



Atomistic simulation of the step mobility at the Al–Si(1 1 1) crystal–melt interface using molecular dynamics



P. Saidi*, J.J. Hoyt

Department of Materials Science and Engineering, McMaster University, Hamilton, ON, Canada

ARTICLE INFO

Article history:

Received 17 July 2015

Received in revised form 14 September 2015

Accepted 15 September 2015

Available online 27 September 2015

Keywords:

Mobility

Molecular dynamics

Al–Si

AEAM

ABSTRACT

Molecular dynamics simulations and an angular embedded atom method description of interatomic forces have been utilized to compute the mobility of steps on the faceted (1 1 1) crystal–melt interface in the binary alloy Al–Si. Two systems were studied: an Al–90%Si alloy in the temperature range of 1560–1580 K and Al–60%Si at $T = 1190$ – 1220 K. It was determined that the higher Si content alloy exhibited attachment controlled growth of steps whereas for the lower temperature, higher Al concentration alloy step growth is characterized by a diffusion controlled or mixed mode mechanism. The step mobility, which is the proportionality constant between the velocity and driving force, was determined for the Al–90%Si alloy as a function of temperature and composition. It was found that mobility decreases fairly rapidly with the addition of Al solute. Also, from the variation with temperature, it appears the mobility is proportional to the interdiffusion coefficient in the liquid.

© 2015 Elsevier B.V. All rights reserved.

1. Introduction

In the quantitative morphological modelling of solidification, one of the crucial materials parameters is the mobility of interface crystalline. Theories of interface motion in crystal growth classify the rate controlling parameter according to two systems: diffusion-controlled, and/or interface-controlled. In the case of a diffusion-controlled system, solute transfer to the surface may be limited by mass transfer or diffusion through the bulk solution. For such systems many analytical solutions are available [1–6]. However, in the case of an interface-controlled system, some rate-controlling events occur on the surface which control the mobility of the interface during solidification [7–9], and thus it is essential to investigate different properties of crystal–melt interfaces. Predictions of the so-called microscopic solvability theory of dendritic growth shows that the anisotropy of interfacial free energies and kinetic coefficients has a significant effect on growth velocity and dendrite tip radius [10,11].

Despite the importance of the kinetic coefficient in the modelling of solidification microstructures, very few experiments have successfully measured this term in metals and alloys [12]. Most of our knowledge about the physics of crystallization kinetics has been obtained from molecular dynamics simulations [13–25] using Lennard-Jones, hard-sphere, repulsive power-law and embedded

atom method potentials, as well as Stillinger–Weber for pure Silicon. Of the different methods, free-solidification is known as the most convenient technique [18]. In nearly all these studies the interface is rough, and atom attachment to the growing phase occurs readily at any point on the boundary. Central force Embedded Atom Method (EAM) [26] potentials have been used extensively in the study of metallic systems. However, in some systems a crystal will grow with a faceted interface from its melt, which means atomic attachment occurs at steps which are energetically favourable. Thermodynamic, structure and kinetic properties of steps at crystal–vapour interfaces [27] and crystal–melt interfaces [28,25] have been considered in some studies.

More specifically for the case of silicon, Landman et al. [29] observed the formation of {1 1 1} faceted planes and short range ordering in the melt close to the interfaces using MD simulations. In a different study, Luedtke [30] observed discontinuous growth of {1 1 1} layers in Si. In a related field of research Jackson [31] investigated the growth of silicon crystals from the amorphous phase. In addition, Buta et al. [25] used non equilibrium MD simulations of crystal growth to calculate the step kinetic coefficient at crystal melt interfaces, as well as the effect of step separation on the kinetic coefficient. In all these studies, the Stillinger–Weber (SW) potential [32] has been used to approximate the interactions between silicon atoms.

Although a detailed understanding of the properties of solid–liquid interfaces between chemically dissimilar materials is important for the description of a number of technologically

* Corresponding author.

E-mail address: saidip@mcmaster.ca (P. Saidi).

important phenomena, relatively few fundamental studies have been carried out on alloying systems [33,34]. Furthermore, to the knowledge of the authors, there have been no molecular dynamics study of the kinetic coefficient for a mixed type bonding system, such as a metallic species alloyed with a covalently bonded element. The main reason for the lack of data in such systems is that the classical MD interatomic potentials for mixed systems are difficult to model. However, in 2009, Dongare et al. [35] developed the angular-EAM (AEAM) interatomic potential model, which is specifically designed to model alloys of a metal species combined with covalently bonded materials such as silicon. In 2014 Saidi et al. used this model for the case of Al–Si to develop a potential which reproduces quite accurately the enthalpy of mixing for different compositions and ground state energies of the L12 and B1 structures. Moreover, the predicted phase diagram is very close to the experimental Al–Si phase diagram. For example, the eutectic concentration for the liquid agrees with experiment within 4 at.% and the eutectic temperature differs from experiment by just 13 K. Another important feature of this potential is zero solubility of Si in Al (and vice versa) in the solid state for the whole range of temperature. An advantage of this potential is that, in contrast to the original Stillinger–Weber (SW) potential where pure silicon stabilizes in the correct diamond cubic structure, a recent MD simulation of nucleation of solid circular islands on a liquid terrace frolov [36] showed the formation of the wurtzite crystal structure.

