## **Supporting Information**

## Self-Assembly of Inverse Patchy Colloids with Tunable Patch Coverage

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## 1. Effect of temperature on the surface morphology of the IPC particles:

Figure S1 shows the effect of temperature on the structure of the IPC particles spread at air-water interface. As the operating temperature of polystyrene particles at the air-water interface is close to the glass transition temperature of polystyrene (~95°C), the particles undergo transformation from spherical-to-non-spherical (Figure S1 A) shape. However, as the temperature of the system is reduced, the spherical shape of the particles is retained but remain fused (Figure S1 B) due to their less liquid-like behaviour. Finally, when the temperature is brought down to 60°C, the particles show resistance to flow as the operating temperature fall well below the T<sub>G</sub>. At this temperature, the particle retains their shape and remain non-fused (Figure S1 C). Hence, we find optimum temperature to be around 60°C. Additionally, in order to ascertain that the particle did not deform during heating, we measured the size of the particles after synthesis. The average size of IPC particles measured from 50 representative HRSEM images is found to be 2.1 ± 0.1 µm, which is in close agreement with amidine functionalized bare PS particles (2.2 ± 0.1 µm). This measurement reiterates that the particles remain intact even after heating at 60°C during the hydrolysis process. Figure S1 D displays the size distribution of assynthesized IPC particles.



**Figure S1.** HRSEM images showing the state of IPC particles spread at air-water interface at A) 80°C, B) 70°C, C) 60°C. The histogram to show the distribution of size of as-synthesized IPC particles is depicted in D. The scale bar corresponds to 2  $\mu$ m.

## 2. HRSEM images showing the effect of surfactant concentration on the quality of nanoparticle deposition.

Figure S2 displays the representative HRSEM images to show the effect of surfactant concentration on the contact angle of the particles. It is clear from Figure S2 that as the concentration of surfactant increases, the contact angle of the PS particles spread at the interface decreases. The observed trend is in line with the theoretical prediction based on Young-Dupre equation.



**Figure S2.** The representative HRSEM images revealing the effect of surfactant concentration used in the subphase. The surfactant concentration (expressed in c/cmc) used are A) 0, B) 0.2, C) 1 and D) 10. The scale bar corresponds to 2  $\mu$ m.