

# Solute sorption, diffusion, and advection in macro–mesoporous materials: Toward a realistic bottom-up simulation strategy

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A general bottom-up approach to the multiscale simulation of sorption, diffusion, and advection in hierarchical, macro–mesoporous adsorbents (e.g., particle-packed beds of micrometer-sized, mesoporous particles used for liquid chromatographic separations) is presented. Gained insight promises a to date unknown understanding of how the interfacial dynamics shapes macroscale transport and the separation efficiency as a function of the fluid flow velocity. In particular, the approach allows to assess individual contributions to longitudinal and transverse dispersion coefficients and the related chromatographic plate heights (Figure 1). These include (i) sorption and partitioning at the surface, (ii) effective diffusion in the mesopore space, and (iii) fluid flow and the engendered hydrodynamic dispersion in the macropore space. Efficient linker schemes guarantee integration of the (detailed molecular-level) interfacial dynamics into the hierarchical porosity and multiscale transport models. At the bottom, molecular dynamics (MD) simulations address the interfacial dynamics (particularly sorption, partitioning, and surface diffusion) at the single-mesopore level. The information obtained for the distribution and mobility of solute molecules is subsequently embedded into Brownian dynamics (BD) simulations of the effective diffusion coefficient in the mesopore space. At the top, longitudinal and transverse plate heights are derived by lattice-Boltzmann (LB) simulations of fluid flow combined with BD simulations to resolve the advection–diffusion problem in the macropore space. The plate heights ultimately account for the interfacial dynamics from the MD simulations (by considering mass transfer of solute molecules between mesopore space and macropore space and sorption at the external surface of the mesoporous domain) as well as for diffusion and flow in realistic geometrical models of mesopore space and macropore space based on physical reconstruction by electron tomography and focused ion-beam scanning electron microscopy, respectively [1].

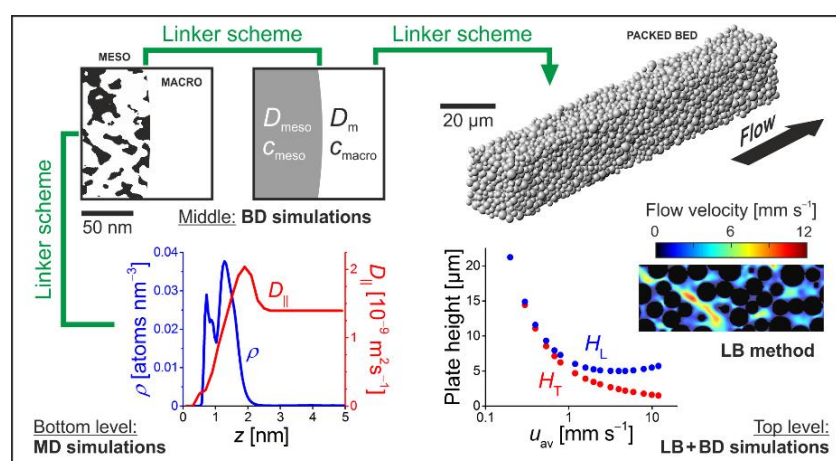


Figure 1: Bottom-up approach to the simulation of solute transport in macro–mesoporous adsorbents. At the bottom, MD simulations address the interfacial dynamics at the single-mesopore level. Information about the distribution and mobility of solute molecules, expressed by  $\rho(z)$  and  $D_{||}(z)$ , is subsequently embedded into BD simulations of the effective diffusion coefficient in the mesopore space,  $D_{\text{meso}}$ . At the top, longitudinal and transverse plate heights  $H_L$  and  $H_T$  are derived by LB simulations of fluid flow in the macropore space combined with BD (in particular, random-walk particle-tracking) simulations.

## References

- [1] U. Tallarek, D. Hlushkou, A. Höltzel: *Solute sorption, diffusion, and advection in macro–mesoporous materials: Toward a realistic bottom-up simulation strategy*. J. Phys. Chem. C **126**, 2336–2348 (2022).