OFFICIAL ABSTRACT and CERTIFICATION

Improving CO2 hydrogenation: Guanidine treatment over bimetallic Fe-Co catalysts

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CO2 hydrogenation can simultaneously address climate change and fossil fuel depletion by converting CO2 into fossil fuel derivatives, such as light olefins. However, many challenges remain in carbide synthesis for CO2 hydrogenation catalysts, particularly in selectivity for desired products. Guanidine could serve as a carbon source to induce carburization. The purpose of this study was to investigate the effect of guanidine treatment on Fe-Co catalysts and possible reaction intermediates over these catalysts. Performance testing of Fe2-Co6-CeO2 and guanidine-treated Fe2-Co6-CeO2-G was conducted using gas chromatography (GC) over a flow-bed reactor. In-situ diffuse reflectance infrared Fourier transform spectroscopy (DRIFTS) and residual gas analysis (RGA) were used to analyze the surface chemistry of Fe2-Co6-CeO2-G. No CO2 conversion was observed on Fe2-Co6-CeO2. On Fe2-Co6-CeO2-G, peak CO2 conversion activity occurred at 300oC at 12.05% and peak light olefin selectivity occurred at 350oC at 8.720%. This improved activity suggests the carburization may have occurred. RGA and DRIFTS showed stepwise increases in CO, H2O, and CH4 with increased temperature, suggesting the occurrence of reverse water-gas shift (RWGS) and Fischer-Tropsch synthesis. Carbonate and formate correlated with RWGS activity, indicating the simultaneous occurrence of the carbonate and formate pathways of RWGS over Fe2-Co6-CeO2-G. These results provide insight on how RWGS occurs over Fe-Co catalysts, and suggests guanidine treatment is promising for CO2 hydrogenation over these catalysts. Future studies should focus on elucidating the phase of this catalyst through X-ray photoelectron spectroscopy (XPS) and X-ray diffraction (XRD) and confirming the occurrence of formate and carbonate pathways through XPS.

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