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Discrete Dislocation Plasticity Modeling of Hydrides in Zirconium under Thermal Cycling

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ABSTRACT

Understanding the ratcheting effect of hydrogen and hydride accumulation in response to thermal cycling is important in establishing a failure criterion for zirconium alloy nuclear fuel cladding. We propose a simple discrete dislocation plasticity model to study the evolution of the dislocation content that arises as a micro-hydride repeatedly precipitates and dissolves over a series of thermal cycles. With each progressive thermal cycle, we find a steady growth in the residual dislocation density in the vicinity of the hydride nucleation site; this corresponds to a gradual increase in the hydrogen concentration and, consequently, the hydride population. The simulated ratcheting in the dislocation density is consistent with experimental observations concerning the hysteresis in the terminal solid solubility of hydrogen in zirconium, which can be correlated to the plastic relaxation of hydrides.

INTRODUCTION

Zirconium alloys are widely used as nuclear fuel cladding in various reactor designs but they are susceptible to a time-dependent failure mechanism known as delayed hydride cracking (DHC). Since the earliest commercial reactors, intensive research has been undertaken to address the risk of DHC failure as it has long-term consequences on both safety and economics in the nuclear industry. In the reactor environment, zirconium components may undergo an aqueous corrosion process that leads to the ingress of hydrogen. The hydrogen atoms diffuse under gradients of stress, temperature and concentration to accumulate ahead of stress-raisers such as cracks or notches. In these hydrogen-enhanced regions, zirconium hydride precipitates are nucleated if the critical solvus is exceeded. The hydride phases are significantly more brittle than the parent metal, which has deleterious effects on the mechanical properties of the zirconium components. Throughout its typical operation lifetime, the zirconium cladding is aged by the moderately high-temperature, high-pressure and high-neutron flux conditions; in particular, the aggravating temperature and loading regimes can instigate DHC by causing an increase in the brittle hydride population in the vicinity of the crack. The reduction in the fracture toughness ultimately leads to crack propagation in the hydride-rich zone. In the nuclear community, DHC universally serves as an umbrella term for the complexity associated with the repetition of the multiscale processes of atomic diffusion, microscale precipitation and fracture, as well as the coupled sub-processes therein, which can compromise the structural integrity of the zirconium alloy cladding [1–4].

Continuum scale multi-field modelling is most routinely used in industrial practice to include the effect of DHC on the failure criterion for cladding. In such multi-field methodologies, the hydride volume fraction is represented by a spatial field, which is governed

by partial differential equations [2]. The hydrides are, therefore, not treated explicitly. Moreover, one of the most important aspects of safeguarding against DHC failure is the understanding of thermal ratcheting. Over a series of thermomechanical cycles during which hydrides dissolve and precipitate, the hydride volume fraction tends to increase; this corresponds to a greater possibility of DHC initiation [1,2]. In continuum scale modeling, the ratcheting process is phenomenologically incorporated into the governing equations. Most simply, ratcheting is described by assuming a two-solvus system for hydrogen [2]. The terminal solid solubility (TSS) of hydrogen in zirconium defines the concentration and temperature boundary for phase transition from hydrogen in solid solution to matrix plus hydride phases. The premise of a two-solvus system originates in experimental studies demonstrating a hysteresis in the TSS between dissolution (TSSd) on heating, and precipitation (TSSp) upon cooling of around 60K [5]. As the diffusible hydrogen concentration is always predefined to be greater at a given temperature for TSSp than for TSSd, inputting the hysteresis naturally results in a growth in the hydride volume fraction over subsequent thermal cycles [2]. However, more fundamentally, it has been postulated that the TSSd/p hysteresis is connected to the dislocation-based plastic relaxation of newly formed misfitting hydrides [5,6]. Some dislocations punched out by precipitates to relax the elastic stresses remain in the matrix when precipitates are dissolved on heating. These dislocations trap hydrogen and hence the overall hydrogen content increases with each cycle. Only a discrete dislocation treatment can capture the trapping and thereby explain the ratcheting.

