Supporting Information

Organic/inorganic heterostructures templated by interfacial

instability-driven BCP colloids in deformable emulsion droplets

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Fig. S1. (a) TEM and (b) SEM images of the highly uniform PS_{38k} -*b*-P4VP_{82k} spherical clusters at relatively low magnification. (c) Magnified TEM image of PS_{38k} -*b*-P4VP_{82k} spherical clusters, in which the P4VP domains are selectively stained by I₂ vapor. (d) The size distribution of the PS_{38k} -*b*-P4VP_{82k} spherical colloids by DLS measurement. The time autocorrelation function can be expressed as the equation of $g(2)(\tau) = \frac{\langle I(t) \cdot I(t+\tau) \rangle}{\langle I(t) \rangle^2}$, in which I(t) represents the intensity of scattered light at a

given time t, while $\langle I(t) I(t+\tau) \rangle$ is the average of the product of light intensities at times t and $t+\tau$. $\langle I(t) \rangle^2$ denotes the square of the mean intensity of the light. The attenuation characterization of this autocorrelation function, which varies with the delay time (τ) , can be used to calculate the diffusion coefficient of the particle, thus generating the size distribution of the particles.



Fig. S2. SEM images of PS_{38k}-*b*-P4VP_{82k} assemblies emulsified with SDS aqueous solution of different concentrations. (a) 0.1 mg/mL; (b) 0.2 mg/mL; (c) 0.5 mg/mL; (d) 3 mg/mL.



Fig. S3. A series of BCP aggregates assembled from (a) PS_{35k} -*b*-P4VP_{2.7k}; (b) PS_{165k} -*b*-P4VP_{35k}; (c) PS_{75k} -*b*-P4VP_{25k}; and (d) PS_{102k} -*b*-P2VP_{97k}.



Fig. S4. The plot showing the interfacial tension values of the PS_{38k} -*b*-P4VP_{82k} chloroform solution of different BCP concentrations in 3.0 mg/mL of SDS aqueous solution.



Fig. S5. (a) STEM and (b-d) TEM images of the hybrid heterostructures. Magnified (e) SEM and (f) TEM images of PS_{38k}-P4VP_{82k}/SiO₂ heterostructures.



Fig. S6. A series of TEM images of PS_{38k} -*b*-P4VP_{82k}/SiO₂ heterostructures with varied SiO₂ layer thickness generated by addition different amount of TEOS precursor. (a) 1 μ L; (b) 3 μ L; (c) 4 μ L; (d) 5 μ L.