## **Supporting Information**

## Micrometer-sized gold-silica Janus particles as particulate emulsifiers

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### Experimental

#### Materials

1-Undecanol (purity 99%), poly(ethylene glycol) (PEG:  $M_n$  285-315), ethylene glycol (purity 99%), diiodo methane (purity 99%) and glycerol (purity 99%) were purchased from Sigma-Aldrich. Methyl myristate (purity 95%) was purchased from Wako and was used as received.

#### Critical surface tension

A critical surface tension of Au substrate was determined by Zisman plot using contact angle measurements of various liquids of known surface tension (in mNm<sup>-1</sup>): PFS (25.7), styrene (33.1), *n*-dodecane (30.1), methyl mryistate (29.6) [1], 1-undecanol (28.5) [1], PEG (43.5) [2], ethylene glycol (47.7) [2], diiodo methane (50.8) [2], glycerol (63.4) [2] and water (72.8) [2].

## Au surface

Au surface is categorized to have a "high" surface energy and it is well known that Au surface is readily contaminated by adsorption of organic materials, whose chemical structures are not clear at this stage, in order to decrease surface energy [3]. Several techniques have been developed to obtain clean metal surface: refreezing of melted gold in vacuum [4,5], electrochemical treatment [6], and strong chemical cleaning [7].

### References

- (1) Binks, B. P.; Clint, J. H. Langumur 2002, 18, 1270-1273.
- (2) Fox, H. W.; Zisman, W. A. J. Colloid Sci. 1950, 5, 514-531.
- (3) Butt, J. H.; Graf, K.; Kappl, M. Physics and Chemistry of Interfaces, 2nd ed.; Wiley-VCH Verlag GmbH & Co. KGaA: Weinheim, **2006**
- (4) Gardner, J. R.; Woods, R. J. Electroanal. Chem. 1977, 81, 285-290.
- (5) Smith, T. J. Colloid Interface Sci. 1980, 75, 51-55.
- (6) Schrader, M. E. J. Colloid Interface Sci. 1984, 100, 372–380.
- (7) Dote, J. L.; Mowery, R. L. J. Phys. Chem. 1988, 92, 1571-1575.

**Figure S1** (a-c) Digital photographs, (d-f) optical microscopy images and (g-i) SEM images of (a,d) 2D SiO<sub>2</sub> and (b,e) Au-SiO<sub>2</sub> Janus particle colloidal crystal on glass substrate and (c,f) glass substrate after removal of the Au-SiO<sub>2</sub> Janus particles.

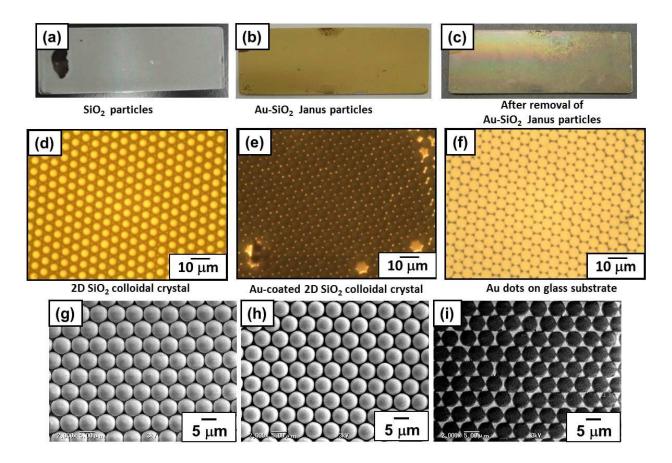
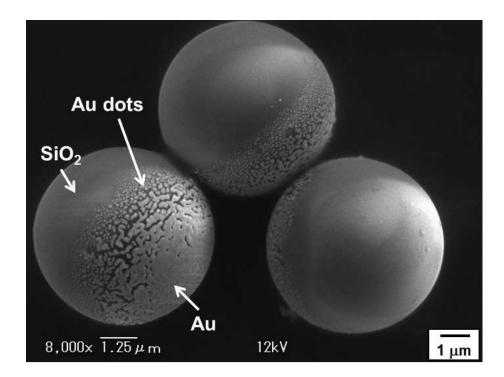
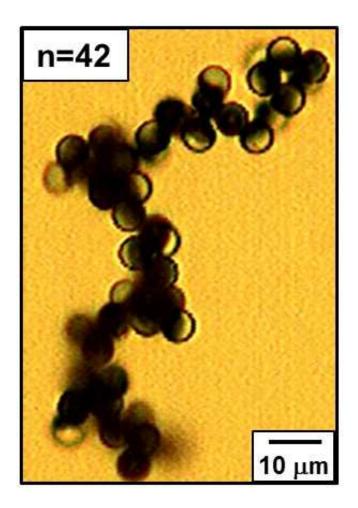


