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A Rationale for Modeling Hydrogen Effects on Plastic Deformation across Scales in FCC metals

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1. Abstract

Although there have been many investigations on the effects of hydrogen on the plastic deformation of metals, an intense debate continues about the physical mechanisms responsible. Most puzzling is the fact that hydrogen appears to be able to both harden and soften FCC metals, depending on the loading conditions. In addition, experiments have shown that hydrogen affects slip system activity differentially, resulting in shear localization of plastic deformation. The work reported in this paper employs a physics-based crystal plasticity model to reproduce the macroscopic response of hydrogen-charged FCC metals through the hydrogen effects on dislocation interactions proposed herein. Different micro-scale mechanisms by which hydrogen may affect plastic deformation are considered, and their resulting macroscopic stress-strain responses under monotonic and cyclic loading are compared. The results support the conclusion that hydrogen screening of dislocations alone cannot explain all the observed macroscopic responses. Instead, it is argued that hydrogen can promote hardening or softening through an increase in glide activation energy and a reduction in dislocation line tension.

2. Introduction

Mechanical failure projections for critical components require an understanding of their mechanical response under various service conditions and in various environments. Most approaches to predict the mechanical response of metals rely on correlations between phenomenological formulations and macroscopic measurements, which require time-consuming experimental programs. Furthermore, the

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predictive power of phenomenological approaches is limited as model fidelity rapidly decreases when the required loading conditions depart from those in the experimental database.

Beside the loading history, the environment plays an important role in determining the mechanical response of metals. In particular, hydrogen absorbed as a result of surface corrosion reactions or available in the surrounding environment is a leading concern in many applications. However, very limited experimental data are available to calibrate phenomenological deformation models for metals with hydrogen content. This dearth of data increases the need for physics-based models that can support confident predictions of metal plastic response beyond limited test data. Developing the needed physics-based models requires a thorough understanding of the mechanisms that lead to plastic deformation and their interaction with hydrogen at multiple scales.

The work herein reported discusses the role of hydrogen on plastic deformation in FCC metals inferred from a wide range of experimental data in the literature and multiscale crystal plasticity models. Our goal is to attribute the macroscopic effects of hydrogen on mechanical properties to specific mechanisms, by informing the effects of hydrogen on micro and mesoscale dislocation interactions.

3. Effect of Hydrogen on Plastic Deformation

The precise mechanisms by which hydrogen affects the deformation and failure of FCC metals are still a matter of conjecture in the scientific community. The Hydrogen Enhanced Decohesion (HEDE) theory (Oriani, 1970) proposes that hydrogen reduces the strength of metallic bonds and promotes cleavage failure. The Hydrogen Enhanced Localized Plasticity (HELP) theory (Birnbaum and Sofronis, 1994; Sofronis et al., 2001) proposes that hydrogen enhances dislocation mobility (Tabata and Birnbaum, 1983) due to the screening effects of hydrogen on dislocations. In reality, both mechanisms may be active: HELP operates in most metals by reducing the ductile fracture resistance, while HEDE reduces the brittle fracture resistance (Gerberich et al., 2008).

In contrast to the lack of consensus for explaining the mechanisms by which hydrogen affects plastic deformation, experiments have shown a few systematic and reproducible effects of hydrogen in many FCC metals, appearing from the microscopic to the macroscopic scales. Most studies to date on the effects of hydrogen on plastic deformation have focused on monotonic loading. For example, Figure 1 presents experimental results from Yagodzinskyy et al., (2008b) that are representative of the effect of hydrogen on FCC single crystals under monotonic loading. In this Figure, hydrogen seems to increase the yield stress and delay the onset of Stage II (Kocks and Mecking, 2003) during

monotonic loading. Similarly, hydrogen in polycrystalline FCC metals increases the yield stress and reduces the initial hardening rate of specimens subject to monotonic loads (Abraham and Altstetter, 1995; 1995; Brass and Chêne, 2006; Yagodzinskyy et al., 2008a).

