Modeling zeolite catalysis at operating conditions: an advanced molecular dynamics benchmark study

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In the last decades, significant progress has been made in the field of computational chemistry. This leads to the possible application of theoretical modeling as a guide for the design of catalysts.[1] Herein, we will compare ab initio advanced molecular dynamics techniques to the earlier used static approach to model zeolite catalysis. These dynamic methods enable us to mimick the reaction conditions as close as possible, taking into account temperature effects, topology and guest molecules.[2] In this benchmark study, several DFT-based dynamical approaches will be compared, for example metadynamics[3], umbrella sampling[4],…

As a case study for the benchmarking, the methylation of hexamethylbenzene (HMB) is chosen, because it is a model reaction for the methanol-to-olefin (MTO) process.[5, 6] The already available results show a clear difference for the reaction profile calculated statically or dynamically (metadynamics), as shown in Figure 1a. These show a clear difference in the product state, due to the insufficient description of the diffusional freedom of the formed water in the static simulation, as shown in Figure 1b. This emphasizes the importance of the use of different advanced molecular dynamics simulations in tackling the problem of further elucidating the reaction mechanism of the MTO process and analyzing the effect of possible guest molecules like water.

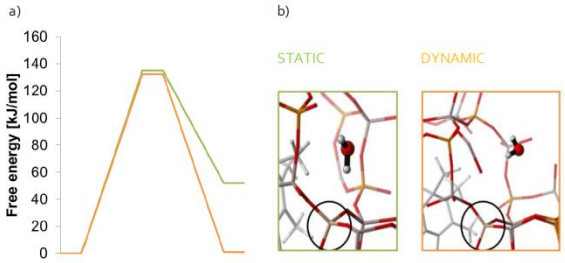

Figure 1: Reaction profile (a) and snapshots (b) of the methylation of HMB using static and dynamic ab initio simulations.

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