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Quantifying the dynamic transition of hydrogenated castor oil gels measured via multiple particle tracking microrheology†

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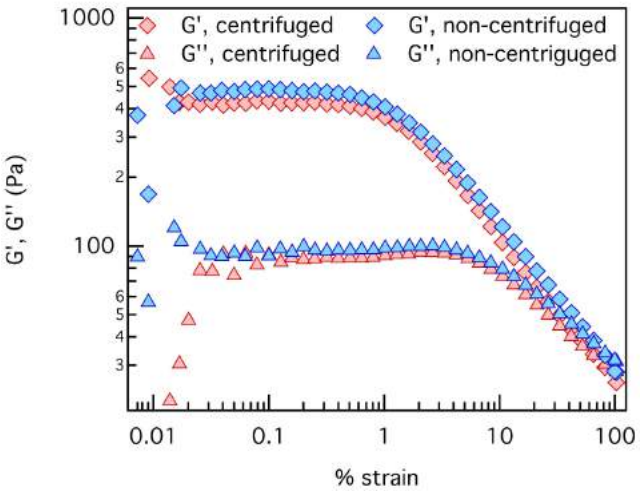

Fig. 1 Strain sweep a 4 wt% HCO gel, with and without centrifugation. No discernible changes to structure of the gel or fibers are apparent from centrifugation. G′ is greater than G″ at all strains below 10%, ensuring that the system is a gel at 4 wt%.

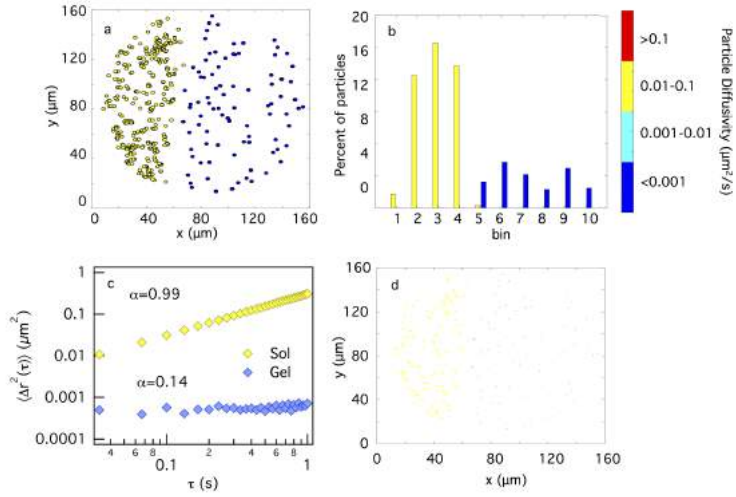

Fig. 2 Experiment to verify clustering of particles using F-test with 95% confidence interval comparing variances of single particle van Hove correlation functions. A photpolymerizable gel made of 3 wt% poly(ethylene glycol) (PEG) dithiol and 0.63 wt %PEG-acrylate is exposed to a strong UV light source. The exposure is limited spatially, and only exposes half of the sample to light. The result is half of the sample is a gel (right of sample), and the other a sol (left of sample). Data are analyzed using techniques developed to identify heterogeneity. (a) Clusters separated by probe diffusivity, with the gel on the right, with minimal probe diffusivity, and sol on the left, with large probe diffusivity. (b) Histogram of clustered particles in bins, each bin is a spatial slice taken along the $x-$axis. (c) Calculated ensemble averaged mean squared displacements (MSD) of the two microenvironments. (d) Trajectory map to visualize the two states within the field of view, yellow is the sol and blue is the gel.

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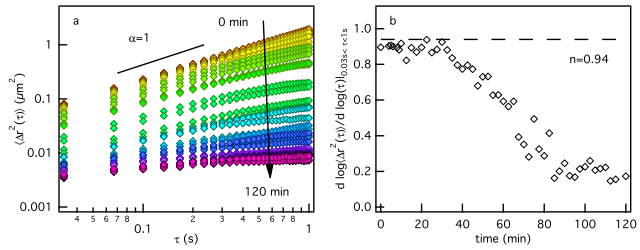

Fig. 3 (a) MSD curves of a contraction experiment of a 0.125 wt% hydrogenated castor oil (HCO) gel. The contraction experiment is completed after two hours when the material is completely contracted into a gel. (a) MSDs decrease as the HCO associates into a gel. (b) Logarithmic slopes of the MSD curves throughout the transition showing the decrease as the HCO solution becomes an associated colloidal gel. The HCO solution starts with a logarithmic slope of the MSD of 0.9 and ends with a logarithmic slope of the MSD of 0.2.

