Applied Surface Science 437 (2018) 103–109

Contents lists available at ScienceDirect

Applied Surface Science

journal homepage: www.elsevier.com/locate/apsusc

Full Length Article

Effects of Ti, Si, Mg and Cu additions on interfacial properties and electronic structure of Al(111)/4H-SiC(0001) interface: A first-principles study

Xin-Yu Xu, Hui-Yuan Wang, Min Zha, Cheng Wang*, Zhi-Zheng Yang, Qi-Chuan Jiang

Key Laboratory of Automobile Materials of Ministry of Education & School of Materials Science and Engineering, Nanling Campus, Jilin University, No. 5988 Renmin Street, Changchun, 130025, PR China

ARTICLE INFO

Article history:
Received 25 August 2017
Received in revised form
11 December 2017
Accepted 12 December 2017
Available online 16 December 2017

Keywords:
Composites
Surface energy
Interfacial adhesion
Electronic structure
First-principles calculation

ABSTRACT

In this study, the structural stability and electronic properties of Al(111)/4H-SiC(0001) interface, as well as the effects of Ti, Si, Mg and Cu additions on the interfacial adhesive strength are investigated via the first-principles method. Surface energy calculations show that 4H-SiC(0001) with C-termination is more active than Si-termination. Moreover, polar covalent bonds are formed across Al/4H-SiC interface. The results of separation energies suggest that the introduction of Ti and Si can improve the adhesive strength of C-terminated Al(111)/4H-SiC(0001) interface, whereas Mg and Cu have the opposite effect. According to the analysis of interfacial electronic structure, the covalent bonds across Al/4H-SiC interface are further enhanced with the addition of Ti and Si atoms, contributing to the improved adhesive strength in this case. However, the interactions between interfacial atoms are weakened with Mg and Cu doped in C-terminated interface, which results in the decrease of interfacial adhesion strength. Our calculations provide a guide to experiments on the design of 4H-SiC reinforced Al-matrix composites with tailored properties.

1. Introduction

SiC reinforced Al-matrix composites are widely used in industry due to their excellent physical and chemical properties. For the fabrication of Al/SiC composites, interfacial properties play a crucial role in determining the final mechanical behavior of composites [1]. Unfortunately, the wettability of SiC by Al was usually reported to be poor [2,3]. In order to improve the interfacial adhesion of Al/SiC interfaces, adoptable strategies include metal coating on SiC surfaces [4,5] and element alloyings in Al matrix [6]. Hereinto, because of simplicity and low-cost, elements additive technology has become one of the most extensively used techniques to fabricate Al/SiC composites with excellent performance.

Several researchers have studied the effects of different alloying elements on the interfacial properties between Al and SiC interface by experiments [6–8]. However, due to the rapid oxidation of Al and SiC as well as the easy evaporation of the alloying elements [9], it is difficult to experimentally fabricate pure Al/SiC interface, and to investigate the natural character of Al/SiC interfacial structure. The stability of pure Al/SiC interface and the real effects of dopants are still interesting for us to know.

The first-principles calculation is a powerful tool to study the interfacial information under the atomic level or even electronic level, which accurately estimate the interfacial stability and effects of dopants and so on [10–12]. In recent years, the properties of Al/SiC interface have been studied by the first-principles calculations [13–15]. However, most of these studies were confined to the Al/3C-SiC interfaces. Only few studies have been performed on Al/4H-SiC interfaces which are more commonly used for industrial purposes because of their complex and versatile structure. Moreover, the contents mainly focus on the interfacial bonding nature and features relating to the atomic structure, however, the effects of alloying elements on the adhesive strength of clean Al/4H-SiC interfaces are still not clear.

In this paper, the atomic structure, work of separation, and electronic properties of the Al(111)/4H-SiC(0001) interface are investigated using the first-principles method. Furthermore, we systematically examine the effects of different alloying elements (including Mg, Ti, Cu and Si) on the separation work and interfacial strength of Al/4H-SiC interfaces. The calculated results could provide a profound understanding of the mechanism of alloying elements that improve the stability and adhesive strength of Al/4H-

---

* Corresponding author.
E-mail address: chengwang@jlu.edu.cn (C. Wang).

https://doi.org/10.1016/j.apsusc.2017.12.103

SiC interfaces, and therefore providing references for the screening of potential alloying elements in preparation of Al/SiC composites with excellent performance.

2. Calculation details

Our calculations were performed by the CASTEP package [16,17], based on the density-functional theory (DFT). The electron exchange and correlation energies were calculated with the generalized gradient approximation (GGA) parametrized by the Perdew, Burke and Ernzefhof (PBE) [18]. The plane-wave cutoff energy was set to 350 eV. The Brillouin zone was sampled with Monkhorst-Pack k-point mesh. Specifically, the k-point of 8 × 8 × 8, 9 × 9 × 2 and 10 × 10 × 1 was adopted for bulk Al, 4H-SiC and Al/4H-SiC interface, respectively. The convergence criteria were as follows: the energy change converged to less than 10^{-5} eV/atom, maximum stress lower than 0.05 GPa, maximum force less than 0.03 eV/Å and maximum displacements within 0.001 Å.

The bulk properties of Al and 4H-SiC were calculated prior to the interface optimization. The calculated lattice constant for bulk fcc-Al is a = 4.044 Å, which is in good agreement with the previous calculated values (4.05 Å [19], 4.039 Å [20]) and the experimental data (4.05 Å [19]). For 4H-SiC, the calculated lattice constants a = b = 3.078 Å, c = 10.046 Å, also accord well with the experimental values, i.e. a = b = 3.073 Å, c = 10.053 Å [21]. These results show that the adopted parameters in our calculations can ensure enough precision.