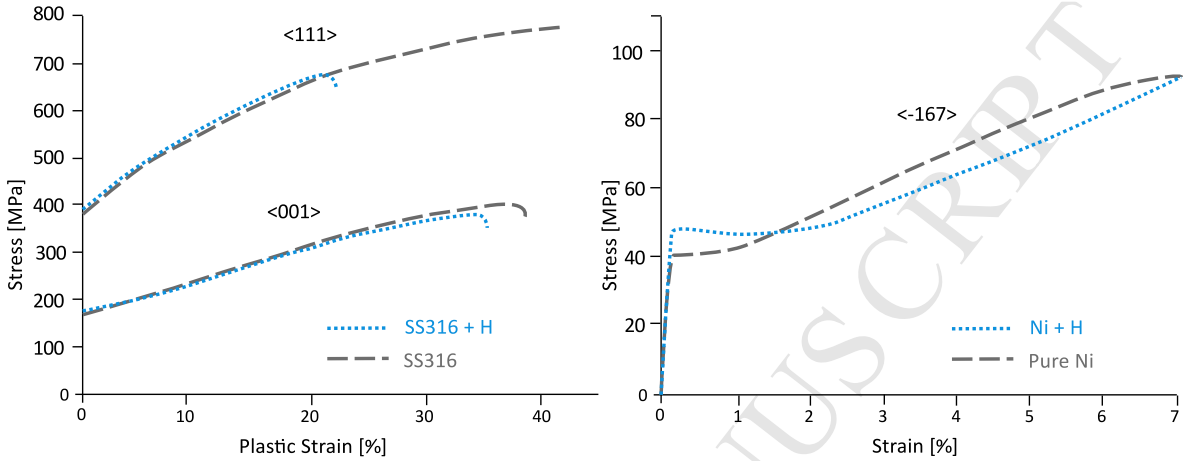


Figure 1. Monotonic stress-strain curves for SS316 (Left) and Ni (Right) single crystals oriented along [167] (Yagodzinskyy et al., 2008b). Gray: Hydrogen-free. Blue: Hydrogen-charged

Magnin et al., (2001) studied the cyclic hardening of single crystal Ni with and without hydrogen (23 ppm and less than 1ppm by weight, respectively). Their results, summarized in Figure 2, show during the first few cycles that under strain control the peak shear stress is higher for the hydrogen-charged specimen (Figure 2, left), which agrees with a higher yield stress and hardening rate for the hydrogen charged samples under monotonic loads. However, upon reaching cyclic saturation, hydrogen charged specimens exhibit lower peak shear stresses than pure Ni. For example, Figure 2 (right) shows the stress amplitude after saturation for different applied plastic strain amplitudes in Ni single crystals. The saturated cyclic stress for the hydrogen charged specimen decreases relative to that in the uncharged specimen for a given plastic shear strain amplitude and both responses exhibit a plateau similar to that in pure FCC metals.

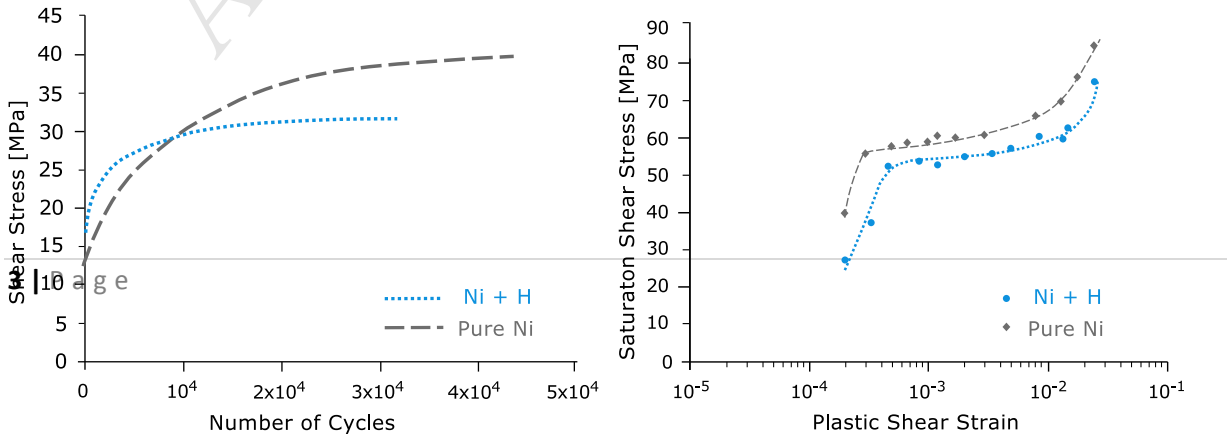


Figure 2. Influence of hydrogen on cyclic hardening curves of a Ni single crystal oriented for single slip (Magnin et al., 2001). (Left) Cyclic Shear stress as a function of number of cycles. (Right) Cyclic stress-strain curve (saturated state).

Mesoscale response

Much research has shown that hydrogen can induce phase transformations, as for example, the austenitic-martensitic transformation. This transformation, which depends on the applied stress, loading rate and temperature, has a significant impact on mechanical properties and the diffusion of hydrogen (Matsunaga and Noda, 2011; Tsong-Pyng and Altstetter, 1986). However, this transformation is usually not seen above room temperature in metallic materials with medium to high stacking fault energies.

Under monotonic loading, when hydrogen-induced martensite is absent, experiments show that mesoscale dislocation structures do not seem to be significantly affected by hydrogen (McInteer et al., 1980; Robertson and Birnbaum, 1984), except for a delay in the onset of multiple slip (i.e., cell structures) in single crystals oriented for single slip (Delafosse, 2012; Girardin et al., 2015). The similarities in dislocation structures with and without added hydrogen under these conditions suggest that the same mechanisms that formed the dislocation structures dominate whether or not hydrogen is present.

Very limited TEM characterization has been performed on single crystal specimens subjected to cyclic deformation in the presence of added hydrogen. However, it is well known that the organization of dislocations in persistent slip band (PSB) structures leads to a stress plateau in single crystals oriented for single slip. Since the morphology of the stress plateau is not affected by hydrogen (Figure 2 right), it is inferred that dislocation structures after cyclic saturation are not strongly altered by hydrogen, although the transients prior to saturation or during progressive directional cyclic strain accumulation ratcheting may be. Elevated hydrogen content may well impact the local stresses and strains, but most likely not the formation of dislocation structures that follow a similitude relation.

