Tandem Carbon Capture and Catalysis over Amine-Functionalized Metal-Organic Frameworks for CO_2 Hydrogenation to Methanol

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Methanol is one of the most critical chemicals, with a broad range of industrial applications. When produced form CO_2, methanol promises to be a key molecule to reaching carbon neutrality. Current catalysis for CO_2 hydrogenation to methanol is carried out at high temperatures and pressures over heterogeneous catalysts. The demand for a high-purity CO_2 feed at high pressures implicates high energy and, therefore, environmental costs. Current scrubbing technologies for CO_2 capture often require high temperatures for sorbent regeneration, further adding energy penalties to the cycle. Carbon capture processes using solid sorbents, in general, hold many promising advantages compared to conventional aqueous amine scrubbers.

In this work, we report bi-functional systems that combine CO_2 adsorption at low partial pressures with its conversion to methanol by hydrogenation with H_2. The materials are made from amine-functionalized metal-organic frameworks (MOFs) that can adsorb and activate CO_2 in form of carbamates. The captured CO_2 is subsequently converted to methanol by means of PNP- or PNN-pincer transition metal complexes. Preliminary tests at low CO_2 partial pressures have shown that amine-functionalized MOFs produce methanol with H_2 at low temperature and mild pressures in the presence of the PNP-pincer ruthenium complex $RuH(BH_4)(CO)(Ph_2PCH_2CH_2NHCH_2CH_2PPh_2)$ (Ru-MACHO-BH).