In Situ Interfacial Surface Modification of Hydrophilic Silica Nanoparticles by Two Organosilanes Leading to Stable Pickering Emulsions

Supporting Information

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Author Contribution:

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1. Pickering emulsions stabilized by silica.

Droplet size analysis of *in situ* functionalized silica-stabilized toluene-in-water emulsion was done over the time span of 4 weeks by measuring each week the droplet diameters from confocal microscopy images. The analysis showed a broader distribution indicative of droplet coalescence occurring with time, although the majority of the droplets analyzed still remained at the 2-20 μm size range 4 weeks after preparation (Figure S1). Accordingly, the average droplet size diameter of the freshly prepared emulsion, 11.4 ± 6.4 μm, increased to 13.2 ± 12.0 μm 4 weeks after preparation.

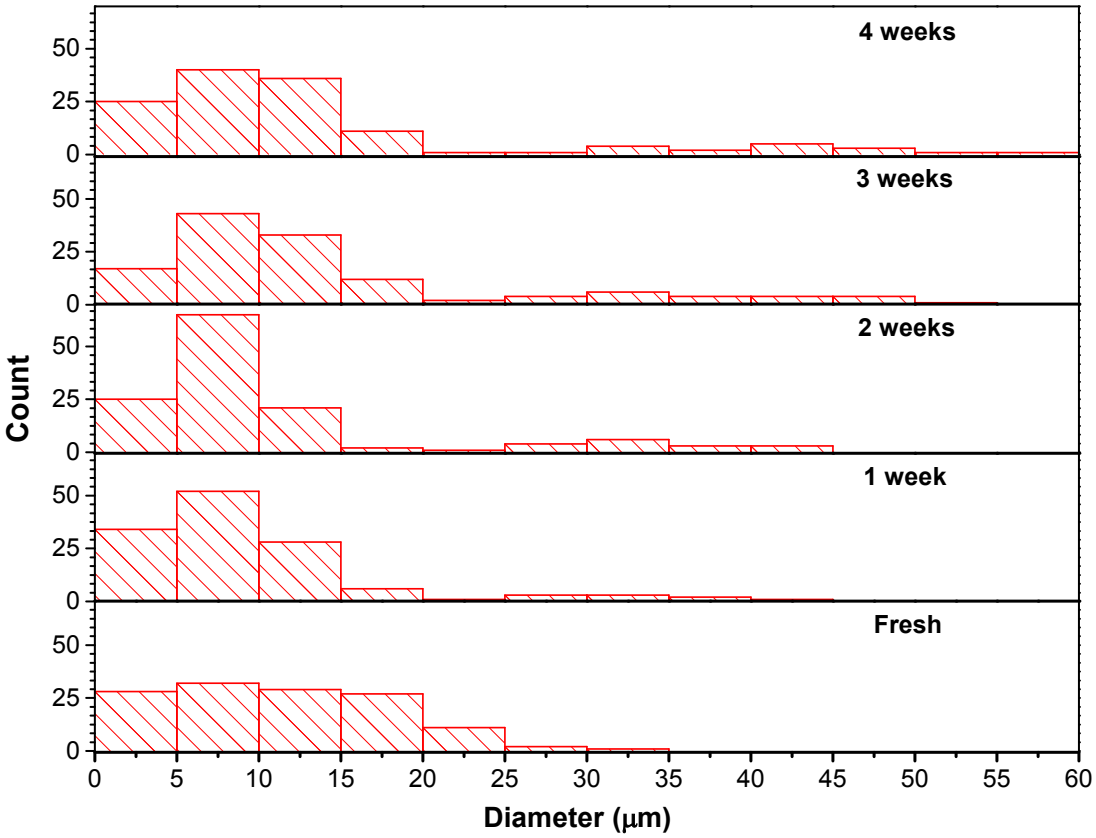

Figure S1. Droplet size distribution of 1:1 v/v toluene-in-water (o/w) emulsion stabilized by 1% w/v *in situ* modified silica nanoparticles, analyzed over a time span of 4 weeks after preparation (fresh – 4 weeks).

2. Silica nanoparticles at the oil-water interface.

The stabilization of the toluene droplets in water by silica was visualized in confocal fluorescence microscopy using a z-series stack, by tracing the localization of 6-aminofluorescein-labelled silica nanoparticles at the toluene-water interface and in the major water phase, and of Nile Red in the minor toluene phase, confirming oil-in-water Pickering emulsion (Figure S2).