3. Results and discussion

3.1. Surface properties

In order to insure that the Al(111) slab and 4H-SiC(0001) slab are thick enough to exhibit bulk-like interiors, convergence tests are conducted on Al(111) and 4H-SiC(0001) slabs in preparation for their use in interface calculations.

The surface energies of Al (111) slab are ascertained with the method proposed by Bottgeret al. [22,23]. For Al (111), the slabs with more than 7 layers can converge to 0.75 J/m^2. Thus, symmetric Al(111) slab with 7 atomic layers is adopted in the following interface geometries to insure the bulk-like interior.

4H-SiC(0001) is the classical polar surface containing only one species of atoms in surface layers, i.e. Si- or C- termination. Furthermore, the upper and lower 4H-SiC(0001) surfaces present different bonding modes, as shown in Fig. 1. The upper surface layer possesses only one dangling bond, named as Si-I or C-I surface. However, there are three dangling bonds for the atoms in lower surface layers, namely, Si-II or C-II surface. The convergence test of 4H-SiC(0001) surface is conducted using the method presented in Refs. [20,24]. The changes in interlayer distance for 4H-SiC(0001) surfaces after relaxation are summarized as a percentage of the bulk spacing, as shown in Table 1. It is found that the interlayer relaxations for the Si(C)-II surfaces are larger than those for the Si(C)-I surfaces, implying that the Si(C)-I surface is more stable than the Si(C)-II surface, and thus has a larger probability to expose. Therefore, the 4H-SiC(0001) slab with one dangling bond was employed in the following calculation. It is also revealed that the interlayer relaxations are mainly localized within the top three atomic layers. When the slab thickness (N) is more than 13, the interlayer relaxations for both the Si- and C-terminated surfaces are well converged.

The atomic species of the 4H-SiC surface play an important role in determining the interfacial adhesion. The stability of surfaces with Si and C terminations is investigated by calculating surface energies, respectively. The Si and C chemical potentials should be taken into account in the calculations of surface energy due to the non-stoichiometric feature of the adopted 4H-SiC supercells with 13 atomic layers. The surface energy of 4H-SiC, can be given by

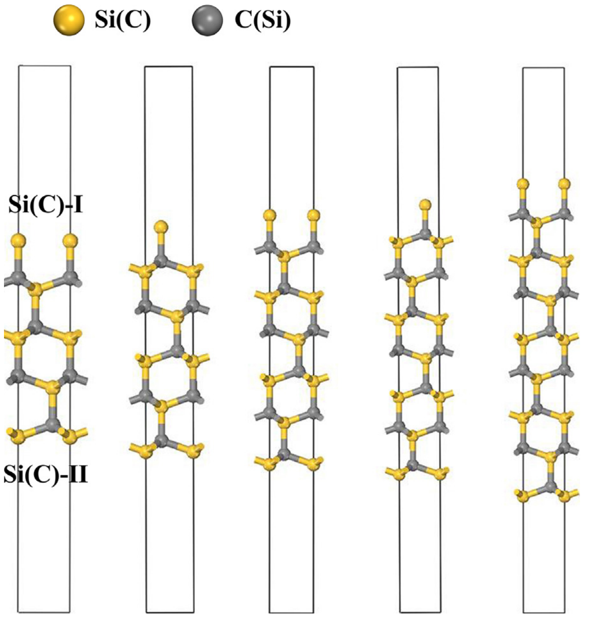

Fig. 1. Surface models of 1 × 1 4H-SiC(0001) slabs with increasing layers, in which the upper and lower atomic layers possess the same type of elements, i.e. Si- or C-termination.

$$\sigma_{\mathrm{SiC}} = \left[E_{slab} - N_C\mu_{\mathrm{SiC}}^{bulk} + (N_C - N_{\mathrm{Si}})\mu_{\mathrm{Si}}\right]/(2A_{surface}) \qquad (1)$$

where $A_{surface}$ is the surface area. E_{slab} is the total energy of a relaxed slab. N_{Si} and N_C are the number of Si and C atoms in the surface slab, respectively. μ_{SiC}^{bulk} stands for the chemical potential of bulk SiC, while μ_{Si} represents the chemical potential of Si in the surface slab.

The details of calculation method were presented in references [24,25]. The surface energies for Si- and C-terminated SiC(0001) surfaces are presented as a function of the Si chemical potential ($\mu_{Si} - \mu_{Si}^{bulk}$), i.e., the difference of Si chemical potential in surface slab and in pure Si bulk. The range of the Si chemical potential can be derived as

$$\Delta H_f^0 \le \mu_{Si} \le \mu_{Si}^{bulk} \le 0 \qquad (2)$$

The value of ΔH_f^0 is the formation enthalpy of bulk 4H-SiC, which is calculated to be 0.6591eV. In Fig. 2, the surface energy of the C-terminated (7.783–8.426 J/m^2) 4H-SiC (0001) slab is much

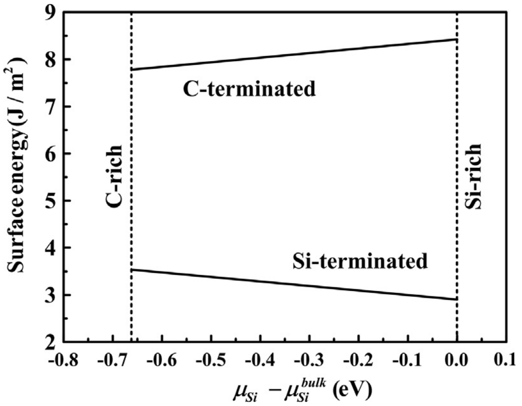

Fig. 2. Surface energies of the Si-terminated and C-terminated SiC(0001) slabs as a function of ($\mu_{Si} - \mu_{Si}^{bulk}$).