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Creating insights in Ni-Fe hydrotalcites as catalysts for CO_2-propane dehydrogenation

Aim

This thesis experimentally assesses the structure-activity relation of Ni-Fe hydrotalcite materials for the oxidative dehydrogenation of propane with CO_2.

Justification

Propylene is among the most important chemical building blocks in industry. Its production mainly stems from steam cracking, an energy-intensive process in which propylene is only produced as a by-product. Propane dehydrogenation is an alternative and on-purpose propylene production technology that has found its way to industry. However, thermodynamic limitations, coke formation and catalyst deactivation are challenges that remain to be tackled.

The addition of an oxidizing agent such as CO_2 can overcome thermodynamic limitations and can oxidize deposited coke, but a suitable catalyst for this process is yet to be found. Nickel-iron catalysts are already known for activating C-C bonds, but tuning their properties in a controlled way towards the desired C-H bond scission remains challenging.

Hydrotalcites (HTLs) belong to the class of Layered Double Hydroxides. The extensive property tuneability of these materials makes them interesting to explore them as Ni-Fe containing catalysts. Their general structure is shown in Figure 1. Divalent M^{2+} cations that are octahedrally coordinated with OH^- anions form brucite-like sheet structures. When trivalent M^{3+} cations replace part of the M^{2+} cations, the brucite-like sheets gain a net positive charge. Interaction with water and anions to compensate for this charge leads to the formation of a stacked structure of brucite-type sheets, alternated with anion-water interlayers. Upon heating, this structure collapses and yields a mixed oxide material.

The above properties introduce extra challenges in assessing the exact behaviour of HTLs in a certain catalytic system. With each use or treatment, their surface area, porosity, redox properties etc. could change, affecting their activity. Therefore, controlling the material properties through synthesis and linking those properties to specific catalyst behaviour is essential when searching for an effective catalyst.
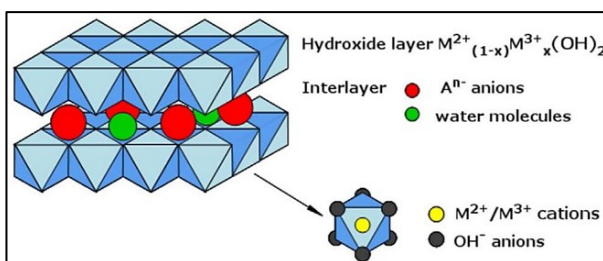

Figure 1: General structure of Layered Double Hydroxides. [1]

Program

- Literature study on hydrotalcite materials and dehydrogenation of hydrocarbons with CO_2.
- Synthesis of hydrotalcite materials with systematic variation of synthesis parameters.
- Catalyst characterization by means of Transmission electron microscopy (TEM), scanning electron microscopy (SEM), energy-dispersive X-ray spectroscopy (EDX), X-ray diffraction (XRD), H_2 temperature programmed reduction (H_2 TPR) and N_2 physisorption.
- Systematic activity tests on the synthesized catalysts during CO_2-hydrocarbon dehydrogenation reactions, with a focus on propane as hydrocarbon. Steady state and transient kinetic techniques (Temporal Analysis of Products, TAP) can be combined with the aforementioned material characterization techniques to determine the catalyst structure and its interplay with the reaction conditions.

[1] M. V. Bukhtiyarova, "A review on effect of synthesis conditions on the formation of layered double hydroxides," *J. Solid State Chem.*, vol. 269, no. June 2018, pp. 494–506, 2019.