Electrokinetic Mass Transport Control in Gels Via Immobilized 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 laboratory measurements the use of gel-based enzymatic studies is compromised by the lack of mass transport control which leads to errors in the determination of kinetic rate constants. 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 convection as a way to accelerate mass transfer in cross-linked polyacrylamide gels. The gels are doped with negatively charged, silica nanoparticles that drive local electroosmotic flow in response to externally applied electric fields. 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 amino-methylcoumarin as a function of particle loading, applied field strength and field direction. Diffusion properties of the gel are shown to be unaltered by the incorporation of silica particles in control experiments. Application of dc 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 dc 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. We demonstrate the feasibility for the convective mixing inside the gels by the measurement of fluorescein mass transport in particle loaded gels under the application of ac electric fields. Florescent recovery after photobleaching measured through fluorescent microscopy is used to test electrokinetic mixing. Contributions of electroosmotic and electrophoretic enhancement provide a convection mechanism for the enhancement of mass transport and are examined with control diffusion experiments. Faster spot recovery is observed and the transport of tracer is demonstrated to be controlled by electrokinetic flows and the ac electric field frequency.