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An improved charge transfer ionic-embedded atom method potential for aluminum/alumina interface system based on damped shifted force method

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ABSTRACT

An improved charge transfer ionic-embedded atom method (CTI + EAM) potential for aluminum/alumina interface system based on damped shifted force (DSF) method was proposed in this paper. To determine the effectiveness of the proposed potential, this study first investigated the effectiveness of the DSF method in the proposed potential. To assess the effectiveness of the DSF method in simulating each region of the interface system, several models were constructed, including bulk models, surface models and interface models. By systematically analyzing the effectiveness of the DSF method in predicting the atomic charges, the forces acting on the atoms and the systematic potential energy, the range of α for an effective DSF method in the proposed potential was determined: from 0.2 to 0.225 $Å^{-1}$. Then, the effectiveness of the proposed potential was validated. By relaxing of the aluminum/alumina interface model using the standard and improved CTI + EAM potential, and analyzing the accuracy of the predicted work of adhesion, the atomic configuration and the atomic charges, the range of α was shown to be reasonable and the optimum value of α was determined to be 0.2 $Å^{-1}$. In addition, it was revealed that to obtain suitable value of α in the DSF method, the priority level for the adoption of models is interface > surface > bulk for metal/metal oxide material systems.

1. Introduction

Aluminum/alumina composites are of great interest in many fields due to their excellent mechanical properties [1,2]. Maximizing the properties of these composites requires a detailed knowledge of the structure and properties of the interface between aluminum and alumina. So far, both experimental and theoretical studies on the structure of the interface have been conducted.

The crystalline orientation relationships between the aluminum and alumina phases in aluminum/alumina composites fabricated via heteroepitaxial growth have been studied by high-resolution transmission electron microscopy (HRTEM) [3,4]. The interfacial layer was also identified and investigated by HRTEM, and an interface thickness of approx. 1 nm was measured. Theoretical calculations (especially quantum calculations and molecular dynamics simulations) have been employed to determine the microstructure of such aluminum/alumina interfaces. Based on first-principle calculations, the configuration of the aluminum/alumina interface atoms was obtained [5–7]. However, first-principle calculations are impractical for handling large scale ($N > 10^2$) problems. In comparison, molecular dynamics simulations can be used to solve problems at a larger scale. In addition, an interface model usually contains a large number of atoms. Therefore molecular dynamics simulations should be more suitable for studying the aluminum/alumina interface. The accuracy of molecular dynamics simulations depends on the adopted potential, which must be determined first.

The standard charge transfer ionic and embedded atom method (CTI + EAM) potential proposed by Zhou et al. [8] has been demonstrated to be able to effectively and accurately simulate the interface between aluminum and alumina. This potential was revised from the electrostatic and embedded atom method (ES + EAM) potential by Streitz and Mintmire [9] and addressed the problem that the ES + EAM potential could not accurately describe the charge transfer in models under pressure. In our previous work, the standard CTI + EAM potential has been used to successfully simulate the interfacial configuration between aluminum and alumina and to reproduce experimental results [10].

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To further study mechanical behaviors around the interface, interactions between defects and the interface, and defects that source from the interface, large-scale (10,000+ atoms) models need to be adopted. However, the standard CTI + EAM potential is not efficient to handle such models. The low efficiency is mainly attributed to the method employed to calculate the long-range electrostatic energy $\left(\frac{1}{2}\mathbb{K}_e\sum_{i=1}^{N}\sum_{j=1}^{N}\frac{q_iq_j}{r_{ij}}\right)$, i.e., the standard Ewald summation method [11,12], which is the most time-consuming part of the simulations and has the complexity $O(N^2)$. The low efficiency limits the application of the standard CTI + EAM potential. Hence, finding a more feasible approach to replace the standard Ewald summation which offers a suitable accuracy and relatively low overhead costs was highly desirable.

There were some alternatives to the standard Ewald summation, including the simplified version of the standard Ewald method with complexity $O\left(N^{\frac{3}{2}}\right)$ [13], the Fourier transform-based Ewald methods with complexity $O(N\log(N))$ (for instance, the particle-particle-particle-mesh (PPPM) method [14], the particle-mesh Ewald (PME) method [15] and the fast Fourier Poisson (FFP) method [16]), and the damped shifted force (DSF) method with complexity $O(N)$ [17].

