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METAL-ORGANIC FRAMEWORKS AS CANDIDATES FOR CATALYTIC HYDROGENATION OF CARBON DIOXIDE TO HIGH VALUE CHEMICALS

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Carbon dioxide (CO_2) is an abundant C1 building block for organic reactions due to its free availability, nontoxicity and it is renewability. Recently, intensive research has been focused on developing methods that can capture CO_2 from the atmosphere and efficient conversion of the greenhouse gas to high value chemicals. This is meant to address environmental challenges associated with the emission of CO_2 into the atmosphere and provide starting materials for chemical industries in countries that are devoid of fossil fuels.

Metal-Organic Frameworks (MOFs) are a new class of crystalline materials which have attracted tremendous attention recently. This has been mainly attributed to the useful properties such as gas storage, catalysis, sensing and drug delivery exhibited by these materials. A lot of work has been devoted on the design and synthesis of MOFs for CO_2 capture and little has been done on the utilisation of the greenhouse gas using this class of materials.

In this contribution we present strategies for the design of novel MOFs that can capture and catalytically convert CO_2 to useful chemicals. In our preliminary studies, we have prepared chemically and thermally stable MOF using a lanthanide metal and pyridyl carboxylate linkers. Structural analysis of the framework revealed unprecedented network topologies constructed from rods that are linked by the pyridyl carboxylate linkers. PXRD studies confirmed that the structural integrity of the MOF is maintained upon activation and encapsulation of active sites in the MOF. The chemical stability of the MOF in bases that are normally used for hydrogenation studies was investigated using FITR and PXRD studies. Intriguingly, the MOF exhibits exceptional stability in bases dissolved in ethanol. Experimental results show that the activated MOF can reduce CO_2 to formate acid under mild conditions. Upon encapsulation of the catalytically active centres within the MOFs, the TON of formate production increased by significantly.

This work is significant in elucidating novel materials that can be used for conversion of CO_2 under mild conditions.

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