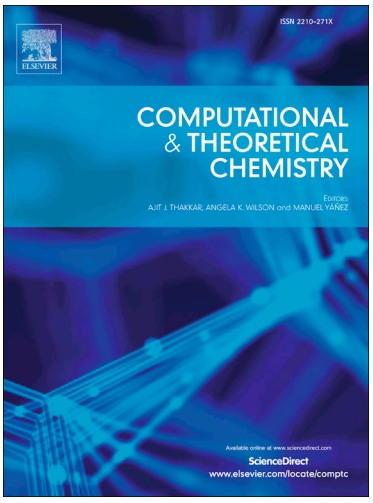

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Theoretical insights into the surface structure of In_2O_3(110) surface and its effect on methanol synthesis from CO_2 hydrogenation

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Abstract

The surface structure of In_2O_3(110) surface and its effect on methanol synthesis from CO_2 hydrogenation via HCOO route have been studied using combined density functional theory calculations and the atomistic thermodynamics method. The thermal desorption of surface oxygen atoms on In_2O_3(110) surface are random and not affected by the adjacent vacancies. The temperature and components of gas atmosphere have pronounced effect on the structure of In_2O_3(110) surface while the effect of pressure is inapparent. Under actual methanol synthesis conditions, the surface structure of In_2O_3(110) surface is in the state of few surface oxygen atoms on the surface. The In_2O_3(110) surface under lower surface reduction degree is favorable for methanol synthesis since the excessive formed oxygen vacancies around the active site on the surface can prohibit the dissociation of molecule H_2, hydrogenation of adsorbed CO_2 and protonation of H_3CO species, and is not facile for the stability of adsorbed CO_2 and dissociatively adsorbed H_2. The fundamental effect of the surface reduction degree is the transformation of the micro surface environment of the active site with the change of surface reduction degree of In_2O_3(110) surface. It is essential to maintain the balance between surface oxygen atoms and vacancies with a lower surface reduction degree on In_2O_3 surface to obtain a higher performance for methanol synthesis from CO_2 hydrogenation.

Key words: In_2O_3, methanol synthesis, surface structure, structure activity relationship, DFT, atmostic thermodynamics

1. Introduction

Since it is of great significance on mitigating global climate change and closing carbon cycle, the direct catalytic conversion of CO_2 into monoxide [1], alcohols [2-4], fuels [5-7] and other valued chemical products [8-12] has attracted considerable interest. Among them, the direct methanol synthesis from CO_2 hydrogenation has obtained great progress using heterogeneous supported metal or metal oxide catalysts in recent years. Consequently, it can be widely utilized as feedstock for the production of other chemicals [13-15]. Previous studies demonstrate that though the Cu [16-19] and Pd [20, 21] catalysts exhibit the potential to form methanol, the CO is the major