



Atomistic simulation study of the hydrogen diffusion in nickel

E. Torres^{a,*}, J. Pencer^{a,b}, D.D. Radford^a

^a Canadian Nuclear Laboratories, Chalk River Laboratories, Chalk River, ON K0J 1J0, Canada

^b Department of Engineering Physics, McMaster University, 1280 Main Street West, Hamilton, ON L8S 4L7, Canada



ARTICLE INFO

Keywords:

Hydrogen diffusion
Diffusion anisotropy
Hydrogen embrittlement
Nickel

ABSTRACT

The fundamental mechanisms and the conditions in which hydrogen embrittlement (HE) occurs in pure nickel and its alloys has not been fully determined. Several models associated with hydrogen-induced deformation and fracture modes have been proposed. In these models, the transport and concentration of hydrogen play the rate-controlling role in delayed HE. In particular, the kinetics of the embrittlement process is driven by the diffusion of hydrogen. Extensive experimental studies have been performed to elucidate the diffusion of hydrogen in nickel. These investigations have determined a significant anisotropy in the diffusivity of hydrogen. However, the nature of the anisotropy is unclear and still needs to be clarified. In the present work, the diffusion of hydrogen in nickel is investigated using a combined approach involving density functional theory (DFT) and molecular dynamics (MD). The temperature-dependent diffusion coefficients of hydrogen in nickel single crystal, determined from simulations, is in excellent agreement with experimental data. Moreover, it is demonstrated that for a single crystal nickel, with no imposed stress, the computed diffusivities in the $\langle 100 \rangle$, $\langle 110 \rangle$ and $\langle 111 \rangle$ directions did not show significant differences. The reported simulation results accurately describe the diffusion of hydrogen in nickel, and also suggest that stress fields may be the primary contributor to experimentally observed diffusion anisotropy.

1. Introduction

It is well known that the absorption, diffusion and concentration of hydrogen, even from the environment, eventually leads to degradation of the mechanical properties of metals and their alloys. The effect manifests itself as a delayed hydrogen-induced embrittlement [1–3]. A number of mechanisms to explain hydrogen embrittlement (HE) in metals have been proposed [4–6]. In particular, embrittlement in metals, in which the formation of brittle hydrides are not favorable, has been associated with the influence of hydrogen as an interstitial “alloying” element [7]. The hydrogen-enhanced decohesion (HEDE), where H atoms concentrated at the crack tip weaken the metal bond strength promoting crack growth [5,7], and hydrogen-enhanced local plasticity (HELP), in which H accumulates ahead the crack tip enhancing the local plasticity and facilitating the propagation of the fracture [4,8], are among the more prominent mechanisms for HE in non-hydride forming materials. Experimental investigations have determined that enhanced transport and trapping of hydrogen control the solute-dependent kinetics of the HE process in metals and alloys [3,9]. As a result, considerable experimental and theoretical effort has been devoted to understand the behavior of hydrogen in metals [10–14].

Ni-based alloys are used in components in nuclear power plants, and

for oil and gas industrial applications due to their superior mechanical properties and corrosion resistance. However, pure nickel and nickel-based alloys, even exposed to low levels of hydrogen, are highly susceptible to hydrogen induced embrittlement [15–17]. As determined from experiments, the formation of a hydride phase in bulk nickel is not favorable [3,15]. Therefore, considerable experimental effort has been devoted to elucidate the mechanism of HE in nickel and nickel-based alloys, i.e., [13,15–19]. In particular, models in which hydrogen locally enhances the plasticity in nickel have been proposed as the mechanism for promoting the intergranular failure mode [13,15].

The macroscopic diffusion constant of impurities in polycrystalline materials is usually considered isotropic. However, the mechanisms that lead to HE occur at the nano-scale, and therefore may be influenced by the local properties of hydrogen transport. For instance, the geometry of the crystal lattice may have an influence on the hydrogen mobility. In fact, experimental and theoretical studies indicate that under stress fields the diffusion of hydrogen in metals is anisotropic [20,21]. The anisotropy of hydrogen diffusion is of fundamental interest in the understanding of the degradation effect at atomistic scale resolution. The diffusivity of hydrogen in nickel has been the focus of several experimental investigations, see Refs. [10,20]. Moreover, it has been recently suggested that the concentration of hydrogen may have a

* Corresponding author.

E-mail address: edmanuel.torres@cnl.ca (E. Torres).

significant effect on hydrogen diffusion [20]. These results indicate that a self-induced stress has a significant influence on the anisotropy of hydrogen diffusivity in nickel, as was predicted by Dederichs and Schroeder [21]. This effect has been also observed in Fe-Al intermetallic compounds [22].

Atomistic scale simulations can provide insight into the behavior of hydrogen, which complement experimental studies [23]. First-principles calculations are very accurate and can be applied to arbitrary configurations of atoms. However, the number of atoms that can be explicitly considered using first-principles calculations is very limited (~ 100 atoms). Studies of hydrogen diffusion and interaction with lattice defects in nickel using density functional theory (DFT) have been reported [24–29]. In particular, Wimmer et al. [24], using transition state theory in combination with DFT calculations, obtained the temperature-dependent coefficients for interstitial diffusion of hydrogen in nickel. It was reported, however, that an error factor between 2 and 3 times was estimated from the absence of lattice vibrations in their model. Atomistic studies involving vast numbers of atoms at the nanoscale can be performed using Molecular dynamics (MD) simulations. However, to compute meaningful results, the interactions between the atoms in the system must be accurately described by the interatomic potentials used in the simulations. Many MD studies, of hydrogen-nickel systems, have been based on the embedded atom method (EAM) interatomic potential by Baskes et al. [30,31], referred hereafter only as the EAM potential. More recently, this EAM potential was modified to better describe hydrogen impurities in the vicinity of Ni $\Sigma 5(120)(100)$ grain boundaries [32]. In preliminary studies, however, we found discrepancies in the description of the impurities and interstitial migration of hydrogen atoms in nickel using this EAM potential. Therefore, the available H-Ni EAM potential was found unsuitable for the modeling of hydrogen diffusion in nickel.

