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Direct quantification of solute effects on grain boundary motion by atomistic simulations

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ABSTRACT

Direct quantification of grain boundary mobility with the presence of impurities poses a great challenge for investigating thermal stability of nanocrystalline alloys. By applying the interface random-walk method, we investigate the dopant segregation and precipitation from direct molecular dynamics simulations. It is found that existing atomistic simulation methods based on pure grain boundaries can be readily extended to extract the mobility of impure grain boundaries. Furthermore, it is confirmed that the grain boundary motion is controlled by the diffusion of segregated dopants at the interface as assumed by many theoretical models, but the grain boundary mobility is directly related to the impurity diffusion in the direction perpendicular to the boundary plane. By directly quantifying the mobility of impure grain boundary under experimental conditions, a correction to the well-accepted solute drag model is proposed.

1. Introduction

Nanocrystalline materials are generally unstable and suffer from rapid grain growth when exposed to high temperatures [1–5], which poses great challenge to maintain their superior properties over their coarse-grained counterparts during thermal processing or service. In recent years, many controlled experiments [1,6], analytical modeling [6,7], and atomistic simulations [8,9] have suggested alloying as an effective approach to stabilize grain boundary (GB) networks through the segregation of selected alloying elements at GBs. In particular, a regular solution model originally proposed by Cahn, Lücke and Stüwe (referred as CLS model) [10–12] has been widely used to study the solute drag effects on migrating GBs; it is assumed in the CLS model that the GB migration is controlled by the diffusion of segregated dopant atoms that move along with the migrating interface. However, although experimental studies on the mobility of GBs containing impurities have been made in the past [2], those studies have been mainly limited to extremely low concentrations, e.g., less than 10 ppm [2]. A direct quantification of the GB mobility with the presence of dopants over several orders of magnitude from either experiments or simulations, which is the key input to validate the CLS model, is still missing.

With the rapid development of high performance computation, various methods based on molecular dynamics (MD) have been developed to extract GB mobility [13–17]. Nevertheless, almost all past simulations were limited to pure systems; only a few qualitative studies have been done on the stabilization of GB networks in nanocrystalline materials by addition of impurities. For example, Millett et al. [8,9,18] have studied bulk nanocrystalline Cu with impurities segregated in the GB regions to investigate their influences on grain growth during annealing at constant temperature of 800 K. The GB energy was found to decrease with the increase of the impurity concentration and the atom radius mismatch between impurity and bulk atom. However, a quantitative prediction between the GB mobility and impurity concentration was not established. One main reason is that most MD methods require unrealistically high driving forces to enable atom transfer across the boundary so that the overall GB migration can be detected in typical MD time scale [13–16]. On the other hand, the rate of boundary migration in impure systems is mainly determined by the diffusional process of the impurities [10–12,19–21], which is too slow to be studied by MD. In addition, the simulation cell must be large enough to form a steady-state impurity profile normal to the GB plane [20]. However, such limitations in MD may be overcome by using the interface-random-walk (IRWalk) method [17,22,23], which is capable of extracting GB mobilities based on purely thermal fluctuations according to:

$$D_{GB} = \frac{d\langle \vec{d}^2 \rangle}{t} \sim \frac{2MkT}{A} \quad (1)$$

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where D_{GB} is the "diffusion" coefficient of the GB plane, T is temperature, A is the interface area, M is the GB mobility. To date, however, this method has been only applied to study pure GBs.

IRWalk method has been successfully applied in various pure GB systems to extract mobilities that were consistent with those extracted based on the driven motion methods at the low driving force limit in the past [17,23,24]. However, no attempt has been made yet to extend IRWalk method to investigate GBs containing impurities. The purpose of this work is thus to extend the IRWalk method to impure systems and directly quantify the mobilities of impure GBs under experimental conditions. It is also hoped to quantitatively validate the CLS model and fill the gap between MD simulations and analytical modeling in the past.

