Implementation and Validation of the Hydride Nucleation-Growth-Dissolution (HNGD) model in BISON

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ABSTRACT

During the operation of a light water reactor, a fraction of the hydrogen produced by waterside corrosion is absorbed into the nuclear fuel cladding. When the hydrogen concentration reaches its solubility limit, a brittle zirconium hydride phase precipitates, leading to a loss of ductility of the cladding. To assess cladding integrity, an accurate simulation tool is needed to predict hydrogen distribution within the cladding and hydride precipitation. Recent studies have developed an improved understanding of the physical processes involved in hydrogen redistribution and hydride precipitation and dissolution. This research led to the development of a new model, called Hydride Nucleation-Growth-Dissolution (HNGD). The present work describes the implementation of HNGD into the fuel performance code BISON, developed at Idaho National Laboratory. The main innovative feature of the HNGD model is that it accounts for hydride nucleation and growth as two distinct precipitation components, using the Johnson-Mehl-Avrami-Kolmogorov model to describe hydride growth kinetics. Each step of the model implementation into BISON was systematically verified, and simulations of experiments performed for validation, showing that the HNGD model provides improved predictions, and captures some experimentally observed physical phenomena related to hydride growth that the previous model could not.

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1. Introduction

During operation of a light water reactor, waterside corrosion leads to hydrogen pickup into the Zr alloy cladding tube, as illustrated schematically in the first panel of Figure 1. The hydrogen redistributes in the cladding following temperature, stress and concentration gradients into regions of low temperature, high stress and low concentrations (Fig. 1, second panel). When the local concentration reaches the solubility limit, a zirconium hydride phase precipitates [1].

This brittle hydride phase can compromise cladding integrity, so simulation tools able to predict accurately hydrogen redistribution and hydride precipitation and dissolution are needed. A simplified model was implemented into the fuel performance code BISON developed at Idaho National Laboratory (INL) [2,3], but recent work brought an improved understanding of the physics involved in hydride precipitation and dissolution [4]. Following this new understanding, a new model called Hydride Nucleation-Growth-Dissolution (HNGD) has been developed. Figure 2 shows a schematic comparison between HNGD and the model previously used in BISON. The precipitation and dissolution of hydrides depends on the temperature and on the hydrogen content in solid solution. In the previous model, precipitation occurs when the matrix is oversaturated with hydrogen, but stops when the precipitation limit is reached. The limit considered here is the so called Terminal Solid Solubility for Precipitation (TSSP). Hydrides dissolve if they exist in a matrix that is undersaturated in hydrogen. The saturation limit is known as the Terminal Solid Solubility for Dissolution (TSSD), and the dissolution is considered instantaneous. According to that model, between TSSP and TSSD there is a hysteresis region in which neither hydride precipitation nor dissolution occurs.

In the HNGD model however, hydride precipitation is divided into nucleation of new hydrides and growth of existing ones. Because of this a marked difference occurs when there are pre-existing hydrides upon cooling and when all the hydrogen is dissolved: experimental evidence shows that hydrides can grow while being held in the hysteresis region at constant temperature [5]. Nucleation of new hydrides occurs when the solid solution content is above the TSSP. The TSSD is identified as the...
thermodynamic solubility limit while the TSS\textsubscript{P} is identified as the supersolubility limit \cite{4}. The growth of existing hydrides occurs when the solid solution content is above the TSS\textsubscript{P}. The kinetics of precipitation by hydride growth are described using the Johnson-Mehl-Avram-Kolmogorov (JMAK) model \cite{6}. Both of these precipitation mechanisms occur at specific rates, determined by kinetic parameters. Further, in the HNGD model hydride dissolution is no longer considered instantaneous \cite{4}. These three phenomena are described by Equations (1) to (3):

\begin{align}
\text{Dissolution} : \quad \frac{\partial C_{SS}}{\partial t} &= -K_D (C_{SS} - TSS_D) \tag{1} \\
\text{Nucleation} : \quad \frac{\partial C_{SS}}{\partial t} &= -K_N (C_{SS} - TSS_P) \tag{2} \\
\text{Growth} : \quad \frac{\partial C_{SS}}{\partial t} &= -K_C (C_{cat} - TSS_D) p(1-x)(-\ln(1-x))^{1-1/p} \tag{3}
\end{align}

