First Principle-Based Dft and Microkinetic Simulation of Co-Conversion of Carbon Dioxide and Methane on Single Iridium Atom Doped Hematite with Surface Oxygen Defect

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Abstract : The catalytic co-conversion of CO_2 and CH_4 to value-added compounds has become one of the promising approaches to addressing global climate change by having valuable fossil fuels. Thedirect co-conversion of CO_2 and CH_4 to value-added compounds is attractive but tremendously challenging because of both molecules #39; thermodynamic stability and kinetic inertness. In the present study, a single iridium atom doped and a single oxygen atom defect hematite (110)surface model catalyst, which can comprehend direct C-O coupling based on simultaneous activation of CO2 and CH4 was studied using density functional theory plus U (DFT + U)calculations. The presence of dual active sites on the $Ir/Fe_2O_3(110)$ -OV surface catalyst enables CO_2 activation on the Ir site and CH_4 activation at the defect site. The electron analysis for the theco-adsorption of CO_2 and CH_4 deals with the electron redistribution on the surface and clearly shows the synergistic effect for simultaneous CO_2 and CH_4 activation on Ir/α - $Fe_2O_3(110)$ -OV surface. The microkinetic analysis shows that the dissociation of CH4 to CH3 * and H^* plays an excellent role in the C-O coupling. The coverage analysis for the intermediate products of the microkinetic simulation results indicates that C-O coupling is the reaction limiting step. Finally, after the CH_3O^* intermediate product species is produced, the radical hydrogen species spontaneously diffuse to the $CH3O^*$ intermediate product to form methanol at around 490 [K]. The present work provides mechanistic and kinetic insights into the direct C-O coupling of CO_2 and CH_4 , which could help design more-efficient catalysts.

Keywords : co-conversion, C-O coupling, doping, oxygen vacancy, microkinetic

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