Synthesis of Hydrotalcite-Like Compounds as Selective Membrane Layer for High Temperature Carbon Dioxide Separation

Lucas Bünger, Hans Leibold, Krassimir Garbev, Dieter Stapf Karlsruhe Institute of Technology (KIT), Karlsruhe/Germany

The removal and subsequent utilization of carbon dioxide (Carbon Capture and Utilization) requires an efficient separation technology and preferably a supply of highly concentrated CO₂. Concentrated sources, e.g. the exhaust streams of cement, syngas, or power plants provide CO₂ at high temperatures (>300°C) for further utilization. Sorbent processes, e.g. chemical looping with calcium oxide as high temperature CO₂-adsorbent, require additional capital and energy for subsequent desorption of CO₂.

This work presents a novel approach for separating CO₂ continuously at high temperatures. Dense ceramic membranes selective for CO₂ are particularly advantageous for this application.

The membrane layer consists of Hydrotalcite, an Al/Mg Layered Double Hydroxide (LDH) and meets the requirements. This material has been characterized comprehensively for CO₂ adsorption at high temperatures by pressure swing adsorption [1]. However, synthesis of robust active membrane layers remains critical. This work presents a way to manufacture a CO₂-selective membrane where the active layer has a strong adhesion to the support material with the LDH-crystals in the magnitude below microns.

In order to achieve strong permanent adherence of the membrane layer to the support material, using the support material for both, mechanical stabilization and as the substrate for one compound of the active membrane layer, was most beneficial. The LDHs contain alumina as one compound in the brucite-like layer. Therefore, a γ -Al₂O₃ layer is deposited onto the microporous α -Al₂O₃ support as alumina source by a Sol-Gel process in the first step [2, 3]. Subsequently, a combination of urea decomposition reaction and a synthesis of LDHs with intercalated methoxide is carried out. This approach combines the advantage of the temperature controlled decomposition of urea to provide a slow supersaturation for the precipitation of LDHs with a highly positively charged brucite-like layer and the small crystallite size produced by the intercalation of methoxides by using methanol. [4, 5]

So produced membrane layers are depicted in Figure 1 where the active separation layer is 1 μ m thick and adheres strongly to the graduated corundum (α -Al₂O₃) support. The membrane synthesis will be presented and discussed in detail.

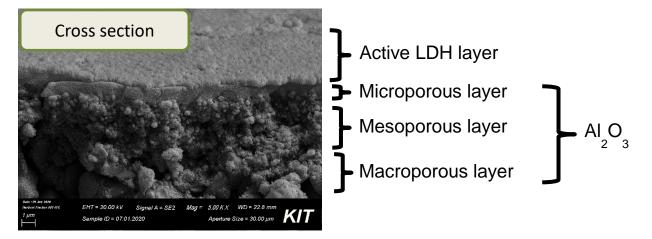


Figure 1: Scanning electron microscopy cross section image of the active LDH layer on a graduated α -Al₂O₃ support</sub> Furthermore, the characterization of the synthesized membranes will be addressed. The experimental setup for the characterization of the synthesized membranes, a membrane reactor with temperature resistant sealing up to 500°C and pressure differences up to 10 bar, will be explained.

Eliminating pinholes and cracks, changing the active layer thickness and the synthesis parameters (reaction time and temperature, support structure) as well as altering the composition of the LDH (i.e. the Al/Mg-ratio) are items of ongoing research.

References

- M. Bublinski. CO₂-Abtrennung aus Synthesegasen mit Hydrotalciten unter Hochtemperatur-Hochdruckbedingungen. Dissertation. Universität Stuttgart; 2017.
- [2] A.F.M. Leenaars, K. Keizer, A.J. Burggraaf. The Preparation and characterization of alumina membranes with ultra-fine pores. Journal of Materials Science. 1984: 1077– 1088.
- B.E. Yoldas. Alumina Sol Preparation from Alkoxides. American Ceramic Bulletin. 1975: 289–290.
- [4] E. Gardner, K. M. Huntoon, T. J. Pinnavaia. Direct Synthesis of Alkoxide-Intercalated Derivates of Hydrotalcite-like Layered Double Hydroxides: Precursors for the Formation of Colloidal Layered Double Hydroxide Suspensions and Transparent Films. Advanced Materials; 2001: 1263–1266.
- [5] U. Costantiono, F. Marmottini, M. Nocchetti, R. Vivani. New Synthetic Routes to Hydrotalcite-Like Compounds - Characterisation and Properties of the Obtained Materials. Journal of Inorganic Chemistry; 1998: 1439–1446.