

Novel Fe catalysts and their CO₂ to hydrocarbon conversions.

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CO₂ waste conversion into energy rich hydrocarbons via reductive hydrogenation with solar H₂ poses an attractive concept to help combat global warming. It would provide desirable transformation of renewable solar-H₂ into more readily managed hydrocarbon fuel and, also, reliable net zero CO₂ emission hydrocarbon-based power generation if coupled with CO₂ capture (i.e. to provide a closed loop). Such value addition to solar-H₂ might help to realise CO₂ capture as an investment, rather than a burden. Catalysts based on transition and noble metals (Fe, Co, Cu, Ni, Ru) and their composites are prospective for such heterogeneous catalysis applications [1]; materials with high surface area and high metal dispersion being preferred.

Recently, metal organic frameworks (MOFs) have been targeted as progenitors for the production of high activity heterogeneous catalysts suitable for gas-phase reactions at elevated temperatures (300-500 °C) [2, 3]. Their targeting is motivated by the potential to transfer in a way their exemplary high surface area and high metal dispersion properties [3]. The present work focuses on the controlled decomposition of MIL-100(Fe), and the characterisation and activity determination of Fe-based catalysts derived from it. Silica incorporation was employed to assist transfer of the parent MOFs high Fe dispersion, to minimize sintering, to provide a means of coke removal (catalyst regeneration) [2] and to minimise negative impacts of water vapor [4].

MIL-100(Fe) was decomposed via pyrolysis of both 'as prepared' samples and after infiltration with tetraethylorthosilicate. The silica loading was varied by repeating the infiltration steps. Silica composites were also subsequently calcined. He density, N₂ sorption at 77 K, powder x-ray diffraction (PXRD), thermogravimetry, scanning electron microscopy (SEM) and magnetic response measures provided for physico-chemical characterization of the products. Their potential to catalyse reductive hydrogenation of CO₂ was characterised via mass spectrometry of exhaust gases from 50 – 400 °C (up to 40 bar). Complimentary physico-chemical characterization of catalysis tested products was also completed. The synthetic methods employed led to the preparation of stable high surface area nanoporous products. Their catalytic activities generally improved with Fe content, temperature and pressure; the direct pyrolysis product exhibited the highest activity.

References

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