where $C_{SS}$ is the hydrogen content in solid solution, $K_D$, $K_N$, $K_C$ are the kinetic parameters for dissolution, nucleation and growth respectively, $x = \frac{C_{SS} - C_{cat}}{C_{SS} - TSS_D}$ is a measure of the advancement of the precipitation reaction, with $C_{cat}$ the sum of the hydrogen contents in solid solution and in hydrides, and $p$ is the dimensionality of the growth (in the JMAK model $p \sim 2.5$ for platelets). \cite{6}

In the zirconium alloy cladding, hydrogen atoms migrate principally driven by a concentration gradient (Fick’s diffusion) and by a temperature gradient (Soret effect). These two effects modify the hydrogen concentration in solid solution as per Equation (4):

\begin{align}
\frac{\partial C_{SS}}{\partial t} &= -\nabla \cdot \left(-D \nabla C_{SS} - \frac{DQ^*C_{SS}^2}{RT^2} \nabla T \right) \tag{4}
\end{align}

where $D$ is the hydrogen diffusion coefficient in the material (following an Arrhenius law), $Q^*$ is the heat of transport of hydrogen in zirconium (assumed not to vary with temperature) and $R$ is the ideal gas constant.
2. HNGD Parameters

In the HNGD model the solubility limits discussed above are fitted using Arrhenius-type laws (Eq. (5))

$$\text{TSS}_D = \text{TSS}_{D0} \times \exp \left( \frac{-Q_0}{RT} \right)$$

$$\text{TSS}_p = \text{TSS}_{P0} \times \exp \left( \frac{-Q_0}{RT} \right)$$

where the preexponential factors TSS$_{D0}$, TSS$_{P0}$ and activation energies Q$_0$, Q$_0$ are determined empirically [7-12].

The hydride dissolution and nucleation K$_D$ and K$_N$ kinetic parameters also follow Arrhenius laws. Hydride growth can be limited by two factors: hydrogen diffusion (subscript 'mob' for mobility) and matrix/hydrate interface reaction (subscript 'th' for thermodynamics). Each of these contributions follows an Arrhenius law. These three factors are described in Equations (6) to (8). [4]

Dissolution: $K_D = K_{D0}\exp(-\frac{E_D}{RT})$  

Nucleation: $K_N = K_{N0}f_0\exp(-\frac{E_N}{RT})$  

Growth: Diffusion controlled: $K_{mob} = K_{mob0}f_0v_0\exp(-\frac{E_D}{RT})$

Reaction controlled: $K_{th} = K_{th0}f_0v_0\exp(-\frac{E_N}{RT})$

$$K_G = (1/K_{mob} + 1/K_{th})^{-1}$$

where $K_{D0}$, $K_{N0}$, $K_{mob0}$, $K_{th0}$ are the preexponential factors of the Arrhenius laws for dissolution, nucleation, diffusion-controlled growth, and reaction-controlled growth; $E_D$ is the activation energy for diffusion of hydrogen in Zircaloy; $E_N$ is the activation energy for hydride growth by accretion of hydrogen atoms to the particle by diffusion, the coefficients $f_0$ and $v_0$ are defined below, and $E_{th}$ is the formation energy of the hydride, fitted with a degree 3 polynomial (Eq. (9)). The coefficients are given in Table 1.

$$E_{th} = -E_{th0} + E_{th1}T - E_{th2}T^2 + E_{th3}T^3$$

The dependencies on hydride content are described by:

• Volume fraction of $\alpha$ phase:

$$f_\alpha = 1 - \frac{x_{prec}}{x_\delta}$$

• Atomic fraction of hydride (with $M_H$ and $M_Z$ the atomic weights of hydrogen and zirconium, $x_{prec}$ the precipitated hydride content, and $C_{tot}$ the total hydrogen content in wt. ppm): $x_{prec} = \frac{C_{prec}}{M_H(\frac{C_{tot}}{M_H} + \frac{10^5 - C_{tot}}{M_Z})}$

