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Direct CO_2 Hydrogenation to Light Olefins over Iron Oxide Catalysts with Tailored Morphologies

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Renewable energy is projected to account for nearly half of the global electricity supply by 2030 [1]. Nonetheless, fossil fuel demand is expected to remain at its peak until 2050, mainly fueled by rising consumption in developing regions, even as usage declines in more advanced economies. The IPCC emphasizes the urgent need to reduce CO_2 emissions, with 2023 already recorded as the warmest year in history. Using CO_2 as a raw material for chemical production is gaining attention as a more efficient and sustainable alternative to conventional carbon capture and storage (CCS) technologies, offering improved energy security by replacing fossil-based chemicals. However, the overall benefits of CO_2 utilization depend on carefully balancing the emissions avoided with those generated during the process, heavily influenced by the sources of CO_2, hydrogen, and energy. When CO_2 is captured from flue gases and hydrogen is produced through renewable methods, the carbon footprint can be significantly reduced. Among the different CO_2 hydrogenation routes, the production of light olefins (C_2-$C_4^{=}$) which are critical for the chemical, polymer, and pharmaceutical industries is heavily researched [2,3]. In light of the above, modified Fischer-Tropsch synthesis (mFTS) using Fe-based catalysts has emerged as a promising approach, although maintaining stable active iron phases during reaction remains a major challenge.

In this work, a series of bare iron catalysts, synthesized through different procedures, were studied to simplify the catalytic system and better understand active phase effects without the interference of support materials or alkali promoters. For this purpose, iron oxide nanoparticles were hydrothermally synthesized to obtain distinct morphologies, such as iron nanoclusters, nanospheres and nanopolyhedra. The as-prepared samples were characterized by various techniques and evaluated in the hydrogenation reaction of CO_2 (WHSV = 6 $L \cdot g^{-1} \cdot h^{-1}$, H_2:CO_2 = 3, P = 20 bar). The results revealed that Fe_3O_4 was the dominant phase before and after reaction, ensuring catalytic stability, while the formation of Hägg carbide (Fe_5C_2) correlated with enhanced activity. Among the different catalysts, Fe nanopolyhedra exhibited the highest CO_2 conversion and C_2-$C_4^{=}$ yield (11.5%), with CO pretreatment further improving performance due to iron carburization. Finally, stability tests confirmed that the catalysts maintained consistent CO_2 conversion and product selectivity over extended operation.

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