ENGINEERING FE-ZEOLITE CATALYSTS FOR CO_2 HYDROGENATION: THE CRITICAL ROLES OF AL PRECURSOR, IMPREGNATION MEDIUM, FE CONTENT, AND NA ADDITION

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Abstract -The conversion of carbon dioxide (CO_2) into valuable hydrocarbons represents a crucial strategy for mitigating greenhouse gas emissions and advancing a sustainable, carbon-neutral economy. This study presents a comprehensive investigation into the catalytic performance of iron-based ZSM-5 zeolites for the direct hydrogenation of CO_2, focusing on a series of strategic modifications to enhance both catalytic activity and product selectivity. The foundational ZSM-5 support was first synthesized using various aluminum sources including aluminum sulfate, aluminum nitrate, sodium aluminate, and aluminum hydroxide, with a key finding being that aluminum hydroxide yielded a catalyst with significantly improved intrinsic properties, resulting in a remarkable hydrocarbon selectivity of up to 73%. Building on this optimized support, iron was incorporated using different solvent types including aniline, ethanol, acetone, and water, revealing that water provided the most effective dispersion and catalytic performance. Further systematic optimization involved a comparative study of iron loading and incorporation methods. Wet impregnation with varying iron percentages (10, 20, and 30 wt%) was evaluated alongside a dry-mixing method utilizing 30 wt% hematite iron. The results demonstrated a clear trade-off between activity and selectivity: the catalyst with a 10 wt% iron loading prepared via wet impregnation achieved the highest CO_2 conversion, while the 30 wt% iron catalyst prepared by the dry-mixing method exhibited superior overall selectivity towards hydrocarbons. Finally, the impact of a chemical promoter was explored by adding 2 wt% sodium using sodium nitrate ($NaNO_3$) via a solid-grinding and mixing method with the iron-based ZSM-5 catalysts. This modification was found to be highly effective, particularly in the dry-mixed catalyst system, where it not only led to the highest overall CO_2 conversion but also dramatically enhanced the selectivity to light olefins. A striking improvement was observed, with light olefin selectivity increasing from a marginal 0.78% to a substantial 18.27%. These findings highlight the potential for precise control over catalytic properties through strategic material synthesis and modification, paving the way for the design of highly efficient catalysts for CO_2 valorization into high-value fuels and chemicals.

Keywords - CO_2 Hydrogenation; Iron Based Catalysts; Methanation; Protonation; ZSM-5.