

## Electroosmotic Mixing inside Polyacrylamide Gels Via Immobilized Silica Nanoparticles

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The dynamics of biosensor response to a change in analyte concentration represent a problem of chemical reaction with coupled mass transfer. The designs of biosensor devices often call for biomolecular recognition elements, such as enzymes, antibodies, or oligonucleotides, to be immobilized in the sensor substrate.<sup>[1-3]</sup> Immobilization in gels is commonly used to protect the sensing elements in applications involving exposure to harsh or fouling environments. This introduces significant barriers to the mass transfer of the analyte molecules to the sensing elements. Additionally, the structure of the gel makes mechanical mixing schemes inappropriate in devices based on molecular recognition within gels.

We present an internal pumping strategy to enhance solute fluxes in polymer gels. The method is based on electroosmotic flow driven by an electric field applied across a polyacrylamide gel that has been doped with charged colloidal silica inclusions. The homogeneity of the charged particle distribution in the gels was previously investigated by small angle neutron scattering.<sup>[4]</sup> Comparisons of the scattering intensity of gels with silica particles with the scattering from dilute aqueous suspensions indicated that silica particles did not aggregate in the gels. The appearance of a local minimum in the scattering intensity and the structure factor for the particle-laden gels suggested that the particle positions may be correlated in the gel. The effect of the particle volume fraction on the gel density was studied by a gravimetric analysis and we found that an increase in osmotic pressure results in lower polymer volume fractions with increasing particle load as shown in Figure 1.

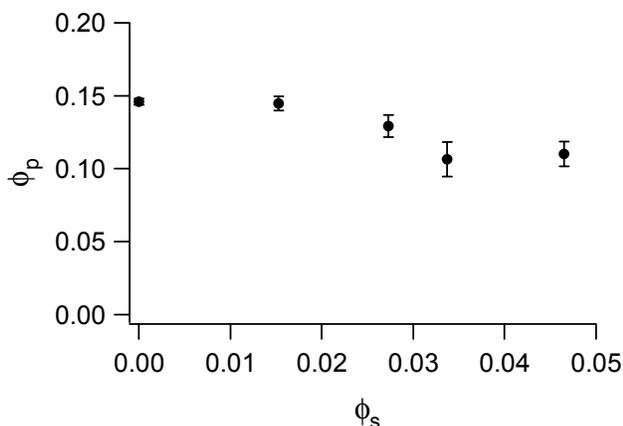


Figure 1: Gel swelling increased with increasing loading of 7 nm silica particles in the gel, as indicated by decreasing polymer volume fractions.

We demonstrated the feasibility for convective mixing inside the gels by the measurement of fluorescein mass transport in particle

loaded gels under the application of nonuniform ac electric fields. Fluorescence recovery after photobleaching (FRAP) was used to test electrokinetic mixing. Electrokinetic phenomena provided convection mechanisms for the enhancement of mass transport. To generate time varying, spatially non-uniform electric fields in the gel, electrodes were served by two phase-locked function generators. The wave form was created specifically to improve mixing of the photobleached spot.

The photobleached spot recoveries under the action of this applied electric field waveform were compared for gels with  $\phi_s = 0$  and 0.05 under oscillatory fields with a frequency  $\omega = 0.1$  Hz. In gels with no particles there was no evident enhancement of the spot recovery as shown in Figure 2. The shape of the photobleached spot showed no distortion. This indicates that in spite of the charge on the FITC tracer, electrophoretic migration was not significant under these applied field conditions.

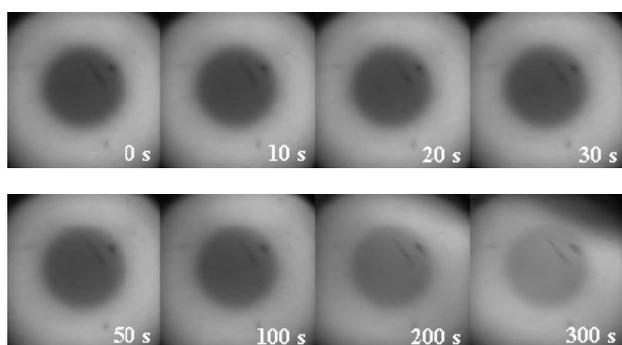


Figure 2. Photobleached spot recovery as a function of time in gels with no particles under applied electric fields with  $\omega = 0.1$  Hz.

For the gel with  $\phi_s = 0.05$ , the fluorescence recovery was significantly faster under this applied field at 0.1 Hz. As shown in Figure 3, the spot was barely observable after just 30 s. This recovery was faster than either the particle-free gel under the applied field or the gel with  $\phi_s = 0.05$  under simple diffusive conditions with no applied field. Deformation of the spot was visibly evident resulting in mixing and faster recovery as shown in Figures 4. The spot deformation in gels with silica particles under designed oscillatory electric potential gradients provided an electroosmotic mixing mechanism that enhanced the spot recovery rate sometimes by more than an order of magnitude when compared to electrophoretic mixing in gels that did not contain silica inclusions. The development of this novel electroosmotic pumping technique can potentially impact the design of new biosensing techniques or biomedical devices.

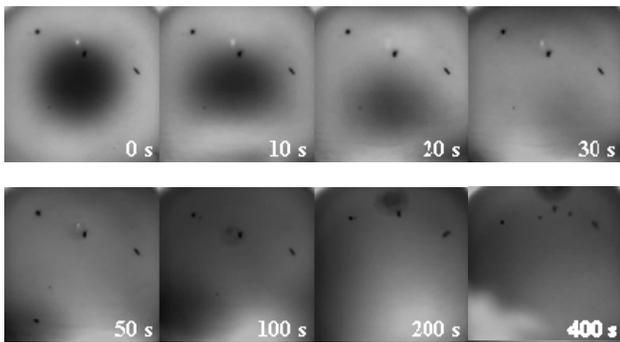


Figure 3. Photobleached spot recovery as a function of time in gels with 5 vol% silica particles under applied electric fields with  $\omega = 0.1$  Hz. Note the spot deformation.

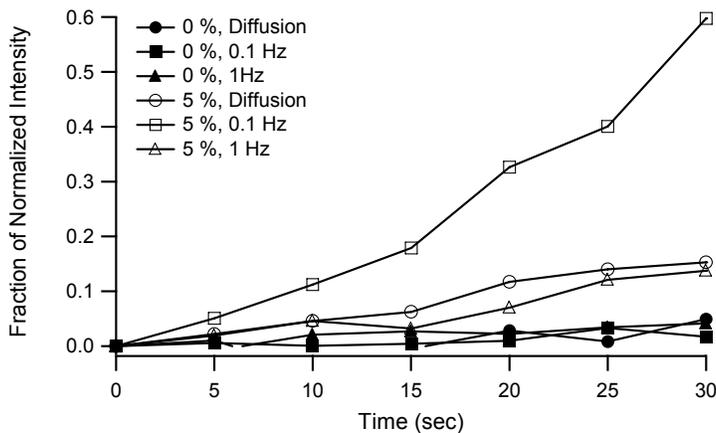


Figure 4. Fraction of normalized intensity for the spot recovery as a function of time and frequency for gels with 0 % and 5 vol% of silica particles under applied electric fields

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