First-principles calculation of the interface stability of 3C-SiC(111)/Mg(0001)

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Abstract: The first-principles plane wave pseudopotential method based on density functional theory was used to study four SiC(111)/Mg(0001) interface models, and the ideal work of adhesion, charge density distribution, charge density difference and Mulliken population of the four interface models were calculated. The calculation results show that the structure of four interface models has not changed significantly, except for the more or less decrease in the interface spacing after geometry optimization. The Center-site interface of the C-terminated has the largest work of adhesion and the smallest interface spacing, which is the most stable structure among the four models. The calculation of the electronic structure shows that interfacial bonding of these two terminated interfaces of Center-site is a mixture of covalent and ionic bonds.

1. Introduction
Magnesium-based composite materials have the characteristics of high specific strength, high specific stiffness, high specific modulus, etc. and it is considered to be one of the most promising light metal materials in the high-tech industry in the 21st century. It is widely used in automobile manufacturing, electronic equipment, aerospace, defense equipment and other fields [1]. DFT (Density functional theory) has made outstanding contributions to the research of interface performance. It can show the atomic position in the interface intuitively, and it can analyse the combined mechanism of interface at the electronic level [2-3]. With the rapid development of computer technology in recent years, computer simulation technology has gradually become increasingly important in the development and research of materials, and the research and development mode of materials has gradually changed from traditional experimental method to theoretical simulation. Among them, the first-principles calculation method is the most reliable. For example, Shingo Tanaka et al. [4] performed a first-principles pseudopotential calculation on the 3C-SiC(111)/Al interface. The binding energy of the C-terminated interface of 3C-SiC(111) is greater than that of the Si-terminated interface. Rui Liu et al. [5] studied the interface structure of the Mg(0001)/TiB2(0001) interface and found that when Mg atoms combined with the TiB2 surface of Ti-terminated and the TiB2 surface of B-terminated, metallic/covalent mixed bonds and ionic bonds were formed respectively. The interface energy of the Mg/TiB2 interface is much larger than that of the α-Mg/Mg melt interface.

The bonding performance of the interface has an important influence on the composite material. However, there is no report on the deeper level of the bonding performance of the SiC/Mg interface. The first principles based on density functional theory have advantages for the research of the interface characteristics. In terms of studying the mechanism of action and interface characteristics, it can satisfy the study of the interface bonding mechanism of SiC/Mg metal-ceramic composites. In this
paper, 4 kinds of 3C-SiC(111)/Mg(0001) interface models are studied through first principles, and a stable structure is obtained by analyzing ideal work of adhesion and electronic structure.

2. Calculation method and model
All calculations in this paper were run by the CASTEP module of MATERIALS STUDIO software based on density function theory [6]. GGA-PBE was selected as the exchange correlation potential functional describing the interaction between electrons and electrons, using the BFGS algorithm, and the plane wave super soft pseudopotential was used as the pseudopotential. The self-consistent field method (SCF) was adopted, the convergence threshold of the self-consistent field was set as energy for 1.0×10^{-6} eV/atom, the energy cut-off point was selected to 380eV, and the maximum number of self-consistent field allowed for energy calculation is 100. The maximum force, the maximum stress and the maximum displacement was selected as 0.03eV/Å, 0.05GPa and 0.001Å respectively, which was used for geometric optimization. The K value of the first Brillouin zone was set as 6×6×1.

The Mg cell is composed of two Mg atoms, the space group is P63/mmc (No.194), and the lattice parameters are a=b=3.2094, c=5.2105, α=β=90°, γ=120°. 3C-SiC, also known as β-SiC, has a zinc blende structure and the space group is F-43m, and the lattice parameter is a=4.348Å. Its primitive cell is composed of 4 Si atoms and 4 C atoms. The atom in the middle is surrounded by four close atoms of different types to form a regular tetrahedron. Mg(0001) surface can be cut into two types of structures. SiC(111) surface is a polar surface, which can be cut into two types of structures, Si-terminated and C-terminated.

After calculating the surface energy of the two terminal structures of the SiC(111) surface, the results show that the surface energy of the Si-terminated is almost the same as that of the C-terminated. So, four surface models can be formed by combining two structures of SiC(111) and two structures of Mg(0001).

![Fig 1. Four structures of Si-terminated and C-terminated SiC(111)/Mg(0001).](image-url)
3. Results and discussion

3.1. Ideal work of adhesion

The interfacial bonding strength of the metal/ceramic is usually expressed by the physical quantity of work of separation. The opposite of work of separation is work of adhesion. Work of adhesion refers to the energy released when two free surfaces are combined to form an interface. The calculation formula is below [7]:

\[
W_{ad} = \frac{E_{SiC}^{slab} + E_{Mg}^{slab} - E_{SiC/Mg}^{total}}{A}
\]  

(1)

Where \(E_{SiC}^{slab}\) and \(E_{Mg}^{slab}\) are the energy of SiC(111) and Mg(0001) after geometry optimization respectively. \(E_{SiC/Mg}^{total}\) is the energy of SiC(111)/Mg(0001) after geometry optimization. A is the surface area of the SiC(111)/Mg(0001) interface.