The length scale of nucleation sites can be inferred from micrographs of typical hydride morphologies, which show the existence of large macroscopic hydride structures that can extend to be hundreds of microns in length [1,3]. On closer inspection, these macro-hydrides are actually assemblies of small microscopic hydride packets whose geometry resembles that of platelets or needles with lengths on the order of a micron [4,6]. So the substructure of the macro-hydrides consists of sequential layers of parent metal and hydride phases; the micro-hydride stacking is attributed to an autocatalytic nucleation process [1,4,7]. Hence, the length scale of interest for investigating nucleation is intermediate to those of the macro- and micro-hydrides.

The micro-hydride particles can be treated as pure hydride phases, which themselves can be differentiated by their respective stoichiometric and crystallographic properties as well as their preferred habit planes [1]. Transmission electron microscopy of bulk hydrides shows the existence of dislocation networks around needle-shaped γ -hydrides residing on the (0001) basal planes [6]. This local dislocation microstructure is presumed to accompany the accommodation of hydride volume misfit within the α -phase matrix. The generation and motion of these dislocations, that is, the punch-out of prismatic loops implies the dissipation of plastic strain energy into the zirconium matrix. The non-recoverability of this strain energy is hypothesized to be the reason for the TSS hysteresis [1,2,5]. Supporting this theory is the observation that highly irradiated materials exhibit a greater hysteresis; conversely, annealing out irradiation damage shifts the TSSd/p values to their unirradiated counterparts [5]. Therefore, given that dislocations play a significant role in DHC, it is imperative to establish a framework to study the dislocation microstructure around a hydride needle subject to thermal cycling.

In this study, we investigate the origin of the TSS hysteresis between precipitation and dissolution and the evolution of the dislocation microstructure at the scale of a single nucleation site. We do so using a simplistic model of hydrides embedded within a 2D microscale modeling technique known as discrete dislocation plasticity (DDP) [8] to definitively show that a residual strain energy remains after precipitate dissolution, making subsequent precipitation more likely.

THEORY

In the following, we present a simple planar DDP methodology to demonstrate plastic relaxation associated with hydride precipitation and the non-recoverability of this plastic strain energy upon hydride dissolution. As this work concerns bulk hydrides, it is justifiable to suppose an infinite continuum but also restrict our attention to the scale of an individual nucleation site by working within a $5\mu\text{m} \times 5\mu\text{m}$ simulation cell around one micro-hydride platelet. Following the work of Perovic [7], the strain fields of a micro-hydride can be approximated by a prismatic dislocation loop whose Burgers vector is chosen to mimic the elastic misfit. In the planar DDP framework, this approximation translates to an edge dislocation dipole in an interstitial loop configuration as shown in Figure 1. Supposing a coherent interface and surface diffusion of hydrogen atoms to and from the long edges of the hydride-matrix interface, it is possible to depict the hydride thickening (or thinning) by increasing (or decreasing) the hydride Burgers vector b_H . Corresponding to the size of experimentally observed micro-hydrides, the hydride length L_H is assumed to be $1\mu\text{m}$ and the hydride is assumed to grow to a maximum thickness of $|b_H|^{\text{max}} = 100b$ where b is the Burgers vector of prismatic edge dislocations in zirconium.