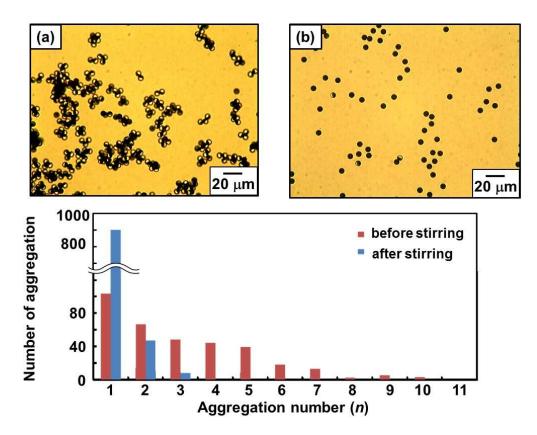
Figure S2 SEM image of Au-SiO<sub>2</sub> Janus particles

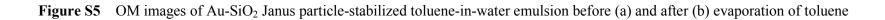


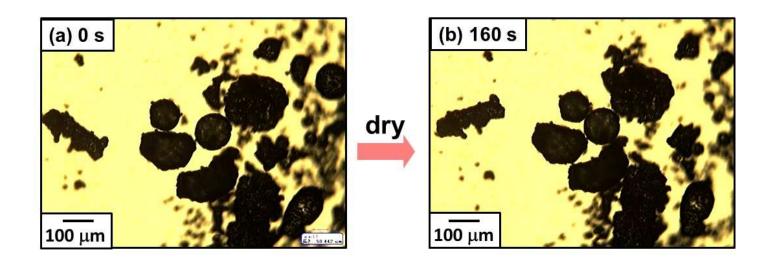
**Figure S3** OM image of Au-SiO<sub>2</sub> Janus particle clusters formed by hydrophobic interactions in water media with an aggregation number (n) of 42.



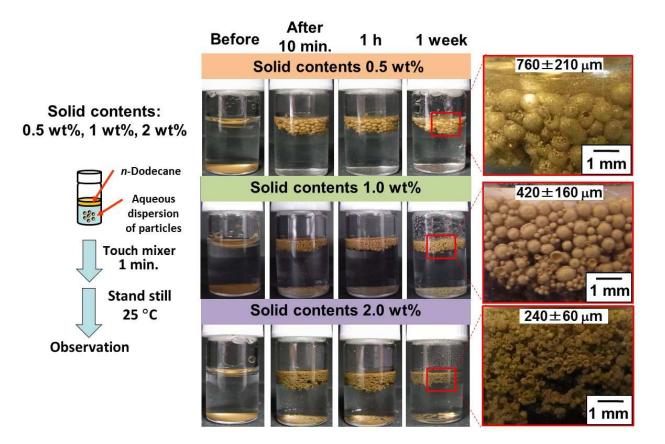
**Figure S4** OM images of Au-SiO<sub>2</sub> Janus particles dispersed in aqueous media: (a) before and (b) after stirring using a touch mixer (Vortex Genius 3,  $IKA^{\text{(B)}}$ ) at 3,000 rpm.



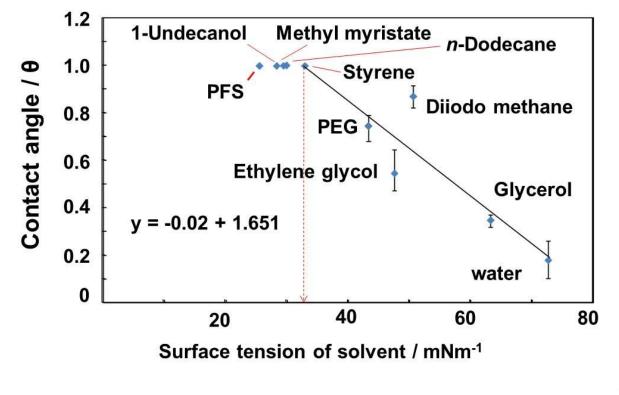




**Figure S6** Digital camera images of "Pickering-type" emulsions stabilized using Au-SiO<sub>2</sub> Janus particles at different Janus particle concentrations (0.5, 1.0 and 2.0 wt%)