Another mesoscale effect induced by hydrogen is the increase in plastic strain localization, which can be quantified by measuring the height and frequency of extrusions at the surface of specimens. Several experimental efforts (Abraham and Altstetter, 1995; Aubert et al., 2012, 2010) have observed evidence of a hydrogen-induced enhancement of plastic deformation. Specifically, they have shown

that hydrogen localizes plastic deformation by producing more intense extrusions, both under monotonic and cyclic loading.

Atomistic mechanisms

The HELP theory, one of the most widely accepted hypotheses for the effect of hydrogen on plastic deformation, proposes that plastic localization arises from the enhancement of dislocation mobility (Tabata and Birnbaum, 1983) due to the screening effects of hydrogen on the forces between dislocations. This concept was supported by TEM analyses (Ferreira et al., 1998; Robertson, 1999), which suggest that in SS310 dislocation spacing in pile-ups are decreased in the presence of added hydrogen.

More recently, atomistic simulations investigating the influence of dissolved hydrogen on dislocations have suggested that the dislocation-hydrogen interaction is too weak to significantly screen the forces between dislocations (Taketomi et al., 2008). Some simulations indeed suggest that over some range of stresses and strain rates, hydrogen may interact with metal vacancies to increase dislocation drag stresses, resulting in hardening (Li et al., 2015). Conversely, under other loading conditions, hydrogen may induce softening due to dislocation breakaway from relatively immobile atmospheres of hydrogen-vacancy complexes (Tehranchi et al., 2017). These contrasting hypotheses may both operate under different loading conditions, suggesting that hydrogen may play a variable, hardening or softening role. There is a clear need for further investigations on the role of hydrogen at the atomic scale.

4. Crystal plasticity model

To identify which mechanisms can potentially explain the effects of hydrogen on macroscopic mechanical properties, we consider a multiscale crystal plasticity model that does not rely on a correlation with macroscopic data, but rather on the parametrization of micro- and meso-scale mechanisms. By explicitly considering parameters that depend only on atomic level and mesoscale attributes, the specific mechanisms are sought which can explain the macroscopic effects of hydrogen on stress and strain.

Hence, the approach introduced earlier by Castelluccio and McDowell (2017) is followed, in which the flow rule specifies the shearing rate of the 12 slip systems in an FCC crystal according to the parametrization proposed by Kocks et al., 1975 , i.e.,

$$\dot{\gamma}^{\alpha} = \rho_m^s \bar{l} b \nu_G \exp\left(-\frac{\Delta G\left(\tau_{eff}^{\alpha}\right)}{k_B T}\right) \mathrm{sgn}\left(\tau^{\alpha} - B^{\alpha}\right). \quad (1)$$

In Equation (1), ρ_m^s is the density of mobile dislocations, $\bar{l}$ is the mean advance distance of a dislocation between obstacle bypass events, b is the Burgers vector ~2.5^{-10} m, and ν_G is an estimate of the attempt frequency ~10^{12} s^{-1}. Also, ΔG is the energy required to bypass dislocation barriers, τ_{eff}^{α} is the effective stress, B^{α} is the directional intragranular contribution to the slip system back stress, k_B is Boltzmann constant, and T is the absolute temperature in Kelvin.

The activation enthalpy, ΔG, pertaining to the mean effective barrier, has been parameterized (Kocks et al., 1975; Kocks and Mecking, 2003) in the form

$$\Delta G\left(\tau_{eff}^{\alpha}\right) = F_0 \left(1 - \left[\frac{\tau_{eff}^{\alpha}}{s_t^0 \frac{\mu}{\mu_0}}\right]^p\right)^q \quad (2)$$

Here F_0 and p, q are the activation energy and profile parameters, respectively, for dislocation barrier bypass at τ_{eff}^{α}=0, s_t^0 is thermal slip resistance at 0 K, and μ and μ_0 represent the shear modulus (C_{44}) at temperatures T and 0K, respectively. In addition, the effective stress that drives the barrier bypass at the slip system level is given by,

$$\tau_{eff}^{\alpha} = \left\langle \left| \tau^{\alpha} - B^{\alpha} \right| - S^{\alpha} \right\rangle \quad (3)$$

where τ^{α} is the local slip system resolved shear stress and S^{α} is the non-directional threshold stress. The Macaulay brackets are defined as follows: $\langle g \rangle$ = 0 if $g \le 0$, $\langle g \rangle$ = g if g > 0.

The threshold stress required to bow a dislocation against a dislocation pile-up between dislocation walls is given by

$$S^{\alpha} = S_0^{\alpha} + \alpha_{LE} \frac{\mu b}{(1 - f_w) d_{struct}} + \mu b \left(A_{ii} \rho^{\alpha}\right)^{1/2}, \quad (4)$$

in which S_0^α corresponds to an intrinsic lattice friction stress, the second term corresponds to the stress required to bow out a dislocation, which depends on the dislocation line energy and dislocation wall fraction (f_w); and the third term corresponds to the strength interaction between collinear dislocations in pile-ups, which depends on the strength of the self-interaction coefficient, A_{ii} .

