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 CO_2 methanation is a promising CO_2 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 ($BaTiO_3$) 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 CO_2 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 NH_3 synthesis, the role of hydride anion in CO_2 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 $BaTiO_3$ improve the adsorption (or decrease the chemisorption energy) of CO_2 molecular on the oxyhydride surface?

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

1/ To build the stable slab structures of $BaTiO_3$ and $BaTiO_{3-}$ H ;

2/ Put CO_2 molecular onto different oxide or oxyhdyride surface and perform the DFT calculations to obtain the chemisorption energy;

$$_{chem} = {}_{2+slab} - {}_{2} - {}_{slab}$$

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