In this paper, an atomistic simulation study of hydrogen in nickel is presented. We first investigate the formation energy of impurities and the energy barriers for interstitial migration of hydrogen in nickel using DFT. Then, based on DFT reference data, we parameterize a set of interatomic potentials, using the Beck potential function [33], to describe H-H and H-Ni interactions in MD simulations. Energetics of hydrogen impurities and migration barriers obtained with the EAM potential are also included for comparison. The developed potentials are shown to accurately reproduce the formation energies of hydrogen defects and energy barriers for interstitial migration in nickel, as determined from DFT calculations, thereby demonstrating the suitability for the study of hydrogen diffusion performed in this work. The computed temperature-dependent diffusion coefficient and the activation energy, obtained from MD simulations of hydrogen in nickel, were found in excellent agreement with experimental data. Subsequently, we investigate the anisotropy of hydrogen diffusion in nickel for various crystallographic directions. The present results provide details of the behavior of hydrogen in perfect bulk nickel with atomistic scale resolution. In order to shed light on the fundamental aspects to consider in advanced theoretical studies, our results are discussed from the perspective of the experimental findings.

2. Computational procedures

2.1. Density functional theory calculations

Density functional theory calculations were performed using the plane-wave code PWscf distributed with the Quantum ESPRESSO integrated suite [34]. Spin polarized total energy calculations were performed within the generalized-gradient approximation (GGA), using the exchange and correlation functional by the Perdew-Burke-Ernzerhof (PBE) [35]. The electronic wavefunction was expanded in a plane-wave basis set determined by the kinetic energy cut-off of 680 eV. The electron smearing was modeled using the Methfessel-Paxton broadening scheme of first-order with $\sigma = 0.2$ eV. A Γ -point centered $8 \times 8 \times 8$ k -

point grid, generated using the Monkhorst-Pack method [36], was used for the sampling of the Brillouin zone. The error threshold of 0.01 meV per atom, used to control the convergence in total energy, was determined accurate for structure optimizations. The properties of bulk nickel and defect formation energies below a 1.0 meV were determined in our DFT calculations. Zero-point energy contributions were not included in the estimation of defect formation energies.

The ground state properties of bulk nickel, and insertion energies of hydrogen impurity atoms in nickel, were calculated using a $3 \times 3 \times 3$ periodic supercell of the conventional fcc unit cell. The supercell with perfect single crystal nickel is composed of 108 nickel atoms. The cores of nickel atoms were represented by a scalar relativistic ultrasoft pseudopotential, that include a non-linear core correction, and the cores of hydrogen atoms were described by a non-relativistic ultrasoft pseudopotential [37]. The magnetic moments on the nickel atoms were configured to start the calculations in a ferromagnetic (fm) ordered state. However, in total energy and structure optimizations, the magnetic moments were left free to relax.

The energy of an isolated hydrogen atom was computed in a cubic supercell with a side of 20 Å. The calculation only included the Γ -point and considering spin-polarization in the calculation. It was determined that the supercell size was sufficient to eliminate self-interactions, and therefore to obtain the reference state energy of a hydrogen atom within 0.1 meV of accuracy. The barriers for the interstitial migration of a hydrogen atom in nickel were evaluated using the Nudged elastic band (NEB) method. The NEB calculations, at the DFT level of theory, were performed with the PWneb code distributed with Quantum ESPRESSO [34]. NEB calculations were computed using a $3 \times 3 \times 3$ nickel supercell, employing the same parameters determined for the computation of formation energies and the force tolerance of 0.1 eV/Å to control convergence.

2.2. Molecular dynamics simulations

The MD simulations were performed with the large-scale atomic/molecular massively parallel simulator (LAMMPS) code [38], using the GPU package for accelerated MD simulations developed by Brown et al. [39]. All the simulations were performed with periodic boundary conditions imposed in all directions. The interactions between nickel atoms were described using the Ni potential developed by Bonny et al. [40]. This potential accurately reproduces the mechanical properties of bulk nickel, but also correctly describes lattice point defects. The interactions between H-H and H-Ni were described by interatomic potentials developed in this work. We found an accurate representation of the interactions of hydrogen atoms in the metal lattice using the Beck potential [33]. Therefore, Beck interatomic potentials, for H-H and H-Ni, were parameterized for the study of hydrogen diffusion in nickel.

The parameters for the Ni-H and H-H interatomic potentials were determined using an iterative random-walk based optimization approach. The details of the parameter optimization procedure are outlined in Ref. [41]. The formation energies of hydrogen defects were determined from the structure minimization using the Polak-Ribiere formulation of the conjugate gradient method as implemented in LAMMPS. The convergence was determined by the force tolerance of 1.0×10^{-6} eV/Å. The accuracy of MD static predictions were validated against DFT reference data.

2.3. Definition of the energy calculations

There are three interstitial sites with high symmetry and one substitutional site in the fcc crystal structure of nickel, as indicated in Fig. 1. In the following, we refer to these sites as octahedral (O), tetrahedral (T), crowdion (C) and substitutional (S). The insertion energy (E_{int}) of a single interstitial hydrogen impurity at interstitial sites was evaluated using the following expression

Download English Version:

<https://daneshyari.com/en/article/7957246>

Download Persian Version:

<https://daneshyari.com/article/7957246>

[Daneshyari.com](https://daneshyari.com)