Materialia 000 (2018) 1–11

Contents lists available at ScienceDirect

Materialia

journal homepage: www.elsevier.com/locate/mtla

Atomistic simulations of the strengthening effect of high-density bubble formation in helium irradiated single crystalline copper

Anupam Neogi, Hesam Askari, Niaz Abdolrahim*

Department of Mechanical Engineering, University of Rochester, Rochester, NY, 14636, USA

ARTICLE INFO

Keywords:
Solid He formation
Molecular dynamics
Mechanical strengthening

ABSTRACT

We performed atomistic simulations to study the strengthening effect of He bubbles with different He atom concentrations within single crystal copper matrix. Uniaxial tensile deformation test demonstrated mechanical strengthening and enhanced ductility of single crystal copper hosting such He bubbles containing high-density He atoms. The yield strength and failure strain have been observed to increase up to vacancy/He ratio of 1:3, followed by a sharp decrease at the vacancy/He ratio of 1:4. Formation of 3D regions with distinct crystallographic orientation has been characterized during the evolution of high-density He bubbles. Considerable number of sessile dislocations have been observed to form in the boundary of those 3D regions for the sample with vacancy to He atom ratio of 1:2 and 1:3 during equilibration. These sessile dislocations contribute towards the observed mechanical strengthening along with Cu/He interfacial energetics. Higher He atom concentration also results into enhancement of total dislocation density with a substantial reduction in mobility of dislocations in copper which promotes homogeneous plastic deformation. During plastic flow, He bubbles undergo significant decrystallization, detwinning, and formation of Shockley partials, while copper exhibits ordinary dislocation-mediated plasticity. The rate of growth of the He bubbles shows significant influence on plastic deformation via coalescence of the He bubbles in the solid region and leads to ultimate failure of the sample.

1. Introduction

The detrimental consequences of high energy alpha (doubly ionized helium nuclei, He^{2+}) irradiation are referred to as radiation damage of materials. It may result in to generation of materials defects such as dislocation loops [1], voids [2], stacking fault tetrahedron (SFT) [3], extended precipitation or segregation of elements [4], etc. These are commonly observed events in component materials deployed in nuclear fission or fusion reactors [5–9]. The microstructural changes due to these defects prevailing in metals have a significant role in their mechanical response which typically is known as 'macroscopic radiation damage' [10–12] and is a serious concern of nuclear power reactors [13]. More specifically, these radiation-induced material defects serve as fracture sites and promote brittle-like failure which is known as 'radiation-induced embrittlement' [10–12] of the structures.

In addition, a significant amount of helium (He) atoms can be produced as a consequence of high energy alpha irradiation in metals or alloys through (n; α) transmutation reactions. These He atoms in metals attract each other and form He nano-clusters, which consequently can grow and coalesce to appear as a nano-metric He-gas bubbles. The mass density and internal He bubble pressure may vary significantly depending on the intensity of radiation, temperature and matrix element. Low density He nanobubbles (with negligible internal bubble pressure) are generally equilibrium in nature, *i.e.*, the radial stress at the bubble surface is equal to the hydrostatic stress of the matrix. At elevated temperature (below 35% of melting temperature) and higher intensity of radiation dose ($\geq 2\times10^{17}$ He ions/cm^2), the mass density of entrapped He atoms increases significantly and leads to formation of pressurized He bubbles. Unlike low density He bubbles, these pressurized high densities He bubbles exhibit nonequilibrium state, i.e., internal bubble pressure is higher than hydrostatic stress of the matrix. These high density He bubbles (formed at different radiation intensity) exhibit distinct deformability and plastic behavior. Besides, the role of those high-density He superlattices over the mechanical response of the irradiated host metals would be significantly different as compared with that of low-density He-gas bubbles and other defects, produced during irradiation.

Gradual enhancement of mass density of He atoms in nano-bubbles may result in to nucleation of solid phase of He. For instance, in 2009, Haghighat et al. [14] reported the formation of a solid phase of He within body-centered cubic (BCC)-Fe lattice using molecular dynamics simulations, and they concluded that the morphology of the observed solid He is a function of the surface energy at low pressure or the elastic-plastic properties of Fe at high pressure.