• Atomic fraction of hydrogen at the ($\delta/\alpha + \delta$) boundary: [4,5]

$$x_\delta(T) \approx -9.93 \times 10^{-11}T^3 + 8.48 \times 10^{-8}T^2 - 5.73 \times 10^{-5}T + 0.623$$

• Atomic fraction of hydrogen in the $\alpha$ phase:

$$\frac{x_\alpha}{x_\delta} = \frac{C_{tot}}{M_H(\frac{C_{tot}}{M_H} + \frac{10^5 - C_{tot}}{M_Z})}$$

• Atomic fraction of hydrogen in the $\alpha$ phase:

$$x_\alpha = \text{TSS}_p$$

The default values of the parameters are shown up in Table 1. These come from the original development of the HNGD model in [4,5].

3. BISON update

3.1. HNGD Model Verification

3.1.1. Analytical Verification

The HNGD model described in the previous section is implemented in BISON, the finite element based nuclear fuel performance code developed at INL. As mentioned earlier, each phenomenon was systematically verified. Hypothetical cases are used to check the mathematical behavior of the model.

The hydride dissolution verification case assumes a uniform sample of Zircaloy loaded with $C_{tot} = 250$ wt. ppm of hydrogen, maintained at a constant temperature of 550K. At this temperature, the equilibrium concentration of hydrogen in solid solution is $TSS_{D0} = 44$ wt. ppm. Using the initial condition $C_{SS}(0) = 0$ wt. ppm (i.e. all hydrogen is in hydride particles) the analytical solution of Equation 1 is given by Equation 10:

$$C_{SS}(t) = TSS_{D0} (1 - e^{-K_{th}t})$$

Because the concentration in solid solution is below the solubility limit, the hydrides dissolve and $C_{SS}$ increases exponentially to reach the equilibrium value. Figure 3a shows the difference between the analytical solution in Equation 10 and the computed result for various time step values. The computed solution follows well the measured precipitation kinetics and approaches the predicted equilibrium value from the analytical solution. Also, as expected, the difference decreases when the time step is reduced. 

