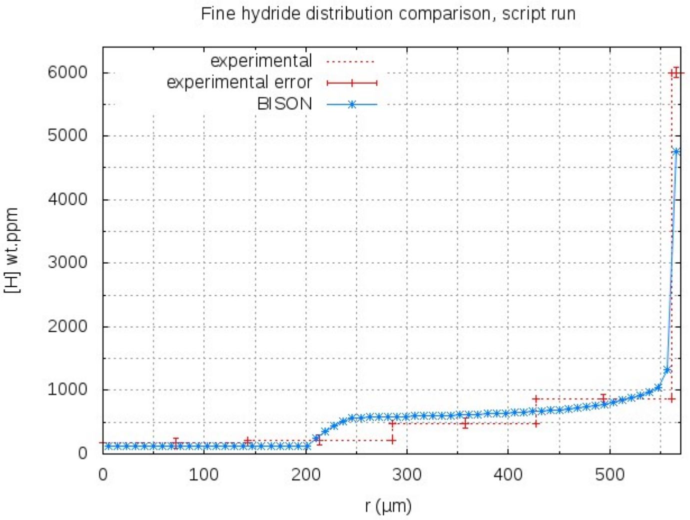

Figure 3: *Hydrogen distribution comparison between experimental data and BISON calculations on a 64 layer mesh*

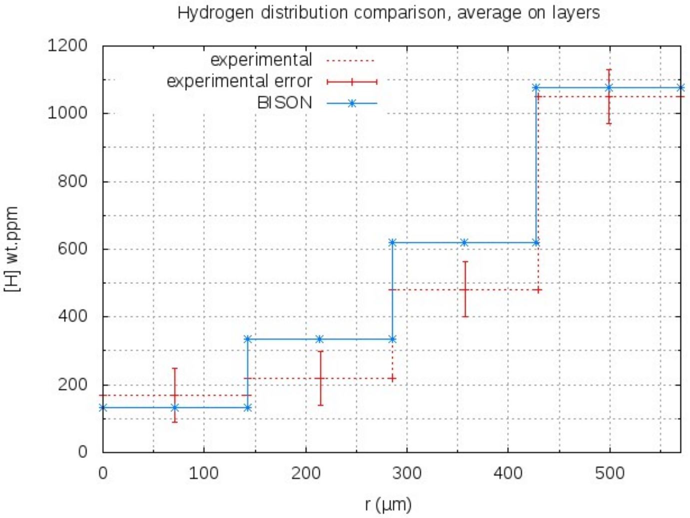

Figure 4: *Hydrogen distribution comparison between experimental data averaged on the 4 experimental layers*

It is important to keep in mind that the numerical model cannot compute the maximum hydrogen content in the metallic matrix at operating conditions. A "clamp parameter" has been put in place in BISON and has been set so that the maximal total hydrogen content (hydride + solid solution) is 6,000 wt.ppm, to match the experimental hydrogen content in the rim. The resulting radial hydride distribution is given by figures 3 and 4, and the previous experimental results have been added (with the extra data point provided by the zirconium hydride rim) for comparison. We can see that the hydrogen distribution calculated by BISON is very close to that observed experimentally.

The plot in figure 3 shows that the hydride rim thickness is well estimated by BISON although its hydrogen content is not the same, Zhang [3] showed that the hydrogen content in those samples was 6,000 wt. ppm. whereas BISON calculates it to be 4,750 wt.ppm. and the overall hydrogen distribution matches the experimental results reasonably well. It is considered that the results obtained are satisfying, given the paucity of data on operation conditions. However, when just looking at the hydrogen content in all four layers, we see that the hydride distribution given by the graph on figure 4, shows very good agreement with the experimental results.

Figures 3 and 4 also show a high concentration of zirconium hydrides in the cold regions (near the cladding/coolant interface), and a lower concentration of these species in the hot regions (near the cladding/gap or cladding/pellet interface), as predicted by the Soret effect, showing that hydrogen migrates from hot regions to cold regions. When the TSS_p is reached, hydrogen starts precipitating as zirconium hydrides and forms a rim. This rim can be observed in figure 3. As hydrogen migrates to the cold zones, hydrogen continues to precipitate until it can no longer do so, either because of mobility issues or because no more precipitation sites are accessible.

Discussion

As can be seen in the previous figures, the hydrogen distribution calculated by BISON is very close to that found experimentally. This difference in hydride content in the rim may come from the fact that this distribution is made without any model of nucleation, germination and growth of the zirconium hydrides. If the physics of that were to be implemented in the model, a difference in the distribution may be seen. Also, even though the stress gradient induced migration is negligible with respect to the Soret effect, this phenomenon coupled to the nucleation model described may be enough to make up for the slight mismatch in the hydride rim content observed in this study.

Even though this study shows promising results on the model's capability to predict zirconium hydride distribution, it is important to perform further benchmarking. To that effect a series of experiments will be held by the authors, that will allow to test the model with different kind of temperature gradients.

Conclusion

This study documents the benchmarking of the model implemented in BISON for hydrogen transport and precipitation. It is also the first comparison between the model calculations and in-reactor experimental data. Thus allowed to have a better understanding of the physics put in place during reactor shutdown, such as the precipitation kinetics that are dependent on the temperature of the clad [3,4].

Overall, the model showed a very good match with experimental results even though a mismatch can be observed in the rim's hydrogen content. The most important part of this type of simulation is to accurately predict the hydrogen content in the weakest point and thus, near the colder part of the cladding where the hydride rim forms. The size of the hydride rim is the same as that observed experimentally but the hydride content differs by about 20% from the experimental data.

In conclusion, the hydrogen distribution shows very good agreement between the experimental and numerical data. It can also be seen that the hydrogen model shows good agreement with the $\mathrm{TSS_p}$ and the $\mathrm{TSS_d}$ values and a slight over saturation can be observed in the kinetics during shutdown. In order to continue benchmarking the model, it has been decided to create an experiment which will consist of putting a ZIRLO tube under a controlled temperature gradient, this way, a very well defined environment will be established and the benchmarking of the 3D finite element code BISON will be possible. Once this task will be done, BISON will be used coupled with other high fidelity codes, such as DeCART and Cobra TF to compute the hydrogen distribution in more complex areas of the reactor such as near the mixing vanes.

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