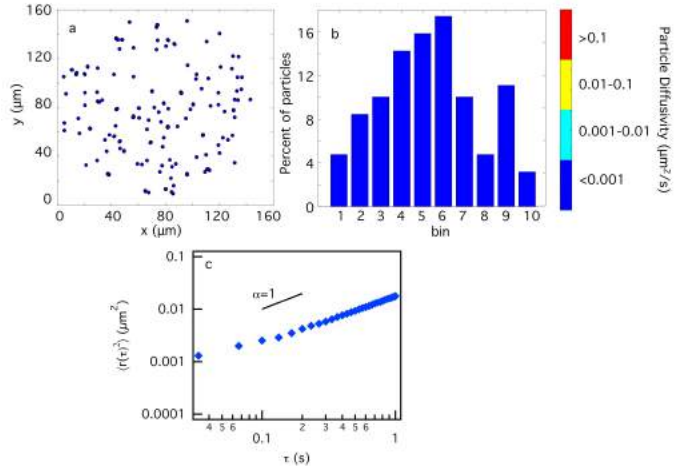

Fig. 4 (a) A single cluster showing probe diffusivity, entirely in sol phase, yet with small diffusivity (b) Histogram of clustered particles in bins, each bin is a spatial slice taken along the $x-$axis. (c) Calculated ensemble averaged mean squared displacement of the microenvironment.

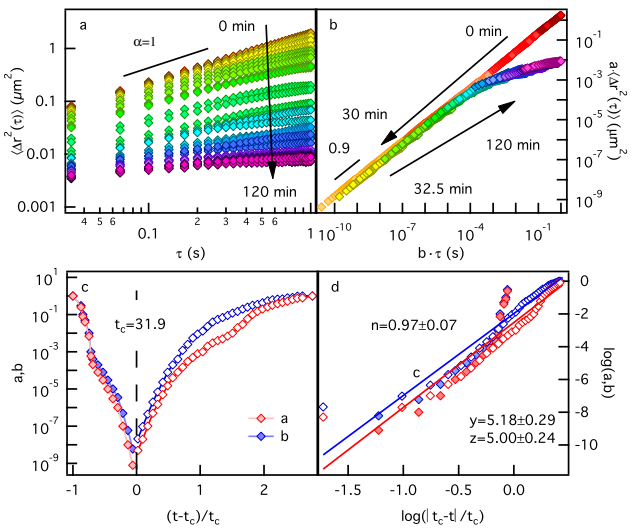

Fig. 5 (a) Measured ensemble averaged MSD curves of a contraction experiment through time. (b) Time-cure superposition is used to shift MSD curves into sol and gel master curves. (c) Shift factors along the time and MSD axes,a and b respectively, diverge at the critical transition time $t_c = 31.9$ min. (d) The critical relaxation exponent, $n = 0.97 \pm 0.07$, is determined by fitting the log of the shift factors to the log of the distance away from the critical transition time.

