Online Supplementary Information

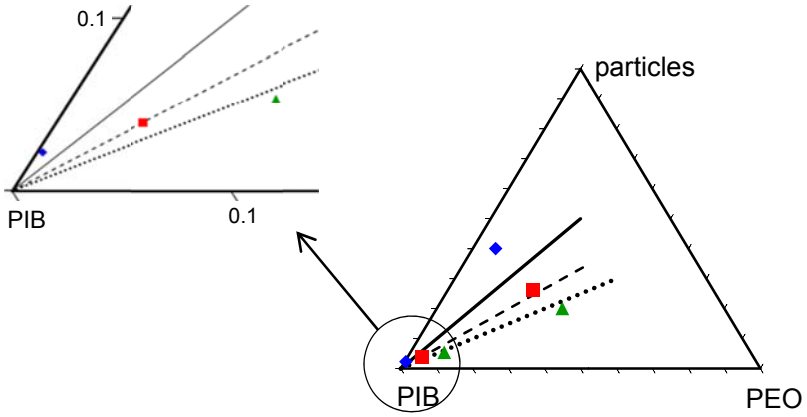

Fig. S1: The compositions examined in this paper represented on a ternary composition diagram. The upper left image is a magnified version of the PIB-rich vertex of the composition diagram. These same compositions are shown in Fig. 1 in rectilinear form. The three lines correspond to $\varrho = 0.5$ (solid), $\varrho = 1$ (dashed), and $\varrho = 1.5$ (dotted) respectively. These three lines are not intended to separate the three symbols; they are for illustration only.

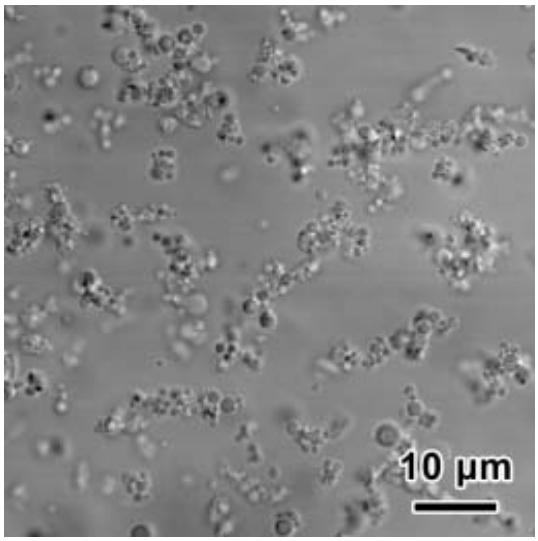

Fig. S2: DIC microscopy image of pendular aggregates corresponding to $\phi_p = 0.022$ and $\varrho = 0.16$.

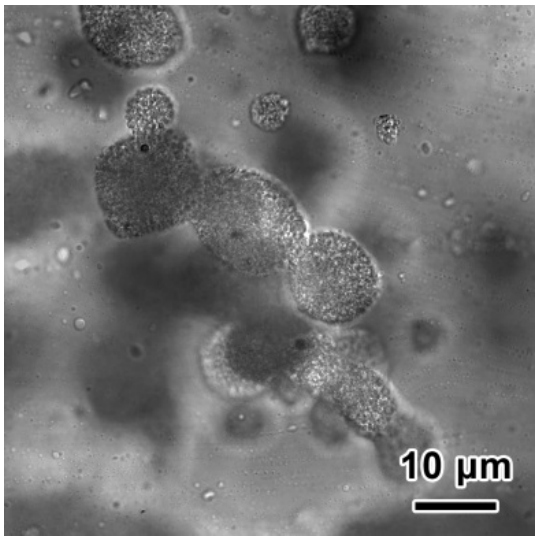

Fig. S3: DIC microscopy image of capillary aggregates corresponding to $\phi_p = 0.04$ and $\varrho = 1$.

