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Molecular dynamics study of the role of symmetric tilt grain boundaries on the helium distribution in nickel

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ABSTRACT

Helium impurities, from either direct implantation or transmutation reactions, have been associated with embrittlement in nickel-based alloys. Helium has very low solubility in nickel, and has been found to aggregate at lattice defects such as vacancies, dislocations, and grain boundaries. The retention and precipitation of helium in nickel-based alloys have deleterious effects on the material mechanical properties. However, the underlying mechanisms that lead to helium effects in the host metal are not fully understood. In the present work, we investigate the role of symmetric tilt grain boundary (STGB) structures on the distribution of helium in nickel using molecular dynamics simulations. We investigate the family of STGBs specific to the ⟨110⟩ tilt axis. The present results indicate that accumulation of helium at the grain boundary may be modulated by details of grain boundary geometry. A plausible correlation between the grain boundary energy and misorientation with the accumulation and mobility of helium is proposed. Small clusters with up to 6 helium atoms show significant interstitial mobility in the nickel bulk, but also become sites for nucleation and grow of more stable helium clusters. High-energy GBs are found mainly populated with small helium clusters. The high mobility of small clusters along the GBs indicates the role of these GBs as fast two-dimensional channels for diffusion. In contrast, the accumulation of helium in large helium clusters at low-energy STGB creates a favorable environment for the formation of large helium bubbles, indicating a potential role for low-energy STGB in promoting helium-induced GB embrittlement.

1. Introduction

Metallic structural materials in nuclear fission and fusion reactors are subject to damage as the result of the presence of helium impurities, either produced through (n,α) reactions with certain isotopes in the materials or injected into the materials as a result of exposure to fusion plasma [1]. The insolubility of helium in these structural materials, e.g., nickel, iron, or tungsten-based alloys, leads to clustering of helium and formation helium bubbles inside grains and at grain boundaries. This behavior has been observed both experimentally and through simulation in, e.g., pure metals and alloys of iron, nickel, tungsten, and alloys of copper-niobium [1–6]. In these systems, it is generally assumed that intergranular embrittlement occurs due to the formation of bubbles at grain boundaries [1,7]. The conditions for helium trapping and retention are still a matter of research [6,8].

An understanding of the fundamental mechanisms of helium-embrittlement and the conditions in which material degradation occurs, are essential in the development of novel approaches to reduce the detrimental effects of helium in the alloys. Methods to overcome helium embrittlement in alloys have been proposed as early as 30 years ago [9], and novel methods have been devised more recently, such as helium trapping in nanochannels [6]. Considerable experimental effort has been devoted to understand the behavior of helium in austenitic steels and nickel-based alloys [10,11]. However, the mechanism of the observed embrittlement caused by helium in irradiated metals is still unclear [3,11]. In addition, while the generic behavior of helium may be similar among various metal compositions (e.g. Ni, Fe, W) and geometries (e.g., bcc, fcc), details such as helium diffusion pathways, diffusion rates, lowest energy locations, etc., depend on and vary greatly with composition and local geometry.

The behavior of helium in nickel is of particular interest because

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of the use of nickel alloys for spacers in heavy water reactors (HWR) and as structural materials in other contemporary and advanced fission reactors ([12], and references therein). Atomistic modeling of helium in nickel contributes to the development of better predictive models for helium diffusion, clustering and bubble formation in nickel, which can then be used in higher length scale finite element and continuum based models for mechanical behavior.

In the temperature range where helium is mobile, He atoms diffuse and precipitate at various structural trapping sites. In experimental investigations, helium-induced features in nickel are frequently studied using transmission electron microscopy (TEM), small-angle neutron scattering (SANS), positron annihilation spectroscopy (PAS), and thermal desorption spectroscopy (TDS) [3,10,11,13]. Mechanical tests are performed to characterize the associated effects of helium with the mechanical performance of the materials [11,14]. Nevertheless, not all the helium present in the matrix contribute to the observed material embrittlement.

The trapping and diffusion properties of helium in nickel have been discussed in experimental studies [15–17]. However, the reported rates and mechanisms for helium diffusion have been a source of controversy [18]. Moreover, large discrepancies between experimental activation energy for helium thermal migration in nickel reported in a recent review indicates that this still an open question [19]. In particular, experimental observations have found that He accumulated at the grain boundaries (GBs) is associated with embrittlement [13,20,21], potentially leading to intergranular mechanical failure under load conditions [11]. In fact, helium-induced GB embrittlement is manifested by observable intergranular fracture paths [3,20]. As such, the origin of helium-embrittlement at GBs in nickel-based alloys is of fundamental importance.