The back stress evolution is based on an Eshelby mean field approach that incorporates dislocation substructure effects (Castelluccio and McDowell, 2017),

$$\dot{B}^\alpha = \frac{f_w}{1-f_w}\frac{2\mu(1-2S_{1212})}{1+4S_{1212}\mu f_{Hill}^S}\dot{\gamma}^\alpha \tag{5}$$

Here,

$$S_{1212} = \frac{\pi\eta^2 + (\eta^2 - 1.75 - 2\nu_p\eta^2 + 2\nu_p)C_{12}}{8\pi(1-\nu_p)(\eta^2-1)} \tag{6}$$

$$C_{12} = \frac{2\pi\eta\left(\eta\sqrt{(\eta^2-1)} - \cosh^{-1}(\eta)\right)}{\sqrt{(\eta^2-1)^3}} \tag{7}$$

$$\nu_p = \frac{\nu + \frac{2}{3}\mu(1+\nu) f_{Hill}^S}{1+\frac{4}{3}\mu(1+\nu) f_{Hill}^S}. \tag{8}$$

in which η corresponds to a normalized distance that a dislocation can glide along the dominant slip system relative to the distance between walls.

This back stress evolution relation represents the homogenized response of a quasiperiodic dislocation structure that induces residual stress in the material due to the coexistence of a plastically deformable matrix surrounded by an essentially linear elastic structure of dense dislocation walls.

The mean free path for dislocation glide, $\bar{l}$, depends on the local strain range and it is computed following the scheme proposed by Castelluccio and McDowell, 2017. Thus, the mean advance distance is determined by identifying the appropriate dislocation mesoscale substructures.

$$\bar{l} \approx l_{struct} = \eta d_{struct} \tag{9}$$

Here, d_{struct} is the characteristic dislocation substructure length scale that follows similitude (Kuhlmann-Wilsdorf, 1962; Sauzay and Kubin, 2011), i.e.,

$$d_{struct} = \frac{K_{struct}\mu(T)b}{\tau}. \tag{10}$$

Sauzay and Kubin, (2011) discussed similitude relations and showed that the constant, K_{struct}, ranges from 2 to 4 for a large number of metals and a wide range of cyclic loading conditions.

Another characteristic distance associated with the dislocation structure is the wall thickness, which along with the structure spacing and length, determines the wall volume fraction, f_w. We employ a parametrization of f_w based on the maximum plastic shear strain range per cycle among all slip systems, which resembles the approach by Estrin et al., 1998, i.e.,

$$f_w = f_{inf} + \left(f_0 - f_{inf} \right) \exp\left(\frac{-\Delta\gamma^{\max}/2}{g_p} \right) \tag{11}$$

where, f_{inf}, f_0, and g_p are material constants that can be estimated by computing f_w from TEM images.

The mobile dislocation density on slip system α results from a balance between dislocation multiplication, annihilation and cross slip (Arsenlis and Parks, 2002; Essmann and Mughrabi, 1979; Patra and McDowell, 2012), i.e.,

$$\dot{\rho}_m^{s\alpha} = \dot{\rho}_{mult}^{s\alpha} - \dot{\rho}_{annih}^{s\alpha} + \dot{\rho}_{cs}^{s}\Big|^{\zeta\to\alpha} - \dot{\rho}_{cs}^{s}\Big|^{\alpha\to\zeta} \tag{12}$$

The multiplication rate of mobile screw dislocations for slip system α is computed as

$$\dot{\rho}_{mult}^{s\alpha} = \frac{k_{multi}}{b l_{struct}} \left| \dot{\gamma}^{\alpha} \right| \tag{13}$$

Here $k_{multi}=1$ for cell structures and $k_{multi}=2$ for parallel walls such as PSBs and labyrinth. The annihilation rate of mobile screw dislocations is given by

$$\dot{\rho}^{s}_{annih}{}^{\alpha}=\frac{2y_s^{edge}}{b}\rho_m^{s\alpha}\left|\dot{\gamma}^{\alpha}\right|-\Upsilon\frac{2\left|\dot{\tau}^{\alpha}-\dot{B}^{\alpha}\right|}{\mu b d_{struct}}\Bigg|^{Unload}, \tag{14}$$

where $\Upsilon=1$ if $\mathrm{sgn}\left(\dot{\tau}^{\alpha}-\dot{B}^{\alpha}\right)\neq\mathrm{sgn}\left(\tau^{\alpha}-B^{\alpha}\right)$ or $\dot{\gamma}^{\alpha}=0$; otherwise $\Upsilon=0$ and $\dot{\rho}^{s}_{annih}{}^{\alpha}\Big|^{Unload}=0$. The first term considers the annihilation mechanisms that involve plastic deformation without cross slip (e.g., edge dislocation annihilation), and the second term in Equation (14) represents the annihilation of dislocations due to anelastic deformation upon reversing the local shear stress.

