From reaction-controlled uptake to uphill diffusion: unprecedented insight into mass transfer by micro-imaging

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As an omnipresent phenomenon in nature, diffusion is among the rate-determining processes in many technological processes. This is in particular true for mass separation and catalytic conversion in nanoporous materials [1]. The talk illustrates the possibilities of exploring mass transfer in such "host" materials by microscopic measurement, i.e. by the direct observation of intrinsic diffusion phenomena. Microscopic diffusion measurements include the observation of the propagation pattern of guest molecules by the pulsed field gradient technique (PFG) of NMR, notably by recording the probability density of molecular displacements as a function of the observation time (referred to as the mean propagator [2]). Such type of measurement, which recently has been shown to be applicable to also exchangeable cations [3], provides direct access to intracrystalline diffusivities and to both their impediment (by additional transport resistances due to intrinsic and/or surface barriers [4]) and enhancement (by mesoporosity [5,6]).

Though providing, at least in principle, the potentials of directly recording intracrystalline guest diffusion under also non-equilibrium conditions, following the example given in ref. [7] most of the applications of MR imaging to nanoporous materials did, so far, deal with the measurement of long-range diffusion in zeolite assemblages. Thus, our potentials for diffusion studies with nanoporous materials have been notably reinforced by the recent advent of the techniques of microimaging [8], notably by interference microscopy (IFM) and IR microscopy (IRM). In this way, transient guest profiles, well known as theoretical patterns from the Crank and Carslaw-Jaeger text-books, became accessible by immediate experimental observation. Examples of insight so far inaccessible (if not even unthinkable) include the quantification of transport resistances on the surface of the individual crystallites [9], the exploration of the interrelation between tracer (or self-) diffusion and transport (or chemical or collective) diffusion of guest molecules [10] and the observation of guest-induced variations in the host lattice [8]. As particular highlights in the exploitation of these novel options, microimaging was most recently successfully applied to the first ***direct*** measurement of the effectiveness factor of catalytic reactions [11] and to the observation of uphill diffusion, i.e. of diffusive fluxes into the direction of increasing concentration.

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