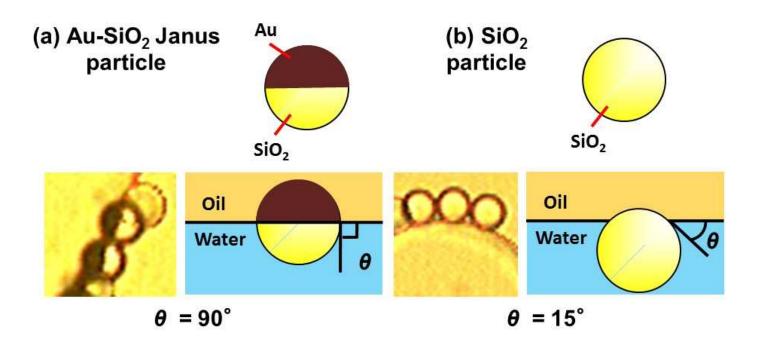


**Figure S7** Zisman plot of contact angle versus surface tension of solvent for various liquids on Au-coated glass. The critical surface tension  $\gamma_{Au} = 32.6 \text{ mNm}^{-1}$  is determined by extrapolating the liquid data to of contact angle = 1.

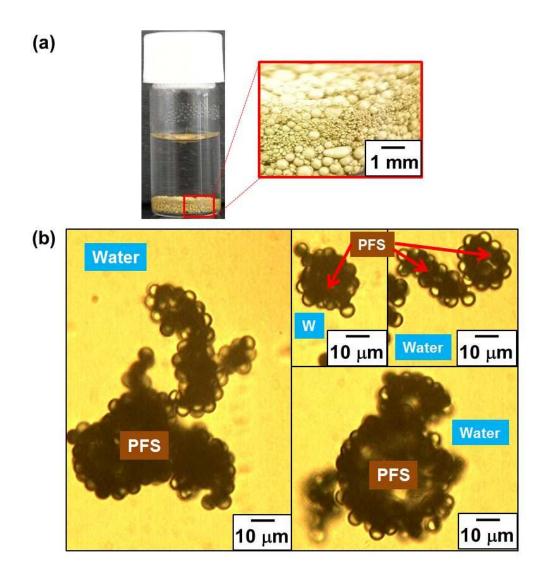


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Figure S8 Contact angles of (a) Au-SiO<sub>2</sub> Janus particle and (b) SiO<sub>2</sub> particle at 2,3,4,5,6-pentafluorostyrene (PFS)-water interface



**Figure S9** (a) Digital photograph and (b) OM images of 2,3,4,5,6-pentafluorostyrene (PFS)-in-water "Pickering-type" emulsions stabilized with Au-SiO<sub>2</sub> Janus particles.



**Figure S10** SEM image of cross section of the PPFS microspheres carrying Au femto-litter cups on their surface. Inset shows magnified image.

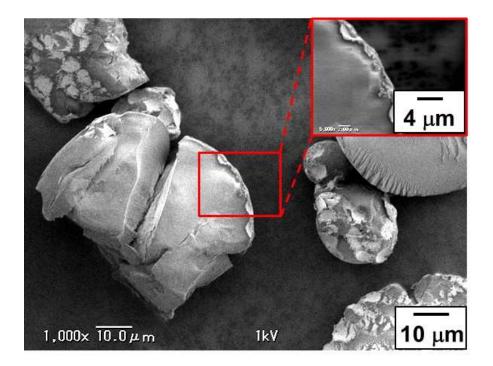
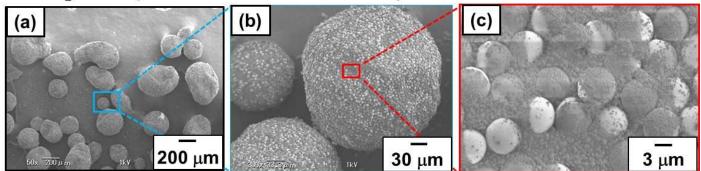


Figure S11 (a-e) SEM images of the Janus particle-stabilized PS microspheres. (d,e) Cross section image of the microsphere.



## Au-SiO<sub>2</sub> Janus particles-stabilized PS microspheres

# **Cross-section images of the PS microsphere**