2. Methodology

2.1. MD simulation geometry and procedure

An inclined Σ5 GB in three types of Al-based alloys was simulated by MD using LAMMPS [25] with embedded-atom method potentials including Al–Ni [26], Al–Pb [27], and Al–Ti [28] with time step of 5 fs. The misorientation angle of the GB is 53.16° and the lattice orientations of the two grains are shown in Fig. 1(a). Various concentrations of impurity atoms, e.g., 2, 5, 10, 20, 41, 61 Ni, Pb, or Ti atoms, were introduced into the model by randomly replacing Al atoms at the GB region prior to the simulations, which correspond to 0.1%, 0.25%, 0.5%, 2.05%, 3.05% coverage of the GB by impurity atoms respectively. The initial simulation cell was set to be 5.69 nm × 5.69 nm × 10.48 nm in x, y and z directions (Fig. 1), respectively, accommodating 20,608 atoms. The geometry of this simulation cell is similar to the reference case proposed by Mendelev et al. for studying GB motions [24]. Periodic boundary conditions were applied along the x and y directions while the two surfaces perpendicular to the z-axis were free. Isothermal–isobaric ensemble (NPT) was used for the first 100 ps to relax the model and canonical ensemble (NVT) was used afterwards for all simulations. It needs to be mentioned that although the relaxation method used in this study may not be able to result in the physically equilibrated structure of the GBs, the interface random walk behavior of the GBs due to thermal fluctuation should not be significantly influenced [22,23]. For each alloy system of a specific concentration, 20 simulations (different random velocity initialization) up to 10 ns were performed. Atomic configurations of each alloy system were visualized by AtomEye [29]. The GB position can be determined by using an order parameter which depends on the local lattice orientation [22,23]. The system temperature was kept at 850 K so that significant GB fluctuation can be observed in these alloys. It has been tested at this temperature that a time step of 5 fs can well maintain the energy conservation in an NVE (micro-canonical ensemble) simulation, which ensures that the time step of 5 fs used in this study will speed up the simulation while maintaining the same degree of accuracy as compared to simulations with shorter time step such as 1 or 2 fs.

2.2. Determination of GB mobility by IRWalk method

According to the interface-random-walk method which was originally proposed by Trautt et al. [17], the GB mobility is correlated to the variance of GB displacements among a large number of independent simulations as described by Eq. (1). For each of

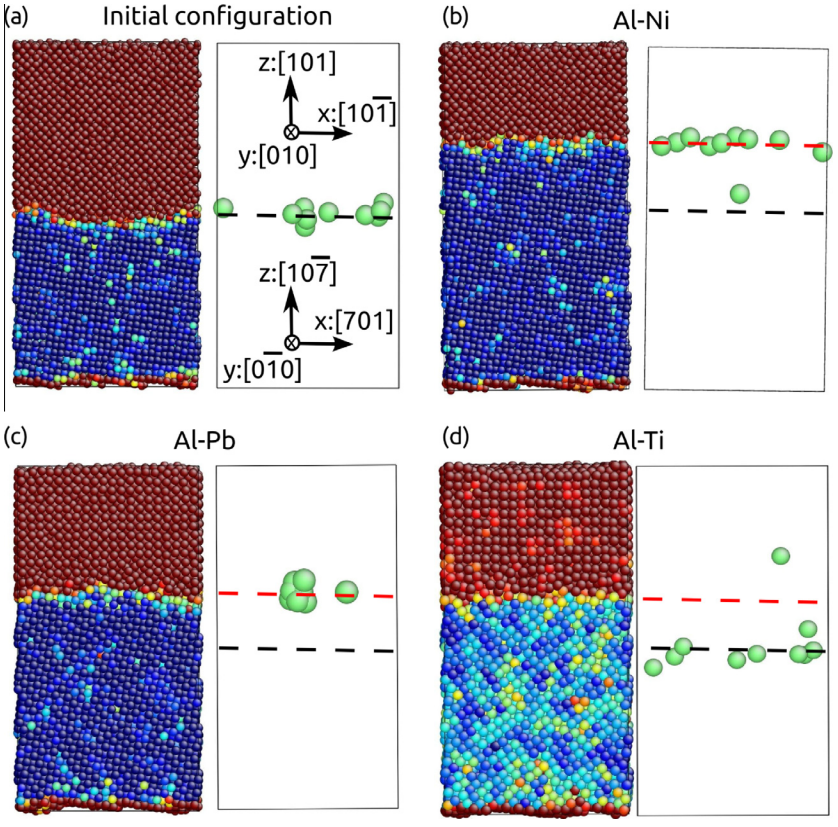

Fig. 1. (a) The initial atomistic configurations of the simulation cell in Al–Ni. The current atomistic configurations of the simulation cell during the GB thermal fluctuation are in (b) Al–Ni, (c) Al–Pb, and (d) Al–Ti. On the left the atom color corresponds to an order parameter depending on the local lattice orientation and on the right only dopant atoms are shown. The black and red dashed lines mark the initial and current GB positions, respectively. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)