Selective permeability in gels: Beyond the solution-diffusion model

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Permeability, a measure of potential transport of macromolecules through crowded media such as hydrogels, determines important control parameters in bio-soft functional material applications, e.g., for filtration, drug release, and transport of reactants in responsive nano-reactors [1-7]. Tuning permeability is thus of great importance since it enables selective barrier crossings in molecular transport. We develop a model of semi-flexible cross-linked polymer gel networks by means of extensive coarse-grained simulations and scaling theories. The gel system consists of randomly formed tetra-functional network regions and also bulk regions where the macromolecular cosolutes diffuse in both regions, enabling a quantitative study of partitioning [1-5], diffusivity [1-4], and permeability [1,3]. The gel undergoes a sharp volume transition upon changing inter- and intra-particle interactions [1,2], yielding a rich topology of the partitioning phase landscape [2] which is highly correlated with the cosolute diffusivity. Moreover, we find that resultant permeability is largely maximized or minimized at an optimal gel density and inter-particle couplings between the networks and the cosolutes. This nontrivial phenomenon is triggered by a competition between partitioning and diffusion, resulting in a large anti-correlation [1]. It is revealed that permeability can be highly selective by tuning the coupling interactions and the solvent quality. By applying external driving forces, we show this selectiveness of permeability beyond the linear response regime based on the solution-diffusion model. Finally we present scaling theories for partitioning, diffusion and thus permeability in crowded systems.

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