

Structured Liquid-in-Liquid Emulsion Stabilized by Surface-Engineered Liquid Metal Droplets as "Mutant" Pickering Emulsifiers

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Liquid metals (LMs), i.e., metals and alloys that exist in a liquid state at room temperature, have recently attracted considerable attention owing to their electronic and rheological properties useful in various cutting-edge technologies. In this study, eutectic Ga-In (EGaIn), one of the most studied LMs, in its microdroplet form was engineered to produce a novel droplet-type surfactant that can stabilize a liquid- *in* -liquid emulsion with unique functionality and processability. Dual-engineered LM droplets with a robust SiO₂ encapsulation shell and adsorbed amphiphilic cetyltrimethylammonium bromide (CTAB) exhibited high chemical stability against water-mediated oxidation while possessing excellent oil-water interfacial activity. These engineered droplet-type surfactants densely adsorb at microscale water-oil interfaces and prevent coalescence of the dispersed oil droplets in the liquid continuous phase, thereby producing long-term stable oil- *in* -water (O/W) emulsions; these droplet-type surfactants were named as "mutant" Pickering emulsifiers, indicating *particle* emulsifiers (different from *molecular* surfactants) that are similar to conventional Pickering emulsifiers, but consisting of the *liquid* core instead of the *solid*. The resulting emulsions exhibited enhanced yield stresses with viscoelastic properties, even at a minimal LM load, and showed remarkable sedimentation stability, indicating significant implications in terms of processability in versatile applications. The "structured" nature of such emulsions combined with the excellent photothermal conversion ability of LM droplets adsorbed at O/W interfaces resulted in the extremely localized photothermal heating effect. Furthermore, photothermally responsive phase-change oil droplets were produced using engineered LM droplets, which can be used for the on-demand release of valuable oil-soluble cargo as a potential application. We expect that engineered LM droplets as novel droplet-type emulsifiers of immiscible liquids in colloidal multidisperse systems will provide unique opportunities for various applications.

Keywords:

Liquid Metal, EGaIn, Pickering Emulsion, Droplet-Type Surfactant, Photothermal Conversion