Modeling the Hydrogen Effect on the Constitutive Response of a Low Carbon Steel in Cyclic Loading

Hydrogen-accelerated fatigue crack growth is a most severe manifestation of hydrogen embrittlement. A mechanistic and predictive model is still lacking partly due to the lack of a descriptive constitutive model of the hydrogen/material interaction at the macroscale under cyclic loading. Such a model could be used to assess the nature of the stress and strain fields in the neighborhood of a crack, a development that could potentially lead to the association of these fields with proper macroscopic parameters. Toward this goal, a constitutive model for cyclic response should be capable of capturing hardening or softening under cyclic straining or ratcheting under stress-controlled testing. In this work, we attempt a constitutive description by using data from uniaxial strain-controlled cyclic loading and stress-controlled ratcheting tests with a low carbon steel, Japanese Industrial Standard (JIS) SM490YB, conducted in air and 1 MPa H2 gas environment at room temperature. We explore the Chaboche constitutive model which is a nonlinear kinematic hardening model that was developed as an extension to the Frederick and Armstrong model, and propose an approach to calibrate the parameters involved. From the combined experimental data and the calibrated Chaboche model, we may conclude that hydrogen decreases the yield stress and the amount of cyclic hardening. On the other hand, hydrogen increases ratcheting, the rate of cyclic hardening, and promotes stronger recovery.

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1 Introduction

Materials loaded in the presence of hydrogen experience mechanical degradation which can be demonstrated as reduced elongation and reduction in area, decreased fracture toughness, accelerated fatigue crack growth, and shortened cyclic fatigue life [1–12]. Such hydrogen-induced degradation of mechanical properties is usually accompanied by transition of deformation processes and fractography [1–12].

Under monotonically increasing or sustained loading [13–21], the hydrogen effect has been investigated extensively and the fundamental hydrogen embrittlement phenomenology has been fairly well analyzed from detailed microstructural observations [18,19], fractographic studies [15], and mechanistic models [13,14,16]. The hydrogen effect on fatigue life and the associated microstructural characteristics of fracture have been investigated under uniaxial cycling loading [10,22–27]. Investigating the response of an annealed medium carbon steel under uniaxial stress-controlled loading, Uyama et al. [22] observed that hydrogen decreases the strain range under a given stress amplitude while it has no effect on fatigue life. On the other hand, Tsuchida et al. [23] carrying out strain-controlled tests with a low carbon S10 ferritic-pearlitic steel under cathodic charging observed that the fatigue life decreases in the presence of hydrogen. Such fatigue life decrease in hydrogen was also observed in other studies [10,24–26]. Martin et al. [25] investigated the fatigue life of a 4130 steel under uniaxial strain-controlled cyclic loading in an effort to develop “accurate fracture mechanics modeling” of fatigue in the presence of hydrogen. They observed that the fatigue life in air is approximately two orders of magnitude longer than in 18 MPa hydrogen. Martin et al. [25] also used the cyclic stress–strain response to fit the Morrow damage model [28]. Das and Singh [26] investigated the effect of hydrogen on P91 steel under strain-controlled cyclic loading. Using the cyclic stress–strain data to fit the Masing model curve, they concluded that the strength of the hydrogen-charged specimen is lower than that of the uncharged. Notably, Das and Singh [26] used the stress–strain data from a single cycle with strain amplitude of 1% to calibrate the Chaboche model for the material constitutive response. However, on the basis of the reported calibrated parameters, it seems that only the first kinematic hardening rule was treated as affected by hydrogen while the second and third kinematic hardening rules were considered to be the same in the hydrogen-charged and uncharged material. Das and Singh [26] also observed cyclic softening in both hydrogen-charged and uncharged specimens with the rate of softening being higher in the presence of hydrogen. Similar hydrogen effect on the rate of cyclic softening was also reported by Mansilla et al. [27] for high strength steel. In summary, with the exception of the work of the Das and Singh [26], the above studies, while they address the hydrogen effect on fatigue life in uniaxial cycling loading, do not provide a framework for the development of a constitutive model that can be used to quantify the local stress and strain fields under cycling loading in the presence of hydrogen.

Despite the fact that hydrogen-induced fatigue crack growth data as well as relevant metallographic [3,4] and fractographic results [1,11,12] have become available for a number of metals and alloys, the salient mechanisms responsible for hydrogen-accelerated fatigue crack growth [1,9,11,12,20] still remain poorly understood, in part because of the complexities inherent in the growth of fatigue cracks. This lack of mechanistic understanding and hydrogen-informed constitutive models limited the field of fatigue crack growth modeling to mainly phenomenological approaches as in cases involving modification of the Paris law parameters or the use of von Mises plasticity with Ramberg-Osgood hardening. It is worth noting that the classical von Mises plasticity model with associated flow rule fails to address such features of the cyclic response as hardening/softening or ratcheting [29].

Given the lack of predictive mechanistic models for hydrogen-accelerated fatigue crack growth, the development of a well characterized constitutive law is essential to quantifying local stress and strain fields under cyclic loading relative to macroscopically measured parameters. We note that full characterization of the stress and strain fields at a crack tip have been the landmark breakthroughs that enabled the successful development of the linear elastic and elastoplastic fracture mechanics, and we view such characterization to be of the same importance in the case of fatigue in the presence of hydrogen. As two noted researchers in the field of mechanics of materials [30] emphasized “Fracture mechanics is the judicious interpretation of stress and strain field ahead of the crack tip.” In this context, the objective of this work is to present precisely an approach for the development of a proper constitutive model specifically for material response under cycling loading.

Most metals and alloys subjected to strain-controlled cyclic loading exhibit either cyclic hardening or cyclic softening with subsequent stabilization after a certain number of cycles [28,31]. In a three-dimensional setting, this can be interpreted in terms of a stabilized yield surface after a certain number of cycles. Another cyclic loading feature is ratcheting under stress-controlled testing with nonzero mean stress. It has been shown that material models formulated with nonlinear kinematic hardening rules exhibit ratcheting response [32,33]. In these models, the stabilization of the yield surface is addressed by adding an isotropic hardening/softening increment to the flow stress. Seminal in this area is the nonlinear kinematic hardening model developed by Armstrong and Frederick [34]. Subsequent to this work, several models were proposed based on Armstrong and Frederick’s approach to improve the capability to predict ratcheting [32,33,35–41]. Notable among these revised approaches was that of Chaboche and Nouailhas [33]. The model is capable of capturing many prominent features induced by cyclic loading such as plastic shakedown, ratcheting, and the relaxation of the mean stress.

