Supplementary Information for

Wettability-Tuned Silica Particles for Emulsion-Templated Microcapsules

Nicholas C. Starvaggi,¹ B. Jack Bradford,² Cameron D.L. Taylor,² Emily B. Pentzer*1,²

¹Dept. of Chemistry, Texas A&M University, College Station, TX 77843

²Dept. of Materials Science & Engineering, Texas A&M University, College Station, TX 77843 *Correspondence: <u>emilypentzer@tamu.edu</u>

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Table S1. Amounts of diamine and diisocyanate used for interfacial polymerization in different emulsion systems.

Microcapsule Type	Continuous Phase	Ethylenediamine Amount (mmol)	1,6-hexanediisocyanate Amount (mmol)
PAO ₄₃₂	Water	1.7	1.3
[BMIM][PF ₆]	Toluene	1.3	1.7
DMF	Octane	1.3	1.7

Table S2. Average diameters of polyurea/silica microcapsules as determined by laser diffraction.

Microcapsule Type	Method	Average Diameter (μm)
PAO ₄₃₂		34.8 ± 25.8
[BMIM][PF ₆]	Laser Diffraction	13.8 ± 6.3
DMF		27.5 ± 17.1

Table S3. Average weight percent encapsulated material as determined via mass difference or quantitative ¹H NMR using mesitylene as an internal standard.

Microcapsule Type	Method	Weight Percent Core
PAO ₄₃₂	Mass Difference	82.5 ± 2.4
[BMIM][PF ₆]	¹ H NMR	68 ± 0.009
DMF	N/A	N/A



Figure S1. Representative ¹H-NMR spectrum of extracted [BMIM][PF₆] in deuterated acetone with mesitylene as an internal standard. The relative integration of peaks shaded in green and blue was used to determine weight% core.



Figure S2. Particle size distributions for modified silica particles.



Figure S3. Contact angle determination for (A) SiO_2 -Pristine, (B) SiO_2 -NH₂, and (C) SiO_2 -C1. Due to high hydrophilicity of all three substrates, no measurement could be recorded.



Figure S4. Optical microscopy of additional emulsion systems. (A) PAO₄₃₂-in-water stabilized by SiO₂-C1; (B) *N*,*N*-dimethylformamide-in-octane stabilized by SiO₂-C8.



Figure S5. Optical microscopy results of varied surfactant loading (0.5 wt% - 5 wt%) for PAO₄₃₂-in-H₂O (A), [BMIM][PF₆]-in-toluene (B), and DMF-in-octane (C) emulsion systems. Total emulsion volume was held at 3 mL.



Figure S6. Optical microscopy results of varied phase volume (1:9 - 9:1) for PAO₄₃₂-in-H₂O emulsion system. SiO₂-NH₂ loading was maintained at 3 wt% across all emulsions; total emulsion volume was held at 3 mL.



Figure S7. Additional SEM images of prepared microcapsules: (A) PAO₄₃₂, (B) [BMIM][PF₆], and (C) DMF.