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Selective conversion of CO₂ to lower olefins over bifunctional catalysts composed of promoted In₂O₃/ZrO₂ and Zeolites

Aim

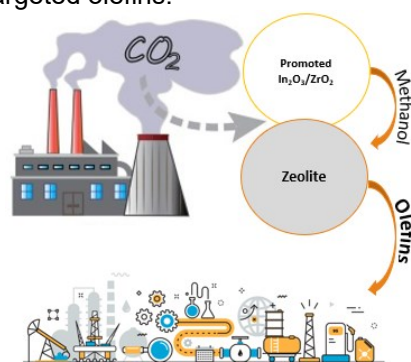
Studying the effect of operating conditions, promoted indium/zirconia catalysts and zeolite type in CO₂ hydrogenation to lower olefins over a tandem catalyst

Justification

The steady increase of carbon dioxide emissions into the atmosphere contributes to global warming and climate change. In this respect, converting CO₂ to value-added products such as methanol, formic acid, dimethyl ether, methane, and hydrocarbons mitigates global climate change. Among these routes, CO₂ transformation to methanol and consequently to lower olefins (C₂-C₄=) over a tandem catalyst can provide an environmentally-friendly raw material for the polymer industries. However, low selectivity is an obstacle in this reaction that has driven interest in the design of an efficient hydrogenation catalyst. The tandem catalyst is a combination of zeolites and metal-oxides-based methanol synthesis catalyst. In this process, CO₂ is mostly hydrogenated to methanol followed by methanol conversion to olefins (MTO) on the bifunctional active sites. The remaining challenge is the conflicting temperature requirement of these two reaction steps. In methanol synthesis reaction, high temperature favors the competing RWGS reaction. However, olefin production is more feasible at high temperatures due to the rapid kinetics of C-C bond formation. Therefore, a compromise of the reaction temperatures needs to be achieved.

Metal oxide catalysts such as indium-supported zirconia have exhibited good selectivity to methanol at high reaction temperatures (>300°C). On the other hand, small pore zeotypes and zeolites (3–4 Å), with large cavities, can convert methanol to light olefins. The selection of zeolite with appropriate topology can stabilize active intermediates and tune the selectivity towards targeted olefins.

The objective of this master thesis will be promoting indium/zirconia-based catalyst and studying the olefin production from CO₂ hydrogenation in a single reactor in the presence of the synthesized catalyst in combination with a commercial zeolite (HZSM-5, SAPO-34, SSZ-13). The crucial target will be tuning the reaction conditions and catalyst's properties to increase lower olefins selectivity, decrease paraffin production and reduce CO and coke formation. Characterization of the synthesized catalysts will be done to investigate the structure-activity relationship.



Program

1. Literature study: Studying tandem CO₂ hydrogenation reaction to lower olefins.
2. Synthesising promoted indium/zirconia catalysts.
3. Characterization of the developed catalysts.
4. Investigation of the effect of operating conditions, promoted catalysts and zeolite type in olefin formation process to increase olefin selectivity and reduce CO and coke formation