

Tandem Carbon Capture and Catalysis over Amine-Functionalized Metal-Organic Frameworks for CO₂ Hydrogenation to Methanol

F. A. Peixoto Esteves^{1,2}, J. A. van Bokhoven^{1,2}, M. Ranocchiari^{1,3*}

¹Laboratory for Catalysis and Sustainable Chemistry, Paul Scherrer Institut, Forschungsstrasse 111, 5232 Villigen PSI, Switzerland, ²Department of Chemistry and Applied Biosciences, ETH Zürich, Vladimir-Prelog-Weg 1-5/10, 8093 Zurich, Switzerland, ³Energy System Integration, Paul Scherrer Institut, Forschungsstrasse 111, 5232 Villigen PSI, Switzerland

Methanol is one of the most critical chemicals, with a broad range of industrial applications. When produced from CO₂, methanol promises to be a key molecule to reaching carbon neutrality. Current catalysis for CO₂ hydrogenation to methanol is carried out at high temperatures and pressures over heterogeneous catalysts. The demand for a high-purity CO₂ feed at high pressures implicates high energy and, therefore, environmental costs. Current scrubbing technologies for CO₂ 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 CO₂ adsorption at low partial pressures with its conversion to methanol by hydrogenation with H₂. The materials are made from amine-functionalized metal-organic frameworks (MOFs) that can adsorb and activate CO₂ in form of carbamates. The captured CO₂ is subsequently converted to methanol by means of PNP- or PNN-pincer transition metal complexes. Preliminary tests at low CO₂ partial pressures have shown that amine-functionalized MOFs produce methanol with H₂ at low temperature and mild pressures in the presence of the PNP-pincer ruthenium complex RuH(BH₄)(CO)(Ph₂PCH₂CH₂NHCH₂CH₂PPh₂) (Ru-MACHO-BH).