The present work is a combined experimental and theoretical attempt to explore and model the hydrogen effect on the constitutive response of materials subjected to cyclic loading. To accomplish this, we rely on the Chaboche constitutive model which we calibrate.
using a sequence of experimental data from uniaxial strain-controlled cyclic loading tests and uniaxial stress-controlled ratcheting tests with a low carbon steel, Japanese Industrial Standard (JIS) SM490YB, in the absence and presence of hydrogen.

2 Experiment

2.1 Material. The material used in this study was the JIS G 3106 (2015) Grade SM490YB steel, which is a low carbon rolled steel used for welded structures. The chemical composition is shown in Table 1. A small amount of Nb was added to refine the grains. A slab was reheated to austenite phase and was hot rolled into a steel plate shape of thickness of 16 mm.

The surface of the sample was etched with 3% Nital to reveal the microstructure for the optical microscopy and scanning electron microscopy (SEM) studies. A Hitachi S–4000 field-emission scanning electron microscope operating at an accelerating voltage of 15 kV was used for the examination of the microstructure. The microstructure of the steel at the quarter section of the steel plate and perpendicular to the rolling direction is shown in Fig. 1. It exhibits a mixed ferrite and pearlite colony microstructure with ferrite mean grain size of \( \sim 15 \mu m \) and pearlite volume fraction of \( \sim 25\% \). The pearlite was formed in steak-like bundles along the rolling direction, which suggests that the austenite phase was first transformed into ferrite phase in bundles along the rolling direction during cooling after hot rolling and then the remainder of the austenite with a higher carbon concentration was transformed into the pearlite phase.

The tensile properties of the steel in the absence of hydrogen are summarized in Table 2. The tensile specimens were machined on a lathe from the steel plate, with the center of the specimens at the middle thickness portion of the plate and perpendicular to the rolling direction. The stress–strain curve showed clear yielding behavior with an ultimate tensile strength of 552 MPa. Further, the hardness distribution along the thickness of the plate, as determined using a standard 98.1 N Vickers indent, exhibited a bowl shape, \textit{i.e.}, lowest at the middle portion of the plate (HV10 = 144) and highest at the surface portion (HV10 = 156). This reflects the distribution of microstructure along the thickness of the plate with the surface section having the most refined grains.

2.2 Strain- and Stress-Controlled Fatigue Tests. A schematic illustration of the specimen used for strain- and stress-controlled fatigue tests is shown in Fig. 2. The specimen is a cylindrical bar and the diameter and the gauge length of the reduced section are 4.0 and 15 mm, respectively. From previous studies [42] on hydrogen uptake in steels exposed to gaseous hydrogen, it is known that natural oxide film covering the specimen surface prevents hydrogen uptake. Moreover, Pd plating accelerates hydrogen uptake in steels by transitioning its endothermic reaction into exothermic reaction as well as by lowering both the hydrogen adsorption and absorption energies that represent the energy barriers from the physisorption state to the chemisorption state and from the chemisorption state to the sub-surface state, respectively [42,43]. Hence, all the specimens charged with hydrogen were electroplated with Pd of \( \sim 74 \text{ nm} \) thickness.

The experimental apparatus for the strain- and stress-controlled fatigue tests is shown in Fig. 3. These fatigue tests were carried out under tension and compression loading both in air and 1 MPa hydrogen gas. For the strain-controlled fatigue tests, strain was measured by a strain-gauge type extensometer. The strain rate was controlled at \( 1.3 \times 10^{-3} \text{ s}^{-1} \). Failure of the specimen was identified with a 25% reduction of the maximum stress from the initial maximum stress. The stress-controlled fatigue tests were conducted under
load control with a constant stress amplitude and two levels of mean stress. The stress amplitude and mean stress were selected so that the maximum stress was sufficiently higher than the yield stress and ratcheting behavior was observed. The stress amplitude $\sigma_a = (\sigma_{\text{max}} - \sigma_{\text{min}})/2$ was 420 MPa, where $\sigma_{\text{max}}$ and $\sigma_{\text{min}}$ are maximum and minimum stress, respectively. The values of the mean stress $\sigma_{\text{mean}} = (\sigma_{\text{max}} + \sigma_{\text{min}})/2$ were 20 and 40 MPa.

The atmospheric temperature of air and hydrogen gas was controlled at 20 °C. The tests in hydrogen were started 24 h after the 1 MPa hydrogen gas environment was established in order for a uniform hydrogen distribution throughout the specimen diameter prior to testing be established. Calculation of the transient hydrogen distribution throughout the cylindrical part of the gauge section [44] yields a saturation time equal to $Dt/a^2 = 1.0$ which implies that a uniform hydrogen distribution was established throughout the specimen in 1.1 h for $D = 10^{-9} \text{m}^2/\text{s}$ [45] and radius $a = 2.0 \text{ mm}$.

A thermal desorption analysis spectrum from the Pd-plated specimen that was exposed to 1 MPa hydrogen gas for 24 h without loading is presented in Fig. 4. The sample was heated at a constant rate of 100 °C/h from room temperature to 500 °C to evolve the hydrogen in a controlled manner. The hydrogen-charged sample shows two broad peaks with prominent desorption peak rates at 120 and 440 °C. The first peak that appears in the thermal desorption analysis spectrum is defined as peak 1 hydrogen associated with hydrogen atoms trapped at low energy traps such as dislocations and ferrite/cementite interfaces in the pearlite colonies [19,46]. The peak 1 hydrogen content in Fig. 4 was 0.16 mass ppm. The peak 2 hydrogen content was much smaller than the associated with the peak 1 and corresponds to hydrogen trapped at higher energy traps such as grain boundaries [19].

