CO_2 conversion to methane using a Ru-modified MIL-140C metal-organic framework

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Abstract:

A variety of MIL-140C-related metal-organic frameworks (MOFs) have been investigated as gas phase heterogeneous catalysts for the conversion of CO_2 to valuable hydrocarbon fuels. For these particular MOFs, the catalytic activity is, in theory, achieved by binding an active metal species to coordination sites located on the linkers. MIL-140C, nominally containing 10 % bipyridine linkers and with coordinated $Ru(CO)_2Cl_2$, was successfully tested as a catalyst for CO_2 reduction with hydrogen. If pre-treated at 450 °C, a highly active catalyst develops, which is able to convert CO_2 into methane at 350 °C. The so-derived catalyst was found to be stable for up to six days of continuous operation and yielded an almost stoichiometric conversion at an overall space velocity of around 1000 ml/(g catalyst · min). Without the 450 °C pre-treatment, a steady increase in hydrogen consumption from zero to 60 % over seven days was observed at 255 °C, implying that in-situ activation was occurring gradually and that this was accelerated by the high-temperature treatment at 450 °C. Powder X-ray diffraction results before and after catalytic experiments showed that the starting MOF had decomposed to a new species. The BET surface area was also reduced. Transmission electron microscopy studies show that the Ru is well dispersed and elemental analysis shows that the Ru content influences the catalytic performance as well as activation towards the active catalyst. XPS results on fresh and spent samples support the hypothesis of MOF decomposition and indicate the presence of reduced Ru species in the active catalyst.

Biographical Statement of speaker:

Christian Vogt has a degree in Chemical Engineering and a PhD in Chemistry. His interests are in fossil fuel upgrading, carbon dioxide capture and conversion to valuable chemicals as well as analytical chemistry in general.