Table 1

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Symbol</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Preexponential and activation energy of the supersolubility Arrhenius law (Eq. (5))</td>
<td>$TSS_{D0}$</td>
<td>$3.08 \times 10^4$ wt. ppm</td>
</tr>
<tr>
<td>Preexponential and activation energy of the solubility Arrhenius law (Eq. (5))</td>
<td>$Q_D$</td>
<td>$0.26 \text{ eV \cdot atm}^{-1}$</td>
</tr>
<tr>
<td>Preexponential and activation energy of the diffusion coefficient Arrhenius law</td>
<td>$Q_0$</td>
<td>$1.02 \times 10^3$ wt. ppm</td>
</tr>
<tr>
<td>Preexponential terms of the nucleation, dissolution reaction-controlled growth and diffusion-controlled growth kinetics Arrhenius laws (Eq. (6) to (8))</td>
<td>$D_0$</td>
<td>$0.37 \text{ eV \cdot atm}^{-1}$</td>
</tr>
<tr>
<td>Coefficients of the 3rd degree polynomial used to express the formation energy of $\delta$ hydrides (Eq. (9))</td>
<td>$E_D$</td>
<td>$0.46 \text{ eV \cdot atm}^{-1}$</td>
</tr>
<tr>
<td>Activation energy of the diffusion-controlled growth kinetics Arrhenius laws (Eq. (3))</td>
<td>$K_D$</td>
<td>$1.11 \times 10^3$ s$^{-1}$</td>
</tr>
<tr>
<td>Heat of transport of hydrogen in zirconium.</td>
<td>$K_N$</td>
<td>$2.75 \times 10^5$ s$^{-1}$</td>
</tr>
<tr>
<td></td>
<td>$K_{mob}$</td>
<td>$5.35 \times 10^3$ s$^{-1}$</td>
</tr>
<tr>
<td></td>
<td>$K_{th}$</td>
<td>$1.6 \times 10^3$ s$^{-1}$</td>
</tr>
<tr>
<td></td>
<td>$E_D$</td>
<td>$5.66 \times 10^{-3} \text{ eV \cdot atm}^{-1}$</td>
</tr>
<tr>
<td></td>
<td>$E_{th1}$</td>
<td>$4 \times 10^{-4} \text{ eV \cdot atm}^{-1}$</td>
</tr>
<tr>
<td></td>
<td>$E_{th2}$</td>
<td>$2 \times 10^{-7} \text{ eV \cdot atm}^{-1}$</td>
</tr>
<tr>
<td></td>
<td>$E_{th3}$</td>
<td>$3 \times 10^{-10} \text{ eV \cdot atm}^{-1}$</td>
</tr>
<tr>
<td></td>
<td>$E_C$</td>
<td>$0.9 \text{ eV \cdot atm}^{-1}$</td>
</tr>
<tr>
<td></td>
<td>$Q^*$</td>
<td>$0.26 \text{ eV \cdot atm}^{-1}$</td>
</tr>
</tbody>
</table>
The hydride nucleation verification studies a case of Zircaloy uniformly loaded with $C_{fat} = 540 \text{wt. ppm}$ of hydrogen, maintained at a constant temperature of $600K$. At this temperature, the equilibrium concentration of hydrogen in solid solution for nucleation only is $TSS_{p}^{600} = 195 \text{wt. ppm}$ (we turn off the growth term in the model for this calculation so no growth is computed here). At this hydrogen level, the factor $f_{a}$ from Equation 7 is close to 1 and does not vary significantly during the computation. Assuming this value is constant and equal to 1 and using the initial condition $C_{SS}(0) = C_{fat}$ (i.e. all hydrogen is in solid solution) the analytical solution to Equation 2 is given by Equation 11:

$$C_{SS}(t) = TSS_{p}^{600} + (C_{fat} - TSS_{p}^{600}) e^{-k_{tot} t}$$  \hspace{1cm} (11)

Because the hydrogen concentration in solid solution is above the $TSS_{p}$, the solid solution content decreases exponentially until it reaches the $TSS_{p}$. As previously, Figure 3b shows the computed results and the analytical solution, illustrating that the calculated results reproduce the nucleation well and the difference between analytical and calculated hydrogen content decreases when the time step is reduced.

The hydride growth verification case assumes a uniform sample of Zircaloy loaded with $C_{fat} = 288 \text{wt. ppm}$ of hydrogen, maintained at a constant temperature of $650K$. At this temperature, the equilibrium concentration of hydrogen in solid solution is $TSS_{p}^{650} = 144 \text{wt. ppm}$. As before, $f_{a}$ and $V_{0}$ (in Eq. 8) are assumed to be equal to 1. Using the initial condition $C_{SS}(0) = C_{fat}$ (i.e. all hydrogen is in solid solution) the analytical solution to Equation 3 is given by Equation 12. In this case, the hydride nucleation computation is disabled, so in the code the hydride content is actually initialized at $10^{-6} \text{wt. ppm}$ so the algorithm triggers the hydride growth computation, and we have hydride precipitation by pure hydride growth.

$$C_{SS}(t) = TSS_{p}^{650} + (C_{fat} - TSS_{p}^{650}) e^{-k_{tot} t}$$  \hspace{1cm} (12)

The convergence of the computed results of hydrogen in solid solution with decreasing time step values is illustrated in Figure 3c. The kinetics of hydride precipitation by hydride growth only is well reproduced by the model. Again, the difference between simulation and analytical solution decreases with decreasing time step values.

3.1.2 Uniform-Temperature Sample Behavior

Each of the verification cases described above features one phenomenon of the HNGD model, occurring at one temperature. This section shows the behavior of a sample uniformly loaded with 200wt. ppm of hydrogen, which is subjected to a heat treatment consisting of a heat-up followed by a cool-down. Two cases are differentiated, whereby hydrides are completely dissolved at the end.
of the heat-up phase or only partially. Each case was computed using a range of values for the cooling rate.

