Electronic supplementary information for

Toward theta-shaped polymer composite particles by seeded emulsion polymerization

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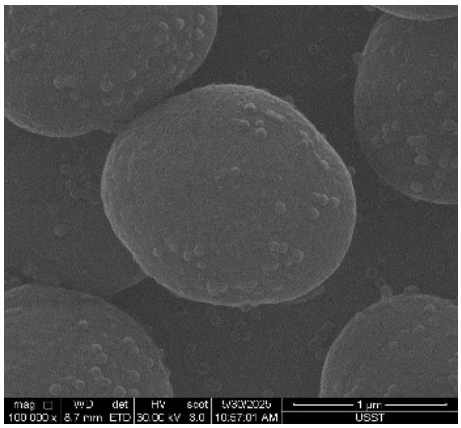

Fig.S1 SEM image of PS-PGMA composite particles synthesized without DBP (entry 8 in Table 1).

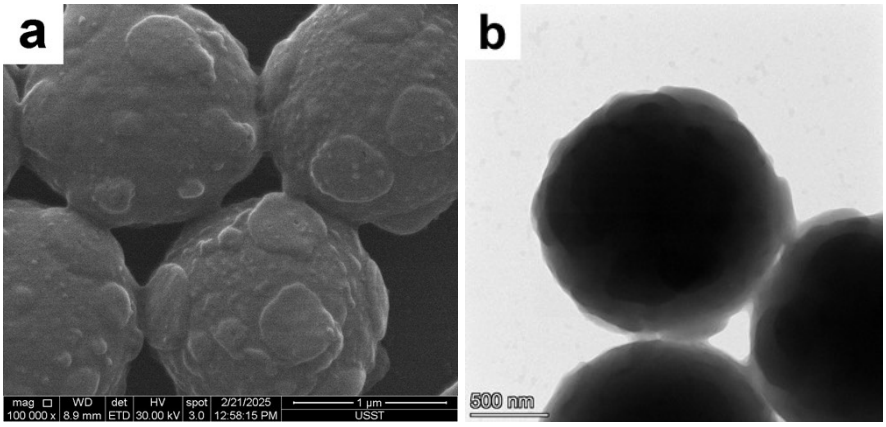

Fig. S2 (a) SEM and (b) TEM images of PS-PGMA composite particles synthesized at a G_s/G_p mass ratio of 1.5: 1.5 and a GMA/DBP ratio of 3/1 (entries 13 in Table 1).

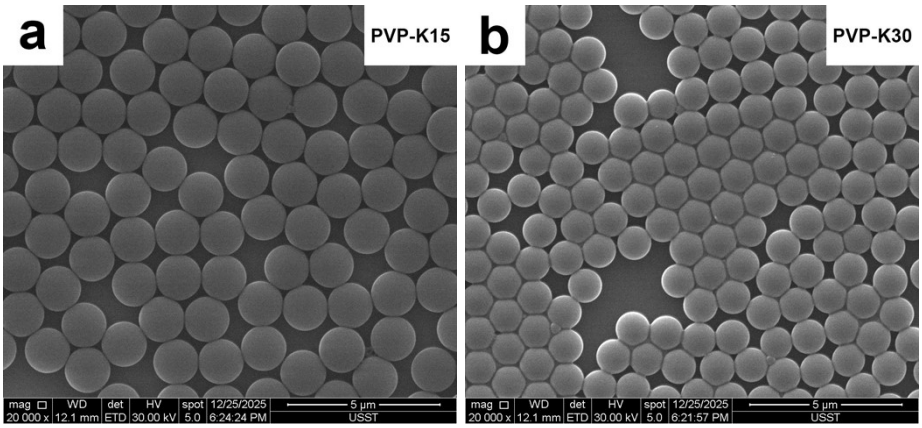

Fig. S3 SEM images of the PS particles prepared using PVP with different *K*-values: (a) 15 K; (b) 40 K.

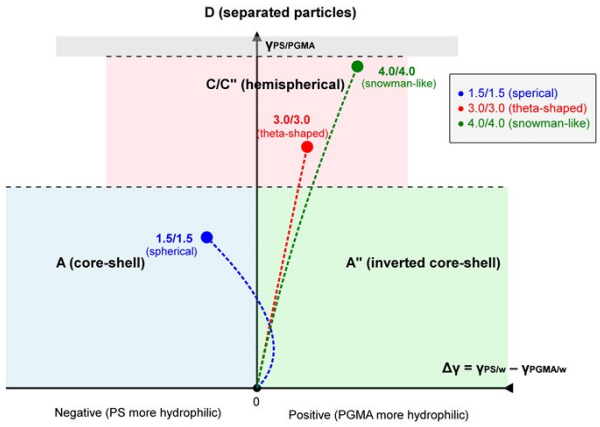

Fig. S4 Conceptual phase diagram for PS-PGMA composite particles based on the thermodynamic model of El-Aasser et al. (ref.19). The x-axis represents the interfacial tension difference $\Delta\gamma = \gamma_{PS/water} - \gamma_{PGMA/water}$, and the y-axis represents the polymer-polymer interfacial tension γ_{12} . Colored regions schematically indicate the stability domains for core-shell (A), inverted core-shell (A"), partially engulfed (hemispherical, C/C"), and separated particle (D) morphologies. In our system, the partially engulfed region manifests as two distinct morphologies depending on the G_s/G_p ratio. Theta-shaped particles (3.0/3.0), which can be viewed as a symmetric double-hemisphere structure with PGMA lobes at both poles, and snowman-like particles (4.0/4.0), corresponding to a single-hemisphere configuration. Colored dashed arrows represent the evolution paths of the

effective interfacial tension $\gamma_{PS/PGMA}^{eff}$ during polymerization for different G_s/G_p ratios, starting from near zero at the origin (due to DBP compatibilization) and increasing as polymerization proceeds. The solid circles mark the estimated final positions corresponding to different G_s/G_p ratios and their resulting morphologies.