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Structural and mechanical properties of homogeneous solid-liquid interface of Al modelled with COMB3 potential

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ABSTRACT

In this study, the homogeneous (1 0 0), (1 1 0) and (1 1 1) interface between solid Al and liquid Al are examined using a recently developed third generation of charge-optimized many body (COMB3) potential and molecular-dynamics simulation (MD). The estimated melting point is about 22% higher than the experimental value, but the reproduced radial distribution function of liquid Al agrees pretty well with experimental result. The interfacial widths (δ) of the (1 0 0), (1 1 0) and (1 1 1) interface derived from smoothed density profiles are about 4, 6 and 4 crystal layers, respectively. The stress profiles are highly orientation dependent and the magnitudes of the stress peaks near the homogeneous interfaces are weaker than those near heterogeneous interfaces. Excess interfacial stress and excess interfacial potential energy are also obtained. The information about the homogeneous interfaces obtained in this study can enrich the understanding of homogeneous solid-liquid interface and will serve as a useful reference for future studies of Al-ceramic interfaces using COMB3 potentials.

1. Introduction

The atomistic-scale structure and properties of solid-liquid interfaces are significantly important for the understanding of a wide range of phenomena, such as nucleation, casting and additive manufacturing techniques [1,2]. In the casting of aluminium and its alloys, grain refiners are added to the melt to improve the performance of final products by promoting heterogeneous nucleation. However, how the grain refiners work is still in debate [3,4]. To elucidate the underlying mechanism, it requires to investigate and compare the structures and properties of solid-liquid interfaces between liquid Al and different substrates [5]. Unfortunately, experimental characterization of solid-liquid interfaces at an atomistic length scale is extremely difficult. Until now two techniques, X-ray scattering method [6–8] and high-resolution transmission electron microscopy (HRTEM) [9–12] are available for such study. The material systems used in those experiments are few, with Al_2O_3-Al system being the most popular one. Hence molecular simulations of Al_2O_3-Al solid-liquid interface are desired, as they can provide quantities that cannot be obtained through experiments, such as the diffusion constant at the interface and interfacial free energy.

The most important task in molecular simulations of Al_2O_3-Al solid-liquid interface is to choose an appropriate interatomic potential for the system. Simple fixed charge empirical potentials [13,14] fall short of accurately modelling the complicated bonding environments, such as the Al_2O_3-Al solid-liquid interface [15]. The recently developed COMB3 potentials [16] can model many kinds of materials including Al_2O_3 and Al. In the development of COMB3 potentials, bond-order concept and a dynamic charge equilibration scheme have been introduced for descriptions of strong short-range interactions and electrostatic energies. These two features make COMB3 potentials seem to be suitable for molecular simulations of heterogeneous Al_2O_3-Al solid-liquid interface. However, in order to perform simulations of Al_2O_3-Al solid-liquid interface, some parameters reproduced by the COMB3 potential must be known in advance. One example is the melting point of Al predicted by the potential. This value will help to determine the temperature range that should be used in simulations of Al_2O_3-Al system and the superheating/undercooling of the Al melt. In addition, it has been demonstrated that the COMB3 potential for Al is able to accurately predict a variety of properties of solid Al at low temperatures [17], but properties related to liquid Al have not been reported so far. Previous studies

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[18,19] have shown that the solid-liquid interface structure has a strong correlation with the liquid structure. Therefore, it is necessary to calculate the liquid Al properties using the COMB3 potential. The degree of agreement between the calculated properties and their experimental counterparts will show whether the COMB3 potentials are suitable for simulations of heterogeneous solid-liquid interfaces.

In the current study, we first employ the COMB3 potential to calculate the melting point and radial distribution function (RDF) of liquid Al, which is a reflection of the liquid structure, and compare them with experimental data. This will directly facilitate the future studies of Al_2O_3-Al solid-liquid interfaces as described above. We then examine the interfacial layering and mechanical properties of the (1 0 0), (1 1 0) and (1 1 1) interface between solid and liquid Al in this study. The homogeneous Al-Al solid-liquid interfaces are of interest for two reasons. First, Al crystal can be regarded as a very effective nucleant for melt Al, while Al_2O_3 is an ineffective nucleant [5]. The information about the Al-Al interfaces obtained in this study can be compared with that of the Al_2O_3-Al interfaces to investigate the influence of substrates on liquid Al. Second, a full understanding of homogeneous interfaces is still lacking. Most previous simulations of homogeneous solid-liquid interfaces used model potentials, for example hard-sphere potential [20], Lennard-Jones potential [19], and inverse-power potential [21]. Although these model potentials can reveal basic trends, they are only a cartoon of real interatomic interactions. Recently more realistic EAM potentials [18,22–24] were used in studies of the Al-Al solid-liquid interface structure, but three of them [18,23,24] pinned solid atoms at their equilibrium sites. Pinning solid atoms at equilibrium sites makes simulations easier but creates an unrealistic interface, because the real interface position is mobile as demonstrated in Refs. [20,25,26].

