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Traction–separation relationships for hydrogen induced grain boundary embrittlement in nickel via molecular dynamics simulations

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ABSTRACT

A statistical approach combined with molecular dynamics simulations is used to study the influence of hydrogen on intergranular decohesion. This methodology is applied to a Ni $\Sigma3(112)[1\bar{1}0]$ symmetric tilt grain boundary. Hydrogenated grain boundaries with different H concentrations are constructed using an energy minimization technique with initial H atom positions guided by Monte Carlo simulation results. Decohesion behavior is assessed through extraction of a traction–separation relationship during steady-state crack propagation in a statistically meaningful approach, building upon prior work employing atomistic cohesive zone volume elements (CZVEs). A sensitivity analysis is performed on the numerical approach used to extract the traction–separation relationships, clarifying the role of CZVE size, threshold parameters necessary to differentiate elastic and decohesion responses, and the numerical averaging technique. Results show that increasing H coverage at the Ni $\Sigma3(112)[1\bar{1}0]$ grain boundary asymmetrically influences the crack tip velocity during propagation, leads to a general decrease in the work of separation required for crack propagation, and provides a reduction in the peak stress in the extracted traction–separation relationship. The present framework offers a meaningful vehicle to pass atomistically derived interfacial behavior to higher length scale formulations for intergranular fracture.

1. Introduction

Embrittlement effects due to the uptake of atomic species from the environment (e.g. H, Cs, or I) are a well-known factor influencing stress corrosion cracking. Atomic species, such as H or I, can be transported easily by a combination of interstitial and intergranular diffusion [1–3], which in turn degrades the strength and ductility of the metal. However, the precise mechanisms by which hydrogen causes embrittlement have been the subject of considerable debate. Some of the proposed mechanisms include: (i) failure due to hydride formation [4,5], and in non-hydride forming metals (ii) hydrogen-enhanced localized plasticity (HELP) [6–8] and (iii) hydrogen-induced decohesion (HID) [9–11]. While researchers have successfully developed empirical continuum models for stress corrosion cracking [12–15], insufficient understanding persists regarding the fundamental coupled role of compositional and microstructural evolution at the atomic scale.

Hydrogen-induced decohesion, also known as hydrogen-enhanced decohesion (HEDE), refers to the reduction of the cohesive force and surface formation energy in the presence of hydrogen [10,11]. Although direct experimental evidence of the HID/HEDE mechanism has yet to be obtained [16], its role in hydrogen embrittlement is supported by observations of failure without significant plastic deformation [17], reduction of fracture strength [18], and by thermodynamic considerations [19,20]. The HELP mechanism is generally described as an increase in plasticity in a localized region ahead of a crack tip [8] resulting from an increase in dislocation nucleation and mobility and has been extensively studied [21–23]. Experimentally, HELP is justified by observations of localized ductile crack propagation rather than cleavage fracture ahead of a propagating crack tip [24] and it was shown through high resolution fractography of hydrogenated Ni that significant plasticity was localized to the fracture surfaces [22].

However, given the contradictory evidence presented in the literature, a consensus has yet to be reached on whether HELP or HID is the governing mechanism for hydrogen embrittlement, or to what extent both mechanisms contribute simultaneously to embrittlement. For example, in direct opposition to atomistic simulations indicating that the presence of H increases edge

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dislocation mobility in α-Fe [25], Song and Curtin [26] showed that H leads to a resistance in dislocation motion. It is likely that no single mechanism conclusively accounts for premature failure due to hydrogen embrittlement and that the interplay of these mechanisms will further depend on microstructural factors, such as the local structure of the grain boundary or the lattice region ahead of the crack tip. These microstructural details can be ascertained from atomistic simulations.

For pristine grain boundaries, significant physical insight regarding the fracture process comes from atomistic studies of bicrystal grain boundaries with carefully chosen orientations of the applied load, crack front and lattice slip systems [27–30]. These simulations have helped clarify the effects of the bicrystallography, generalized stacking fault energies, temperature and other factors on dislocation nucleation, twinning and other processes occurring at the grain boundary crack tip. For example, Yamakov et al. [29] provided great details on the dynamics of intergranular crack propagation and different mechanisms of energy dissipation. One of the promising outcomes of the work of Yamakov et al. [29] and Spearot et al. [30] was the extraction of traction–separation relationships which can provide atomistic-based input for cohesive-zone models used in continuum fracture simulations [29,31–33].