The rate of cross slip of mobile screw dislocations from plane α to ζ $\left(\dot{\rho}^{s}_{cs}\Big|^{\alpha\to\zeta}\right)$ is estimated using the probabilistic formulation proposed by several researchers (Kubin et al., 1992; Verdier et al., 1998; Zhou et al., 2010), but considering the local shear stress to account for screening effects from dislocation structures, i.e.,

$$\dot{\rho}_{cs}^{s\alpha}=-\nu_G\frac{l_{CS}}{d_0}\rho_m^{s\alpha}e^{\left(-V_{CS}\frac{\tau_{III}-\left|\tau^{\zeta}-B^{\zeta}\right|}{k_BT}\right)}. \tag{15}$$

Here, ν_G and $l_{CS}\approx d_{struct}$ are the cross slip characteristic frequency and dislocation length, respectively, τ_{III} is the critical cross slip stress, and $|\tau^{\zeta}-B^{\zeta}|$ is the magnitude of the local stress on the cross slip plane of a screw dislocation in system α. The cross slip activation volume, V_{CS}, depends on the dislocation constraints, and its statistical average has been shown to be inversely proportional to the slip system shear stress, i.e.,

$$V_{CS}^{\alpha}=\frac{V_{CS}^{0}}{\left|\tau^{\alpha}\right|}, \tag{16}$$

with $V_{CS}^{0}\approx 1000b^{3}MPa$ (Armstrong and Rodriguez, 2006). The critical cross slip stress, τ_{III}, which has been estimated between 30 to 80MPa (Armstrong and Rodriguez, 2006; Mitchell, 1964; Zhou et al., 2010) for Ni, is related to the location of the cyclic shear stress-strain plateau, which is at about 50 MPa

(Blochwitz and Veit, 1982; Buque, 2001). Here we compute the critical cross slip stress value following Brown (Brown, 2002), i.e.,

$$\tau_{III} = \frac{\mu b}{4\pi y_s^{screw}} \tag{17}$$

where y_s^{screw} corresponds to the annihilation distance of screw dislocations, which is controlled by the activation energy of cross slip.

In summary, the evolution of screw dislocation density is written as

$$\dot{\rho}_m^{s\alpha} = \frac{k_{multi}}{b l_{struct}} \left|\dot{\gamma}^{\alpha}\right| - \frac{2 y_s^{edge}}{b} \rho_m^{s\alpha} \left|\dot{\gamma}^{\alpha}\right| - \frac{2\pi}{\mu b d_{struct}} \left|\dot{\tau}_{eff}\right|^{Unload} + \nu_G \frac{d_{struct}}{d_0} \left(\phi_{CS} \sum_{\zeta=1(\zeta\neq\alpha)}^{N} \rho_m^{s\zeta} e^{\left(-V_{CS}^{\alpha} \frac{\frac{\mu b}{4\pi y_s^{screw}} - \left|\tau^{\alpha} - B^{\alpha}\right|}{k_B T} \right)} - (1-\phi_{CS}) \sum_{\zeta=1(\zeta\neq\alpha)}^{N} \rho_m^{s\alpha} e^{\left(-V_{CS}^{\zeta} \frac{\frac{\mu b}{4\pi y_s^{screw}} - \left|\tau^{\zeta} - B^{\zeta}\right|}{k_B T} \right)} \right) \tag{18}$$

Hypotheses of Hydrogen Effects on Plastic Deformation

As discussed before, hydrogen seems to affect local stresses and strains, with less of an effect on the formation of dislocation structures that follow similitude. We proceed with the hypothesis that hydrogen affects mostly the behavior of dislocations at the atomistic level with negligible effects on the mesoscale structure. Therefore, our simulations will consider precisely the same mesoscale attributes as those in Castelluccio and McDowell, 2017, and we will assume that the role of hydrogen is only to affect parameters associated with atomic scale unit processes. A fully hydrogen-dependent crystal plasticity model would require atomistic parametrizations that depend on the local concentration of hydrogen and vacancies. Here, we proceed by introducing effects of hydrogen on model parameters in order to qualitatively match the trends produced by added hydrogen on the mechanical response.

Since there is no consensus on the role of hydrogen at the scale of atomistically resolved unit process for dislocations, we identified a few physically-based cases that could explain much of the experimental literature from the microscopic scales. These scenarios are as follows:

- Case A follows the argument that hydrogen screens forces between dislocations, reducing the strength of dislocation interactions. Hence, we propose that hydrogen reduces the strength of the self-hardening coefficient (A_{ii}) in Equation (4), which could explain the hydrogen-induced reduction in dislocation spacing in pile-ups. Given the negligible screening effect shown by atomistic simulations, we expect that such a reduction should be modest, so our initial guess is that A_{ii} should be on the same order of magnitude regardless of hydrogen content. Hence, we propose that hydrogen reduces A_{ii} by 25% in all slip systems to match Figure 1 and Figure 2. Note that this assumed reduction follows from the explanation proposed to justify the change in dislocation spacing within pile-ups observed in TEM analyses (Ferreira et al., 1998; Robertson, 1999), but it is not predicted by atomistic simulations.
- Case B assumes that hydrogen increases the drag stress opposing dislocation motion by virtue of increasing the activation energy (F_0) and thermal stress (s_t^0) in Equation (2), as suggested by atomistic simulations. Furthermore, based on the experimental observation of an increase in bow-out diameter found by Miyata (2003), and theoretical arguments regarding atomic segregation to defects (Barnoush et al., 2012; Barnoush and Vehoff, 2010; Kirchheim, 2007), it is argued that hydrogen reduces the stress to bow out a dislocation due to a reduction in the line energy of edge dislocations. Hence, in Case B an increase was proposed of 10% in both F_0 and s_t^0, and a 15% reduction in the line tension coefficient, α_{LE}, in Equation (4). In addition, it was assumed that hydrogen reduces the annihilation distance of screw dislocations by 30%. These combined effects of hydrogen on material parameters were applied uniformly to all slip systems.
- Case C is similar to Case B, except that the effects of hydrogen assumed in case B are confined exclusively to the most active slip system, while the parameters on the remaining slip systems remain unchanged. In so doing, we argue that hydrogen will be preferentially trapped in the vicinity of vacancies, which are a by-product of plastic deformation. Hence, at relatively low temperatures at which vacancy mobility is limited, it is likely that vacancy concentration is higher along the most active slip planes where most of them originated. By associating the most

active slip system with the strongest hydrogen effects, one can explore the potential role of vacancy-induced localization of strain.

These cases were implemented in finite element simulations, and their results were compared.

5. Constitutive model implementation

Following the methodology introduced earlier by Castelluccio and McDowell, 2017, the constitutive model described herein was implemented using an ABAQUS UMAT and UEXTERNALDB subroutines (*FEM software 2016, Simulia Corp., Providence, RI, USA*, 2016). Simulations employed a simple smooth mesh containing 500 "brick" (C3D8R) elements. Displacements are specified along an axis on which periodic boundary conditions are imposed while the other faces of the mesh are unconstrained. Mesoscale structures are determined based on the plastic shear ranges per cycle at the slip system level. Further details about meshes, time scale decoupling, and model implementation may be found in Castelluccio and McDowell, (2017).

Material parameters and physical constants

Different types of parameters important in crystal plasticity should be differentiated: First, direct parameters are considered, which are associated with intrinsic physical properties related to the crystal lattice and to lattice defect interactions, and which do not depend on the mesoscopic response of a statistical ensemble (e.g., temperature, elastic potential). These parameters are presented in Table 1.

Table 1. Direct parameters for pure Ni.

T	ν_G	b	C_{11}	C_{12}	C_{44}	C_{44} @0K	μ
300K	$10^{12}s^{-1}$	$2.5x10^{-10}$	*249GPa*	*155GPa*	*114GPa*	*127.6GPa*	*80.6GPa*

Additional parameters are associated with mechanisms or theoretical models that describe the behavior of unit processes related to dislocation statistical ensembles. The values related to atomistic scale unit process mechanisms are presented in Table 2. Those associated with mesoscale structures appear in Table 3. Table 4 summarizes the values employed to initialize internal state variables.

Finally, Table 5 presents the changes proposed to various parameter values in order to reproduce the effects of hydrogen on slip distributions for Cases A, B. and C.

Table 2. Parameters related to atomistic scale unit processes for pure Ni.

F_0	s_t^0	P	q	y_e	y_s	S_0^α	A_{ii}	α_{LE}	V_{CS}^0	ϕ_{CS}
0.985ev	*100MPa*	*0.9*	*1.5*	*3nm*	*13nm*	*4MPa*	*0.1*	*1*	*1000b3*	*0.5*

Table 3. Parameters associated with mesoscale dislocation structures for pure Ni.

η_{cell}	η_{labyr}	η_{PSB}	η_{veins}	K_{struct}	k_{multi}	k_{multi}^{Cell}	f_{inf}	f_0	g_p
1	*2*	*20*	*50*	*3.75*	*2*	*1*	*0.24*	*0.4*	*1.7*

Table 4. Initial values for internal state variables for pure Ni

ρ_m^s	d_{struct}	η	η_{veins}	f_w^0	V_{CS}
$10^{-10}m^{-2}$	$10\mu m$	η_{veins}	*50*	*0.25*	$1000b^3$

Table 5. Effect of hydrogen on Ni

Case A	*Cases B and C*			
A_{ii}	α_{LE}	F_0	s_t^0	y_s
0.075	*0.85*	*1.08ev*	*110MPa*	*10nm*

6. Simulation Results

Figure 3 compares the stress-strain response of a single crystal of Ni under monotonic straining, with and without added hydrogen for Model Cases A and B. The constitutive model for monotonic loading is similar to that described for cyclic loading (Castelluccio and McDowell, 2017), except that $\eta_{struct} \approx 1$,

$K_{struct} \approx 12$, and the wall thickness evolution behavior is taken from Estrin et al. (Estrin et al., 1998). The simulations for pure Ni match the experimental results in Figure 1 in terms of the yield strength and hardening slope. They slightly underestimate the extent of Stage I deformation and the corresponding delay in the activation of multiple slip (which depends on the mesoscale structures). Full validation of the monotonic response model requires the quantification of the mean free glide path of dislocations. However, this is not necessary for determining the yield strength of a pristine annealed metal with low initial dislocation density, effectively turning the grain size the mean free path.