* Corresponding author.
E-mail address: niaz@rochester.edu (N. Abdolrahim).

https://doi.org/10.1016/j.mtla.2018.04.004
Received 15 April 2018; Received in revised form 18 April 2018; Accepted 23 April 2018
Available online xxx
2589-1529/

The role of low-density He-gas bubbles and irradiation-induced defects, over the mechanical properties of various irradiated metals and alloys, has been investigated by numerous experiments [15–19] and as well as atomistic simulations [20,21]. The atomistic mechanism of the interaction of a dislocation gliding through irradiation-induced defects such as voids and precipitates (e.g., He bubble), has been studied extensively by previous researchers [22–26]. It is widely reported that irradiated materials exhibit strengthening primarily by pinning of mobile dislocations at He bubbles. The pinning strength of He bubbles has been observed to depend on He atom density (i.e., internal pressure) in the bubbles. At much higher He density such as vacancy/He ratio of 1:5 [26], He bubble distorts the surrounding lattice structure by initiating dislocation loops that exert additional impedance to the glide of mobile dislocation.

A similar type of irradiation-induced He-gas bubble formation and consequent strengthening and embrittlement at the interface of a nanolayered composite of Cu/Mo, Cu/V, and Cu/Nb has been characterized by Li et al. [27]. However, none of the studies discussed above, have considered the formation of solid phase of He and its role over the mechanical response of the materials explicitly. Therefore, the behavior of low density He-gas bubble under mechanical loading has been studied for decades, but very little is known about high density solid phase of He and their effect on irradiated metals. However, a recent experimental study by Ding et al. [28] reported enhancement of ductility of pressurized He-nanobubbled submicron-sized single crystal copper. As one reason for this, they conjectured that the He-gas bubbles act like potential dislocation sources which promote storage of dislocations and thus, lead to more homogeneous and stable plastic flow and ultimate ductile failure.

In a recent in-situ transmission electron microscopy (TEM) compression test by Zhang-Jie et al. [29] similar type of mechanical strengthening of irradiated copper, hosting He superlattices [30–33], has been observed. They characterized formation of He superlattices during He implantation in nanotwined (dose rate of 2×10^{17} and 1×10^{18} ions/cm^2) and single crystalline (dose rate of 2×10^{17} ions/cm^2) nanopillar copper. They observed twin-like transformation and disordering (via random dislocation motion) of He superlattices, when the host metal (copper) undergoes deformation twining. They studied the effect of He dose on the mechanical strength and critical resolved shear stress (CRSS), and concluded that yield stress and CRSS are larger at high irradiation doses.

However, the atomic-scale mechanisms behind this experimentally observed strengthening phenomena and other plastic mechanisms that may occur in irradiated single crystal FCC copper is still unclear.

Therefore, this present work aims to explore the effect of He atom concentration on the mechanical response of the irradiated FCC host metals by using atomistic simulations. We have considered different vacancy/He ratio, e.g., 1:0 (nano-voids without He atom), 1:1, 1:2, 1:3, and 1:4, to explore the effect of He atoms concentration on mechanical properties of FCC–Cu hosting such irradiation induced He bubbles. The average pressures of the He-bubbles are 0.121, 1.262, 2.452 and 4.88 GPa for the vacancy/He ratio of 1:1, 1:2, 1:3 and 1:4, respectively. The details of the simulations performed in this work has been described in Section 2. Results have been discussed in Section 3, while conclusions have been made in Section 4.

2. Simulation methodologies

In their experimental study on the mechanical behaviour of irradiated single crystal copper containing He superlattices, Zhang-Jie et al. [29], observed that the irradiation-induced helium bubbles are not in full equilibrium with the matrix. So, the He bubbles are pressurized within the host metal. Thus, to consider this experimentally observed 'non-equilibrium nature' of the He bubbles, we have performed equilibration of the initial atomistic models employing microcanonical (NVE) constant volume, constant energy ensemble. This ensures confinement of He atoms (as the volume is constant) within the host metal matrix without relaxation of the residual stress. During this NVE equilibration simulations, apart from He superlattice formation, various types of material defects (such as dislocations, SFT, vacancies, etc.) form around the He-bubbles. The resulting microstructures exhibit defects around the He bubbles, and is analogous to a representative volume element (RVE) of a component material of an in-service nuclear shielding structure exposed to the radiation.