An illustration was performed to show the computational time of the mentioned alternatives. Since the Fourier transform-based Ewald methods have the same complexity, the representative PPPM method was chosen for investigation. A single-crystal bulk alumina model was constructed. The model contains 51,840 atoms and has the dimensions $49.447 \times 57.096 \times 155.880\ \text{Å}^3$. The charges of the atoms in the model were preappointed (Atom Al: +3e, atom O: −2e). The calculation of the systematic potential energy and virial stress was performed. Since the computational time of the simplified Ewald method and the PPPM method is related with the accuracy adopted, several typical accuracies were considered. For the DSF method, an typical cutoff of 15 Å was considered. As illustrated in Fig. 1, with the increase of the accuracy in the simplified method and PPPM method, the computational time increased, which was reasonable. In addition, overall, the sequence of the computational time of the methods was consistent with that of the complexity of the methods. Here, we chose the DSF method to substitute the standard Ewald method, since the computational cost of the DSF method is relatively low and the mathematical form of the method is simple.

Based on the above analysis, an improved CTI + EAM potential for aluminum/alumina interface system based on the DSF method was proposed here, which inherits its form and parameters from the standard CTI + EAM potential [8] and the DSF method should be used for calculating the long-range electrostatic potential. The effectiveness of the proposed potential is only dependent on the effectiveness of the DSF method in the potential. The accuracy of the method has been validated, and it was shown to be effective in simulating a variety of systems (liquid water, crystalline water, NaCl crystals and NaCl melts, etc.) compared to the smooth particle-mesh Ewald method [18]. In addition, the DSF method has been used in many fields [19–22]. Although it has already been demonstrated that the DSF method is a reasonable alternative to the standard Ewald sum in common molecular dynamics simulations, it was still necessary to validate the effectiveness of the DSF method in the potential by studying the performance of the DSF method in combination with the standard CTI + EAM potential when simulating the aluminum/alumina interface in comparison to the standard Ewald sum for two reasons:

Firstly, the improved CTI + EAM potential has the ability of predicting the charges of atoms, which is a vital function. Therefore, the effectiveness of the DSF method partially depends on the effectiveness of the DSF method in predicting the charges of atoms. However, the original study did not consider the effectiveness of the DSF method in predicting the charges of atoms [17].

Secondly, according to the original validation, the cutoff values used in the original validation might be unreasonable. Generally, a cutoff value is considered to be reasonable when the predicted results do not change with the increase of a cutoff value. Specifically, according to Fennell and Gezelter [17], for each model and each value of α, three different cutoff values were considered, but the rationality of the cutoff values has not been evaluated. For each model and each value of α, with the increase of the cutoff values, the predicted energy and the predicted values and degrees of the force vectors were found to vary as well, and they were not observed to converge to a certain value. Hence, the values of the parameters were very likely unreasonable, and therefore the results of the validation were unreasonable as well, and it was necessary to choose reasonable cutoff values in the new validation process. Since there was so far no evidence for the existence of the reasonable cutoff value, in this study, the existence of a reasonable cutoff value had to be proven for each model and each value of α first, and then the minimum cutoff value among all reasonable values was chosen and referred to as the optimum cutoff value for the remainder of the investigation.

The main aim of this study was to validate the effectiveness of the improved potential for simulating the aluminum/alumina interface. The paper is structured as follows: in Section 2, the related theory, methods and models are introduced, including the standard CTI + EAM potential, the summation methods, the methods for validation, the adopted parameters for validation and the adopted models; in Section 3, the existence of reasonable cutoffs in predicting physical quantities was validated, the effectiveness of the DSF method in predicting the charges of the atoms, the forces acting on the atoms and the systematic potential energy were evaluated individually, the effectiveness of the improved potential was validated and the characters of the DSF method in the potential were determined.

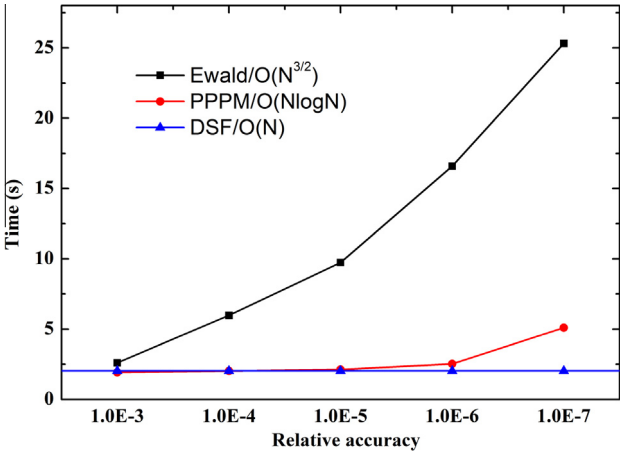

Fig. 1. Time vs. relative accuracy. Relative accuracy refers to the relative root mean square error in per-atom forces. For the DSF method, time is only directly dependent on cutoff.

2. Theory, method and model

2.1. The standard charge transfer ionic-embedded atom method potential

The total energy of the standard CTI + EAM potential model proposed by Zhou et al., which is revised from ES + EAM potential proposed by Streitz and Mintmire [9], can be expressed in the following form, which contains two parts: