

酸水素化物への表面吸着と反応
Surface reactions and adsorption on oxyhydrides

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研究成果概要

Heterogeneous catalytic CO₂ methanation is a promising CO₂ conversion reaction that it typically uses oxide-supported transition metals as the catalysts. For these heterogeneous catalysts, tremendous efforts have been dedicated to tuning the particle size, surface area, or cation compositions of the oxides. We have taken an oxide, barium titanate (BaTiO₃) and converted it to its oxyhydride, BaTiO_{2.4}H_{0.6}. In the presence of Ni or Ru, the presence of hydride enhances the catalytic activity of CO₂ methanation by 2~7 times. We find the hydride here seems to be adequately stabilized in the perovskite oxide lattice despite the water-rich environment. Based on our kinetic analysis, we discuss the reaction mechanisms that are different from the conventional oxides and show that the lattice hydride improves the hydrogen adsorption on metal surface. However, unlike NH₃ synthesis, the role of hydride anion in CO₂ methanation reactions is still unclear. It is not trivial to gain a complete picture of the mechanism, but some simple DFT calculations offer us answers to focused questions. In other words, does the incorporation of hydride anion into BaTiO₃ improve the adsorption (or decrease the chemisorption energy) of CO₂ molecular on the oxyhydride surface?

Hence, I have been using supercomputer for the following works:

- 1/ To build the stable slab structures of BaTiO₃ and BaTiO_{3-x}H_x;
- 2/ Put CO₂ molecular onto different oxide or oxyhydride surface and perform the DFT calculations to obtain the chemisorption energy;

$$E_{chem}^{CO_2} = E_{CO_2+slab} - E_{CO_2} - E_{slab}$$
where, $E_{chem}^{CO_2}$ is the adsorption energy of CO₂ molecular, E_{CO_2+slab} is the energy of CO₂ and slab structures, E_{CO_2} and E_{slab} is the energy of CO₂ molecular and slab structure, respectively. These surface calculations are still on the way.