**3 The Chaboche Constitutive Model for Cyclic Plasticity**

As we mentioned in Sec. 1, we explored the Chaboche constitutive model [32,33,36,38] to describe the cyclic plasticity of JIS SM490YB steel, as this model is arguably the most promising in reproducing features of cyclic response. First, we briefly describe the model and its features, the idea being to associate these features
with the experimental response for subsequent calibration. In this regard, we also present the form of the model in uniaxial cycling loading.

3.1 Formulation. We assume that the deformation rate tensor $D$ is decomposed into elastic $D^e$ and plastic $D^p$ components [47]:

$$D = D^e + D^p$$  \hspace{1cm} (1)

For linear and isotropic elasticity:

$$D^e = \frac{1}{2} \frac{\nu}{1 + \nu} \sigma \delta - \frac{1}{2G} \sigma_{ik} \dot{\delta}$$  \hspace{1cm} (2)

where $\sigma$ is the Jaumann derivative of the Cauchy stress tensor, $\delta$ is the second-order identity tensor whose components $\delta_{ij}$ are the Kronecker delta, and $G$, $\nu$ are, respectively, the shear modulus and Poisson’s ratio. For isotropic yielding

$$f(\sigma, \alpha, R) = \Sigma - \sigma_0 - R = 0$$  \hspace{1cm} (3)

where $\Sigma = \sqrt{3/2} (\sigma^\prime - \alpha) : (\sigma^\prime - \alpha)$ is the effective stress, $\sigma^\prime$ is the deviatoric stress tensor, $\alpha$ is the back stress tensor, $\sigma_0$ is the yield stress, and $R$ describes either isotropic hardening if positive or isotropic softening if negative, and it is the increment to the yield stress that modulates the stabilization of the response in strain-controlled cyclic loading.

The back stress tensor is composed of three parts, $\alpha = \alpha_1 + \alpha_2 + \alpha_3$, with corresponding time rate of change:

$$\dot{\alpha}_i = \frac{2}{3} C_i D^p - \gamma_i \dot{\varepsilon}_p^p, \hspace{0.5cm} i = 1, 2, 3$$  \hspace{1cm} (4)

where $C_i$ and $\gamma_i$ are material constants

$$\dot{\varepsilon}_p^p = \sqrt[3]{2} D_{ij} D_{ij} / \Sigma_e$$  \hspace{1cm} (5)

is the effective plastic strain rate and the standard summation convention is implied over a repeated index. The associated flow rule leads to

$$D^p = \dot{\varepsilon}_p^p N$$  \hspace{1cm} (6)

in which $N = \partial f(\sigma, \alpha, R) / \partial \sigma = 3 (\sigma^\prime - \alpha) / 2 \Sigma$ is the normal to the yield surface and $\sigma^\prime$ is the stress deviator. The time rate
The change of the isotropic hardening/softening parameter $R$ is expressed as

$$\dot{R} = h(Q - R)\dot{\epsilon}_p$$ (7)

where $h$ and $Q$ are constants. Integration of Eq. (7) yields

$$R = Q(1 - e^{-h\epsilon_p})$$ (8)

and Fig. 11 shows a schematic for the variation of $R$ as a function of the accumulated plastic strain $\epsilon_p$. $R$ increases monotonically and levels to a value $Q$ which is positive (negative) for cyclic hardening (softening). It is noted that the parameter $Q$ determines the amount of cyclic hardening/softening during a strain-controlled test. In other words, the change in the maximum stress from the first cycle to the stabilized cycle is controlled by the parameter $Q$.

In summary, the Chaboche model for cyclic plasticity that we adopted in this work involves nine parameters: $C_1$, $C_2$, $C_3$, $\gamma_1$, $\gamma_2$, $\gamma_3$, $Q$, $h$, and $\sigma_0$ that can be calibrated experimentally. The calibration process is outlined in Sec. 4.

### 3.2 Uniaxial Strain-Controlled Cyclic Response

Our experimental results for the stabilized stress–strain hysteresis loop shown in Fig. 12 for the case $\Delta \epsilon = 2.2\%$ show three distinct segments [32,33,41]:

(i) Segment 1: the part of the stable hysteresis curve with high modulus at the onset of yielding and very small plastic strains;

(ii) Segment 2: the transient nonlinear portion of the hysteresis curve at moderate plastic strains;

(iii) Segment 3: the subsequent part of the hysteresis curve that varies almost linearly with plastic strain at large plastic strains.

Recall that the Chaboche nonlinear kinematic hardening model [32,33] involves a superposition of three Armstrong and Frederick [34] hardening rules, i.e., $\alpha = \alpha_1 + \alpha_2 + \alpha_3$ with corresponding evolution rules given by Eq. (4). Each of the three hardening rules controls the behavior of a corresponding segment of the hysteresis loop. The first rule, $\alpha_1$, governs initial hardening with a large modulus at very small plastic strains (segment 1), the second rule, $\alpha_2$, governs the nonlinear behavior at moderate plastic strains (segment 2), and the third hardening rule, $\alpha_3$, governs the nearly linear segment of the hysteresis loop along which the modulus is nearly constant (segment 3). The above three features of the Chaboche model will be further clarified in the discussion for the calibration of the model relative to Fig. 13 in Sec. 4.

First, we consider the uniaxial strain-controlled response under fully reversed tension/compression cyclic loading, i.e., $\epsilon_p^{\text{max}} = -\epsilon_p^{\text{min}}$. As shown in Appendix A, the closed form solution for the stable hysteresis loop is given by

$$\sigma_{xx} = C_1\gamma_1(1 - 2 \exp(-\lambda \gamma_1(\epsilon_p^{\text{max}} - \lambda \epsilon_p^{\text{min}}))) + C_2\gamma_2(1 - 2 \exp(-\lambda \gamma_2(\epsilon_p^{\text{max}} - \lambda \epsilon_p^{\text{min}}))) + C_3\epsilon_p^{\text{max}} + \lambda \sigma_0 + \lambda Q$$ (9)

where $\sigma_{xx}$ is the axial stress. As can be seen from Eq. (9), the parameter $\gamma_3$ does not appear in the solution for the stabilized loop because of its absence from the third hardening rule; see Appendix A for relevant details.

