World Academy of Science, Engineering and Technology
International Journal of Chemical and Materials Engineering
Vol:17, No:01, 2023

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' 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 enablesCO_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)-OVsurface. 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
Conference Title : ICC 2023 : International Conference on Chemistry
Conference Location : New York, United States
Conference Dates : January 30-31, 2023