

KINETIC STUDY OF DESULFURIZATION OVER A METAL ORGANIC FRAMEWORK BASED ON COMPLEX REACTION THEORY

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As improving deep desulfurization is essential for producing ultra-clean sulfur-free fuel, Catalytic Oxidation Desulfurization (CODS) is a practical approach for removing planar sulfur compounds [1]. CODS process includes oxidation and the extraction step for the removal. The porous zirconium terephthalate UiO-66 provides a significant contribution to catalytic purposes because of its large surface area, tunable porosity, and reusability [2]. The insertion of functional groups into organic linkers affect on the ODS performance of UiO-66(Zr) due to electron induction effect [3]. Introduction of nitro-group in BDC linker can reduce the electron density of Zr sites in UiO-66 via the benzene ring conduction and thus enhance the ability to gain electrons and therefore increasing the Lewis acidic nature of zirconium clusters. Ultimately leading to stronger interaction with aromatic sulfur containing compounds. UiO-66-NO₂ exhibited superior catalytic activity in CODS reactions because it has a strong ability to gain electrons [3]. In this study, a systematic procedure [4] is applied to derive the overall rate equation expressions for reactant/product molecules cooperated in the reaction networks based on the theory of complex reactions for DBT removal in CODS reaction over UiO-66-NO₂. Heterogeneous catalytic oxidation is a multi-component reaction procedure that goes through elementary reaction steps. The mathematical procedure of the overall reaction has been employed by MATLAB to justify the overall items of complex kinetic mechanisms. The approach of overall reaction has been applied for this study to investigate the mechanism of CODS of Dibenzothiophene (DBT) over MOF. The proposed combined mechanism consists of 11 elementary reaction steps. CODS reactions were conducted in a reactor and for the model fuel (MF), 1000 ppm of DBT was dissolved in n-dodecane, and acetonitrile was added to the reactor as the polar phase. A specified amount of MOF was added to the reactor; then, hydrogen peroxide was supplied after heating the reactor to a predefined temperature. The aqueous and fuel phase was collected after the reaction had been completed and then analyzed. Predicted values of component yield including DBT, its sulfone and, sulfoxide was in appropriate accordance with the experimental data. Eventually, this study smooths the way for similar research toward the joint of complex reactions and multiple reaction/component mechanisms.

References

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