The maximum stress $\sigma_{MS}$ in the stable hysteresis loop is obtained from Eq. (9) for $\epsilon_p^{\text{max}} = \epsilon_p^{\text{max}}$ and $\lambda = 1$:

$$\sigma_{MS} = \alpha_{MS} + \sigma_0 + Q$$ (10)
slope of segment 1 is controlled by the absence of hydrogen. By construction of the Chaboche model, the maximum stress in cycle \( \Delta \) discerned for effect of hydrogen on the response of SM490YB steel can only be hysteresis loop data for \( d \). Thus, \( \varepsilon \) of the hysteresis curve at small strains, we have \( \varepsilon = \varepsilon_{\text{max}} \) under the assumption that the parameters \( \gamma \) are large so that \( \exp (-\gamma \Delta \varepsilon) \sim 0 \), as discussed in Appendix A.

4 Calibration

In this section, we present our approach to determining the nine parameters of the Chaboche model by using:

(i) The results from the uniaxial strain-controlled tests for the three different strain ranges \( \Delta \varepsilon = 0.92\% \), 1.08\%, and 2.2\% along with Eq. (9) to determine the parameters \( C_1, C_2, C_3, \gamma_1, \gamma_2, Q, h, \) and \( \sigma_0 \).

(ii) The results from the stress-controlled ratcheting tests with stress amplitude 420 MPa and mean stresses 20 MPa and 40 MPa to determine the remaining parameter \( \gamma_3 \).

4.1 Determination of Parameters \( C_1, C_2, C_3, \gamma_1, \gamma_2, Q, \) and \( \sigma_0 \) in the Absence of Hydrogen. As can be observed from Fig. 8, the effect of hydrogen on the response of SM490YB steel can only be discerned for \( \Delta \varepsilon = 2.2\% \) in comparison with \( \Delta \varepsilon = 0.92\% \) and 1.08\%. Hence, for the calibration, we use the stress–strain hysteresis loop data for \( \Delta \varepsilon = 2.2\% \) as plotted in Fig. 12 in the absence of hydrogen. By construction of the Chaboche model, the slope of segment 1 is controlled by the first hardening rule. Hence by neglecting the contributions of \( C_2 \) and \( C_3 \) to the slope of the hysteresis curve at small strains, we have \( d\sigma_c/d\varepsilon^0 \sim C_1 \) at \( \varepsilon^0 = \varepsilon_{\text{max}} \), which along with Fig. 12 yields

\[
C_1 = 3.2 \times 10^3 \text{MPa}
\]

Again, by construction of the Chaboche model, the slope of segment 3 is controlled by the parameter \( C_3 \), which is warranted if the parameters \( \gamma_1 \) and \( \gamma_2 \) are large so that \( \exp (-\gamma \Delta \varepsilon) \sim 0 \). Thus, \( d\sigma_c/d\varepsilon^0 \sim C_3 \) at \( \varepsilon^0 = \varepsilon_{\text{max}} \) and along with Fig. 12 we find

\[
C_3 = 5.3 \times 10^3 \text{MPa}
\]

In order to determine the parameter \( Q \), we use Eq. (A3) to obtain the maximum stress in cycle \( N \)

\[
\sigma_{\text{MN}} = \sigma_{\text{MN}} + \sigma_0 + R_N
\]

where \( \sigma_{\text{MN}} \) is the maximum back stress in the stable cycle given by

\[
\alpha_{\text{MN}} = \frac{C_1}{\gamma_1} + \frac{C_2}{\gamma_2} + \frac{C_3}{\gamma_3} \varepsilon_{\text{max}}
\]

and \( \gamma_1, \gamma_2, \gamma_3 \) are, respectively, the maximum stress, maximum back stress, and the isotropic hardening parameter \( \alpha \) associated with the stable cycle and the first cycle for \( \varepsilon^0 = \varepsilon_{\text{max}} \). Recalling that \( R \) levels to \( Q \) at stabilization, i.e., \( R_N = Q \), and considering that \( R_N \) is small in comparison with \( R_N \), we have

\[
\sigma_{\text{MN}} - \sigma_{\text{MN}} = R_N - R_1
\]

Hence, the parameter \( Q \) is obtained from the difference between the maximum stresses of the stable and first cycle. Figure 5(a) for the case \( \Delta \varepsilon = 2.2\% \) yields this difference as

\[
Q = 47 \text{MPa}
\]
Table 3: Calibrated material parameters for SM490YB steel in air and in 1 MPa H2 gas

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Symbol</th>
<th>air</th>
<th>Hydrogen</th>
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<tbody>
<tr>
<td>Young’s modulus</td>
<td>$E$</td>
<td>200 GPa</td>
<td>200 GPa</td>
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<tr>
<td>Initial yield stress</td>
<td>$\sigma_0$</td>
<td>220 MPa</td>
<td>184 MPa</td>
</tr>
<tr>
<td>Evolution of back stress</td>
<td>$C_i$</td>
<td>$3.2 \times 10^5$ MPa</td>
<td>$3.2 \times 10^5$ MPa</td>
</tr>
<tr>
<td></td>
<td>$C_2$</td>
<td>$6.8 \times 10^6$ MPa</td>
<td>$9 \times 10^6$ MPa</td>
</tr>
<tr>
<td></td>
<td>$C_3$</td>
<td>$5.3 \times 10^5$ MPa</td>
<td>$5.5 \times 10^5$ MPa</td>
</tr>
<tr>
<td>$\gamma_1$</td>
<td>$2 \times 10^4$</td>
<td>3.2 $\times 10^4$</td>
<td></td>
</tr>
<tr>
<td>$\gamma_2$</td>
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<td>570</td>
<td></td>
</tr>
<tr>
<td>$\gamma_3$</td>
<td>12</td>
<td>25</td>
<td></td>
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<tr>
<td>Evolution of isotropic hardening/softening parameter $R$</td>
<td>$h$</td>
<td>2</td>
<td>4</td>
</tr>
<tr>
<td></td>
<td>$Q$</td>
<td>47 MPa</td>
<td>37 MPa</td>
</tr>
</tbody>
</table>

The values of the remaining parameters $C_2$, $\gamma_1$, $\gamma_2$, and $\sigma_0$ are determined by simulating the experimentally measured stable hysteresis loop shown in Fig. 12 through using Eq. (9). Nonlinear least square approximation yields $C_2 = 6.8 \times 10^6$ MPa, $\gamma_1 = 2 \times 10^3$, $\gamma_2 = 530$, and $\sigma_0 = 220$ MPa; see Appendix B. The calibrated parameters are listed in Table 3. As can be seen, the magnitudes of $C_2$ and $\gamma_1$ are, respectively, one and two orders of magnitude less than the magnitude of $C_1$; and this validates the approximation that $C_1$ controls the slope of the hysteresis loop at zero plastic strain. Similarly the parameters $\gamma_1$ and $\gamma_2$ are large enough so that the assumption $\exp(−γ_1/\epsilon_p)$~0 holds and $C_3$ controls the slope of the hysteresis loop close to $\epsilon_p^{\max}$.

