Unraveling the Catalytic Performance of RuO_2(110) for Highly-selective Ethylene Production from Methane at Low Temperature: Insights from First-Principles and Microkinetic Simulations

Santhanamoorthi Nachimuthu* and Jyh-Chiang Jiang

Department of Chemical Engineering, National Taiwan University of Science and Technology, Taipei 106, Taiwan.

Email: santhanamoorthi@mail.ntust.edu.tw

Abstract

Despite significant progress in low-temperature methane (CH_4) activation, commercial viability, specifically obtaining high yields of C_1/C_2 products, remains a challenge. High desorption energy (>2 eV) and overoxidation of the target products are key limitations in CH_4 utilization. Herein, we employ first-principles density functional theory (DFT) coupled with microkinetics simulations to investigate the CH_4 activation and the feasibility of its conversion to ethylene (C_2H_4) on the RuO_2 (110) surface. The C-H activation and CH_4 dehydrogenation processes are thoroughly investigated, with a particular focus on the diffusion of surface intermediates. The results show that the RuO_2 (110) surface exhibits high reactivity in CH_4 activation (E_a = 0.60 eV), with CH_3 and CH_2 are the predominant species, and CH_2 being the most mobile intermediate on the surface. Consequently, self-coupling of CH_2^* species via C-C coupling occurs more readily, yielding C_2H_4, a potential raw material for the chemical industry. More importantly, we demonstrate that the produced C_2H_4 can easily desorb under mild conditions due to its low desorption energy of 0.97 eV. Microkinetic simulations based on the DFT energetics indicate that CH_4 activation can occur at temperatures below 200 K, and C_2H_4 can be desorbed at room temperature. Further, the selectivity analysis predicts that C_2H_4 is the major product at low temperatures (300 - 450 K) with 100% selectivity, then competes with formaldehyde at intermediate temperatures in the CH_4 conversion over RuO_2 (110) surface. The findings in this work provide a deeper understanding of the catalytic performance of the RuO_2 (110) surface towards CH_4 activation and suggest that the RuO_2 (110) surface could be used to produce C_2H_4 under industrially milder conditions.

Biography

Dr. Santhanamoorthi Nachimuthu currently is a senior researcher at the Department of Chemical Engineering, National Taiwan University of Science and Technology, Taipei, Taiwan. With extensive expertise in electronic structure calculations, he specializes in Computational Materials Science, adept at developing and employing computational models to simulate biomolecules and materials crucial for energy conversion and storage. His research also involves a pivotal understanding of the heterogeneous catalytic reactions for various applications.