



An examination of nickel doping effect on the mechanical strength of a tungsten grain boundary[☆]



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ABSTRACT

Grain boundary (GB) embrittlement in nanostructured metals intended for high temperature applications is considered an important detriment. In the present work, embrittlement in a nickel (Ni)-doped tungsten (W) (100)–(210) GB is examined using *ab initio* simulations based on Car Parrinello molecular dynamics (CPMDs) framework. The atomic fraction of substituted Ni atoms in the examined W GB and simulation temperature are varied in order to understand the strength of the W GB as a function of temperature. An increase in the Ni atomic fraction in the W GB from 12.5% to above 25% value leads to a peak in yield strength and reduction in the strain corresponding to the ultimate tensile strength which can be characterized as embrittlement. While the elastic modulus does not show a dependence on Ni atomic fraction variation and temperature, the yield strength, the ultimate tensile strength, and the fracture strength show an appreciable dependence. Addition of Ni atoms adds localized peaks in *f*-orbital electron density of states which is found to contribute to increase in the bond strength with increase in Ni atomic fraction. Based on analyses performed, a relation expressing tensile strength of the examined W GB as a function of W surface energy, Ni atomic fraction, and simulation temperature is derived. The relation is shown to predict temperature dependent strength of examined Ni-doped W GB that fits the simulation data.

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1. Introduction

Intergranular glassy films (IGFs) have been known to affect high temperature creep and fracture behavior of nanostructured ceramics significantly e.g. [1]. Similar to IGFs in nanostructured ceramics, grain boundary (GB) embrittlement in nanostructured metals intended for high temperature applications is considered an important determinant of mechanical strength [2]. GB embrittlement occurs through diffusion of a small atomic fraction of liquid metals such as nickel (Ni) in GBs of intrinsically ductile materials such as tungsten (W) during sintering or during high temperature operation [3]. The problem of W GB embrittlement is not new. Toth and Lockington [4] examined W GBs and found that Ni phase present at the surface of W powder before activated sintering had segregated along the W GBs afterwards. This was referred to as GB segregation, a fundamental contributor to GB embrittlement. Nieh [5] presented an evidence of GB segregation of Ni in W using auger electron spectroscopy (AES). They also demonstrated that such Ni segregation results in a severe embrittlement of W filaments. Similar phenomenon has been analyzed in Ni-sulfur system by Yamaguchi et al. [6] who showed that the embrittlement leads to GB decohesion and reduced material strength. Similar diffusion of impurity elements in GBs leading to material failure had been shown to occur in the case of bismuth-copper system [7].

Gupta et al. [3] used high resolution transmission electron microscopy (HRTEM) and AES to reveal the presence of nanometer-thick, Ni-enriched, disordered films at W GBs for the first time. Hwang et al. [8] advocated a thermodynamics based approach to predict the effect of change in Ni atomic fraction on GB strength in order to demonstrate comparison between pure W and Ni-doped W. They suggested that the solid-state activated sintering of W can occur due to a GB roughening transition induced by Ni segregation. Mishin et al. [9] performed atomistic simulations to confirm that GB mobility depends on the atomic structure in GB, operation temperature, and impurity content. While it is certain that Ni segregation in W GBs leads to embrittlement as well as material strength degradation, the effect of change in Ni atomic fraction in W GBs and the influence of such variation as a function of operation temperature on GB strength has not been characterized. Primary hindrances are the time scale limitations of molecular simulation methods to predict an equilibrium GB structure as a function of operating temperature in the presence of impurities and limitations of spectroscopy as well as microscopy based approaches in predicting atomic composition of nanometer sized GBs as a function of change in temperature. Another issue is the

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inability to predict GB mobility using molecular simulations while taking into account precise chemical composition due to molecular simulation time scale limitations. In this work, such limitations are circumvented by analyzing an experimentally observed $\langle 100 \rangle$ – $\langle 210 \rangle$ GB analyzed in the work of Gupta et al. [3]. This is the only work that has presented GB composition as a function of thickness using a combination of electron microscopy with thermodynamic calculations. Therefore, analyses only consider their work. The focus of the present work is on analyzing the influence of Ni doping in the examined W GB on its mechanical strength as a function of temperature while accounting for the effect of atomic level bond strength and lattice dynamics.

W and W alloys have earlier been analyzed for their tensile strength and fracture strength [10–12]. It has been shown that both temperature and Ni atomic fraction affects mechanical strength of nanostructured W [3,13,14]. However, a relationship describing such has not been established. In this work, GB structure experimentally observed by Gupta et al. [3] is analyzed for mechanical strength as a function of temperature and Ni atomic fraction using *ab initio* simulations based on Car Parrinello molecular dynamics (CPMD). The emphasis is on advancing the understanding of GB strength from the point of view of individual atom behavior. Focus is also on developing an analytical relationship to describe GB strength–temperature–Ni atomic fraction correlation. The amount of Ni in the GB is varied from 12.5% atomic fraction to 50% atomic fraction in order to analyze the role of Ni addition on the mechanical properties of Ni-doped W GB at the atomic scale as a function of temperature from 300 K to 600 K.