Figure 4 shows the calculated results of the hydrogen in solid solution in this sample when heated to 800K (complete dissolution, Fig. 4a) and to 650K (incomplete dissolution, Fig. 4b). Figure 4a shows that when the sample is heated to 800K all hydrogen goes into solid solution \((C_{SS} = 200\text{wt. ppm})\). Subsequent cooling causes no precipitation until the TSS\(P\) is reached. At that point different results accrue depending on the cooling rate. At a 10K/min cooling rate, the concentration of hydrogen in solid solution follows closely the TSS\(P\) curve, indicating that most hydrogen precipitates through hydride nucleation, which is fast enough to maintain its equilibrium at TSS\(P\) while hydride growth is too slow to be significant. At very high cooling rates (100K/min), departure from the equilibrium behavior is clearly observed. In particular, as the hydride nucleation kinetics is not rapid enough to keep up with the temperature decrease, the concentration of hydrogen in solid solution is higher than the TSS\(P\) during the transient. In contrast, for low cooling rate \((0.35\text{K/min} \text{ and below})\), hydrogen precipitation into hydrides occurs mostly by hydride growth. Accordingly, for the low cooling rates the curve deviates significantly from TSS\(P\) and the hydrogen in solid solution decreases upon holding for a long time at a constant temperature.

In contrast, when some hydrides are still present at the highest temperature, hydrogen precipitation by hydride growth starts to occur immediately when the sample starts to be cooled (no need to reach TSS\(P\)). Figure 4b shows the results for such simulation. The sample is heated up to 650K. At this temperature the total hydrogen content is higher than the solubility, so there are still hydrides particles in the sample when the cool down begins (about 55wt.ppm of hydrogen is in precipitated form). The results show that hydrogen precipitation by hydride growth becomes increasingly important as the cooling rate decreases.

Thus the simulations in this section show that the HNGD model behaves as intended, as it is qualitatively consistent with the data presented in [5].

3.2. HNGD Model Validation

Now that the HNGD model is verified, it is tested by comparing its predictions with experiments. Three types of experiments are presented here. The first one uses a Zircaloy sample subjected to a uniform temperature treatment, which validates that the HNGD model is able to predict hydride precipitation. The second one uses a sample loaded with a uniform hydrogen distribution and subjected to a temperature gradient, which validates that the model is able to predict the hydrogen distribution in such setup. The third one uses a sample loaded with hydrogen via a hydride rim formed at one end, subjected to an asymmetric temperature profile. The results of the calculation shows that further work is needed to pre- dict hydride precipitation on a broad region when the hydrogen is not uniformly distributed at the beginning of the experiment.

3.2.1. Transients Benchmark

The first experiment used for validation consist of a series of thermal transients applied to a hydrogen-loaded sample while the evolution of the hydrogen concentration is monitored via synchrotron X-ray diffraction [4]. The temperature changes lead to successive dissolution and precipitation. The 254wt.ppm sample is subjected to the heating and cooling cycle shown by the dashed line in Figure 5, which was designed to highlight the different phenomena occurring in the sample by inducing successive dissolution and precipitation [4]. The sample is initially at room temperature and is heated to 700K, which is enough to dissolve all the hydrides; accordingly the hydrogen in solid solution increases to the full amount. In the following cycle, the temperature is raised.
to 660K, which causes 180wt. ppm to dissolve. The temperature is then held at 560K, causing the hydrogen in solid solution to decrease from 110wt. ppm to 85wt. ppm during a 1000s-temperature hold because of hydride growth. Then the sample is heated again to 700K, causing all the hydride particles to dissolve, and the sample is finally brought back to room temperature. These thermal transients are applied while the hydrogen in solid solution content is measured via the hydride diffraction signal [4]. Figure 5 shows the evolution of hydrogen content in solid solution, comparing the experimental data (red squares) with BISON using the previous model (dotted blue line) and the HNGD model, using two different fits for the solubility. In particular, the plain blue line uses the default fit of the model, while the dashed blue uses a custom TSSD fit. The experimental data shows that there is a total dissolution of the hydrides by the end of the first heat-up. This is predicted by the previous BISON model, but the dissolution is only partial with the HNGD model using its default solubility fit. To provide an additional comparison to the previous BISON model that excludes the effect of a particular parameter, and thus isolate the impact of the new hypotheses of the HNGD model, an Arrhenius fit based on the first heat-up was used in the custom HNGD calculation. Given the variability of the TSSD values in the literature [7–12], this fit is still in the 1σ uncertainty range of the default fit.

