

User Meeting 2020

Contribution ID: 228

Type: Oral

Highly Active Gas Phase Organometallic Catalysis
Supported Within Metal-organic Framework Pores

Thursday, 19 November 2020 15:20 (20 minutes)

Metal-organic Frameworks (MOFs) possess a set of unique attributes including permanent porosity, large internal surface areas and robust crystallinity which has motivated extensive interest in the field of gas-phase catalysis. In particular, MOFs can act as a platform for the heterogenization of molecular catalysts, allowing easy catalyst recovery and a route towards structural elucidation of the active centres immobilised in a crystalline host. We have developed a unique MOF, MnMOF-1 which features vacant N,N-chelation sites which are accessible via the porous channels that penetrate the structure^{1,2}. In the present work, cationic Rhodium(I) norbornadiene (NBD), cyclooctadiene (COD) and bis(ethylene) complexes have been incorporated into the vacant N,N-chelation sites of MnMOF-1 via post-synthetic metalation and facile anion exchange. Exploiting the crystallinity of the host framework, the immobilised Rh(I) centres were structurally characterised using X-ray crystallography.

The activity of the Rh(I) bisethylene complexes MnMOF-1 · [Rh(C₂H₄)₂]BF₄ and MnMOF-1 · [Rh(C₂H₄)₂]Cl in gas phase butene isomerisation was studied using gas-phase NMR spectroscopy. Under one atmosphere of butene at 46 °C, MnMOF-1 · [Rh(C₂H₄)₂]BF₄ rapidly catalyses the conversion of 1-butene to 2-butene with a TOF of ca. 2000hr⁻¹. Notably, the chloride derivative, MnMOF-1 [Rh(C₂H₄)₂]Cl displays negligible activity in comparison, suggesting that interactions between the chloride anion and the Rh centre impact the catalytic activity.

References

1. Peralta, R.A.; Huxley, M.; Young, R.; Linder-Patton, O. M.; Evans, J. D.; Doonan, C. J.; Sumbly, C. J. Faraday Discussions 2020.
2. Peralta R. A.; Huxley, M. T.; Evans, J. D.; Fallon, T.; Cao, H.; He, M.; Zhao, X. S.; Agnoli, S.; Sumbly, C. J.; Doonan, C. J. J. Am. Chem. Soc., 2020.

Primary author: Mr PERALTA, Ricardo (University of Adelaide)

Presenter: Mr PERALTA, Ricardo (University of Adelaide)

Session Classification: Session 8 - Advanced Materials and Hard Matter

Track Classification: Advanced Materials and Hard Matter