# Supplementary Information

**Encapsulation of oil droplets using film-forming Janus nanoparticles**

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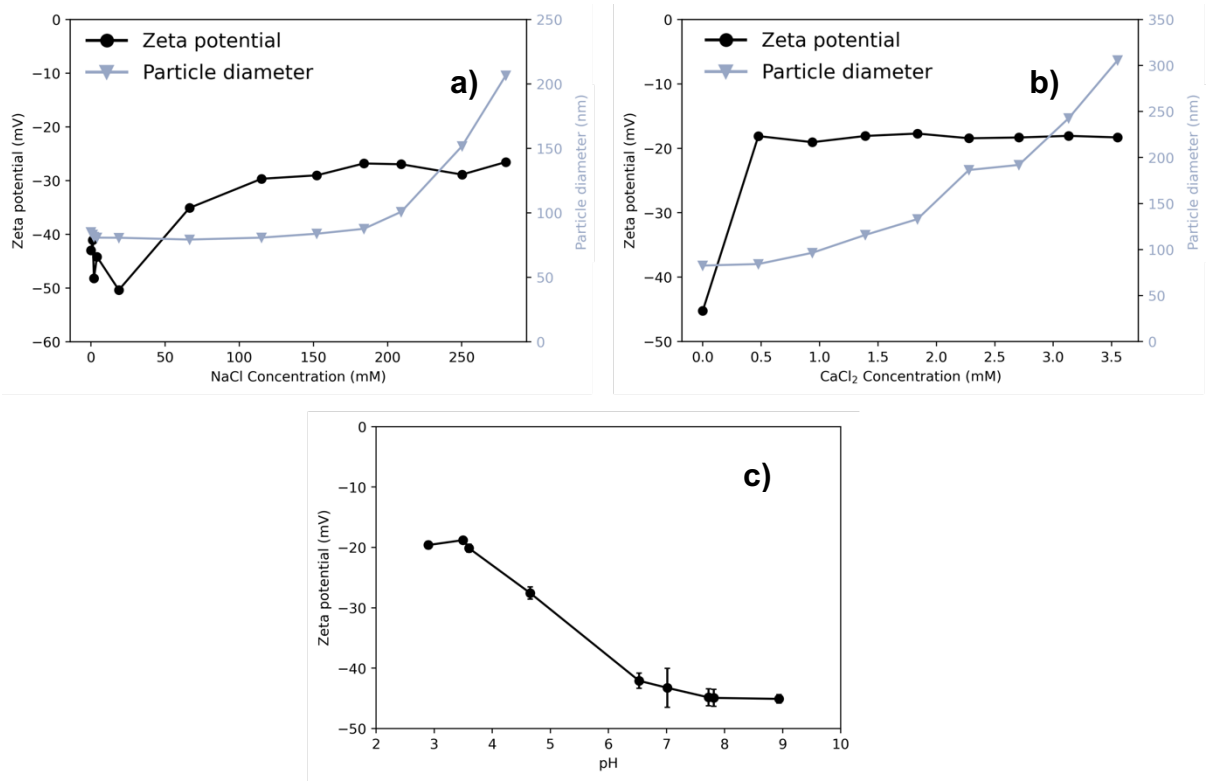


Figure S1.Zeta potential and particle diameter of Metastable Janus nanoparticles (0.17 wt%) as a function (a) NaCl and (b) $CaCl_2$ concentration for suspensions of pH = 8.3. (c) Zeta potential values measured for Janus nanoparticles suspensions (4 wt%) as a function of suspension pH.