2. Formulation

In this study, computational chemistry software, NWChem, is used for performing *ab initio* CPMD simulations [15]. NWChem uses pseudo potential plane-wave (PSPW) basis sets to perform density functional theory (DFT) calculations underlying the CPMD simulations. PSPW basis sets have similar accuracy as the traditional Gaussian function-based approaches for performing DFT calculations. However, such basis sets are much faster and, therefore, can be used to simulate hundreds of atoms [16]. Advantage of CPMD simulations is the ability to simulate temperature dependent dynamics on a ground state potential surface directly at run-time using the Car–Parrinello algorithm. CPMD method's efficiency and accuracy makes it a good candidate for time dependent first principles simulations of complex molecular systems [17].

2.1. Car–Parrinello molecular dynamics

The CPMD method works on the principle of finding the motion of atomic nuclei as a function of time by performing molecular dynamics (MDs) calculations, while calculating *ab initio* quantum mechanical motion of electrons using DFT calculations [18]. In CPMD approach quantum–mechanical adiabatic time-scale separation of fast electronic and slow nuclear motions is performed in the framework of dynamical systems theory [19]. Advantage of CPMD framework is the ability to perform temperature and time dependent molecular calculations using DFT calculations. DFT calculations are based on pseudo potential plane wave (PSPW) formulation. PSPW formulation is one of the most widely used methods for calculating ground state properties within the framework of DFT [20–22]. In PSPW formulation, the core electrons are described by a pseudo-potential. The wave functions of the valence electrons are approximated by a plane wave basis set.

2.1.1. Simulation details

An ultrasoft pseudopotential [23], and plane wave approach by Kresse and Furthmüller [24] were used in all calculations. The plane wave cutoff energy of 350 eV was chosen. This is the same value which Becquart and Domain [25] used for their *ab initio* calculations. The error induced by the chosen cutoff energy was found to be negligible. The variation of the cutoff energy in the range of 350–700 eV did not affect the results indicating that the chosen cutoff energy level is sufficient for simulation results convergence. The time step for the CPMD simulations was set to 5.0 a.u. (0.121 fs, 8271 time steps per pico-second of simulation). Each simulation was run for 2000 time steps. The number of time steps was fixed after observing convergence in temperature, system energy, and pressure as a function of simulation time. Nose–Hoover thermostat scheme [26,27], was used for thermal equilibration of the examined W model system. The electronic fake mass for CPMD calculation is 400 a.u.

Perdew–Burke–Erzerhof [28], version of generalized gradient approximation (GGA) for the exchange–correlation energy functional, and $32 \times 32 \times 32$ Monkhorst–pack *k*-point mesh [29], for sampling of Brillouin zone were chosen in the present calculations. Such parameters were determined from several convergence tests, and the tests showed this choice was sufficient. We calculated the lattice constant of the W nanostructure sample as well as of Ni in equilibrium as a convergence test using different simulation parameters, and compared the results in order to determine the set of parameters that make the simulation results fit the best with the previously reported values. GGA was compared with the results of local density approximation (LDA) [30], for the exchange–correlation energy functional. $16 \times 16 \times 16$, $32 \times 32 \times 32$, $40 \times 40 \times 40$, and $50 \times 50 \times 50$ *k*-point mesh for the integration over the Brillouin zone were compared. Results for fictitious mass values of 400 a.u., 500 a.u., and 600 a.u. for CPMD were compared. Additional simulations showed that this computation methodology reproduces previously reported experimental data well. The lattice constant and Young's Modulus of examined W nanostructure in equilibrium were calculated using the final determined parameters and they were 3.148 Å and 400 GPa, respectively. These results are in good agreement with experimental values of 3.16 Å [31], and 410 GPa [32]. Similarly W–Ni bond length was found to be 2.86 Å in agreement with Scherer et al. [33]. In the case of exchange–correlation functionals, GGA takes into account the gradient of the density for the coordinate and LDA gives accurate result for ground-state energies [34]. There are several options in GGA such as Perdew–Burke–Erzerhof exchange model (PBE96) and xperdew91 [35]. These two approximation methods are frequently used by researchers [36,37]. In tests performed in the present study, PBE96 and xperdew91 produced almost identical convergence patterns.

2.1.2. Phonon dispersion relation calculations

Phonons are essential to understand the elastic properties of solids. Phonon dispersion relation is the connection between the frequency of the atomic vibrations and lattice wave vector. Phonon dispersion relations in this work are calculated by obtaining atomic force constants using the *ab-initio* simulations supplied into the general utility lattice program (GULP) [38]. Before the phonon dispersion calculations of the GB models, a single unit cell of W first examined. The results for W were found to be in good agreement with experiments [39]. While the most of phonon frequencies are positive, some of the GB phonon frequencies were found to be negative (shown later) indicating lattice instabilities. Such lattice instabilities are known to be often observed in the tensional loading simulations as reported by, Einarsdotter et al. [40] Lattice instabilities appear in the phonon dispersion are known to be an indication that limits the theoretical strength of material [41].

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