The effect of added hydrogen on yield strength seems to exhibit opposite trends in Cases A and B. Case A predicts a slightly lower yield stress for hydrogen-charged Ni and a significantly lower hardening rate beyond yield, compared to those for Ni without added hydrogen. On the contrary, Case B predicts an increase in the yield stress and a reduction in the hardening rate for hydrogen-charged Ni compare to pure Ni (note the cross over at 7% strain). Compared to the macroscopic experimental results in Figure 1, only Case B correctly reflects the observed qualitative effects of hydrogen on deformation under monotonic loading.

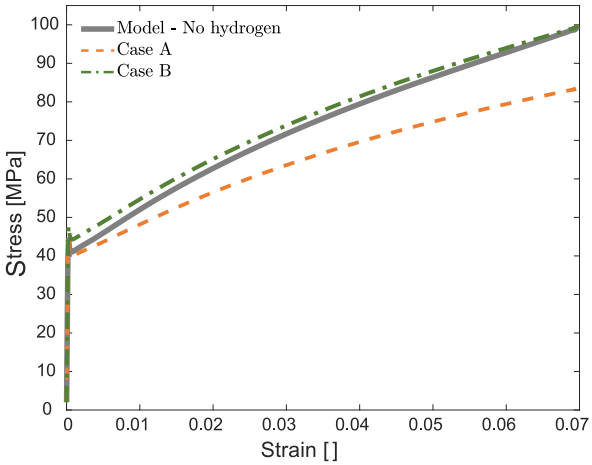


Figure 3. Monotonic stress-strain curve from models and experiments (Yagodzinskyy et al., 2008b), for single crystals loaded along the [167] direction at a strain rate of $9x10^{-5}s^{-1}$.

Figure 4 presents the cyclic stress-strain curves for the Case A and Case B simulations for pure Ni with and without added hydrogen. Just as was shown in previous work (Castelluccio and McDowell, 2017), the current material model can reproduce the response of single crystals of pure

Ni. Both Cases A and B predict a lower cyclic stress amplitude in strain-controlled tests in hydrogen charged samples, which agrees with the softening observed in experiments (Figure 2).

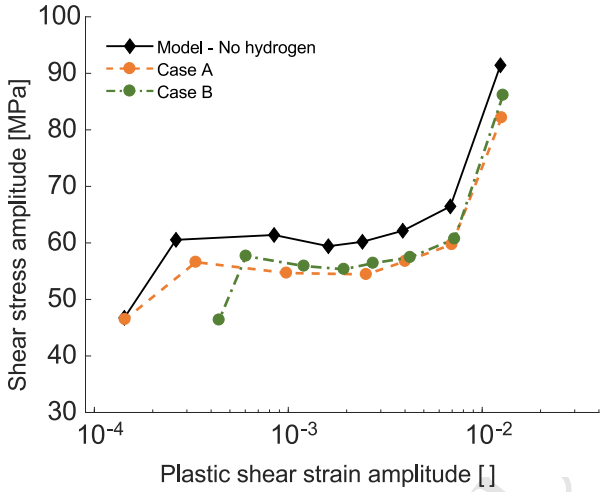


Figure 4. Cyclic stress-strain curve comparison among models with (Cases A and B) and without hydrogen for single crystals under completely reversed strain-controlled uniaxial loading ($R_\varepsilon = -1$) along the [153] direction.

Given that the Case A and Case B simulations give almost indistinguishable results under cyclic loading (Figure 4) and they agree with cyclic experiments (Figure 2), we seek to understand whether both cases would suggest an increase in mesoscale strain localization. Since we do not explicitly model the formation of extrusions from slip bands (which would require very detailed meshes and accounting from point defects (Repetto and Ortiz, 1997)), it is not possible to quantify strain localization from the extrusion height. Instead, we propose to evaluate strain localization by comparing the shear strain range per cycle among all slip systems. Evidence of localization of plastic deformation would manifest in the form of an increase in the shear strain range on the most dominant slip system, and reductions of slip activity on all other slip systems.

Figure 5 presents histograms of shear strain range from the last loading cycle for each of the slip systems, as calculated for pure Ni using the Case A and Case B material models within 500 elements. For clarity, only shear strain ranges larger than $1x10^{-10}$ are presented. This value should be regarded as a threshold for onset of plastic deformation. Hence, slip systems 4, 9, 10 and 11 are seen not to be active, while only a few elements show some activity on slip systems 1, 2, 8 and 12. As expected from the Schmid factor for this crystal orientation, slip system 6 dominates plastic deformation, followed by slip system 3.