## Table S1.Static contact angle of water on films made from Janus particles and PS seed particles.

| Nanoparticle constituent of film | Static contact angle |
|---|---|
| Polystyrene seed film | 83 ± 3° |
| JP film with P(MMA-BA) lobe protruding into air | 74 ± 3° |
| JP film with PS lobe protruding into air | 78 ± 3° |

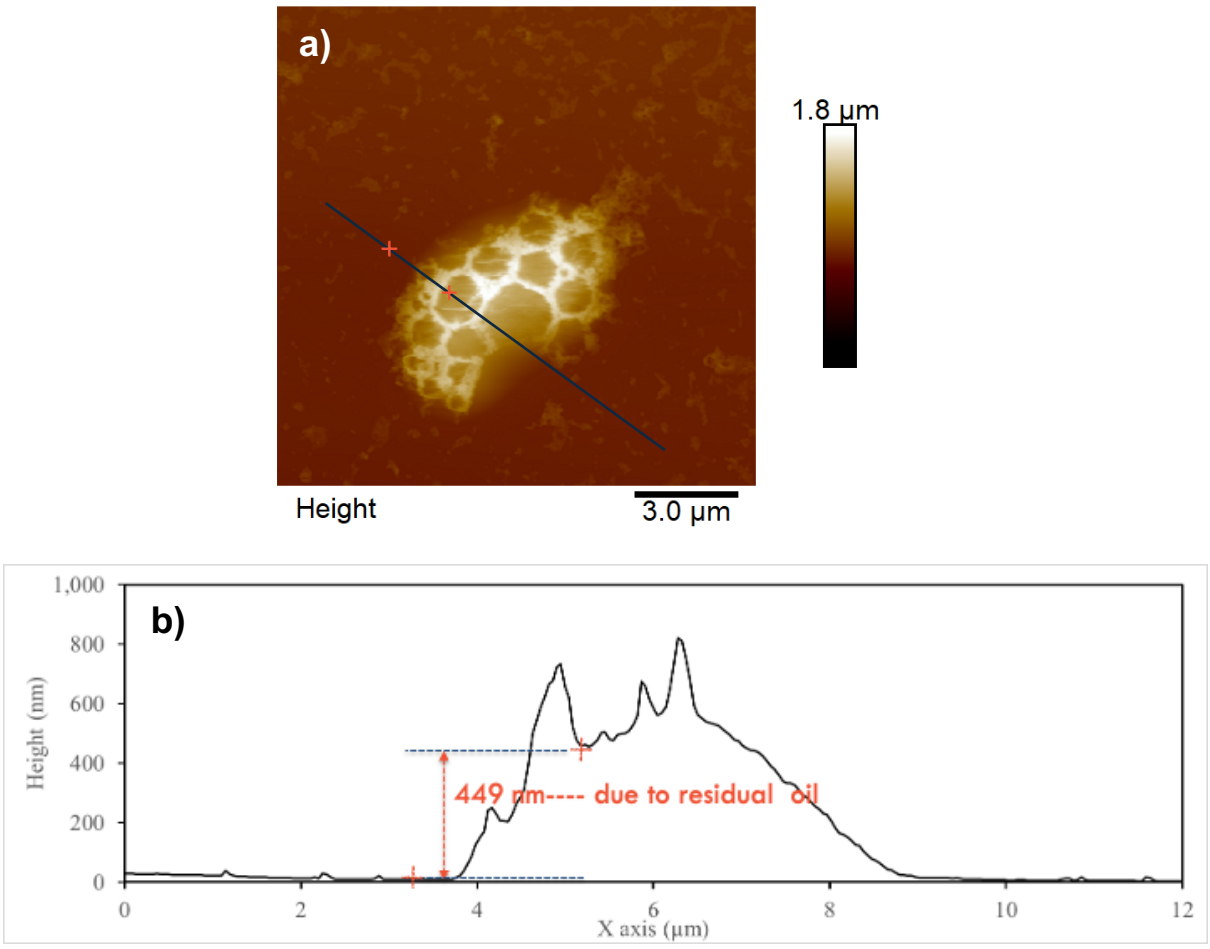


**Figure S2.AFM micrograph of unwashed silicone oil capsules** dried on a silicon wafer, corresponding to Figure 4f) of the main article text (a). Here the residual oil in the capsule is visible inside the capsule. b) Cross-sectional profile corresponds to the red line in AFM micrograph.

