459c Electrokinetic Control of Mass Transfer in Gels Via Embedded Nanoparticle Pumps

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Gels are a common matrix for biosensors. Hindered transport through the polymeric matrix slows down the response rate of such sensors, limiting the applicability of gel-based sensors for processes with relatively rapid dynamics. In addition to their main use as analytical instruments, gel-based biosensors also have been used in the laboratory to measure receptor-ligand and antibody-antigen binding rate constants, yet a lack of control over mass transport limitations has caused orders of magnitude errors in some kinetic rate constant determinations. This motivates the development of techniques to promote mass transfer in gels. The network and mechanical properties of a gel make mechanical mixing schemes inappropriate. We are investigating novel internal pumping strategies based on electrically driven intra-gel convection as a way to accelerate mass transfer in cross-linked polyacrylamide gels. The gels are doped with negatively charged, approximately 10 nm silica colloids that drive local electroosmotic flow in response to externally applied electric fields. Upon application of the field, charged solutes are convected in the electroosmotic flow, but of course they also experience a direct electrophoretic force. The use of particle-driven electroosmotic flows makes it possible to promote mass transfer of neutral molecules as well. The uniformity of the particle dispersion throughout the gels is confirmed by small angle neutron scattering. We use fluorescence spectroscopy to measure the mass transport of a fluorescent dye, amino-methylcoumarin, in these gels as a function of particle loading, applied field strength, and electrolyte concentration. Control experiments show that the silica particle incorporation does not alter the diffusion of dyes through the gel. Application of electric fields to the particle-doped gels significantly enhances dye transport through the gel, and when the field is applied in a direction that causes the electroosmotic flow to run counter to the dye concentration gradient, the dye flux can be halted. Application of similar electric field strengths to gels that contain no embedded particles produces only a modest electrophoretic effect on dye transport, confirming that significant electroosmotic flows originate from the presence of the charged silica particles. Studies of silica particles with different sizes, but embedded in the gel at equivalent total particle surface area, show that the electroosmotic mass transport enhancement is strongest when using smaller silica particles, since this increases the number density of “pumping sites” in the gel.