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QM/MM studies of the Methanol to Hydrocarbons process on zeolite catalysts

S. A. F. Nastase and C. R. A. Catlow
Cardiff Catalysis Institute, School of Chemistry, Cardiff University, CF10 3AT, UK
E-mail: NastaseS@cardiff.ac.uk

We are using state-of-the-art molecular simulations with validation of those results against recent experimental neutron scattering studies, to have a better understanding of the early stages of the highly important methanol to hydrocarbons process(MTH). We are employing the QM/MM embedded cluster approach to provide an improved and less computationally expensive approach over the more conventional periodic DFT methods, to study in depth the adsorption of the methanol to Brønsted acid sites, and the initial methoxylation step of the MTH process.

We are using Chemshell [1] as our computational environment to support the hybrid QM/MM calculations, were several codes that have been proven to have a good chemical accuracy [2] to describe the QM and MM regions are implemented. We are employing these packages to determine different observables describing sorbate-zeolite interactions (including adsorption energy, transition barriers to the methoxylation step and vibrational frequencies) in faujasite (H-Y) and the industrially used H-ZSM-5 and by comparing our calculated values with experimental spectroscopic results [3].

To have a better understanding of the methanol to hydrocarbons process, we will create several realistic models of the reaction environment by inserting a variety of molecules, such as reactants (methanol, methane) or secondary reaction products (water, carbon monoxide), in to the zeolitic environment and model using this accurate, yet more efficient QM/MM methodology, how their interactions with the active site, affect the catalytic process.

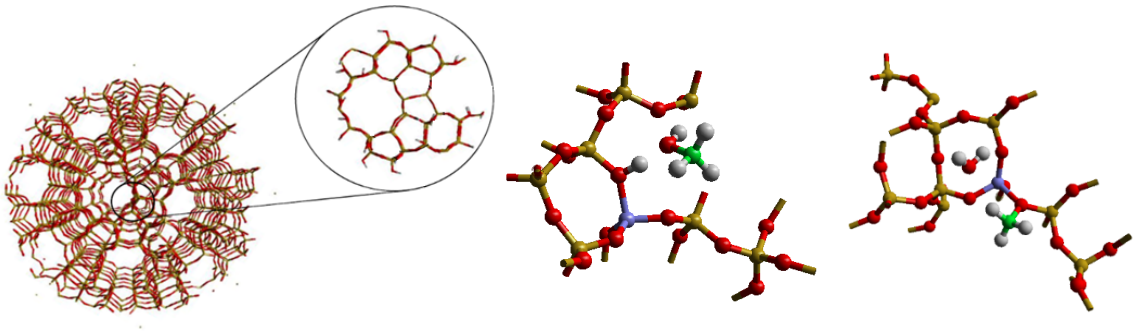

Figure 1: An example of a QM/MM setup for zeolite H-Y, describing the reaction path of methoxylation from the empty cluster (left), a section of the QM region with adsorbed methanol (middle) and a section of the QM region with the methoxylated site (right).

(1) Sokol, A. A.; Bromley, S. T.; French, S. A.; Catlow, C. R. A.; Sherwood, P. *International Journal of Quantum Chemistry* **2004**, *99*, 695.
(2) O'Malley A. J.; Logsdail A. J.; Sokol A. A.; Catlow C. R. A., *Faraday Discussions* **2016**, 188, 235
(3) O'Malley, A. J.; Parker, S.F.; Chutia, A.; Farrow, M.R.; Silverwood, I.P.; Garcia-Sakai, V.G.; Catlow, C. R. A. *Chemical Communications* **2016**, *52,* 2897-2900.