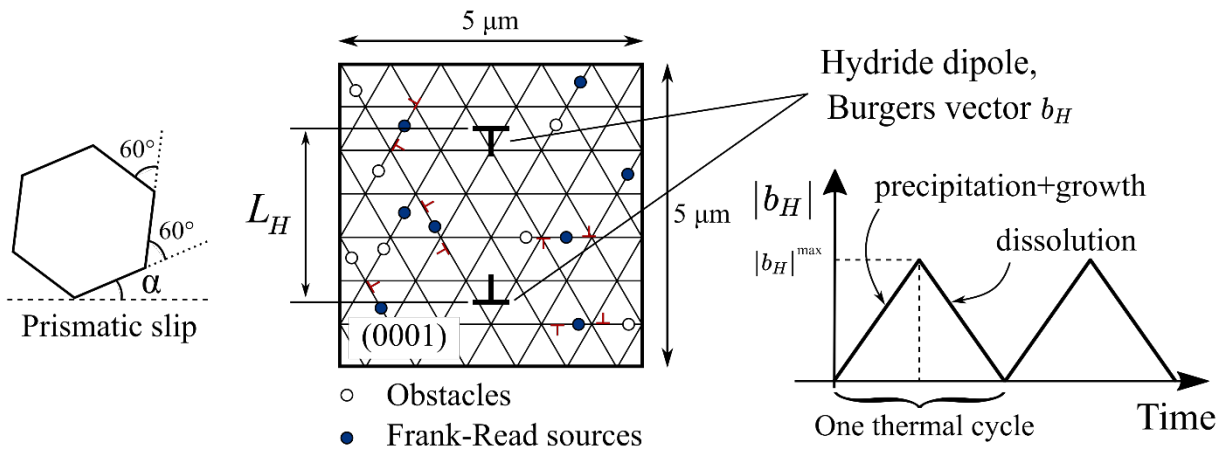


Figure 1. Model formulation of a γ -hydride needle as a prismatic dislocation dipole shrouded by a dislocation network within planar DDP. The evolution of the Burgers vector of the hydride b_H mimics the precipitation and dissolution process.

The matrix surrounding the hydride is assumed to be isotropic linear-elastic. As the hydride grows, the stress rises in the surrounding matrix, eventually leading to plastic deformation. Plasticity occurs through the nucleation and glide of a large number of discrete dislocations on defined slip planes; in this case on the prismatic slip planes assumed to be at 60° to each other with a crystal orientation $\alpha = 0^\circ$ as shown in Figure 1. Point sources representing planar Frank-Read sources are distributed randomly in the simulation cell with a source density $\rho_s = 10\mu\text{m}^{-2}$. It is assumed that the material is initially dislocation free. Dislocation dipoles are nucleated from sources when the critical resolved shear stress on the source exceeds the source nucleation strength τ_s for the duration of the nucleation time $t_{\text{nuc}} = 10\text{ ns}$. The nucleation strengths are prescribed from a Gaussian distribution with mean $\bar{\tau}_s = 110\text{ MPa}$ and standard deviation 20 MPa . Obstacles representing the effect of irradiation damage are also distributed in the matrix with a density $\rho_o = 3 \times \rho_s$ and a strength fixed at $\tau_o = 3 \times \bar{\tau}_s$ for all obstacles in

keeping with prior DDP studies. Dislocations become pinned at obstacles, only to be released when the resolved shear stress on the pinned dislocation exceeds the obstacle strength. The dislocation glide velocity is found using a linear mobility law, $v^{(l)} = f^{(l)}/B$ where $f^{(l)}$ is the glide component of the Peach-Koehler force on dislocation l , accounting for the effect of the hydride dipole and all other dislocations in the system, and $B = 4 \times 10^{-4} \text{ Pa} \cdot \text{s}$ is the assumed edge drag coefficient in zirconium. Further details on DDP as applied to the mechanical behavior of small-scale zirconium can be found in the work of Tarleton *et al.* [9]. Although the hydride-matrix interface could impede dislocation motion in reality, this interaction is not included here for simplicity; the hydride dipole and matrix dislocations are only allowed to interact via their stress fields. The hydride precipitation and dissolution are modeled incrementally with the rate of change of hydride Burgers vector $\dot{b}_H = 10^4 \mu\text{m/s}$, and a time step between increments assumed to be $dt = 0.1 \text{ ns}$. Thermal cycling is represented by repetitions of precipitate growth and dissolution. The results are averaged over three different DDP realizations with the same parameter set. Finally, a parametric study on the effect of model parameters ρ_s , \dot{b}_H and dt showed that the simulation results were qualitatively independent of the parameters used, thus implying that the discussion that follows applies more generally.

