World Academy of Science, Engineering and Technology
International Journal of Chemical and Molecular Engineering
Vol:19, No:05, 2025

A Multi-Scale Study of Potential-Dependent Ammonia Synthesis on IrO_2 (110): DFT, 3D-RISM, and Microkinetic Modeling

Authors : Shih-Huang Pan, Tsuyoshi Miyazaki, Minoru Otani, Santhanamoorthi Nachimuthu, Jyh-Chiang Jiang

Abstract : Ammonia (NH_3) is crucial in renewable energy and agriculture, yet its traditional production via the Haber-Bosch process faces challenges due to the inherent inertness of nitrogen (N_2) and the need for high temperatures and pressures. The electrocatalytic nitrogen reduction (ENRR) presents a more sustainable option, functioning at ambient conditions. However, its advancement is limited by selectivity and efficiency challenges due to the competing hydrogen evolution reaction (HER). The critical roles of protonation of N-species and HER highlight the necessity of selecting optimal catalysts and solvents to enhance ENRR performance. Notably, transition metal oxides, with their adjustable electronic states and excellent chemical and thermal stability, have shown promising ENRR characteristics. In this study, we use density functional theory (DFT) methods to investigate the ENRR mechanisms on IrO_2 (110), a material known for its tunable electronic properties and exceptional chemical and thermal stability. Employing the constant electrode potential (CEP) model, where the electrode - electrolyte interface is treated as a polarizable continuum with implicit solvation, and adjusting electron counts to equalize work functions in the grand canonical ensemble, we further incorporate the advanced 3D Reference Interaction Site Model (3D-RISM) to accurately determine the ENRR limiting potential across various solvents and pH conditions. Our findings reveal that the limiting potential for ENRR on IrO_2 (110) is significantly more favorable than for HER, highlighting the efficiency of the IrO_2 catalyst for converting N_2 to NH_3. This is supported by the optimal $*NH_3$ desorption energy on IrO_2, which enhances the overall reaction efficiency. Microkinetic simulations further predict a promising NH_3 production rate, even at the solution's boiling point¸ reinforcing the catalytic viability of IrO_2 (110). This comprehensive approach provides an atomic-level understanding of the electrode-electrolyte interface in ENRR, demonstrating the practical application of IrO_2 in electrochemical catalysis. The findings provide a foundation for developing more efficient and selective catalytic strategies, potentially revolutionizing industrial NH_3 production.

Keywords : density functional theory, electrocatalyst, nitrogen reduction reaction, electrochemistry
Conference Title : ICMCB 2025 : International Conference on Molecular Chemistry and Biochemistry
Conference Location : Barcelona, Spain
Conference Dates : May 24-25, 2025