

Two novel metal–organic frameworks functionalised with pentamethylcyclopentadienyl iridium(III) chloride for catalytic conversion of carbon dioxide to formate†

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Abstract

Hydrogenation of CO₂ to formate is a vital reaction, because formate is an excellent hydrogen carrier, which yields blue hydrogen. Blue hydrogen is comparatively cheaper and attractive as the world envisions the hydrogen economy. In this work, two isostructural lanthanide-based MOFs (JMS-6 and JMS-7 [Ln(bpdc)_{3/2}(dmf)₂(H₂O)₂]_n) were prepared and used as support materials for molecular catalysts. The bipyridyl MOF backbone were functionalised using pentamethylcyclopentadienyl iridium(III) chloride to give Ir(III)@JMS-6a and Ir(III)@JMS-7a. XPS of the functionalised MOFs show downfield shifts in the N 1s binding energy indicating successful grafting of the complex to the MOF. Hydrogenation experiments in the presence of an organic base showed that the functionalised MOFs were active towards converting CO₂ to formate. Ir(III)@JMS-6a and Ir(III)@JMS-7a exhibited the highest turnover numbers of 813 and 621 respectively. ICP-OES indicated insignificant leaching during catalysis. TEM images and XPS data of the recovered catalyst ruled out the presence of Ir(0), confirming that the activity observed was attributed to the molecular Iridium(III) centres.