Table 1. Work of adhesion and interface spacing of SiC(111)/Mg(0001) interface.

| Stacking  | d0/Å  | Wad/ J/m² |
|-----------|-------|-----------|
| Si-terminated | 1  | Center  | 2.4024 | 1.8053 |
|           | 2  | Top     | 2.7086 | 1.7155 |
| C-terminated | 3  | Center  | 1.7193 | 2.5834 |
|           | 4  | Top     | 2.1950 | 2.4104 |

After geometric optimization, the interface spacing of the four models in the z-axis direction is reduced to varying degrees, and the interface spacing has a close relationship with the interface bonding strength. Among the four SiC(111)/Mg(0001) interface models, the interface spacing of the C-terminated models are smaller than that of the Si-terminated models. In terms of work of adhesion, the C-terminated models are larger. Relatively speaking, C-terminated models are significantly more stable. Among the Si-terminated and C-terminated models, the center-site models are better than the top-site models, with smaller interfacial spacing (2.4024 Å and 1.7193 Å) and bigger work of adhesion (1.8053 J/m² and 2.5834 J/m²).

3.2. Electronic structure analysis

Fig 2. (a) Charge density distribution and (b) charge density difference of Center-site Si-terminated SiC(111)/Mg(0001) interface.
Fig 2(a) and (b) are the charge density distribution and charge density difference of the Center-site Si-terminated SiC(111)/Mg(0001) interface, respectively. In Fig 2(a) and Fig 3(a), there is charge accumulation between Mg side and SiC side. The charge density of SiC side is mainly concentrated near C atom, because C atom has strong electronegativity compared with Si atom. As can be seen from Fig 2(b) and Fig 3(b), the vicinity of Si atom is mainly the blue area where electrons are lost, while the vicinity of C atom is mainly the red area where electrons are gained. There is overlap of electron cloud in the interface, and the overlap of electron cloud in Fig 3(b) is more obvious than that in Fig 2(b). The electrons lost by Mg atoms enter the interface and form ionic bonds/covalent bonds with the charges of Si atoms and C atoms in the interface. The phenomenon of electron accumulation in the interface in Fig 3(b) is more significant than that in Fig 2(b), indicating that the interface bonding of C-terminated structure is more stable than that of C-terminated structure.

| Stacking Sites | Species | s  | p  | d  | Total Charge (e) |
|----------------|---------|----|----|----|------------------|
| Si-terminated Center Interface | Mg | 0.85 | 6.97 | 7.82 | 0.18 |
| Interface | Si | 1.19 | 2.19 | 3.38 | 0.62 |
| 2nd layer | C2 | 1.45 | 3.65 | 5.10 | -1.10 |
| 3rd layer | Si | 1.03 | 1.78 | 2.81 | 1.19 |
| C-terminated Center Interface | Mg | 0.67 | 6.71 | 7.38 | 0.62 |
| Interface | C | 1.51 | 3.77 | 5.28 | -1.28 |
| 2nd layer | Si | 1.08 | 1.95 | 3.04 | 0.96 |
| 3rd layer | C | 1.45 | 3.72 | 5.17 | -1.17 |

| Stacking Sites | Bonds | Populations |
|----------------|-------|-------------|
| Si-terminated Center Si1-Mg1 | 0.20 |
| C2-Si1 | 0.79 |
| Si3-C2 | 0.69 |
| C-terminated Center C1-Mg1 | -0.05 |
| Si2-C1 | 0.87 |
| C3-Si2 | 0.68 |
The population can reflect the distribution of electrons at the interface and measure the bond type and strength. The closer the Mulliken overlapping population is to 0, the iconicity is stronger, and the closer the Mulliken overlapping population is to 1, the covalency is stronger. In the Si-terminated SiC (111)/Mg (0001) interface, Mg atoms at the interface lose part of electrons and become +0.18 valence, and the Mulliken overlapping population with Si atoms at the interface is 0.20, which is in the bonding state. Compared with the Si-terminated, the Mg atom at the interface of C-terminated SiC (111)/Mg (0001) lost more electrons (+0.62), and form a weak anti-bonding state with the C atom at the interface, with Mulliken overlapping population of -0.05. The overlapping population of Si-Mg bond formed by Si-terminated Mg atom and adjacent atom is larger than C-Mg bond formed by C-terminated Mg atom and adjacent atom, and iconicity of Si-Mg bond is weaker than that of C-Mg bond. It can be concluded from the table that the overlapping populations (0.79 and 0.87) of Si-C bonds at Si-terminated and C-terminated SiC (111)/Mg (0001) interface are stronger than that of internal Si-C bonds (0.69 and 0.68), indicating that the interface has changed the covalent bond properties of Si-C.

4. Conclusion
In this paper, the first-principles method is used to calculate the work of adhesion and electronic structure of the SiC(111)/Mg(0001) interface, considering the Center-site and Top-site stacking structures of the interface, and the Si-terminated and the C-terminated. After calculation, it can be concluded that the work of adhesion of the Center-site structure is bigger than that of the Top-site structure, the work of adhesion of the C-terminated is bigger than that of the Si-terminated, and the structure of Center-site C-terminated is the most stable among the four structures. According to the analysis of electronic structure and Mulliken charge, the two interfaces have covalent bonds and ionic bonds.

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