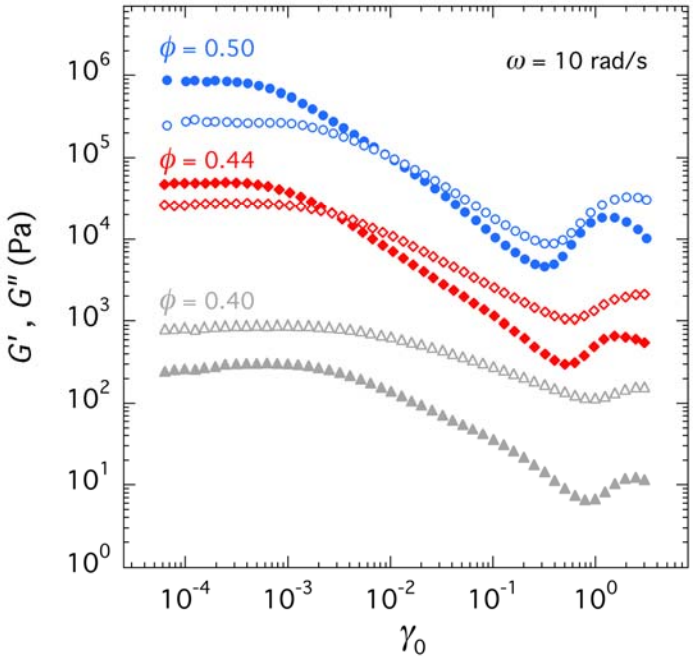

Fig. S4: Large amplitude oscillatory shear (LAOS) rheology of particle-PEO mixtures at 80℃ at a frequency of 10 rad/s. The values of particle volume fraction are listed alongside each dataset. Note that these samples do not contain PIB at all.

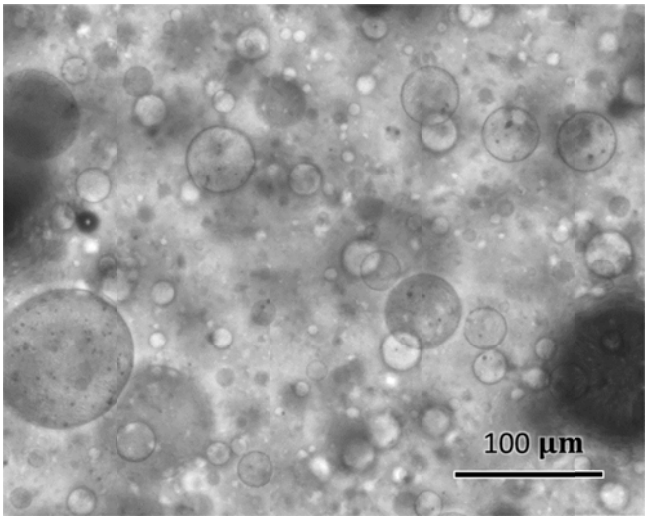

Fig. S5: Brightfield optical microscopy image of a particles-in-drops morphology for a sample with $\phi_p = 0.054$ and $\varrho = 1.75$. The grainy texture of the PEO drops is attributable to particles even though individual particles are not visible at this resolution.

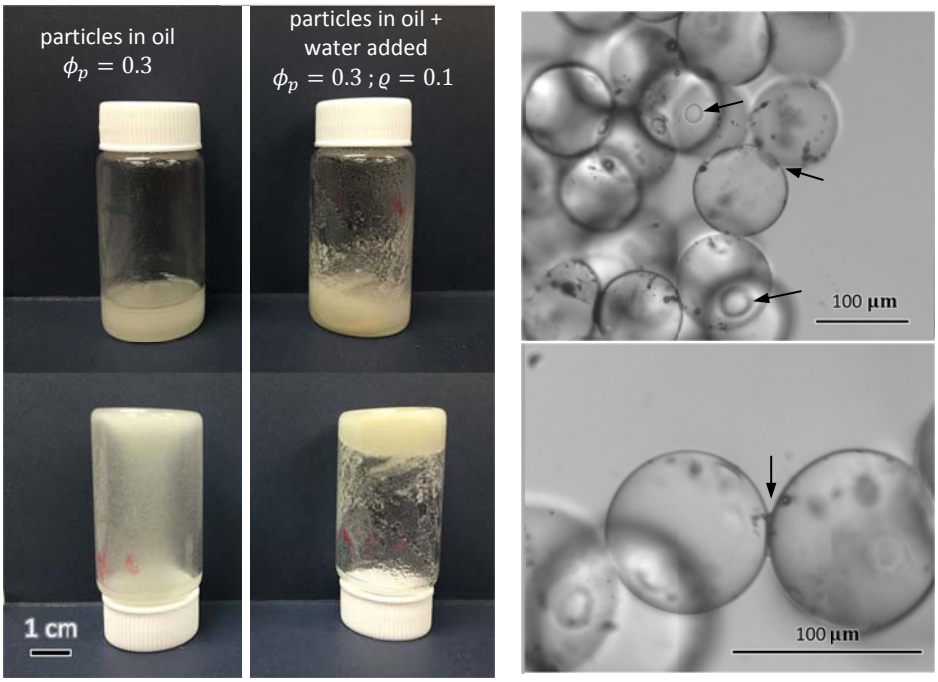

Fig. S6: Photographs of samples of glass particles in oil (leftmost column), and same with 3% water added (middle column). Note that the suspension can flow readily before water is added (leftmost column), but has a yield stress after water is added. Right: Brightfield microscope images of the sample in the middle column with ($\phi_p = 0.3$ and $\varrho = 0.1$) showing water menisci (indicated by arrows) binding the particles together.