For metallic systems with impurities, atomistic simulations including first principles methods have been used to study hydride precipitation, hydrogen segregation, and the embrittling potency of various elements [4,25,34–44]. For example, ab initio methods have been used to study energetic aspects of the interaction between H and defects in metallic materials including: (i) the reduction in cohesive energy due to the presence of H in Ni [38], Al and Fe [37], (ii) the strong binding energy between H and dislocation cores, which could prevent dislocation cross slip and promote planarity of slip in Al [39] and α-Fe [25], and (iii) the reduction in stacking fault energy in Zr [4,40], enabling enhanced plasticity as confirmed by larger scale atomistic simulations [4]. Interatomic potential based atomistic simulations have also been used to explore the role of hydrogen on embrittlement at larger length scales: (i) H lowered the stress intensity factor and energy required to generate new surfaces during crack propagation in single crystal Ni [45], (ii) low constant concentrations of H facilitated dislocation emission at a crack tip [41,42] and crack propagation [43], and (iii) Xu et al. [44] explored the effect of H concentration and location on the fracture mechanisms of single crystal Ni under tension and found that low H content facilitated crack propagation and created localized plasticity around the propagating crack tip. However, insufficient work has been done to explore embrittlement mechanisms in Ni via atomistic simulations for a realistic distribution of H across a representative grain boundary area, which is necessary to further elucidate the role of hydrogen on grain boundary embrittlement in ductile materials.

In the present work, atomistic simulations are performed to explore the influence of H impurities inserted at a Ni $\Sigma3(112)[1\bar{1}0]$ symmetric tilt grain boundary on decohesion mechanisms and strength during fracture. Although a single grain boundary is analyzed in this work, the algorithm presented can be implemented in a high-throughput fashion to analyze a wide range of grain boundary structures to elucidate the dependencies of HELP and HID embrittlement mechanisms on microstructural features. Section 2 provides details regarding the procedure used to build hydrogenated grain boundaries and to insert an intergranular crack of sufficient length for propagation. Decohesion is assessed through extraction of a traction–separation relationship during steady-state crack propagation in a statistically meaningful approach, building upon the work of Yamakov et al. [29] employing cohesive zone volume elements (CZVE) in the atomistic simulation. The first part of the results (Section 3) focuses on the influence of the numerical approach on the sensitivity of the extracted traction–separation relationships. The second part of the results (Section 4) discusses how H coverage influences the details of crack propagation along the $\Sigma3(112)$ symmetric tilt grain boundary, including asymmetries depending on crack propagation direction, and the cohesive strength of this grain boundary.

2. Steady-state crack propagation simulation model

The simulation approach in this study is based on a molecular dynamics (MD) model of crack propagation under time-independent, or steady-state, conditions along a flat grain boundary. The approach is composed of three steps: (i) creation of a hydrogenated grain boundary, (ii) introduction and propagation of an atomically sharp crack, and (iii) data mining to extract the relationship between atomistic simulations and continuum interface separation laws. Steps (i) and (ii) are covered in this section whereas the data mining procedure is discussed in Section 3. All simulations in this work are performed using LAMMPS [46] with an embedded-atom method (EAM) potential for Ni–H [47]. This EAM potential accurately predicts the bulk cohesive energy, elastic constants, the intrinsic stacking fault energy, and vacancy formation energy in Ni. The Ni–H interactions were fit to the solution energy and migration energy of H in Ni. This potential is not designed to capture hydride formation in the Ni–H system [47]; thus, analysis of this mechanism for embrittlement is beyond the scope of this work.

2.1. Creation of hydrogenated grain boundaries

Grain boundaries with $\Sigma3$ misorientations are commonly observed in metallic materials that have been subjected to grain boundary engineering processing routes [48]; the lateral $\Sigma3(112)[1\bar{1}0]$ interface is selected in this work as it provides sufficient free volume for H segregation compared to the coherent $\Sigma3$ twin grain boundary. The construction of a Ni $\Sigma3(112)[1\bar{1}0]$ symmetric tilt grain boundary with varying concentrations of H is done via a three-step process. First, the lowest energy hydrogen-free Ni $\Sigma3(112)[1\bar{1}0]$ symmetric tilt grain boundary structure is determined using a relatively small bicrystal ($<15{,}000$ Ni atoms) with periodic boundary conditions. To determine the minimum energy configuration, one grain is incrementally displaced relative to the other, sampling N_{pos} initial positions ($N_{pos}=100$) within the grain boundary plane, and subjected to energy minimization using the Polak–Ribiere version of the nonlinear conjugate gradient algorithm [49]. The translation corresponding to the minimum energy configuration is used to create larger systems utilized in the remainder of this study. The interface structure is consistent with prior work in the literature for the Ni $\Sigma3(112)[1\bar{1}0]$ grain boundary [47].

Second, using the translation associated with the minimum energy $\Sigma3(112)[1\bar{1}0]$ grain boundary structure, larger simulation models with periodic boundary conditions are created that are comprised of four layers following the methodology proposed by Yamakov et al. [29], as shown in Fig. 1: Two primary grains, between which is the primary (hydrogenated) grain boundary, and two “absorbing layers” [29] (hydrogen-free) on either side of the primary grains. The boundaries between the absorbing layers and the primary grains as well as the grain boundary between the two absorbing layers across the periodic boundary are referred to as “absorbing grain boundaries” [29] and act simply as obstacles to prevent dislocations from passing through the periodic boundary. The dimensions of the atomistic system (ℓ_x, ℓ_y, and ℓ_z) are chosen to be appropriate multiples of the grain boundary displacement shift complete distances [50], ensuring that no artificial boundaries or discontinuities are created as a result of the periodic