Investigation on Absorption and Diffusion of the Oxygen in the Asymmetric Face of the $\beta$-Silicon Carbide through the Born-Oppenheimer Molecular Dynamics

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Abstract. Many methods had been proposed to process the $\beta$-silicon carbide, and the thermal oxidation-assisted polishing was considered as an efficient technique. The critical procedure in the thermal oxidation was absorption and diffusion process of the oxygen in the $\beta$-silicon carbide, especially in the asymmetric face. Therefore, the absorption and diffusion of the oxygen in the asymmetric face of the $\beta$-silicon carbide by born-oppenheimer molecular dynamics were studied. It could be observed from absorption and dissociation process of the O atom with the cell size that absorption of the oxygen in upper Si surface was fast, which was realized when the distance was smaller than 3Å. It could be observed from absorption and dissociation process of O atom with supercell size that absorption and oxidation for the Si surface was quickly, and steady oxide layer with silica was formed. On the contrary, absorption range of the C surface was smaller than that of the Si surface, and the surface recombination was more significant for the C surface. It could be calculated that the bond length of Si-O in the oxide layer was in the 1.65-1.77Å. The research products obtained in this study were propitious to reveal the mechanism in the thermal oxidation of asymmetric face of the $\beta$-silicon carbide with oxygen under ultra-high temperature.

1. Introduction

As one of the common silicon carbide products, hardness of the $\beta$-silicon carbide is close to that of the diamond, and its fineness and polishing performance is obviously better than that of the corundum and that of the $\alpha$-silicon carbide. Moreover, electrical conductivity of the $\beta$-silicon carbide is several times higher than that of the $\alpha$-silicon carbide. Meanwhile, the $\beta$-silicon carbide can obtain excellent thermal conductivity and low expansion coefficient, which indicates that the thermal stress will be very small in the heating and cooling processes. Therefore, many methods had been proposed to process $\beta$-silicon carbide [1, 2], which aimed to improve its machining level.

Thermal oxidation-assisted polishing, which included oxidation of the hard material by the oxygen under ultra-high temperature and polishing of the soft oxide layer by the ceria slurry [3-6], could be considered as an efficient technique to process $\beta$-silicon carbide. The critical procedure in the thermal oxidation was absorption and diffusion process of the oxygen in the $\beta$-silicon carbide, especially in the asymmetric face, because most of the $\beta$-silicon carbide grains in the sintered or boned silicon carbide products were in the asymmetric state [7-9]. Therefore, the absorption and diffusion of the oxygen in the asymmetric face of the $\beta$-silicon carbide through the born-oppenheimer molecular dynamics were investigated in this research, which aimed to reveal the thermal oxidation mechanism.
2. Simulation with cell size
According to the constructed born-oppenheimer molecular dynamics simulation model, absorption and dissociation process of the O atom with the cell size for the asymmetric face is shown in the Figure 1, and the corresponding root mean square deviation curve is shown in the Figure 2.

![Figure 1. Absorption and dissociation process of the O atom with the cell size for asymmetric face.](image1)

![Figure 2. Root mean square deviation curve of the O atom with the cell size for the asymmetric face.](image2)

It could be observed from the Figure 1 that absorption of the oxygen in the upper Si surface was fast, which was realized when the distance was smaller than 3Å. Meanwhile, the dissociation could be finished during 200-300fs. It could be found from Figure 2 that there was continuous position change during 0-800fs, and the variation still existed in the range of 800-3000fs, since there was new oxygen...
absorbed and dissociated. Distribution of the state density of O atom with the cell size for asymmetric face is shown in the Figure 3, and the distribution of the projective state density of the CO molecule is shown in the Figure 4. In the Figure 4, the mark 2 was C atom and the mark 31 was O atom. It could be found that their atomic orbitals almost overlapped, even for the ranges above the Fermi level.

![Figure 3. Distribution of the state density of the O atom with the cell size for the asymmetric face.](image)

![Figure 4. Distribution of the projective state density of the CO molecule.](image)

3. Simulation with supercell size
Absorption and dissociation process of O atom with supercell size for the asymmetric face is shown in the Figure 5, and the corresponding born-oppenheimer molecular dynamics simulation times were 0fs, 65fs, 165fs, 270fs, 810fs, 1850fs, 2391fs, 4951fs, 4999fs, 5505fs, and 5725fs respectively. Size of the supercell in the Figure 5 was a=b=8.69Å, and thickness of the vacuum layer was 20Å. Relative to the simulation model for the cell size, absorption superficial area for supercell size was larger for 4 times.
Figure 5. Absorption and dissociation process of O atom with supercell size for the asymmetric face.
It could be observed from the Figure 5 that the oxygen molecule was absorbed by the C surface at the 65fs, and one oxygen molecule was close to the position to be adsorbed for the Si surface. With the simulation time of 165fs, the oxygen molecule absorbed onto the C surface was decomposed, and one oxygen was absorbed onto the Si surface. It could be found that surface recombination phenomenon for the C surface was more intense than that for the Si surface under the high temperature thermal oxidation. At the simulation time of 270fs, the oxygen molecule absorbed onto the Si surface was decomposed, and the O atom on the C surface began diffusion along the surface. With the simulation time of 810fs, there were more oxygen molecules were absorbed on the upper Si surface, and the O atom onto the C surface swung with the connected C atoms. At the simulation time of 1850fs, one oxygen molecule got into the absorption range of the C surface, and the absorption and decomposition were finished at 2391fs. Until 4951fs, the O atom and the connected C atoms kept swinging. During this period, the Si surface kept absorbing new oxygen molecules until all the bridge locations were completely filled. Comparing the absorption and dissociation process of the O atom with the cell size in the Figure 1 and those with the supercell size in the Figure 5, it could be found that there were many common points between them two. It could be found that the absorption and oxidation for the Si surface was quickly, and steady oxide layer with silica was formed. On the contrary, absorption range of the C surface was smaller than that of the Si surface, and the surface recombination was more significant for the C surface. Judging from the BADER analysis, it could be found that charge of the surface C atom was smaller than that of the inner C atom for 1.5e, and that of surface Si atom was larger than that of the inner Si atom for 1.5e by contrary, which indicated that the generated surface had polarity.

Root mean square deviation curve of the O atom with supercell size for asymmetric face is shown in the Figure 6. It could be observed that diffusion process of the O atom was smooth relatively. The root mean square deviation curve became smooth gradually at the 2750fs, and it was close to flat state at the simulation time of 3750fs. It could be calculated that the bond length of Si-O in the oxide layer was in the 1.65-1.77Å, which was consistent with that of the silica.

![Figure 6. Root mean square deviation curve of the O atom with supercell size for asymmetric face.](attachment:Figure6.png)
4. Conclusions
Absorption and diffusion of the oxygen in the asymmetric face of the $\beta$-silicon carbide through the born-oppenheimer molecular dynamics were conducted in this research. It could be observed from absorption and dissociation process of the O atom with the cell size that absorption of the oxygen in the upper Si surface was fast, which was realized when the distance was smaller than 3Å. Meanwhile, the dissociation could be finished during 200-300fs. It could be observed from absorption and dissociation process of O atom with supercell size that absorption and oxidation for the Si surface was quickly, and steady oxide layer with silica was formed. On the contrary, absorption range of the C surface was smaller than that of the Si surface, and the surface recombination was more significant for the C surface. It could be calculated that the bond length of Si-O in the oxide layer was in the 1.65-1.77Å, which was consistent with that of the silica. The research products obtained in this study were propitious to reveal mechanism in thermal oxidation of the silicon carbide with oxygen under ultra-high temperature.

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