The two models diverge during the first cool-down: at the yellow marker the solid solution concentration reaches the hysteresis region (TSSD < CSS < TSSP) so the previous model stops the hydride precipitation, while the HNGD model continues to compute precipitation by hydride growth. Another difference occurs when the second heat-up starts, marked by the green line and shade. For the previous model the solid solution concentration is still high so it resumes precipitation (CSS > TSSP) until the concentration in solid solution content reaches TSSP. Instead, for the HNGD model, most of the hydrogen is in hydrides at that point, so hydride dissolution occurs (CSS_HNGD < TSSD) until reaching the solubility limit (TSSP). It is clear that the HNGD model provides a better description of the experimental data than the previous model.

The difference between the two solubility fits used by the HNGD model is particularly apparent at high temperature. Experimentally, all hydrogen is in solid solution at 700K, and the custom simulation is consistent with this, but there are still about 20wt. ppm of hydride when using the default parameters. This leads to another difference happening at the beginning of the cool down. In the default simulation, hydride growth occurs as soon as the temperature starts decreasing, whereas the custom simulation must reach TSSP to trigger hydride nucleation and start precipitating.

This validation case shows that the HNGD model is able to predict hydride precipitation and dissolution in a uniformly hydrided sample. The next section shows the ability to predict the hydrogen profile in a sample initially uniformly loaded with hydrogen and subjected to a temperature gradient for a long time.

3.2.2. Sawatzky’s experiments

The second set of cases used for validation is Sawatzky’s two experiments described in [13]: two 2.54cm long Zircaloy-2 samples loaded with hydrogen (130wt. ppm and 63wt. ppm respectively) are subjected to a temperature gradient (57.9K/cm and 116K/cm) for 34 and 41 days respectively. We use the HNGD model in BISON to simulate this couple of experiments and compare the hydrogen profile after 34 and 41 days respectively.

Figure 6 shows the result from BISON computation (blue) compared to the experimental data (red markers) and the analytical solution derived by Sawatzky (red line). This analytical solution was obtained based on the following assumptions: (i) the hydrogen is the only component diffusing, (ii) it diffuses mainly in the matrix, and (iii) the hydrogen in solution is in equilibrium with the hydride particles in the biphasic region (i.e., $CSS = TSSD$). See [13] for complete derivation. In Figure 6a, the additional grey curve represents the results using the previous model [3]. The shape of the profile is much closer to the analytical solution than the data using the HNGD model. The additional profiles [upper][lower] were obtained by using a solubility fit one standard deviation above and under the default value.
When temperature, A54, profile x of solution/precipitation higher, creates build. The temperature Kammenzind’s model simulates this process, where an asymmetric temperature profile, with higher temperatures at the cold side, leads to the precipitation of hydride at the cold side. Hydride precipitation occurs in a domain near the cold end (about 0.5 to 1 cm wide), with a maximum hydrogen content of a few hundreds of wt ppm at the interface. At the end of the experiment, the hydride rim has not completely dissolved: the far most right experimental point still reaches 600 to 3000 wt ppm of hydrogen.

