

192h Modeling Permeation in Nanoporous Media with Lattice Density Functional Theory

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In zeolite membranes, the permeability and the flux of a single component can exhibit maxima with respect to the temperature. This behavior has been explained generally as a competition between gas and surface diffusion, and as a process that depends on the ratio of the diffusive activation energy (E) to the heat of adsorption (Q) - maxima are observed only when $0 < E < Q$. We have reproduced this behavior in simple nanopores by modeling with Lattice Density Functional Theory, a new approach for understanding diffusion and phase behavior on a molecular scale. For transport of supercritical fluids through simple nanopores, the approach gives analytical expressions for the permeability in terms of the bulk densities and the intermolecular interactions. For single-file diffusion with penetrant-wall attractions, the approach gives a new molecular explanation for this permeation maximum: (i) an enhanced permeability while decreasing temperature can be attributed to the energetic barrier between the pore entrance and the upstream bulk phase, i.e., weak attractions increase the permeability because molecules in the pore find it difficult to diffuse back into the feed; (ii) at lower temperatures, a decreased permeability while decreasing temperature can be attributed to the energetic barrier between the pore exit and the downstream bulk phase, i.e., strong attractions reduce the permeability because molecules in the pore find it difficult to escape from the last stage. The approach also was used to model permeation through wider pores, giving qualitatively-correct behavior and demonstrating the competition between gas and surface diffusion.