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From bulk to interface: Critical role of surface-driven nanoparticle localization on the magnetorheological properties of ferrofluid emulsions

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Magnetorheological (MR) fluids are field-responsive materials that undergo millisecond-scale, reversible transitions from a liquid to a semi-solid state upon the application of an external magnetic stimulus [1]. The underlying mechanism lies in the induction of strong magnetostatic inter-particle interactions, leading to the formation of field-aligned columnar microstructures that overcome hydrodynamic drag and shear-induced forces. However, their applicability is constrained by irreversible particle aggregation and sedimentation. Ferrofluid emulsions constitute an alternative class of field-responsive colloidal systems, in which magnetic nanoparticles are confined within dispersed droplets in a continuous non-magnetic carrier liquid, providing enhanced stability and additional interfacial degrees of freedom.

In this research, the magnetorheology of oil-based ferrofluid emulsions was investigated as a function of nanoparticle localization. By tailoring surface chemistry, nanoparticles were strategically partitioned either within the droplet interior (using hydrophobic particles) or at the oil–water interface (using amphiphilic particles). Accordingly, the rheological and magnetorheological properties of these limiting systems, with nanoparticle volumetric concentrations $\phi \leq 0.2\%$ and an optimized droplet size distribution ($\sim$1 μm), were characterized using a magnetorheometer integrated with an MR device generating a magnetic field oriented perpendicular to the flow direction. We demonstrated the formation of highly stable ferrofluid emulsions, with creaming completely suppressed. Likewise, formulations with interfacially active nanoparticles exhibited greater increases in apparent viscosity and enhanced field-induced structuring, which was attributed to the coupling between dipolar assembly and interfacial stress distributions, modifying droplet deformation and interactions under flow. To rationalize this behavior, a coupled adsorption–aggregation–interfacial model was developed by describing dynamic interfacial tension using a Gibbs adsorption framework, incorporating both surfactant-induced Marangoni stresses and magnetic dipolar cohesive interactions within aggregates.

References

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