

Insight of the methanol-to-DME reaction from descriptor-based microkinetic modelling

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Using a descriptor-based approach to study the methanol-to-DME reaction in zeotypes from first-principles, we investigate the different reaction mechanism through a microkinetic model. This theoretical study focuses on Brønsted acidic zeotype catalysts. We demonstrate that the shift in OH stretch frequency with and without CO adsorbed on the site is a good descriptor for the strength of the acid site. Different zeotypes were studied, namely CHA, BEA and MFI zeolite frameworks, where besides conventional Al exchange, we also consider B, Ga and Fe, along with SAPO-34, Mg-AIPO-34, Zn-AIPO-34, and Ti-AIPO-34. These span a wide range of strength of the acidic sites.

The top figure shows the free energy landscape for the two main reaction mechanisms for BEA and B-BEA (which have quite different acidity), namely the concerted where two methanol molecules adsorb simultaneously to form DME and water, and the stepwise where one methanol dissociates to a methyl group, producing water. The bottom figure shows the calculated turn-over frequency (TOF) for the two mechanisms as a function of temperature. This shows that the concerted mechanism is dominating at lower temperatures while the stepwise dominates at higher temperatures for both systems.