Table S2. Concentration and pH of Metastable Janus suspensions used to stabilize silicone oil emulsion droplets.

| Concentration of Janus nanoparticles (wt%) | Preparation age | Suspension pH | Duration of emulsion stability |
|---|---|---|---|
| 0.6% | fresh | 3.5 | Not stable |
| 4.2% | fresh | 4.2 | Not stable |
| 4.3 % | fresh | 6.3 | Not stable |
| 0.6% | fresh | 7.0 | 30 min |
| 4.0% | fresh | 7.7 | 2 days |
| 4.0% | aged | 7.7~8.2 | 14 days |
| 2.1% | aged | 7.9 | 6 days |
| 2.0% | fresh | 8.3 | 4 days |
| 4.1% | fresh | 8.5 | 2 day |
| 7.4% | fresh | 8.5 | 2 days |
| 2.2% (150 mM NaCl) | fresh | | Not stable |

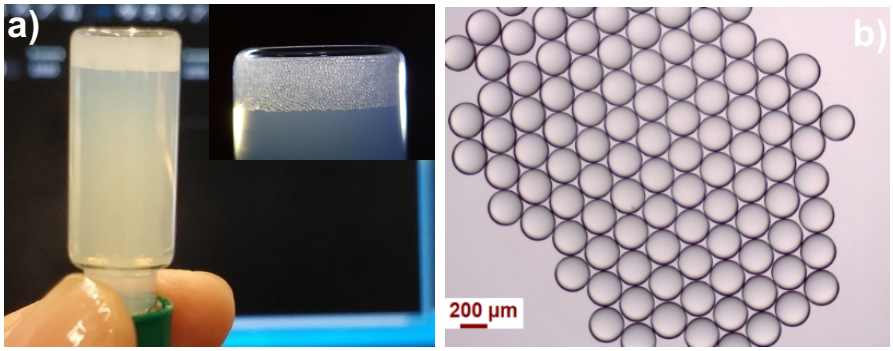


**Figure S3.Silicone oil emulsion droplets generated in the microfluidic channel**. a) Demonstration of collection of droplets with JPs with an upside down vial. b) Micrograph showing the droplets are monodisperse.

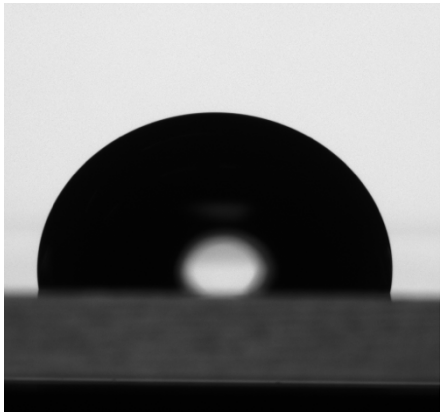

**Figure S4.Demonstration of the modification of the surface of silica upon application of silicone oil.** The water contact angle was 99 ± 1° on this coated glass surface, indicating that PDMS grafting was successful.

