Ab initio study on the influence of water on methanol conversion in H-SAPO-34

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Due to the depleting oil reserves and the increasing demand for base chemicals like ethylene and propylene, the search for processes based on alternative feedstock is an active research area both from industry as from academia. Interesting alternative routes are those which start from methanol, because it can be produced from any gasifiable carbon-rich feedstock. In the methanol-to-olefin (MTO) process, methanol is transformed into valuable base chemicals. The CHA-structured H-SAPO-34 catalyst is of particular industrial interest as it shows a high selectivity to light olefins.[1,2]

Nowadays, reaction mechanisms based on hydrocarbon pool (HP) species that co-catalyze the reactions have been generally accepted for the olefin production via the MTO process. Two main mechanisms based on hexamethylbenzene as a HP species have been proposed for the production of olefins in H-SAPO-34, namely the paring and side-chain mechanism. For the latter mechanism, a complete low barrier cycle has been proven.[3] However to date the influence of water on the overall reaction mechanism is still poorly understood. In this contribution we study the influence of water on elementary steps in the MTO process as the feed of the process might contain water. Moreover, water is a major byproduct of the MTO process.[4] Via ab initio molecular dynamics and metadynamics simulations we assess the influence of the addition of water to elementary steps in the MTO process. In particular, adsorption, proton mobility and reactivity towards olefin elimination in the side-chain mechanism are assessed.

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