Figure 13 shows the stable hysteresis loop predicted by the model, Eq. (9), for $\epsilon_p^{\max} = \epsilon_p^{\min} = 1\%$ along with the corresponding three back stresses, Eqs. (A7) and (A8), in the absence of hydrogen. The first back stress, $\alpha_1$, starts with a large initial slope and quickly levels to $C_i/\gamma_1$ ($−C_i/\gamma_1$) on the loading (unloading) part of the curve. We note again that the first back stress underpins the segment 1 in Fig. 13. Similarly, the second back stress levels to $C_2/\gamma_2$ ($−C_2/\gamma_2$) on the loading (unloading) part of the curve and underpins the segment 2 in Fig. 13, which is the nonlinear portion of the stable hysteresis curve. The third back stress starts from $C_3/\epsilon_p^{\min}$ ($C_3/\epsilon_p^{\max}$) on the loading (unloading) part and passes through the origin. The third back stress underpins the segment 3 in Fig. 13 which is the linear part of the stable hysteresis curve at large plastic strains.

Interestingly, the calibrated parameters provide fairly good agreement between the model and the experimental data for the hysteresis loop at stabilization also for strain ranges $\Delta \epsilon = 1.08\%$ and 0.92%, as shown in Fig. 14. The agreement is remarkable if one considers that the experimental data from these two tests were not used in the calibration process.

### 4.2 Determination of Parameters $C_1$, $C_2$, $C_3$, $\gamma_1$, $\gamma_2$, $Q$, and $\sigma_0$ in the Presence of Hydrogen.

For the calibration in the presence of hydrogen, we used the experimental data from the stable hysteresis loops shown in Fig. 15 for strain range $\Delta \epsilon = 2.2\%$ in 1 MPa hydrogen gas environment and room temperature. Following the same calibration procedure detailed in Sec. 4.1, we determined $C_1$, $C_2$, $C_3$, $\gamma_1$, $\gamma_2$, $Q$, and $\sigma_0$; results are shown in Table 3. The calibrated model predictions are shown as continuous lines superposed on Fig. 15. Remarkably, the model also reproduces the experimental data for $\Delta \epsilon = 1.08\%$ and 0.92% fairly well.

### 4.3 Determination of the Parameter $h$ in the Absence and Presence of Hydrogen.

According to Eq. (8), the parameter $h$ controls the rate of cyclic hardening, that is, how fast the maximum stress in each cycle changes toward the maximum stress in the stabilized cycle. A smaller $h$ is associated with a larger number of cycles till stabilization, which also implies that the number of cycles for the isotropic hardening (softening) parameter $R$ to increase (decrease) from zero to $Q$ is larger.

Given that $\sigma_{MN} = \sigma_{MN}$, Eq. (14) yields for the maximum stresses $\sigma_{M1}$ and $\sigma_{MN}$, respectively, for the $1^{st}$ and $N^{th}$ cycles at $\epsilon_p^{\max}$:

$$\sigma_{MN} - \sigma_{M1} = R_N - R_1$$

where $R_N$ is the value of the parameter $R$ associated with cycle $N$. With $R_1$ being small in comparison with $R_N$, we find

$$\sigma_{MN} - \sigma_{M1} = R_N$$

Combining Eqs. (16) and (19) yields

$$\frac{\sigma_{MN} - \sigma_{M1}}{\sigma_{M1}} = \frac{R_N}{Q}$$

Given that $\Delta \sigma_p$ is the plastic range of a cycle, the total accumulated plastic strain in cycle $N$ is $\epsilon_p^p = 2N\Delta \epsilon_p$ and Eq. (8) gives

$$R_N = Q(1 - e^{-h(2N\Delta \epsilon_p^p)})$$

Fig. 13 Model prediction of the stable stress–strain hysteresis loop along with the associated back stress $\alpha$ for uniaxial strain-controlled cyclic loading with strain range 2.2% in the absence of hydrogen. The calibrated model parameters are listed in Table 3.
Substitution of Eq. (21) in Eq. (20) provides

$$\sigma_{MN} - \sigma_{M1} = \sigma_{MS} - \sigma_{M1} = 1 - e^{-h2N\Delta e_p} \quad (22)$$

The parameter $h$ is calculated by using Eq. (22) to fit the experimental data for the normalized maximum stress difference $(\sigma_{MN} - \sigma_{M1})/(\sigma_{MS} - \sigma_{M1})$ as a function of accumulated plastic strain. As shown in Fig. 16, a fairly good fitting of the experimental data for $\Delta e_p = 2.2\%$ is obtained with $h = 2$ and $h = 4$ for the uncharged and hydrogen-charged specimens, respectively.

### 4.4 Determination of the Parameter $\gamma_3$ in the Absence and Presence of Hydrogen.

The remaining parameter $\gamma_3$ is determined by using the ratcheting data shown in Fig. 10 obtained from uniaxial stress-controlled cyclic loading tests with stress amplitude $\sigma_m = 420$ MPa at mean stress $\sigma_{mean} = 20$ and 40 MPa. As shown in Fig. 10, immediately upon yielding, a long elongation was found which is behavior associated with rapid onset of Lüders bands. We corrected for this elongation by subtracting the maximum strain of the first cycle, $\varepsilon_{max|N=1}$, from the ratcheting data for all other cycles; the ratcheting strain thus calculated as $\varepsilon_{ratchet} = \varepsilon_{max} - \varepsilon_{max|N_{=1}}$. The data are replotted in Fig. 17.