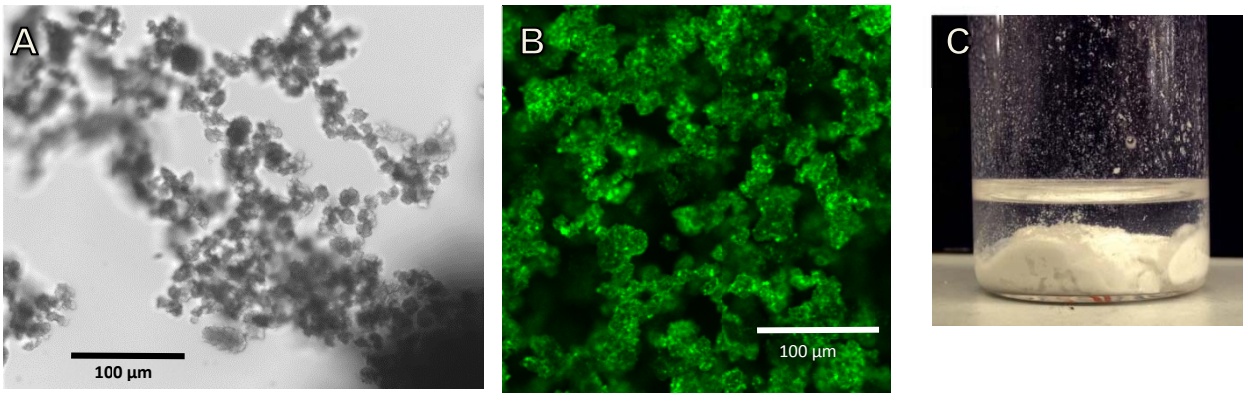

Fig. S7: Brightfield (A) and confocal (B) images of a capillary aggregate network prepared by mixing silica particles, mineral oil, and water with $\phi_p = 0.086$; $\phi_{oil} = 0.8$; $\phi_{water} = 0.114$. Since particles are fully-wetted by water, ϱ is defined as the ratio $\varrho = \phi_{water}/\phi_p = 0.75$. (A) was taken by squeezing the sample into a thin layer on a microscope slide to better-resolve the aggregates. (B) was taken on a thick slab of sample, and hence is more representative of the actual morphology. (C) Same system completely separates into two layers (oil layer atop a particle-in-water layer) at $\varrho = 0.8$.

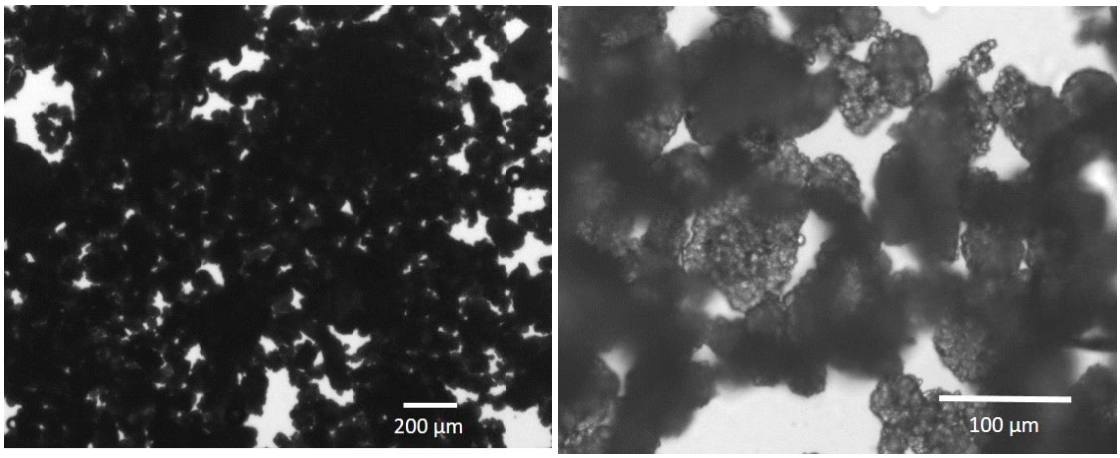

Fig. S8: Brightfield optical microscopy images at two different magnifications of capillary aggregates prepared by mixing polyethylene particles, mineral oil, and methanol with $\phi_p = 0.13$; $\phi_{oil} = 0.067$; $\phi_{methanol} = 0.8$. Since particles are fully-wetted by oil, ϱ is defined as $\varrho = \phi_{oil}/\phi_p = 0.5$.