DISCUSSION

Hydride-Dislocation Networks

A typical realization of the dislocation structure at maximum hydride thickness and upon dissolution is shown in Figure 2. As hydrogen accumulates in the nucleation site, the hydride continues to grow until it reaches a critical size, at which point its misfit accommodation results in plastic relaxation and the dissipation of plastic strain energy into the zirconium matrix. This plastic relaxation is represented by the extensive dislocation network seen in Figure 2(a). In particular, the dislocation pile-ups may be viewed as sections through the concentric loops of prismatic dislocations observed in experiments [6]. Upon dissolution, however, a substantial residual dislocation density remains in this region, as depicted in Figure 2(b), which corresponds to the non-recoverability of the plastic strain energy associated with hydride formation.

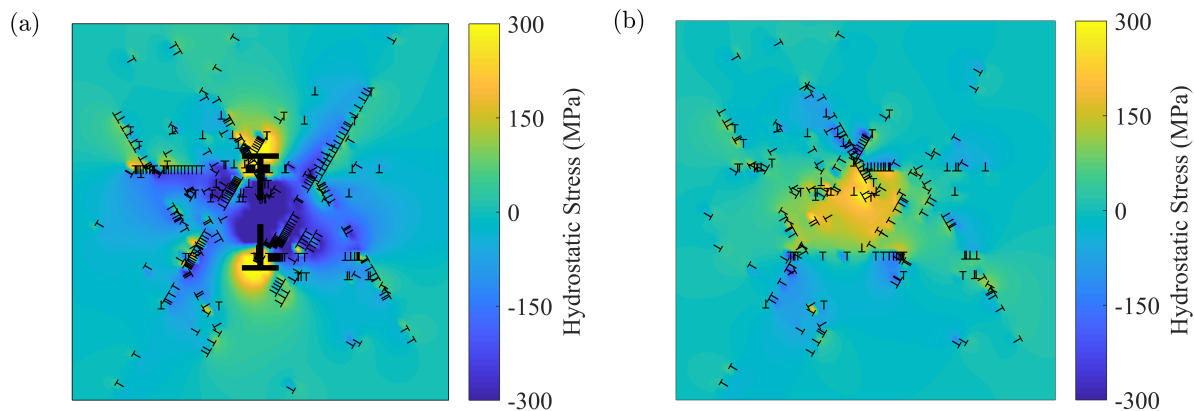


Figure 2. The hydrostatic stress fields of the dislocation structure around a hydride nucleation site following (a) precipitation and (b) subsequent dissolution. The remnants of the dissolution process contribute to a tensile hydrostatic state that would enhance the local hydrogen content.

More importantly, Figure 2(b) also shows that a residual tensile hydrostatic stress state exists at the nucleation site after dissolution, which provides a positive potential for hydrogen diffusion in anisotropic zirconium [10]. This residual tensile state is also obtained for the different realizations. This is because dislocations nucleate and configure so as to minimize the total strain energy of the system by counteracting the compressive field of the hydride. Thus, a residual tensile stress state remains upon dissolution of the compressive field. These dislocation networks may play a role in trapping hydrogen from dissolving hydrides as well as acting as an enhanced stress-raiser in the vicinity of the nucleation site to drive hydrogen diffusion from the bulk, both of which make subsequent re-precipitation more favorable; this is consistent with the observation that hydrides increase in number as thermal cycling progresses.

Dislocation Ratcheting

The evolution of the dislocation density over sequential thermal cycles is shown in Figure 3. The dislocation density reaches a peak at maximum precipitate thickness, after which it begins to decrease as dissolution occurs. However, as noted previously, a residual dislocation density remains even after dissolution. The dislocation density at the end of each thermal cycle is marked on Figure 3(a) and Figure 3(b), which reveals how the residual dislocation density evolves. It can be seen that there is a steady increase in the residual dislocation density following multiple dissolutions. This ratcheting in the dislocation density suggests that over time a nucleation site may become more effective at precipitating hydrides due to the interaction between hydrogen interstitials and an increasing dislocation population.