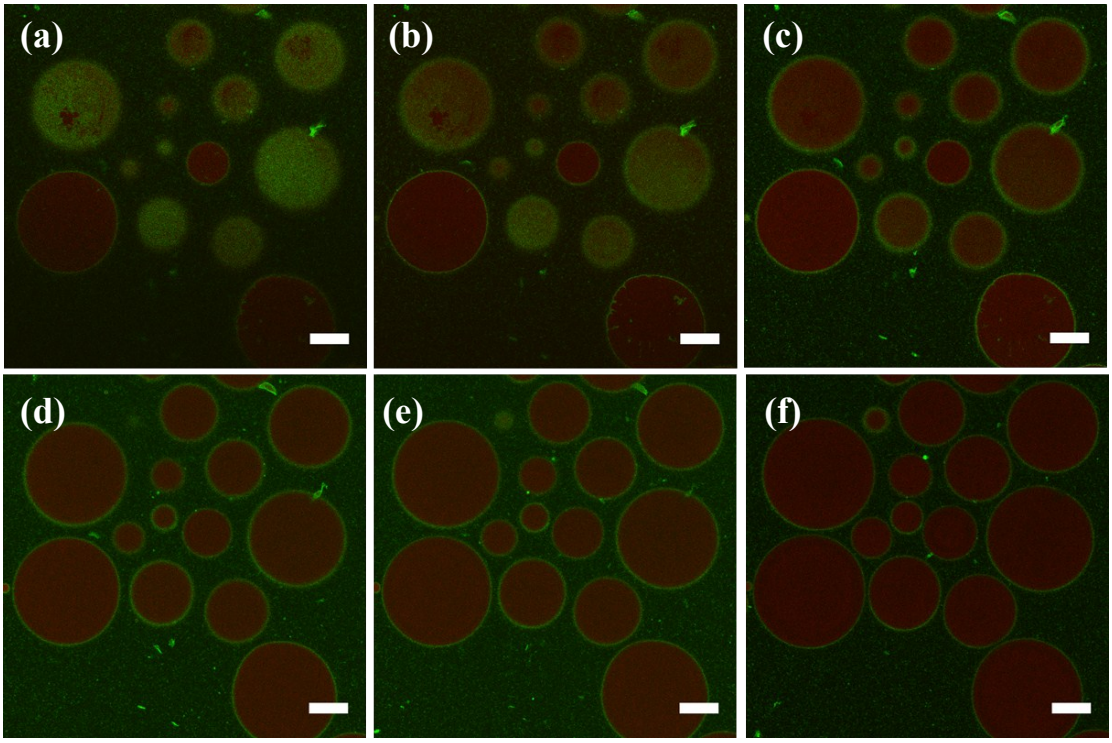

Figure S2. Confocal microscopy z stack overlay images of green and red channels showing the localization of 6-aminofluorescein-labelled SiO_2 nanoparticles at the toluene-water interface. The oil droplets were stained with Nile Red fluorophore (scale bar 20 μm in all images): (a) slice 12; (b) slice 15; (c) slice 18; (d) slice 24; (e) slice 30; (f) slice 33.

3. *In situ* functionalization of silica - control experiments with APTES.

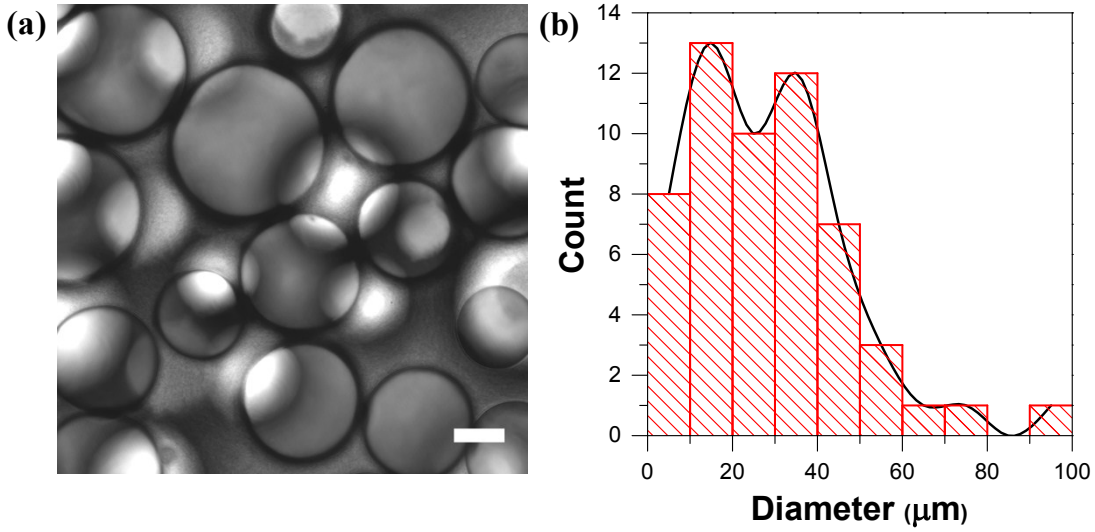

Figure S3. (a) Confocal microscopy image of a 1:1 v/v toluene-in-water (o/w) emulsion, stabilized by 1% w/v Aerosil® 300 silica nanoparticles that were functionalized *in situ* only with APTES (scale bar 20 μm). (b) Droplet size distribution of the o/w emulsion shown in (a). The average droplet diameter was larger than the *in situ* DTES-APTES-modified emulsion (29.0 ± 18.9 μm compared to 11.4 ± 6.4 μm, respectively). The average droplet diameter increased to 46.4 ± 15.4 μm 4 weeks after preparation, indicative of coalescence occurring with time.

4. Aerosil® 300 morphology characterization

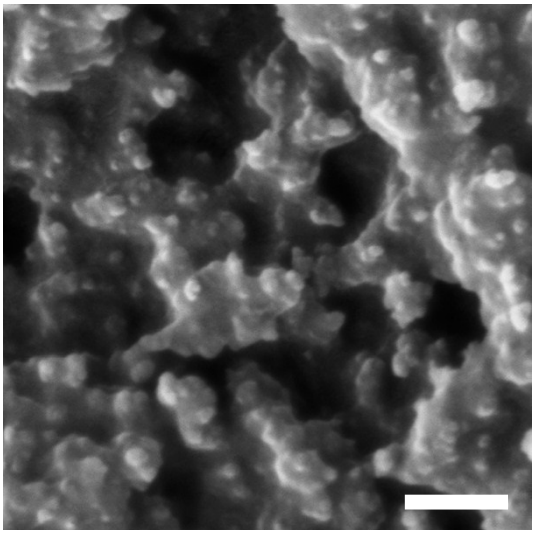

Figure S4. HRSEM micrograph of Aerosil® 300 fumed silica, showing the surface morphology and fine structure of the silica agglomerate. Scale bar is 200 nm.