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

Thomas M. Koller[1], Ulrich Tallarek[2]

[1]Institute of Advanced Optical Technologies – Thermophysical Properties (AOT-TP), Friedrich-Alexander-University Erlangen-Nürnberg (FAU), Erlangen, Germany
[2]Department of Chemistry, Philipps-University Marburg, Marburg, Germany
thomas.m.koller@fau.de, tallarek@staff.uni-marburg.de

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.

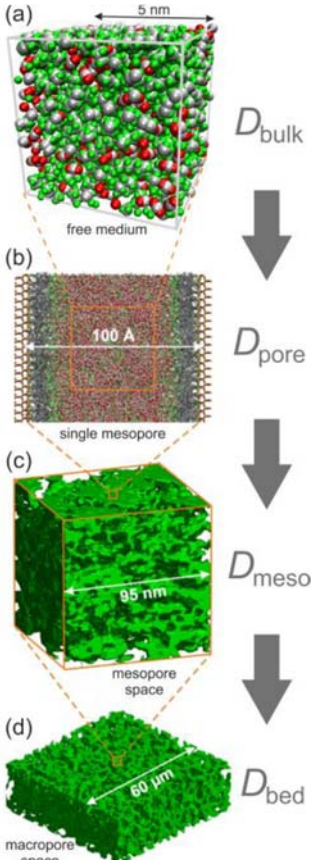

Figure 1: Hierarchical modeling of diffusion in (a) free medium, (b) single pore, (c) mesopore space, and (d) macropore space.

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.

Thomas Manfred Koller is an academic staff member and leader of the group "Molecular Modelling" at the Institute of Advanced Optical Technologies – Thermophysical Properties (AOT-TP) at the Department of Chemical and Biological Engineering (CBI) and at the Erlangen Graduate School in Advanced Optical Technologies (SAOT) of the Friedrich-Alexander-University Erlangen-Nürnberg (FAU), Germany. His research activities focus on the characterisation of working fluids in energy and process engineering via the determination of their thermophysical properties. Here, the main interest is directed towards the study of heterogeneous systems including fluid interfaces. Besides the experimental investigation through conventional techniques and dynamic light scattering, his thermophysical property research involves theoretical approaches including molecular dynamics simulations and the development of prediction models. Since 2018 he is an elected member of the International Association for Transport Properties (IATP) which is a non-profit grouping of scientists devoted to the advancement of the transport properties of materials.

Ulrich Tallarek is full professor of materials and interface science at the Department of Chemistry of the Philipps-Universität Marburg (Marburg, Germany). He studied chemistry at the Eberhard-Karls-Universität Tübingen and obtained his Dr. rer. nat. in 1998 with an NMR imaging study of the fluid dynamics in porous media. As a Marie-Curie postdoctoral fellow from 1998 to 2000 in Wageningen (The Netherlands) he developed and applied NMR tools for *in situ* characterization of transport in microfluidic devices, particularly electrokinetic microfluidics. From 2000 to 2007, he was on the faculty of the Department of Chemical and Process Engineering of the Otto-von-Guericke-Universität Magdeburg, where he completed his habilitation in 2004 (mentor: Prof. Dr. Andreas Seidel-Morgenstern) and received a *venia legendi* for physical chemistry. In 2007, he became professor at the Philipps-Universität and member of the board of directors of the materials science center in Marburg. His research focuses on morphology–functionality–transport relationships in porous media, from solute–surface interactions to macroscale transport. This approach relies on advanced simulation methods to address the different spatiotemporal scales and on the 3D physical reconstruction of the materials. Professor Tallarek has received several honors and awards, including the Desty Memorial Prize for Innovation in Separation Science (2003), the Young Scientist Award from DECHEMA e.V. (2006), and the "Silver Jubilee Medal 2017" from The Chromatographic Society (UK). He was also finalist for the World Technology Awards, category "Environment", presented in association with *TIME*, *Fortune*, CNN, and *Science*, and in 2013 was named as one of the 100 most influential analytical scientists in the world.