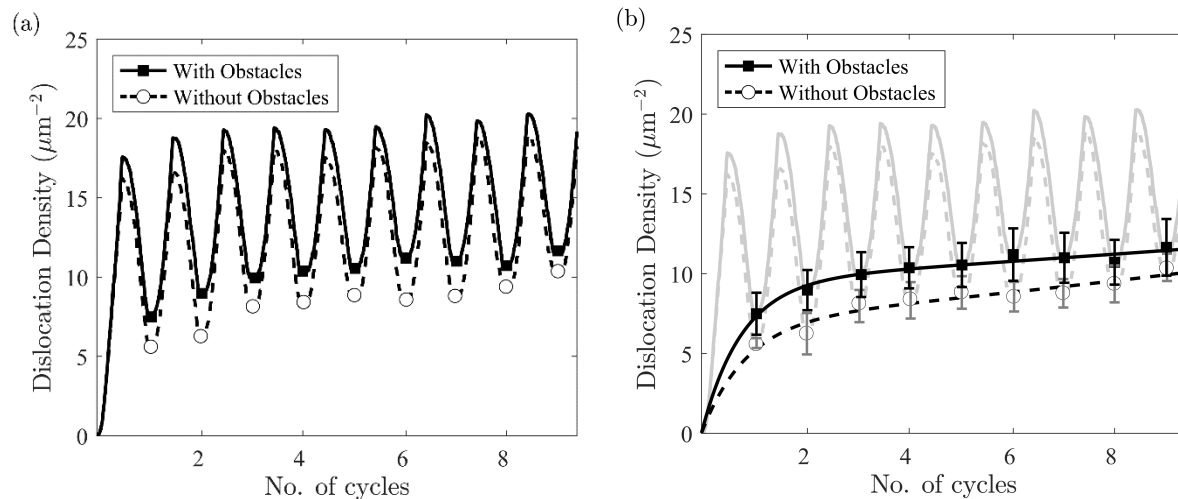


Figure 3. The ratcheting of dislocation density in unirradiated (without obstacles) and irradiated (with obstacles) zirconium for (a) a series of thermal cycles and (b) repeated dissolutions. The results are averaged over three DDP realizations and error bars are shown in (b) for the residual dislocation density.

Furthermore, obstacles were introduced to study the effects of irradiation damage; for example, interstitial clusters can impede dislocation motion. As evident from Figure 3, the dislocation density is higher for the irradiated material as the obstacles help to confine the

dislocation network to a smaller region around the nucleation site. This supports the experimental work that the TSS hysteresis is greater for irradiated zirconium [5].

CONCLUSION

This work investigated micro-hydrides under thermal cycling to understand ratcheting observed in hydride precipitation in zirconium. Microscale DDP simulations were used to study the evolution of the crystallographic dislocation network as it forms a residual hydrostatic tensile state around a hydride nucleation site. The hydride was represented by a prismatic dislocation dipole; its dissolution and precipitation kinetics were treated via changes in the dipole Burgers vector. It is shown that the dislocation density exhibits a ratcheting over heating and cooling cycles, which has implications on the TSSd/p hysteresis as the residual hydrostatic stress influences hydrogen diffusion. As explicit obstacle interactions are inherent to DDP, a comparison of unirradiated and irradiated zirconium is more natural in this model; it is shown the residual dislocation density is higher in the presence of obstacles (irradiation damage), which suggests a larger hysteresis. These trends agree with experimental studies on the TSS.

There are several possible avenues for further work: (i) incorporating the effects of anisotropy as α -zirconium is hexagonal close-packed; (ii) studying the stress-driven diffusion of hydrogen to dislocation cores using linear irreversible thermodynamics, which can be coupled with phase transition modeling to capture precipitation and dissolution; (iii) including more complicated obstacles interactions such as grain boundaries and second-phase particles; and (iv) undertaking a parametric study for a system of multiple micro-hydrides formed via autocatalytic nucleation to replicate macro-hydrides features. Following validation, the proposed formulation will be used as a statistical sub-model for macro-hydride ratcheting within a 2D component-scale framework for simulating DHC in modern fuel performance codes.

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