0.00293



0.00089



0.00309



0.00084



0.00335



0.00056



0.00394



0.00042

combination of an increase in activation energy for dislocations to glide; a reduction of the dislocation line energy, which reduces the stress required for edge dislocations to bow out; and the localization of these effects on the most active slip systems.

7. Discussion

Physics based constitutive models typically require the separate determination of numerous parameter values that inform deformation mechanisms at multiple scales. A comprehensive model validation program would consequently require the quantification of parameters at lower scales with a combination of bottom-up models and experiments, in a manner independent of the macroscopic response of the material. Fitting of model parameter values directly to macroscale data should be performed as a consistency check and validation of the values determined by independent means. Small adjustments to the independently obtained parameter values may then be considered on an informed basis. The engineering trades would need to be evaluated between optimizing the fit of the model to existing data, versus maximizing the predictive value of the model for data outside the range of loading and material conditions in the database.

Fitting parameters to a macroscopic response without an independent estimation from lower scales may result in fortuitous cancelation of offsetting errors in model inputs that may no longer offset under different modeling conditions. This risk is particularly important in crystal plasticity models, which require dozens of experiments to unambiguously determine all parameters. A clear example of this risk is presented here by comparing Case A, B and C, all of which can adequately reproduce the softening induced by hydrogen under cyclic loading, but Case A cannot reproduce the proper response under monotonic loading or plastic localization.

Our systematic construction of the crystal plasticity model relies on proposing mechanisms validated at various scales. For instance, atomistic models suggest which parameters should be sensitive to hydrogen, whereas dislocation dynamics and TEM analyses show the sensitivity of dislocation arrangements to hydrogen concentration. Furthermore, the comparison with macroscopic single crystal experiments mitigates the uncertainty introduced by polycrystalline variability. Therefore, the current approach provides a basis for a robust attribution study of the mechanisms responsible for the macroscopic response of hydrogen.

The comparison of Cases A, B and C are consistent with the finding from atomistic simulations that hydrogen does not significantly screen forces between dislocations in FCC metals. Although this conclusion is in apparent contradiction with the conclusions of Ferreira and Robertson based on their TEM studies (Ferreira et al., 1998; Robertson, 1999), it is argued that the observed reduction in dislocation spacing within a pile-up is not due to a screening effect, but due to an increase in the resulting local stress. Indeed, these TEM in-situ experiments can be roughly classified as displacement-controlled rather than force- or stress-controlled. Hence, the final positions of dislocations within a pre-existing pile-up depend on the kinematic configuration applied. If hydrogen interacts with vacancies in such a way that it increases the drag stress opposing dislocation glide, that interaction would also increase the nominal stress, and in consequence, pile up dislocations more closely. This mechanism could be clarified by measuring the applied in-situ force, which has not been reported. Furthermore, softening effects from the reduction of line tension would not be observable in a pre-existing pile-up.

Hydrogen effects on the dislocation glide activation energy also explain the increase in yield stress under monotonic loading. Initial yielding is controlled by the glide of pre-existing dislocations, which must overcome greater resistance in the presence of hydrogen. Upon further deformation after the initial yield, continued plastic deformation relies on the proliferation of dislocations, a process facilitated by hydrogen through its reduction in dislocation line energy. Therefore, this effect delays the hardening of the material, extends Stage I deformation under monotonic loading, and reduces the saturated stress under cyclic loading. Further research is required to understand the atomistic scale mechanisms that lead to a reduction in line energy, which is a mesoscale concept useful for parameterizing dislocation curvature. In addition, other considerations such as dynamic strain aging may contribute at different temperatures and deformation rates.

8. Conclusions

This work employs a crystal plasticity model informed by independent studies at multiple scales to understand the effects of hydrogen on plastic deformation. Three model cases were analyzed, which are based on physical mechanisms that have been inferred from experiments to explain the macroscopic response of FCC metals under the influence of added hydrogen. First, a reduction of the strength (i.e., hydrogen screening) of dislocation interactions is considered. Second, the results are evaluated of a proposed simultaneous increase in the glide activation energy and a reduction in the dislocation line

tension. Finally, the localization of hydrogen on the dominant slip system is superimposed on the previously hypothesized effects.

The results of the current analysis are consistent with those of atomistic simulations and suggest that hydrogen would not significantly affect the strength of the interaction among dislocations. Instead, the increase in glide activation energy may explain an increase in yield strength under monotonic loading, while a hydrogen-induced reduction in line tension may explain a reduction in stresses at saturation under cyclic loading. Furthermore, a hypothesized localization of hydrogen near dislocation cores gliding on the most active slip system may induce additional softening. Future work should investigate closely the effect of hydrogen-vacancy interactions in relation to bow-out multiplication of dislocations and localization of plasticity.

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Highlights

1. Modeling the effect of Hydrogen on the mechanical response of Ni single crystals.
2. A physics-based constitutive model for FCC metals informed at multiple scales.
3. A rationale to understand the mechanisms by with hydrogen affects plastic deformation.
4. A single crystal plasticity model that can predict the increase in yield stress, softening under cyclic loading and strain localization induced by hydrogen, all compared to experiments.

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A rationale for modeling hydrogen effects on plastic deformation across scales in FCC metals

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