Using the calibrated model, we carried out finite element simulations of the ratcheting tests by varying $\gamma_3$ and keeping all other parameters fixed so as to best reproduce the ratcheting response. The simulated uniaxial tension domain was loaded incrementally by changing the applied stress from $\sigma_{min} = -400$ MPa to $\sigma_{max} = 440$ MPa for mean stress $\sigma_{mean} = 20$ MPa and from $\sigma_{min} = -380$ MPa to $\sigma_{max} = 460$ MPa for $\sigma_{mean} = 40$ MPa. The finite element calculations were carried under axisymmetric conditions and the Chaboche model as outlined in Sec. 3.1 was integrated by following Hosseini et al. [29]. The best match between simulation and experimental results was obtained when $\gamma_3 = 12$ in the absence of hydrogen and $\gamma_3 = 25$ in the presence of hydrogen.

Figures 17(a) and 17(b) show the comparison between the calibrated model predictions and the experimental ratcheting data in the absence and presence of hydrogen, respectively. The agreement between model predictions and experimental data is deemed to be satisfactory. We note that the ratcheting data vary almost linearly with strain and this linearity is captured by the calibrated model. Lastly, in view of the fact that $\varepsilon_{p, max}^{\text{pp}} = 1.0\%$ for the cycle with $2.2\%$ strain range we find $\gamma_3 \varepsilon_{p, max}^{\text{pp}} = 0.12$ and 0.25 in the absence of hydrogen.
and presence of hydrogen, respectively. It should be remembered that $\gamma_3 \varepsilon_{\text{p}}^{\text{max}} < 1$ is compatible with the requirement of nearly linear variation of the third hardening rule as function of the plastic strain.

5 Discussion

We investigated the effect of hydrogen on the constitutive response of the low carbon steel, SM490YB, by carrying out cycling uniaxial strain-controlled and ratcheting tests in 1.0 MPa H2 gas at room temperature. The hysteresis loops in the absence of hydrogen show that the steel is hardened at $\Delta \varepsilon = 2.2\%$ and softened at 0.92\% and 1.08\%. Hydrogen softened the response for all three strain ranges, although the effect is almost negligible for 0.92\% and 1.08\% (Figs. 6–8). On the other hand, a large hydrogen effect was observed on ratcheting as shown in Fig. 10. We

Fig. 16 Comparison of the model prediction of the normalized maximum stress difference ($\sigma_{\text{MN}} - \sigma_{\text{M1}})/\sigma_{\text{MS}} - \sigma_{\text{M1}}$) with the experimental data for uniaxial strain-controlled cyclic loading with strain range 2.2% in the absence and presence of hydrogen. The parameters $\sigma_{\text{MS}}$, $\sigma_{\text{MN}}$, and $\sigma_{\text{M1}}$ denote, respectively, the maximum stress in the stabilized cycle, cycle $N$, and first cycle.

Fig. 17 Model prediction of ratcheting response with (a) $\gamma_3 = 12$ in the absence of hydrogen and (b) $\gamma_3 = 25$ in the presence of hydrogen. The experimental data were obtained with stress amplitude $\sigma_a = 420$ MPa and mean stress $\sigma_{\text{mean}} = 20$ and 40 MPa.

Fig. 18 Experimentally measured stable hysteresis curves in the absence and presence of hydrogen under uniaxial strain-controlled cyclic loading with strain range 2.2%
simulated the experimental response through the Chaboche model which is so constructed to capture material response in cyclic loading. The model involves nine phenomenological parameters which are identified with features of the experimentally measured response. We proposed a calibration procedure for these nine parameters, and based on the calculated values we attempted to associate the hydrogen effect with standard phenomenological descriptors such as hardening, yielding, and ratcheting.

Figure 18 shows the stable hysteresis curves for the charged and uncharged specimens from strain-controlled tests with strain range 2.2% and for which $\varepsilon^{\text{min}} = -1\%$. The two nearly linear parts of the stable hysteresis curves at large plastic strains whose slope is controlled by the parameter $C_3$, i.e., segment 3, are almost parallel to each other. Hence, the hydrogen effect on the parameter $C_3$ is not significant, and this is shown by the close values $5.5 \times 10^3$ and $5.3 \times 10^3$ MPa, respectively, in the presence and absence of hydrogen. Similarly, looking at segment 1, one argues that there is no hydrogen effect on the parameter $C_1$. The parameter $C_1$ is equal to $3.2 \times 10^5$ MPa in both cases of hydrogen and no hydrogen. In summary, whereas hydrogen has a discernible effect on the parameter $C_2$ (slope of segment 2), and this is shown by the values $9 \times 10^4$ and $6.8 \times 10^4$ MPa, respectively, in the presence and absence of hydrogen, its effect on $C_1$ and $C_2$ is not significant. To conclude, additional experimental data for a range of hydrogen pressures are needed to understand the effect of hydrogen on the parameters $C_1$, $C_2$, and $C_3$.

Figure 18 shows that the maximum stresses in the stable hysteresis curves are 435 MPa and 450 MPa in the presence and absence of hydrogen, respectively. From Eq. (11) along with the fact that $C_1/\gamma_1$ is small compared with $C_2/\gamma_2$, $C_3/\gamma_2$, and $C_3$ are larger in hydrogen than in air, we deduce that the lower maximum stress in the presence of hydrogen is associated with a lower yield stress $\sigma_0$ and $Q$. Indeed, the yield stress $\sigma_0 = 184$ MPa in the presence of hydrogen is lower than $\sigma_0 = 220$ MPa in the absence of hydrogen (see Table 3) by 16.4%. Table 3 also shows that the value of the parameter $Q$ in the presence of hydrogen, $Q = 37$ MPa, is by 21% lower than that in the absence of hydrogen, $Q = 47$ MPa. This 21% lower value of $Q$ in the presence of hydrogen indicates less isotropic hardening. Lastly, the calibrated values of $h$ were 4 and 2 in the presence and absence of hydrogen, respectively. Hence, cyclic hardening in the presence of hydrogen is faster, which is associated with a smaller number of cycles to stabilization.
It is worth pointing out that a marked hydrogen effect was observed on the ratcheting response of the JIS SM490YB steel. The parameter $y_1$ which governs the ratcheting response is 108% larger in the presence of hydrogen, $y_1 = 25$, than in the absence, $y_1 = 12$. Figure 19 shows that hydrogen increases ratcheting significantly, especially at the larger mean stress 40 MPa.

