CATL: Division of Catalysis Science and Technology

322 - Toward a more accurate description of adsorption in Brønsted acid zeolites by combining static and dynamic molecular simulations

View Session Detail

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Abstract: Adsorption of the reactants from the gas phase onto the Brønsted acid sites inside the pores is the first step in any zeolite-catalyzed reaction. Hence, a reliable estimate for the corresponding heat of adsorption is necessary for a precise description of chemical pathways and to allow accurate predictions of experimentally observed reaction barriers and rates. In a previous paper, we have benchmarked different DFT methods on a series of hydrogen-bonded alcohol and nitrile complexes [1] for which experimental heats of adsorption on H-ZSM-5 are available in the literature [2]. Static calculations allowed to recover the experimentally observed trends in adsorption enthalpy provided dispersion interactions were properly accounted for. In the literature, similar methods have also been applied to the study of alkene adsorption, [3] however, recent reports have highlighted the significance of temperature effects when no strong interaction between the adsorbate and the zeolite host exists. In these cases, the additional conformational freedom contributes to the experimentally observed adsorption behavior.[4]

In this contribution, static calculations with different state-of-the-art DFT methods are combined with ab initio molecular dynamics simulations to study the adsorption of various guest molecules in H-ZSM-5. A systematic assessment is made of the influence of the zeolite model and DFT method, the conformational freedom of the adsorbate and the flexibility of the zeolite lattice at finite temperatures on the predicted heat of adsorption, depending on the governing interactions in the different adsorption complexes. These insights lead to an improved description of the host-guest interactions in zeolite systems, ultimately enabling a more accurate prediction of apparent barriers and overall rate coefficients of catalytic reactions.

[1] J. Van der Mynsbrugge, K. Hemelsoet, M. Vandichel, M. Waroquier, V. Van Speybroeck, J. Phys. Chem. C 2012, 116, 5499.
[2] C.-C. Lee, R. J. Gorte, W. E. Farneth, J. Phys. Chem. B 1997, 101, 3811.
[3] C. M. Nguyen, B. A. De Moor, G. B. Marin, J. Phys. Chem. C 2012, 116, 18236.
[4] F. Göltl, J. Hafner, Micro. Meso. Mater. 2013, 166, 176.