To simulate this experiment, the initial condition is built assuming that (i) the hydride rim is made of a solid hydride, (ii) the rest of the sample (‘body’) contains a small amount of hydride, and (iii) at room temperature the concentration of hydrogen in solid solution is negligible (all is precipitated). In the code, this is translated as (i) $C_{\text{prec}} = C_{\text{rim}} = 17000 \text{wt ppm}$ on a certain length $\delta$. (ii) $C_{\text{prec}} = C_{\text{body}} = 10 \text{wt ppm}$ in the rest of the sample, and (iii) $C_{SS} = 0 \text{wt ppm}$ everywhere. The value of $\delta$ is determined from conservation of mass. The total hydrogen content of the sample H is derived from the experimental data, and $\delta$ is defined so that $H$ is constant during the simulation:

\[ \text{Experiment} : \quad H = \frac{1}{L} \sum_{i} h_{i}l_{i} \quad (13) \]

\[ \text{Simulation} : \quad H = \frac{1}{L} \int_{0}^{L} C_{\text{prec}} dx = \frac{(L - \delta)C_{\text{body}} + \delta C_{\text{rim}}}{L} \quad (14) \]

where $h_{i}$ and $l_{i}$ are the hydrogen content and length of the slices cut at the end of the experiment and $L$ is the length of the sample. Using Equations 13 and 14 we obtain:

\[ \delta = L \frac{H - C_{\text{body}}}{C_{\text{rim}} - C_{\text{body}}} \quad (15) \]

Figure 8 shows the results of BISON simulations compared with the experimental data. This time the parameter that is modified is the TSSP fit, similarly to the solubility in the previous case.

In the simulation, the sample temperature is initially at 300K and takes an hour to reach the desired annealing profile. During the annealing, the hydride rim dissolves partially, emitting hydrogen into the rest of the sample. The hydrogen in solid solution migrates to the cold side and builds up at the cold end. If $C_{SS}$ reaches the TSSP limit, hydride nucleation is triggered in the first node on the left in the simulation mesh (cold end). Then hydride growth also occurs, creating a depletion of hydrogen in solid solution in the first node. With this depletion, and under Fickian and Soret diffusion, the first node acts as a hydrogen sink: the concentration in solid solution reaches TSSP (equilibrium between the hydrides and the matrix) and all hydrogen that reaches this node is absorbed via hydride growth. Because of this, the hydrogen concentration in the other nodes is lowered and remains below TSSP. As a result, if precipitation occurs in the first node, it does not occur anywhere else in the simulation domain. Whether precipitation occurs in the first node depends on the choice of the TSSP fit, as seen in Figure 8. For each sample simulated, the results overlap when using the default TSSP fit or a higher one. With a lower TSSP, precipitation can occur in the first node only, if anywhere. The lower TSSP allows precipitation in all cases, except for the sample A56 for which all profiles overlap. The model fails to predict the hydride profile in the bipha-

![Fig. 7. Evolution of the hydride distribution in Sawatzky’s first experiment [13]. A dissolution/precipitation cycle occurring at the limit between the one-phase and two-phase regions creates a hydride peak that gets higher and sharper as time passes. The times in the legend are given in seconds.](image-url)
Fig. 8. Simulation of the cases from [14] with three different fits of TSSP: the default fit of the HNGD model, one standard deviation higher (noted 'high'), and one standard deviation lower (noted 'low'), based on the variability of the TSSP fits in the literature [7–12]. TSSP. The results are similar to those obtained with the previous model.

4. Conclusion and future work

This study describes the implementation of the Hydride Nucleation-Growth-Dissolution (HNGD) model into the fuel performance code BISON. As part of this work, we have performed systematic verification calculations for each represented physical mechanism, and validation against data from eight experiments.

The calculated hydrogen distribution and hydride precipitation show significant qualitative and quantitative improvement of the model predictions relative to the simple model previously available in BISON. The model is valid for samples containing up to $\sim 300$ wt. ppm of hydrogen, and it is now available for use by the community of BISON users.

Future work includes predicting the hydrogen profile in experiments where hydrogen is not initially homogeneously distributed.
Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

CRediT authorship contribution statement

Florian Passelaigue: Software, Writing - original draft, Visualization. Evrard Lacroix: Conceptualization, Methodology, Writing - review & editing. Giovanni Pastore: Supervision, Conceptualization, Methodology, Validation, Resources, Writing - review & editing. Arthur T. Motta: Supervision, Conceptualization, Validation, Writing - review & editing, Funding acquisition.

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Supplementary materials


References