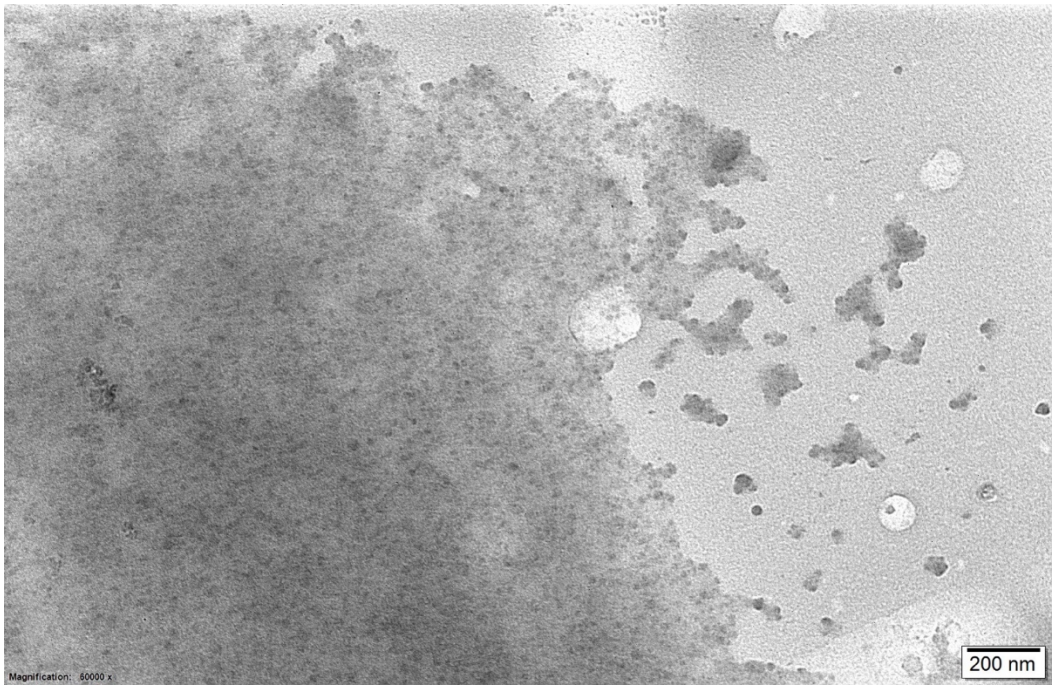


**Figure S5.TEM micrograph of 'Metastable JP'**. The soft lobes of the Metastable JP merge into a continuous film, while the hard PS lobes stand out as distinct particles.

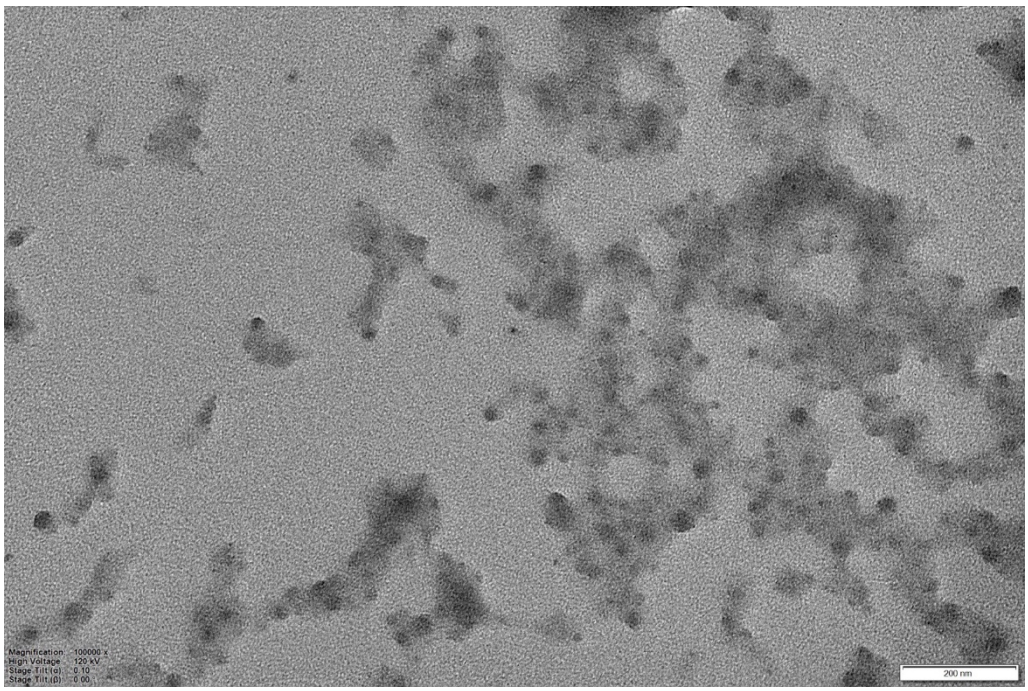


**Figure S6.TEM micrograph of Stable JP.** The soft lobes of the Stable JP merge into patches of film, while the hard PS lobes stand out as distinct particles.

