Supporting Information

Deformation and Stability of Core-shell microgels at Oil/water Interface

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1. The morphologies of PTFMA particles and core-shell microgels

The morphologies of PTFMA particles and core-shell microgels have been characterized by scanning electron microscope (SEM). The particles were dispersed on a silicon substrate by adding 30 μ L of the particle dispersion onto a silicon substrate after been dried at 50 °C for 2 hours. The silicon substrate was sputter-coated with a 5 nm homogeneous gold layer and observed by Hitachi S5500 FE-S(T)EM. Figure S1(a) and (b) shows the PTFMA particles and P(NIPAM-*co*-AAc)@PTFMA core-shell microgels, respectively.



Figure S1. SEM image of PTFMA particles (a) and P(NIPAM-*co*-AAc)@PTFMA core-shell microgels.

2. Interfacial pressure measurement

In this study, the interfacial pressure is measured by a balance connected with a Wilhelmy plate. The Wilhelmy plate is partly immersed blow the water/decane interface. The forces acting on the plate consist of the gravity, downward interfacial force due to the interfacial tension, and buoyancy due to displaced water upward. For a rectangular plate with dimensions (length l_P , width w_p and thickness t_P) and material density ρ_P , immersed to a depth h_l in a liquid of density ρ_l , the net downward force is given by the equation S1:

$$F = \rho_P g l_P w_P t_P + 2\gamma_{ow} (t_P + w_P) \cos\theta + 2\gamma_{ao} (t_P + w_P) \cos\varphi - \rho_l g t_l w_l h_l \qquad (\text{equation}$$

S1)

where g is the gravitational constant, γ_{ow} and γ_{ao} are the interfacial tensions between oil/water and air/oil phases, respectively, θ and φ are water contact angle at two sets of threephase contact regions, i.e. water/oil/Wilhelmy plate and air/oil/Wilhelmy plate. The interfacial tension can be calculated based on equation S1 by the measured force from the balance. The interfacial pressure is the value of the interfacial tension reduction from a clean interface to the interface with a monolayer of microgels. The Wilhelmy plate is made of platinum, assuming it is completely wetted by the water (i.e. $\cos\theta = 1$), so the interfacial pressure is then obtained by the equation S2:

$$\pi = -\Delta \gamma = -\left(\frac{\Delta F}{2(t_P + w_P)}\right) = -\frac{\Delta F}{2W_P}, \text{ if } w_P \gg t_P \qquad (\text{equation S2})$$

3. Compression isotherm of pure P(NIPAM-co-AAc) microgels

Compression isotherm of pure P(NIPAM-*co*-AAc) microgels (PMG) is shown in Figure S2. It has four regimes structure, which is similar as the compression isotherm of the core-shell microgels. The critical desorption interfacial pressure is ~45 mN/m.



Figure S2. The compression isotherm of pure P(NIPAM-co-AAc) (PMG) microgels