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Insights in selective oligomerisation and polymerisation catalysis using stopped flow XAS techniques

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The activation of [MoX3(L)] (with X = Cl, Br; L = tridentate ligands with S3 and SNS donor sets) by AlMe3, analogous to the industrially important [CrCl3(L)] catalysts for selective oligomerisation of alkenes, has been investigated by X-ray absorption (XAS) and UV-visible spectroscopies. Time-resolved stopped flow XAS, as developed by our group for homogeneous catalysis over the last few years, in combination with a newly developed freeze-quench approach, have provided new insights in the activation mechanisms. The complete alkylation of the Mo centres and a slower, stepwise sequence for [MoBr3(L)], has been established, without the evidence for directly bonded or bridged Mo-Mo dimers. Decomposition of the complexes in time resulted in precipitation of particulate Mo, as observed during catalytic tests. Links to the industrially relevant Cr systems will be made, and preliminary results shown. The novel freeze-quench approach, which can trap reaction solutions within 1 second of mixing, opens up a large field of homogeneous catalysis and liquid chemistry to be studied, being able to quench at sub second time scales, while characterisation techniques with long data acquisition can be performed.

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Talk

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Heterogeneous catalysis

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