The current work is devoted to the study of the structure and kinetic properties of steps using an AEAM potential for Al–Si. However, in the first part of this paper we reproduce the results of Buta et al. [25] for pure silicon to assure that, despite the modification of SW potential, the satisfying capabilities of the original SW potential in reproducing silicon properties are preserved. For this purpose, the magnitude of the step kinetic coefficient, M_{step} , is defined as the proportionality constant between crystal growth and undercooling determines for silicon.

In the remainder of the paper, we change the composition of the melt in contact with the silicon crystal. Some important features such as the diffusivity of components in the Al–Si melt and the range of undercooling in which a reaction-controlled scenario is dominant are investigated. The nonequilibrium MD simulations are employed to calculate the effect of composition and step separation on the step kinetic coefficient from interfaces vicinal to the (111) plane. In this case the driving force of the transformation is the difference in chemical potential of Si between the solid and liquid [37], where the chemical potential change due to changes in liquid concentration in the liquid must be accounted for. At each temperature, the driving force as a function of composition is calculated using the Semi-Grand-Canonical technique, whereas the interface velocity during transformation is determined at each composition. The computed kinetic coefficients and related results are presented, followed by a discussion and conclusion.

2. Methodology of atomistic simulation

By definition, the step kinetic coefficient, M_{step} , of a crystal–melt interface is the constant of proportionality between the growth velocity, and the driving force of the transformation. Driving force for the solidification in pure systems is typically expressed in terms of the undercooling $\Delta T = T_m - T$ and for alloying systems it is the difference between the chemical potential at each component from the equilibrium chemical potential $\Delta\mu = \mu_{Eq} - \mu$ [38]. The step velocity, then, can be written as:

$$v_{step}(T, C) = M_{step}\Delta\mu(T, C) \quad (1)$$

where both driving force and velocity are dependent on the temperature and composition of transformation. Thus, to determined the

mobility, the interface velocity and driving force of solidification should be calculated at each point during the transformation.

The interactions between the components of this system are approximated by the angular embedding atom method (AEAM) empirical potential [39]. Morris' coexistence technique [40,41] was employed to prepare systems with coexistence of a pure silicon crystal in contact with a pure or alloying liquid. Since the crystal orientations are high index directions the size of the system should be calculated accurately to avoid very small distances between atoms under periodic boundary conditions. In order to prepare steps with different terrace lengths the crystal is rotated around the $[\bar{1}2\bar{1}]$ or $[10\bar{1}]$ directions, which results in the formation of steps along the $[\bar{1}2\bar{1}]$ and $[10\bar{1}]$, respectively. Table 1 shows the list of the vicinal orientations used in this study.

System preparation starts with the equilibration of pure silicon in the solid state using an *NPT* ensemble at the desired temperature T and pressure $P = 0$. The output of this level of simulation is used as the initial condition for the subsequent step. However, in the case of alloying systems, the type of some atoms changes to reach the corresponding composition of the liquid in contact with solid silicon crystal. The melt–solid system is prepared by employing the $NA_{yz}P_xT$ ensemble. This means that the system size is fixed in the y and z directions, and the pressure is controlled by changes in the x direction, which is perpendicular to the vicinal plane. The portion of the system that is melted increases to a temperature well above the melting temperature, then the temperature decreases gradually to the temperature of interest and is held constant until equilibration is established. Subsequently the system is quenched and the progress of solidification is captured. The amount of undercooling is determined with respect to the liquidus line in the phase diagram of Al–Si, whereas for the pure silicon system it is the melting temperature of silicon ($T_M = 1682 \pm 1$ K). Snapshots of the system were saved every 50 ps during this simulation. The snapshots contained positions of atoms, energies, and stresses, and were used for post processing.

Based on Eq. (1) the interface velocity and the driving force of transformation should be calculated for all independent snapshots. In the case of a pure silicon system, the crystal growth rate in the vicinal direction ($v_{vicinal}$) is proportional to the rate of energy release, $(\frac{dE}{dt})$, during solidification:

$$v_{vicinal} = \frac{1}{2A_{yz}\Delta H_m} \frac{dE}{dt} \quad (2)$$

where ΔH_m is the latent heat of melting per volume in the silicon diamond crystal structure and A_{yz} is the fixed cross sectional area of the simulation box perpendicular to the crystal/melt interface. According to the geometry of the periodic system there are two solid–melt interfaces, and therefore the denominator will carry a factor of 2.

However, in the case of alloying systems, the average velocity of interfaces in the vicinal direction is calculated from the rate of conversion of silicon atoms from liquid state to the solid state by passing through the interface $(\frac{dN_{Si}^{L-S}}{dt})$ [42].

$$v_{vicinal} = \frac{\Omega}{2A_{yz}} \left(\frac{dN_{Si}^{L-S}}{dt} \right) \quad (3)$$

where Ω is the atomic volume of silicon atoms in the solid state at the corresponding temperature. Converting $(\frac{dN_{Si}^{L-S}}{dt})$ using the mass conservation principle for a closed system, $v_{vicinal}$ can be reformulated as:

$$v_{vicinal} = \frac{\Omega N_{Al}}{2A_{yz}} \left(\frac{1}{1-C} \right)^2 \left(\frac{dC}{dt} \right) \quad (4)$$

Download English Version:

<https://daneshyari.com/en/article/7959151>

Download Persian Version:

<https://daneshyari.com/article/7959151>

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