diffusion-fundamentals

The Open-Access Journal for the Basic Principles of Diffusion Theory, Experiment and Application

Transport Diffusivity in Zeolites: Possible Reasons for Misleading Results of Macroscopic Techniques

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1. Introduction

Molecular transport in zeolites is of crucial importance for many industrial applications, e.g. mass separation and catalysis. In the present work we show that conventional uptake techniques broadly used for the characterization of the transport properties, may lead to wrong conclusions on molecular transport. Describing the uptake process with meaningful values is of major importance, e.g., for a further optimization and development of new materials and for explaining the differences in the diffusivities measured by different techniques for the same system.

2. Experimental

Individual zeolite crystals of two host systems (CrAPO-5 and ferrierite) have been studied using Interference and IR microscopy sorption experiments. Interference microscopy (IFM) is based on following the change of the optical density of a zeolite crystal during molecular uptake and provides the possibilty to calculate two-dimensional intracrystalline concentration profiles [1]. The overall sorption rates were also measured by IR microscopy (IRM) mimicing the view of conventional uptake techniques without the spatial information of the intracrytalline concentration profiles.

3. Results and Conclusion

Although the measured overall uptake curves exhibit typical features of a diffusion-controlled system, only from the intracrystalline concentration profiles could be concluded, which model gives the best description of the real uptake process.

In the case of CrAPO-5 the intracrystalline concentration profiles recorded during sorption were compared with the results of dynamic Monte Carlo (MC) simulations. This approach allowed us to investigate seperately the influences of (i) the intracrystalline diffusion, (ii) the transport barriers on the external crystal surface and (iii) the effects of the intergrowth structure on molecular uptake (see fig. 1) [2].

For ferrierite, the overall sorption curves were analyzed based on three different models. They implied, respectively, dominating mass transfer due to diffusion, surface barriers or their combination. Onlv the information on the intracrvstalline concentration

profiles allowed us to figure out the correct model for describing the uptake close to reality (especially, that the uptake proceeded mainly along the smaller 8-ring channels). Without this knowledge, the values derived for the transport diffusivity strongly depend on the applied model and deviate from each other over more than two orders of magnitude [3].

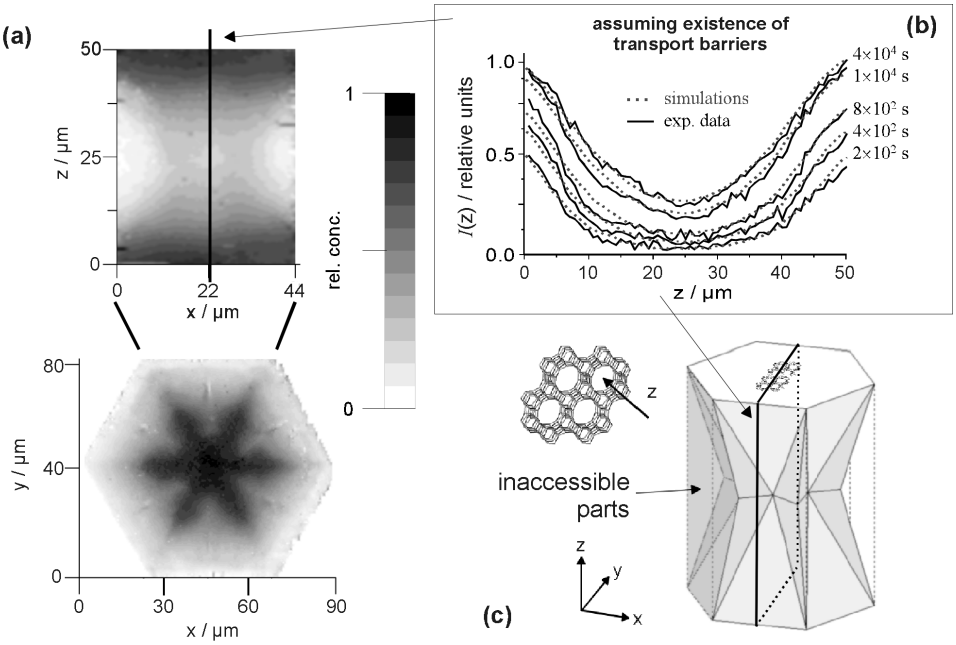

Fig. 1: (a) IFM intracrystalline concentration profile of methanol in a CrAPO-5 crystal at equilibrium. The color intensity is proportional to the integrals of local concentration. (b) Comparison of the one-dimensional IFM concentration profiles, recorded at different times after the start of the methanol adsorption with the results of the dynamic MC simulations. (c) Suggested internal structure. The channel direction coincides with the *z* direction.

The overall uptake curves could be described reasonably by different theoretical models. But only the intracrystalline concentration profiles provided us the deep insight into the uptake process which is necessary for choosing the right theoretical model for data processing including the influence of special crystal features, such as intergrowth structure and surface barriers.

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