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Unraveling the mechanism of methanol synthesis from CO_2 hydrogenation over ZnO-Al_2O_3 based catalysts at high reaction temperatures

Aim

Elucidating the reaction mechanism, intermediates, spectators, and rate-determining steps in methanol synthesis and its side reactions at high reaction temperatures on the surface of ZnO-Al_2O_3 based catalysts using SSITKA + DRIFTS measurement.

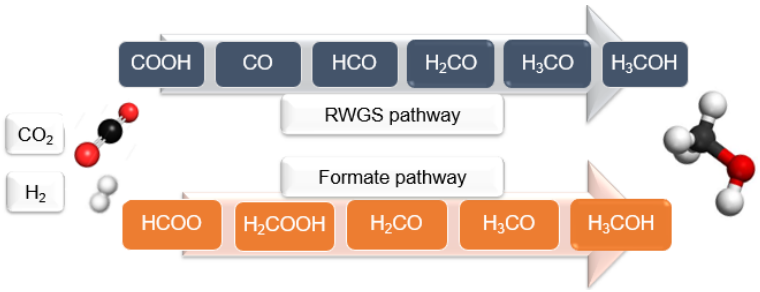
Justification

One way to mitigate the carbon dioxide emissions causing global warming and climate change is heterogeneous catalytic hydrogenation of CO_2 to valuable products, particularly to methanol, owing to the extreme relevance of this alcohol for the chemical industry. However, the lack of understanding of the reaction mechanism, especially at high reaction temperatures (>300ºC), hinders designing a selective catalyst. To date, considerable efforts have been devoted to demonstrating the exact mechanism of this reaction. Most of the intermediates cannot be followed experimentally, therefore their existence must be searched through other studies. Two main mechanisms have been proposed until now. Carbon dioxide is either hydrogenated on the carbon atom, yielding formate and following the formate route (H_2COO, H_2COOH, CH_2O, CH_3O), or hydrogenated on the oxygen atom, yielding COOH and following the reverse water-gas shift pathway.

The existing problem is that, at high reaction temperatures, the thermodynamic equilibrium highly favors the endothermic RWGS reaction over the exothermic hydrogenation to methanol. However, metal oxide catalysts have exhibited good activity and selectivity to methanol at high reaction temperatures (>300ºC). The main challenge is elucidating the predominant reaction mechanism, intermediates, and active sites of metal oxide catalysts at these temperatures.

Steady-state isotopic transient kinetic analysis (SSITKA) has evolved as one of the most powerful techniques allowing measurements at steady state to determine the mean surface lifetimes and abundances of intermediates leading to products. However, SSITKA cannot identify the chemical nature of surface species, a task that can be accomplished by Fourier transform infrared spectroscopy (FTIR).

The objective of this master thesis will be unraveling the mechanism of CO_2 hydrogenation to methanol at high temperatures over ZnO-Al_2O_3 based catalysts using: 1) steady-state isotopic transient kinetic analysis measurements, following the buildup/ disappearance of specific products during the exchange of isotope-labeled reactants, and 2) elucidating the surface intermediate and spectators during the reaction through DRIFTS measurements.



Program

1. Literature study: Mechanistic studies of methanol synthesis

2. SSITKA + DRIFTS measurements of the ZnO-Al_2O_3 based catalysts to determine the reaction mechanism, intermediates, spectators, and rate-determining steps in methanol synthesis and its side reactions at high reaction temperatures