The rest of this paper is organized as follows. In the second section, we introduce the COMB3 potential and the methods used in this study. In the third section, we present the results for the melting point, RDF and the structural and mechanical properties of homogeneous Al-Al solid-liquid interface, which are characterized by the interfacial profiles and interfacial excess quantities. We conclude in the last section.

2. Potential and methods

2.1. COMB3 potential

The potential employed in this study is the COMB3 potential for pure Al which has been recently developed by Choudhary et al. [17]. Here, we only give a brief sketch of the formalism involved, for a full derivation see [16]. In COMB3 potentials, for an atom locating at position r with charge q, its total energy ($U^{tot}(q, r)$) is given by:

$$U^{tot}(q, r) = U^{es}(q, r) + U^{short}(q, r) + U^{vdW}(r) + U^{corr}(r) \quad (1)$$

where $U^{es}(q, r)$ is the electrostatic energy including all the energies associated with charges, whose distribution is determined through a dynamic charge equilibration process. The short-range interaction energy $U^{short}(q, r)$ is expressed in pairwise interactions with bond order concept introduced. The van der Waals interaction $U^{vdW}(r)$ is described by Lennard–Jones potential. The last correction term $U^{corr}(r)$ is in the form of Legendre polynomials [16], mainly responsible for correcting energies arising from bond bending.

The bond order concept and dynamic charge equilibration are two key features introduced into the COMB3 potentials. The bond order reflects the bond strength based on its local environment, therefore giving COMB3 potentials the ability to model a system with different bond types. Dynamic charge equilibration is a process during which the charge associated with each atom is determined dynamically and autonomously according to its local environment. This makes it possible to model heterogeneous environments with changing local conditions, such as at interfaces where charge redistribution is required. These two features make the COMB3 potentials appear suitable for molecular simulations of the Al_2O_3-Al solid-liquid interfaces. In this study we employ the COMB3 potential for pure Al because properties related to liquid Al reproduced by this potential have not been reported so far. Note that the dynamic charge equilibration was not performed in the current study as the influence of change on pure Al is negligible. Not performing dynamic charge equilibrium allows to utilize a larger time step and saves computational time.

2.2. Interface preparation

All MD simulations were carried out using the LAMMPS software [27] with a time step of 1 fs and periodic boundary conditions in three directions. A Nosé–Hoover thermostat with a relaxation time of 100 fs was used to control temperature and a Nosé–Hoover barostat with a relaxation time of 1000 fs was adopted to control pressure. Visualization was performed in another software called VMD [28].

The first step of the simulations is to obtain the equilibrium lattice constant of Al at an estimated melting point by relaxing solid Al in the *NPT* simulation with zero barostat pressure. Then solid and liquid samples having equal cross-sections were prepared separately in the NP_zAT simulations, which allow the box length (z direction) to fluctuate while keep the cross-section area (xy) fixed. The solid samples were equilibrated for 200 ps, while the liquid samples were obtained by melting a crystal with the same cross-section as the solid samples at 2000 K for 200 ps first and then relaxing the liquid at the chosen temperature for another 200 ps. The potential energies of pure solid and pure liquid were recorded, which would be used in the calculation of melting point. Three solid-liquid interfaces: (1 0 0), (1 1 0) and (1 1 1) were examined in this study. With the lattice constant (a) for the model at the chosen simulation temperature determined in the previous *NPT* simulations, the simulation systems could be constructed. The basic information for the three systems is summarized in Table 1.

After the NP_zAT simulations, we chose solid and liquid samples whose dimensions match their average dimensions calculated from the final 10^5 steps of the NP_zAT runs. Then two chosen liquid samples were placed on both sides of a chosen solid sample in the z direction with an gap of 0.5 Å between them and thus formed a coexistence system having two interfaces. Fig. 1 shows the snapshot of the initial configuration of the (1 1 1) coexistence system.

2.3. Calculation of melting point

The melting point and its uncertainty were determined in two steps. In the first step, the solid-liquid coexistence systems with the (1 0 0) interface obtained as described above were equilibrated in the NP_zAT

Table 1
The geometrical parameters and number of atoms for the solid and liquid samples.

Interface	Orientation (xyz)	Dimensions		Number of atoms	
		Solid	Liquid	Solid	Liquid
(1 0 0)	[0 1 0] [0 0 1] [1 0 0]	10 a× 10 a× 20a	10 a× 10 a× 15a	8000	6000
(1 1 0)	[$\bar{1}$ 1 0] [0 0 1][1 1 0]	$7\sqrt{2}$ a× 10 a× $14\sqrt{2}a$	$7\sqrt{2}$ a× 10 a× $10\sqrt{2}a$	7840	5600
(1 1 1)	[10$\bar{1}$][$\bar{1}$2$\bar{1}$] [1 1 1]	$7\sqrt{2}a \times 4\sqrt{6}a \times 12\sqrt{3}a$	$7\sqrt{2}a \times 4\sqrt{6}a \times 10\sqrt{3}a$	8064	6720