Lastly, Fig. 20 shows the comparison of the model predictions with the experimental response for the transient stress–strain hysteresis loop at strain range 2.2% from the first cycle to the stabilized cycle, i.e., cycles 1, 10, and 100 in air and cycles 1, 10, and 50 in the presence of hydrogen. The agreement can be deemed as good if one considers that the experimental data for cycles 1 and 10 were not used in the model calibration process.

Before closing, we wish to point out that we observed a dependence of the cyclic hardening/softening behavior in the experimental data from strain-controlled tests on strain amplitude. The material of this study, SM490YB steel, is one with an annealed microstructure and one might have expected to cyclically harden at all strain amplitudes. However, testing showed softening for strain ranges 0.92% and 1.08%, and hardening for 2.2%. Softening for the strain ranges 0.92% and 1.08% can be attributed to the continuous increase of the plastically deforming volume of material along the gauge length. However, in the test with the highest strain range, 2.2%, the entire gauge length is already plastically deformed after the first quarter of the first cycle and the subsequent development of the dislocation density and arrangement leads to cyclic hardening. This phenomenon of initial cyclic softening and subsequent hardening at higher strains is well known in low carbon steels where cyclic loading effectively removes the upper yield point and Luders' strains [49]. The latter effects are associated microscopically with the removal under cyclic loading of dislocation pinning by interstitial carbon “atmospheres,” which controls the upper yield point under monotonic loads.

Finally, we note that fatigue is generally regarded as the most ubiquitous mode of failure of industrial structures and components, and similarly hydrogen provides a potent source for the degradation of the structural integrity of innumerable metals and alloys. Accordingly, hydrogen-assisted fatigue in many respects represents the perfect storm, yet despite this, theoretical models for this phenomenon are almost non-existent. To comprehend and model the role of hydrogen in fatigue, it is vital to discern the salient hydrogen-induced damage mechanisms and how these translate to the propagation of a fatigue crack under cyclic loading. The first step to achieving this understanding is to develop a model to describe cyclic plasticity, which is the principal driving force for fatigue crack advance, but specifically in the presence of hydrogen.

This has been the prime objective of this work.

6 Conclusions

The description of cyclic plasticity during fatigue crack growth in the presence of hydrogen requires the development of a constitutive model that accounts for the experimentally observed hydrogen effects on material response in cyclic loading. Toward this goal, the Chaboche model was used to characterize constitutively a low carbon SM490YB in air and in 1 MPa hydrogen gas at room temperature. For the calibration of the model which involves nine parameters, uniaxial strain-controlled cyclic and ratcheting tests were used. From the combined experimental and modeling results, we deduce the following:

(a) This low carbon steel shows a lower yield strength in the presence of hydrogen.

(b) At strain amplitude 2.2%, the amount of cyclic hardening is smaller in the presence of hydrogen. This observation is related to a lower value of the parameter $Q$ in the Chaboche model that is used to describe isotropic hardening. Similarly, cyclic hardening in the presence of hydrogen is faster than that in the absence of hydrogen, which is expressed by a higher value of the parameter $h$.

(c) Ratcheting increases in the presence of hydrogen.

(d) Stronger recovery was found in the presence of hydrogen. This observation is reflected in the model by the higher values of the parameters $y_1$ and $y_2$ in the presence of hydrogen.

(e) The parameters $C_1$ and $C_3$ in the presence of hydrogen are almost the same as those in the absence of hydrogen, whereas the parameter $C_2$ is higher in the presence of hydrogen. Additional experimental data are needed to understand the effect of hydrogen as described by the three kinematic hardening rules of the model.

In summary, the objective of this study was to demonstrate the principle of how a constitutive model can be developed based on experimental data in two environments (air and hydrogen). When such a constitutive model is then applied to simulate fatigue crack growth, then there must be more comprehensive data to ensure the accuracy of the constitutive model parameters. Based on the experimental data and the trends teased out from the calibrated Chaboche model, we may conclude that hydrogen decreases the yield strength and the amount of cyclic hardening. On the other hand, hydrogen increases ratcheting, the rate of cyclic hardening, and promotes stronger recovery.

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Conflict of Interest

There are no conflicts of interest.

Data Availability Statement

The datasets generated and supporting the findings of this article are obtainable from the corresponding author upon reasonable request. The authors attest that all data for this study are included in the paper.

Appendix A

Uniaxial Cyclic Response of a Material Charactized by the Chaboche Constitutive Model. To demonstrate the features associated with the Chaboche model with three kinematic hardening rules, we consider the uniaxial strain-controlled response under fully reversed cyclic loading, i.e., $\epsilon^{p}_{\max} = -\epsilon^{p}_{\min}$. The only nonzero stress component is the axial stress $\sigma_{xx}$ and the matrix of the back stress tensor is

$$[\sigma] = \begin{bmatrix} \alpha_{xx} & 0 & 0 \\ 0 & -\alpha_{xx}/2 & 0 \\ 0 & 0 & -\alpha_{xx}/2 \end{bmatrix}$$

where $\alpha_{xx}$ is the normal component in the loading direction $x$. The component $\alpha_{xx}$ is composed of three parts: $\alpha_{xx} = (\alpha_{xx})_1 + (\alpha_{xx})_2 + (\alpha_{xx})_3$, in which $\alpha_{xx}$ is the normal component of the $i$th back stress in the loading direction. The yielding condition as given by Eq. (3) reduces to