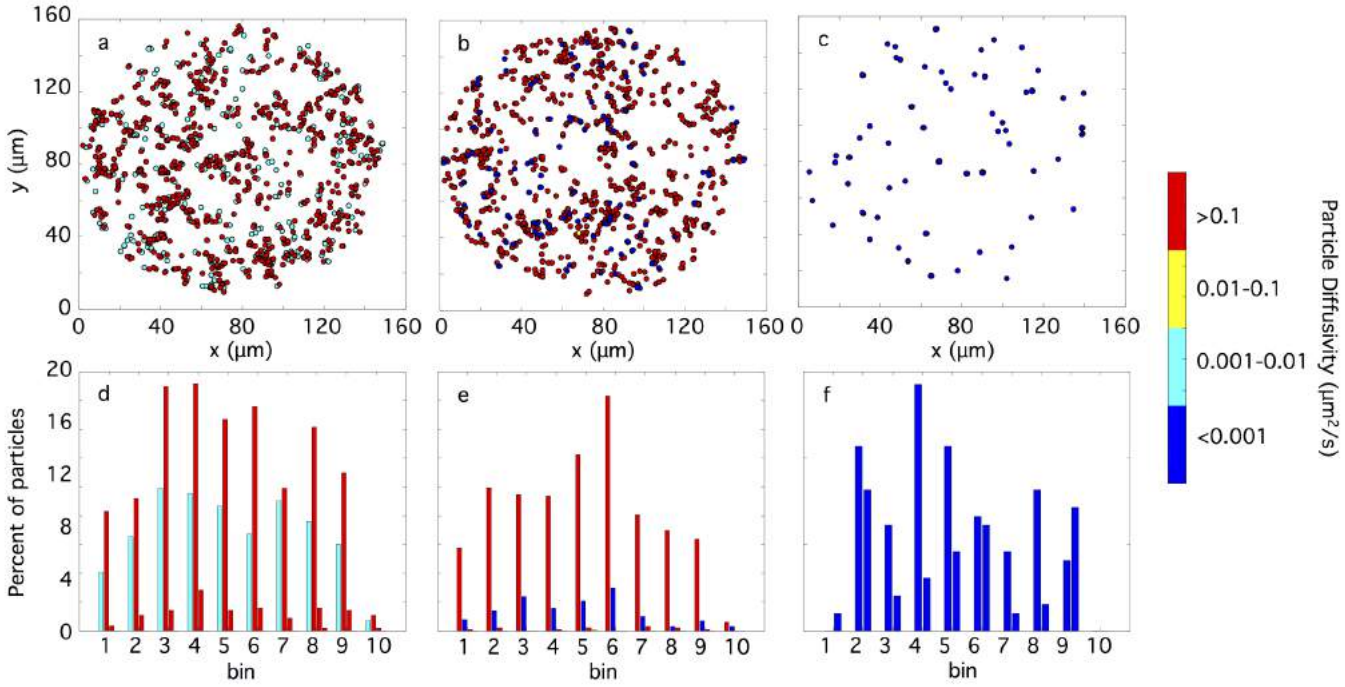

Fig. 6 Rheological (a−c) and spatial (d−f) heterogeneity of probe microenvironments for the sol→gel transition during HCO contraction at (a, d) $t_r = -1.0$, (b, e) $t_r = 0$, and (c, f) $t_r = 2.0$.

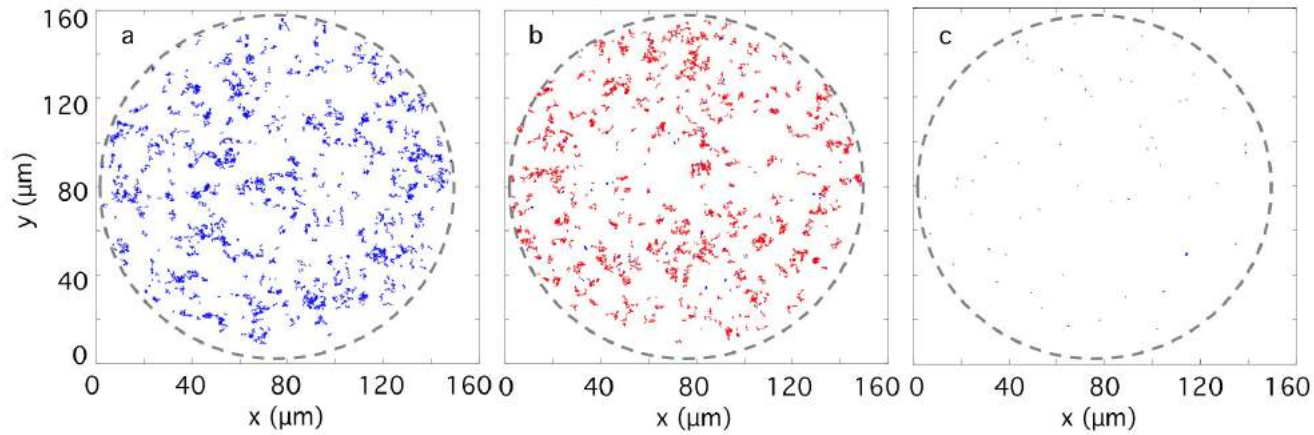

Fig. 7 Trajectory maps for the contraction of an HCO gel at (a) $t_r = -1.0$, (b) $t_r = 0$, and (c) $t_r = 2.0$. Red indicates a particle in a cluster with a logarithmic slope of the MSD greater than the critical relaxation exponent ($n = 0.94$) indicative of a sol, and blue with a slope less than n which is a gel. Due to the high value of n for the contraction experiments, $n = 0.94$, the expanded state probes are in a gel microenvironment in (a), however the slope of the cluster particles fluctuates around the critical relaxation exponent in the expanded phase. (b) During transition both gel and sol microenvironments are present. (c) After transition the gel is completely contracted and all probes are in a gel microenvironment.