Grain boundaries are constituted by the discontinuous interface between two adjacent grains in a polycrystalline material. The complex structure of the GBs can accommodate a large distribution of diverse trapping sites for impurities. Therefore, helium atoms may precipitate at those GB trapping sites. The accumulation of helium in large volumes may lead to significant changes in structural and materials properties. Symmetric tilt grain boundaries (STGB) have very interesting properties and are frequently observed experimentally [22–24]. As a consequence, STGBs may play a fundamental role in modulating helium effects on the material properties.

A significant number of modeling studies of the helium-iron system have contributed to elucidate the mechanism of bubble nucleation and growth [2]. In particular, atomistic simulations studies have been essential to understand the role of grain boundaries on the helium effects [25–30]. While similar studies have also been undertaken to investigate helium in nickel, the interatomic potential used in many atomistic studies of He-Ni system was recently found to incorrectly describe the behavior of helium in nickel [12]. A recently reported He-Ni interatomic potential was shown to correctly describe single He impurities and clusters in nickel. Furthermore, the characterization of nanobubbles were found in good agreement with experimental results [31], and therefore it is used in the present study.

Subnanometer helium bubbles in nickel are of considerable interest because their formation is the precursor of larger helium bubbles and their effects [6,21,32]. However, experimental characterizations of helium bubbles in nickel are typically limited to bubbles with diameters >1 nm and their interpretations are complicated by interactions with point defects also present, generated as a consequence of either recoil damage (in the case of neutron irradiation-induced transmutation) or cascade damage (in the case of implantation) [11,33]. As such, the characterization of subnanometer bubbles, including their association with various microstructural features of the alloys, represents a major challenge in experimental investigations. The behavior of helium in nickel and formation of helium agglomerates can be studied using atomistic scale simulations. Molecular dynamics simulations can aid in the characterization of subnanometer helium clusters, and therefore to shed light on the conditions that lead to the formation of larger helium bubbles.

The characterization of the helium behavior in the presence of STGB structures in nickel may help to elucidate the initial stages of helium embrittlement, thus contributing to the development of higher length scale models for helium-induced embrittlement and strategies to circumvent the conditions that lead to such embrittlement in nickel-based alloys [9,34,35]. In this work, the role of STGBs on the spatial helium distributions in nickel is investigated using molecular dynamics simulations. In order to evaluate the effects of individual GB structures, the simulations are performed using bicrystal model systems of GB in nickel [23,36]. We investigate helium concentrations from 2000 to 6000 appm, which are within the range from medium to high helium concentration levels of experimental investigations [11]. The atomic densities of helium and the spatial distribution of clusters in the grain interior and in the vicinity of the grain boundaries are determined. The characteristics of helium distributions from MD simulations are discussed and compared to experimental findings. The reported simulation results show that the cluster size and density distribution of helium strongly depends on the characteristics of GBs. It is expected that the formation of helium bubbles is dependent on the initial helium distribution, and therefore the reported findings enhance the understanding of helium effects in nickel-based alloys.

2. Computational methods

Molecular dynamics simulations were carried out with the large-scale atomic/molecular massively parallel simulator (LAMMPS) code [37] including the GPU accelerator package [38]. Simulations were performed with periodic boundary conditions imposed in all Cartesian directions. Molecular dynamics simulations of the nickel-helium systems were performed using the Bonny et al. [39], Beck [40] and the Morse-3G [31] potentials.

The Ni-Ni interactions are described using the potential of Bonny et al. [39]. This potential was chosen because it accurately reproduces point defects in nickel. This particular potential function also predicts stacking fault energy and vacancy formation energies quite accurately, and so its use is appropriate to examine planar defects such as grain boundaries. The He-He interactions are modeled using the Beck potential [40]. This potential reproduces the second virial coefficient for helium, and is therefore adequate in the study of helium agglomerates in this work. The He-Ni interactions are described using the Morse-3G interatomic potential [31], which accurately reproduces the barrier for interstitial helium migration, the formation energies of helium point defects, and helium clusters in nickel as determined in density functional theory calculations [31].

In the bicrystal approach, the simulated supercell is divided in two crystalline regions. The initial construction of the GB structure included a rigid body rotation and a translation offset vector of one crystal relative to the other. The interface between the two rotated crystals constitute the grain boundary. In the case that the crystalline regions are rotated by the same angle, but in opposite directions, a bicrystal containing two STGBs is created. The two grain boundary structures are identical and are separated by bulk nickel. The details of the bicrystal method for the construction of GB structures can be found in Refs. [23] and [24]. In the present atomistic simulation study, we followed the steps for the construction of bicrystal models as outlined in Ref. [41]. The