$$f = \lambda (\sigma_{xx} - 3/2 \alpha_{xx}) - \sigma_0 - R = 0$$

where $\lambda$ is the loading/unloading parameter such that $\lambda = 1$ for loading and $\lambda = -1$ for unloading. Setting $\alpha_1 = 3(\alpha_{xx})/2$, we set $3\alpha_{xx}/2 = \alpha_1 + \alpha_2 + \alpha_3 = \alpha$. Then, Eqs. (A2) and (4) reduce,
respectively, to
\[
f = \lambda \sigma_{xx} - \alpha - \sigma_0 - R = 0 \quad (A3)
\]
and
\[
\dot{\alpha}_i = \frac{C_i}{\gamma_i} - \lambda \gamma_i \alpha \dot{R}, \quad i = 1, 2, 3 \quad \text{(no summation over } i) \quad (A4)
\]
Solving Eq. (A4) for the first and second back stress yields
\[
\dot{\alpha}_i = \frac{C_i}{\gamma_i} + \frac{\alpha_0 - \lambda \gamma_i}{\gamma_i} \exp(-\gamma_i (\sigma^p - \sigma^p_{\text{min}})), \quad i = 1, 2 \quad (A5)
\]
where \(\alpha_0\) and \(\sigma^p_0\) are the values of \(\alpha_i\) and \(\sigma^p\) at the start of loading or unloading. We also note that
\[
\alpha_0 = \begin{cases} 
- \frac{C_i}{\gamma_i}, & \sigma^p_0 = \sigma^p_{\text{max}} \quad i = 1, 2 \\
+ \frac{C_i}{\gamma_i}, & \sigma^p_0 = \sigma^p_{\text{min}} \quad i = 1, 2
\end{cases} \quad (A6)
\]
From Eqs. (A5) and (A6), we find
\[
\dot{\alpha}_i = \frac{C_i}{\gamma_i} (1 - 2 \exp(-\gamma_i (\sigma^p - \sigma^p_{\text{min}}))), \quad i = 1, 2 \quad (no \text{ summation over } i) \quad (A7)
\]
The first and second back stresses, \(\alpha_i\), have an initial slope 2\(C_i\) at \(\sigma^p_{\text{min}}\) and level, respectively, to \(C_i/\gamma_i\) with increasing plastic strain, provided that the parameters \(\gamma_i\) are large so that \(\exp(-\gamma_i (\sigma^p - \sigma^p_{\text{min}})) \sim 0\).

The linearity of segment 3 (Fig. 12), which is controlled by the third hardening rule and which should pass through the origin due to the symmetry of the hysteresis loop, is enforced by taking \(\gamma_3\) to be such that \(\gamma_3 \sigma_{\text{max}}^p\) is less than unity. In this case, integration of Eq. (A4) for the third hardening rule \((i = 3)\) yields
\[
\sigma_{xx} = \alpha_3 + \alpha_0 + \lambda_0 \sigma_0 + \lambda Q \quad (A8)
\]
Given that \(R\) levels to \(Q\) at stabilization, substitution of Eqs. (A8) and (A7) in Eq. (A3) provides the closed form solution for the stable hysteresis loop as
\[
\sigma_{xx} = \frac{C_1}{\gamma_1} (1 - 2 \exp(-\gamma_1 (\sigma^p - \sigma^p_{\text{min}}))) + \frac{C_2}{\gamma_2} (1 - 2 \exp(-\gamma_2 (\sigma^p - \sigma^p_{\text{min}}))) + C_3 \sigma^p + \lambda_0 \sigma_0 + \lambda Q \quad (A9)
\]

Appendix B

On the Calibration of the Chaboche Model Parameters \(C_2, \gamma_1, \gamma_2\), and \(\sigma_0\). As presented in Sec. 3, Chaboche’s constitutive model for cyclic plasticity involves nine parameters, \(C_1, C_2, \gamma_1, \gamma_2, \gamma_3, \gamma_0, Q, h, \) and \(\sigma_0\), that need to be calibrated through experiment. The details of the determination of the parameters \(C_1, C_2, \gamma_1, \gamma_2, \gamma_3, \gamma_0, Q, h, \) and \(\sigma_0\) need to be calibrated through experiment. The details of the determination of the parameters \(C_1, C_2, \gamma_1, \gamma_2, \gamma_3, \gamma_0, Q, h, \) and \(\sigma_0\) through using the hysteresis loop from the uniaxial strain-controlled cyclic test are given in Sec. 4. In this appendix, we describe the calibration of the parameters \(C_2, \gamma_1, \gamma_2,\) and \(\sigma_0\).

The closed form solution for uniaxial strain-controlled cyclic loading was found in Sec. 3.2 as
\[
\sigma_{xx} = \frac{C_1}{\gamma_1} (1 - 2 \exp(-\gamma_1 (\sigma^p - \sigma^p_{\text{min}}))) + \frac{C_2}{\gamma_2} (1 - 2 \exp(-\gamma_2 (\sigma^p - \sigma^p_{\text{min}}))) + C_3 \sigma^p + \lambda \sigma_0 + \lambda Q \quad (B1)
\]
where \(\lambda\) is the loading/unloading parameter such that \(\lambda = 1\) for loading and \(\lambda = -1\) for unloading. In this equation, the parameters \(C_1, C_2, Q\), and \(\sigma_0\) are already known from the calibration process discussed in Sec. 4.1. For the calibration of the parameters \(C_2, \gamma_1, \gamma_2,\) and \(\sigma_0\), we used least squares optimization along with Eq. (B1) as follows:

(i) From the stable-stress–strain hysteresis loop data obtained from the uniaxial strain-controlled cyclic loading test, we considered a series of \(n\) stress–strain data \((\sigma^p_i, \varepsilon^p_i)\).

(ii) For a given strain \(\varepsilon^p\), Eq. (B1) provides the model prediction \(\sigma^p_{\text{model}}\). The difference between the model prediction, \(\sigma^p_{\text{model}}\), and the experimentally measured stress, \(\sigma^p_{\text{exp}}\), is the error of the calibrated model prediction. Using the MATLAB optimization toolbox with \(n = 100\), we obtained the parameters \(C_2, \gamma_1, \gamma_2,\) and \(\sigma_0\) by minimizing the sum of the squared errors
\[
SSE = \sum_{i=1}^{n} (\sigma^p_{\text{exp}}(\varepsilon^p_i) - \sigma^p_{\text{model}}(\varepsilon^p_i))^2 \quad (B2)
\]