

1P19 Accelerated quantum chemical molecular dynamics study on the properties of excited state

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【Introduction】

The pollution by environmentally harmful organic compounds is coming up as a serious problem. Some researches have proved that TiO_2 is an effective photooxidation catalyst for such organic compounds. It is proposed that the photogenerated $\text{OH}\cdot$ radicals derived from water adsorbed on the TiO_2 surface are important oxidation agents, which control the kinetics of the overall oxidation process. Thus the identification of $\text{OH}\cdot$ radical species on the surface is important to understand the mechanism of photooxidation of organic compounds. In this study, the dynamic processes of $\text{OH}\cdot$ radical produced on the TiO_2 surface under the light irradiation have been simulated using our accelerated quantum chemical molecular dynamics program.

【Computational methods】

The accelerated quantum chemical molecular dynamics program “Colors”, which is based on our original tight-binding approximations, was used for the simulation. The Amsterdam density functional (ADF) program based on the density functional theory is employed to determine the parameters for “Colors” and to optimize the structure of acetone molecule.

【Results and Discussion】

The energies of the singlet and triplet state of the anatase TiO_2 bulk were calculated by “Colors” program. Furthermore, the excited-states dynamic behaviors of the anatase TiO_2 surface with dissociatively adsorbed water were simulated using “Colors” program. Fig. 1 shows the snapshots of the dynamic processes of the anatase TiO_2 surface with dissociatively adsorbed water at (a) 0 step, and (b) 300 step, respectively. The

light was introduced to this system by promoting the electron from HOMO to LUMO orbital. It was found that $\text{OH}\cdot$ radicals were produced at 300 step (See Fig. 1b). The change of the bond length between surface Ti atom and $\text{OH}\cdot$ radicals was investigated. It is clear that the bond length of Ti-OH increased with the time proceeding. The Ti-OH distance becomes long enough to indicate that the bond of Ti-OH was broken and $\text{OH}\cdot$ radical was formed. Our results support experimental finding that $\text{OH}\cdot$ radicals are the active species in photocatalysis on the TiO_2 surface. In addition, it proves that our “Colors” program is a very effective tool to simulate the dynamic reactions involving the excited states.

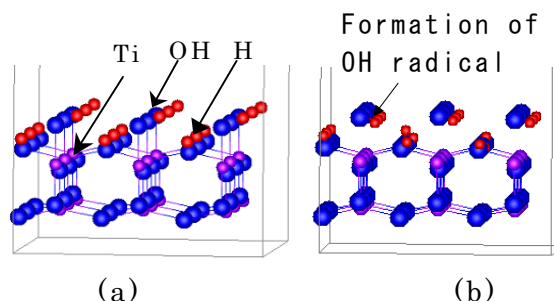


Fig. 1. Snapshots taken from quantum chemical MD simulation of anatase TiO_2 surface with dissociatively adsorbed water at 0 step (a), and 300 step (b).