Magnetite nanoparticles program the assembly, response, and reconfiguration of structured emulsions: supplemental information

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The endoskeletal droplet formulation is adjusted to include a cosolvent that reduces aggregation of magnetic nanoparticles to maximize the magnetic susceptibility of the final droplet mixture. Several cosolvents were screened to determine a suitable additive that significantly reduces the degree of aggregation of magnetic nanoparticles, a quantity we coarsely quantify by observing the mixtures under a microscope. Figure 1(a) is a ferrofluid mixture in the absence of petrolatum, whereas Figure 1(b) is the same mixture following the addition of petrolatum.

Micrographs of dispersions containing aggregated particles like those in Figure 1(b) are thresholded and converted to binary such that only aggregates remain. The fraction of the image that is colored is then compared for different cosolvent mixtures, normalized against a mixture consisting of 10 % EFH3, 30 % hexadecane, and 60 % petrolatum by weight. Figure 2 plots the normalized aggregation extent of magnetite nanoparticles in various mixtures of 10 % EFH3, 15 % cosolvent, 15 % hexadecane, and 60 % petrolatum by weight, as well as the calculated density of each suspension. Simple alkanes reduce aggregation to the greatest extent, with decane performing best. No suspension matched the density of water at the ratios added, so this property is not adjusted; however, it is useful to note the densities because emulsion stability improves as the density mismatch between phases decreases. As a result, magnetically functionalized endoskeletal droplets are expected to be more stable than non-particle-laden droplets due to their greater density.

The new endoskeletal droplet mixture is formulated with 15% decane, 15% hexadecane, 10% EFH3, and 60% petrolatum by weight. This ratio of components is used because it preserves much of the chemical and physical behavior of endoskeletal droplets from previous studies while improving dispersion stability. Moreover, minimizing the addition of species more volatile than hexadecane improves processing reliability in microfluidic devices, where the high temperatures requisite for generating endoskeletal droplets can generate bubbles that destabilize flow. While decane reduces aggregation, it does not prevent it altogether, so clusters that do form are removed by centrifugation of the droplet mixture.



Figure 1: Ferrofluid in hexadecane, on the left, creates a stable dispersion, while adding petrolatum leads to rapid and irreversible aggregation of the magnetite nanoparticles, shown on the right. Scale bar is $50 \,\mu\text{m}$.



Figure 2: Comparison of particle aggregation (\bullet) and suspension density (\triangle) in different cosolvents. Normal alkanes tend to reduce aggregation effectively, while halogenated solvents can come closest to density-matching the droplets with water.