# Supplementary Information

#### Yield stress-enabled for microencapsulation of field responsive microparticle suspensions

Samuel R. Wilson-Whitford<sup>\*a,b</sup>, Maria Chiara Roffin,<sup>a,c</sup> Jinghui Gao,<sup>a</sup> Thitiporn Kaewpetch<sup>d</sup> and James F. Gilchrist<sup>\*a,b</sup>

<sup>a</sup>Department of Chemical and Biomolecular Engineering, Lehigh University, Bethlehem, PA, USA

<sup>b</sup>School of Chemistry, University of Leicester, Leicester, UK

<sup>c</sup>School of Science and Technology, Nottingham Trent University, Nottingham, UK

<sup>d</sup>Department of Packaging and Materials Technology, Faculty of Agro-Industry, Kasetsart University, Bangkok, Thailand

### Experimental

*Materials:* 1 µm silica particles  $(0.951 \pm 0.022 \text{ µm})$  were purchased from Fuso chemical co., Japan. Solsperse 3000 dispersant was donated by Lubrizol Ltd. Dioctyl phthalate (99%), Nile red and Mowiol 8-88 ( $M_w = 67 \text{ g mol}^{-1}$ ) polyvinyl alcohol were purchased from Sigma-Aldrich. Ethyl cellulose (98%) and isophorone diisocyanate (98%) were purchased from MP Biomedicals and diethylene triamine (99%) were purchased from Alfa Aesar.

*Equipment:* Automatic Langmuir-Blodgett deposition set up detailed in previous publication<sup>1</sup>. Suspensions were sonicated using a Fisher Scientific FS20D. Volume measurements were performed with calibrated 100  $\mu$ L and 1000  $\mu$ L Fisherbrand Elite pipettes. Vapor deposition was performed using Eddy SC 20 E-Beam Evaporator. Microscopy of samples was performed on a Visitech International confocal microscope fitted with a VT-eye high speed AOD based point scanner, 20x objective and 100x piezo objective. Imaging was performed at 72 fps. 3D scans were performed using Visitech software and the 100x piezo objective. Image analysis was performed using ImageJ software and accompanying 3D viewer plugin. Additionally, Particle tracking was performed using a Matlab<sup>2</sup> script version of IDL particle tracking software<sup>3</sup> available also through publicly available repositories.<sup>5</sup>

#### Videos

**V1:** Particle motion under magnetic agitation. Particles suspended in 1 wt% EC:DOP gel inside set-up illustrated in Fig. S4. Particle motion is indicative of a viscous environment. 72 fps.

**V2:** Particle motion under magnetic agitation. Particles suspended in 6 wt% EC:DOP gel inside set-up illustrated in Fig. S4. Particle motion is indicative of an elastic environment. 72 fps.

**V3:** Example of particle and particle cluster agitation inside microcapsules under the influence of an external magnetic field. EC:DOP 3 wt%. Scale bar 50 µm. 60 fps.

V4: Microscopy of particle containing capsules during variable speed agitation on top of a magnetic stirrer plate. EC:DOP 3 wt%. Scale bar  $\sim 200 \,\mu m$ . 30 fps.

#### **Bulk confocal imaging**



**S1:** Illustration of well devices used for confocal light scanning microscopy of bulk Janus particle suspensions.

### **Settling Velocity**

$$v = \frac{2}{9} \cdot \frac{(\rho_p - \rho_f)}{\mu} \cdot gr^2$$



**S2:** Apparent settling velocity of particles in ECDOP over 4 h. Values for 1-6 wt% ECDOP taken from data for figure 2. Value for 0 wt% ECDOP (i.e. DOP) calculated from literature.<sup>5</sup>



**S3.** X-direction view of vertical z-axis confocal microscopy of particles trapped at the interface of a dried layer of capsules with cores comprised of pure DOP (0 wt% EC:DOP). Majority of particle accumulation is in the membrane. Scale bar =  $10 \,\mu$ m.

## 6 wt% synthesis and analysis



**S4.** Particle cluster tracking data for particles encapsulated in the yield stress core of a microcapsule. (*a*) Frame 50 from particle tracking analysis of 6 wt% EC:DOP microcapsule under random magnetic oscillation. Scale bar = 50  $\mu$ m. (*b*) Distance travelled by particles over time due to magnetic force (*c*) MSD of tracked particles from magnetic force. (*d*) Total distance travelled by particles under magnetic influence against spatial position of particle relative to the capsule centre of mass.

### **Size Distribution**



**S5:** Size distribution of microcapsules calculated from CLS microscopy over 487 capsules.

## Microscopy of glass surface in bulk



**S6:** CLSM imaging of 1  $\mu$ m particles on the glass substrate 15 min after preparation of samples at ethyl cellulose concentrations in dioctyl phthalate of (*a*) 0 wt.% (*b*) 1 wt.% (*c*) 2 wt.% (*d*) 3 wt.% (*e*) 4 wt.% (*f*) 5 wt.%

## References

- 1. X. Li and J. F. Gilchrist, *Langmuir*, 2016, **32**, 1220–1226.
- Matlab particle tracking repository, https://site.physics.georgetown.edu/matlab/, (accessed Janurary 2020)
- 3. J. C. Cocker and D. G. Grier, J. Colloid Interface Sci., 1996, 179, 298-310.
- Particle Tracking using IDL, https://physics.emory.edu/faculty/weeks/idl/index.html, (accessed January 2020)

5. L. De Lorenzi, M. Fermaglia and G. Torriano, J. Chem. Eng. Data, 1997, 42, 919-923