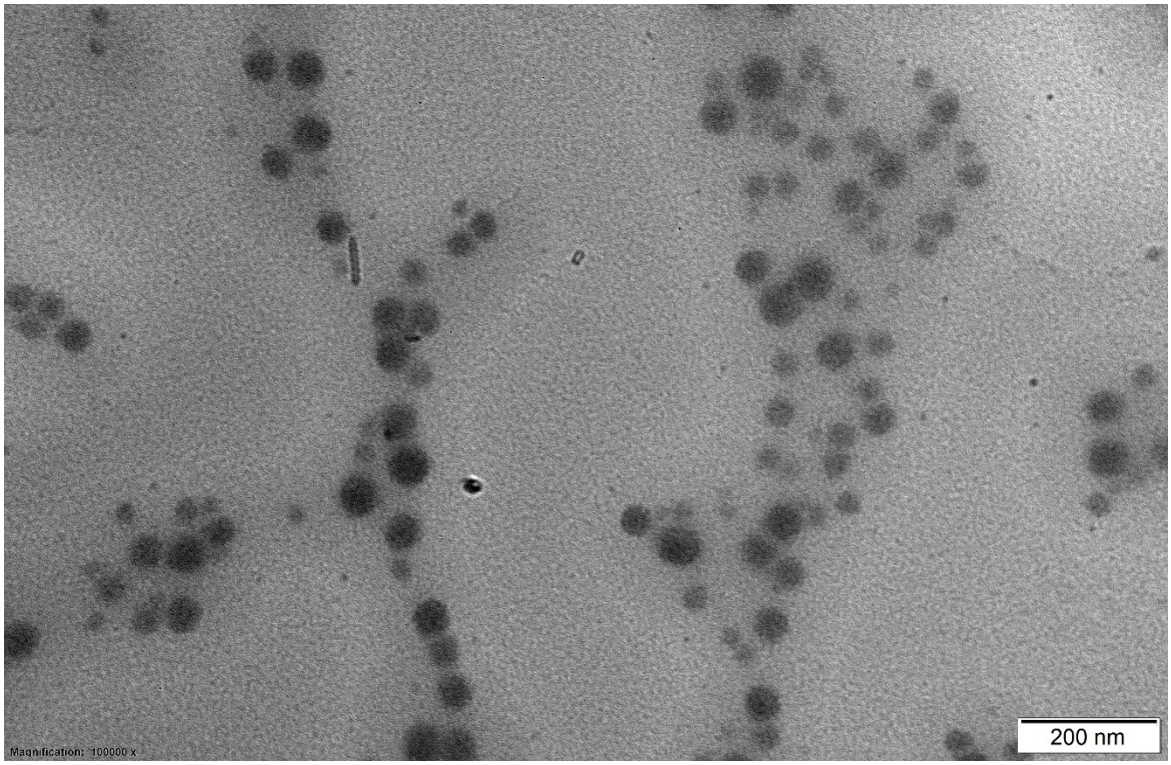


**Figure S7.TEM micrograph of Large JP,** synthesised using triblock DBTTC-poly(Sty)-block-poly(BA-co-AA) macro-RAFT copolymer. The soft lobes of the Large JP merge into patches of film, while the hard PS lobes stand out as distinct particles.

## Aqueous GPC characterization of macro-RAFT DoPAT-poly(HEA-co-AA-co-StS)

The water-soluble macro-RAFT polymer was dissolved in the aqueous GPC eluent (80 v.% 50 mM, pH 7.4 PBS, 20v.% methanol) over 24 hours to the concentration of approximately 4 mg $mL^{-1}$. All GPC samples were filtered through 0.45 μm polyethersulfone PES filters prior to characterization.

The system comprised a DGU-20A5R degasser, an LC-20ADXR UHPLC pump, an SIL-20AHT automatic injector, a CTO-20A column oven, a RID-20A refractive index detector, and an SPD-M20A Shimadzu detector. The setup included a PL aquagel-OH 8μm 50 × 7.8 mm guard column, followed by three columns in the following order: PL Rapide Aqua L 150 x 7.5 mm, PL Rapide Aqua H 150 x 7.5 mm, and PL Aquagel-OH MIXED-H 8μm 300 x 7.5 mm. The flow rate was set at 1 mL $min^{-1}$, and the temperature was maintained at a constant 40 °C. The unit was calibrated using commercially available linear polyethylene oxide (PEO) standards (0.2–1000 kDa, ReadyCal-Kit PEO/PEG, PSS).