In this current work, we have considered the morphology of He bubbles within the Cu matrix in an idealized BCC pattern. To model the structure, we have created a bulk $50a\times50a\times50a$ (where a = lattice constant) supercell of single crystal FCC Cu and equilibrated the sample by utilizing NVE ensemble at ambient temperature (300 K) for 100 ps with a time step of 1 fs. Periodic boundary condition (PBC) is employed in each orthogonal direction of X, Y, and Z. Initial configuration of He-nanobubbled single crystal copper model has been represented in Fig. 1. Spherical regions with an idealized pattern (to set desired inter-bubble distance) have been defined in the equilibrated Cu, and Cu atoms of those regions are deleted to introduce He atoms inside the spheres. The internal He bubble pressure has been adjusted by changing the number of He atoms. We have considered vacancy/He ratio of 1:0 (sample with voids), 1:1, 1:2, 1:3 and 1:4 corresponding to the average bubble pressure of 0 (sample with void), 0.121, 1.262, 2.452 and 4.88 GPa, respectively.

After introducing He atoms, we have performed energy minimization with a typical force tolerance limit of 1×10^8, followed by re-thermalization by utilizing microcanonical (NVE) ensemble at temperature = 300 K for 100–500 ps (models with higher He atom concentration required more sampling time) with a timestep of 1 fs and keeping PBC in all directions. In case of higher He concentration (such as 1:3 and 1:4), significant dislocation density has been characterized in Cu matrix around the He bubbles.

The interatomic interactions of Cu–Cu have been described by using embedded atom method (EAM) potential developed by Mishin et al. [34], while for modelling Cu–He and He–He interactions, we have employed a Lennard–Jones (LJ) potential [35], $V(r)=4\varepsilon[(\sigma/r)^a-(\sigma/r)^b]$, where a = 9, b = 6, $\sigma=3.546$ Å and $\varepsilon=0.000745$ eV, for Cu–He interactions, and a = 12, b = 6, $\sigma=2.280$ Å and $\varepsilon=0.000876$ eV [36], for He-He interactions. To calculate the stress-strain response of the structure, we have simulated tensile deformation (with a constant strain rate of 10^9 s^{-1}) under uniaxial stress condition ($\sigma_{xx}\neq0$, $\sigma_{yy}=\sigma_{zz}=0$) maintaining PBC in all directions. The velocity-Verlet algorithm has been used to integrate the equation of motion of the atoms with a time-step of 1 fs, and to control temperature and pressure during deformation we have employed Nose-Hoover thermostat and barostat in conjunction with NPT ensemble. To investigate the effect of He bubbles on the mechanical response of copper, we extracted per-atom stress components of copper atoms separately, and calculated stress-strain plot exclusively for the copper matrix. We have considered the volume of only copper matrix during the calculation of mean stress of copper atoms. Voronoi tessellation [37] has been performed to compute the volume of copper matrix at a given configuration of applied strain.

To identify the atomic level defects during deformation, various techniques, such as adaptive common neighbour analysis (CNA), centrosymmetry parameter (CSP) estimation, coordination per atom, *etc*. have been used in this work. Dislocation extraction algorithm (DXA) [38] (as implemented in program OVITO, developed by Stukowski and Alexander [39]) which represents the dislocated crystal into a line based representation of dislocation network, has also been utilized. It should be noted here that apart from distinguishing between different dislocation mechanisms, this algorithm has the capability of recognizing true Burgers vector of each dislocation segment and dislocation junctions [38]. Also, this DXA algorithm is successfully applied [40,41] for characterizing distinct types of dislocations in FCC metals. For identifying local lattice environment of the atoms in the deformed samples another program, crystal analysis tool (CAT), developed by Stukowski et al. [42,43] has been utilized. Voronoi tessellation [37] analysis technique