Molecular dynamics (MD) simulation and modeling of diffusion in fluids and porous materials

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Molecular diffusion in fluids and porous materials is important in many areas of chemical and process engineering. Examples include continuous-flow reactors, desalination membranes, battery electrodes, or fixed-bed adsorption and separation columns. The ability to simulate diffusion in hierarchically porous materials is a challenge and of great importance to the understanding and design of functional devices and processes. In recent years, computer simulations and mathematical models have been continuously developed to study diffusion in fluids and porous materials. With these techniques, it is possible to investigate diffusion processes hierarchically over different spatiotemporal scales, while all information obtained from a previous scale can be retained on the next higher one.

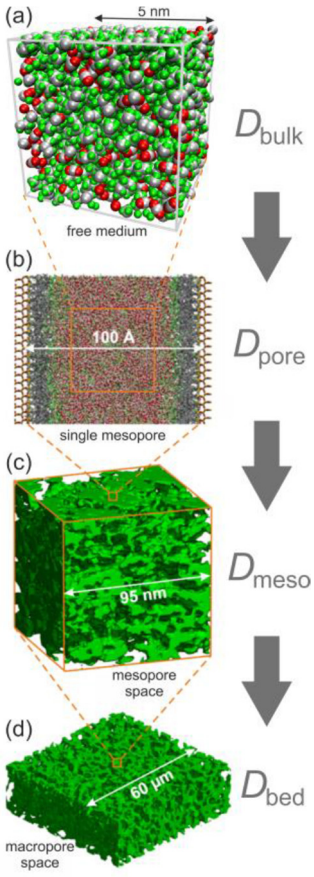

Figure 1: Hierarchical modeling of diffusion in (a) free medium, (b) single pore, (c) mesopore space, and (d) macropore space.

In this workshop, we will guide through the different steps for the hierarchical modeling of diffusion processes in fluids and porous materials. For this, a multiscale reconstruction approach is applied which provides realistic models for surface chemistry and resulting fluid organization on the single-pore level as well as for the 3D morphology of mesopore and macropore spaces in hierarchical materials. As modeling tools, Molecular Dynamics (MD) and Brownian Dynamics (BD) simulations used on different length and time scales will be performed live during the workshop. While MD simulations analyze the dynamics of molecules on the basis of atomistic models and classical Newtonian mechanics, BD simulations also account for friction and random forces in larger mesoscopic systems interacting via simplified models.

In the first step (Fig. 1a), we will study self-, Maxwell-Stefan, and Fickian diffusion in binary fluid mixtures without any confinement (D_{bulk}) in MD simulations on a scale of ~5 nm at gaseous, supercritical, and liquid states [1]. In the second step (Fig. 1b), our MD simulations on the single-mesopore level (~10 nm) take account for pore shape and surface functionalization and allow to quantify distributions as well as mobilities (D_{pore}) for solvent and solutes [2]. In the third step (Fig. 1c), the mesopore network (~100 nm) reconstructed by electron tomography serves as realistic model in the BD simulation of hindered solute diffusion (D_{meso}) using a random-walk approach [3]. At this level, we will show that the MD information obtained on the previous single-pore level is explicitly preserved. In the fourth step (Fig. 1d), we will compute effective bed diffusion coefficients (D_{bed}) on the µm-scale by a BD model which accounts for the solute exchange between the mesopore and macropore space, the latter being reconstructed by confocal laser scanning microscopy. By coupling the flow dynamics in the macropore space, an outlook to the study of effective dispersion coefficients for solvent and solutes will be given.