Supporting Information

1. XRD data of AUC

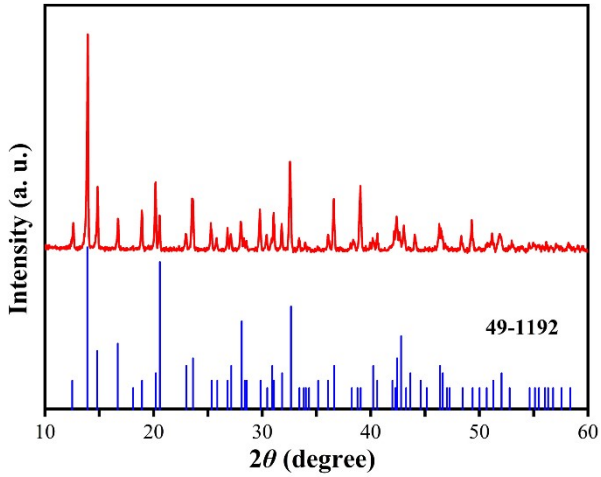

Fig. S1. XRD patterns of the prepared ammonium uranyl carbonate (AUC).

2. Column experiment

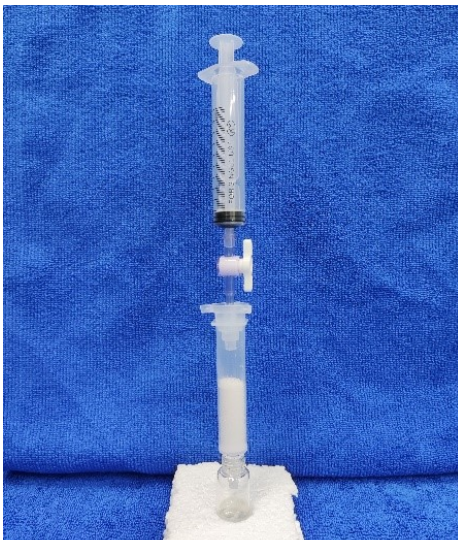

Fig. S2 Photograph of the column experiment.

3. Micelle model with a homogeneous core and a corona with decaying density

The normalized form factor of PS core is expressed as:[1]

$$P_{\text{core}}(q, R_{\text{core}}) = F^2_{\text{core}}(q, R_{\text{core}}) \quad \text{(S1)}$$

$$F_{\text{core}}(q, R_{\text{core}}) = \frac{3[sin(qR_{\text{core}}) - qR_{\text{core}}cos(qR_{\text{core}})]}{(qR_{\text{core}})^3} \quad \text{(S2)}$$

The form factor of the individual PAA chains in the corona was described by a worm-like chain with excluded volume and negligible cross-section as given by Sharp and Bloomfield.[2] It is noted that Pedersen and Schurtenberger[3] replaced the Debye function in the Sharp and Bloomfield expression with one that contains flexible chains and excluded volume effects.

$$P_{\text{chain}}(q, L, b) = S_{\text{exv}}(q, L, b) + C(\frac{b}{L})[\frac{4}{15} + \frac{7}{15u} - (\frac{11}{15} + \frac{7}{15u})\exp(-u)]\frac{b}{L} \quad \text{(S3)}$$
$$u = R_g{}^2 q^2$$

The Kuhn length, *b*, used to describe the stiffness of the PAA chains in the corona, was derived from model fitting. Note that this parameter is not sensitive in the model fitting, because PAA is a flexible polymer and thus has a small Kuhn length (< 1 nm) with several repeating C-C units.

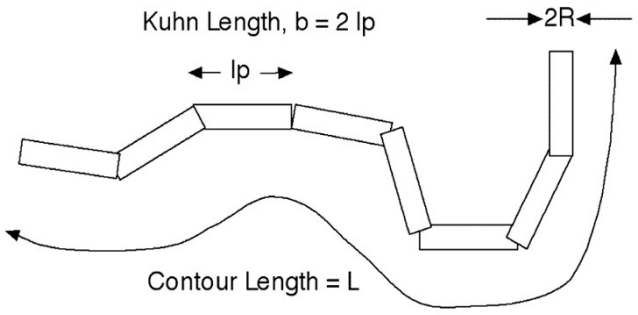

Fig. S3 Worm-like chain model for the individual PAA block in the corona.

The density profile in the PAA corona is decreased as according to:[4]

$$\varphi(r)=\phi_{chain}(\frac{r}{R_{\text{core}}})^{-\alpha} \qquad \text{with} \quad R_{\text{core}}<r<R_{\text{core}}+t$$
$$\alpha=\frac{(D-1)(3v-1)}{2v} \tag{S4}$$

where α is a power exponent that determines the decaying of the density profile in the PAA corona. For stretched PAA chains on PS spheres, the dimension (D) of the curvature of PS core is 3, the Flory exponent (v) is 1, and thus, $\alpha = 2$. As a result, the F_{corona} is expressed as:

$$F_{\text{corona}}(q,t)=\frac{1}{\ln(R_{\text{core}}+t)-\ln(R_{\text{core}})}\int_{R_{\text{core}}}^{R_{\text{core}}+t}\frac{\sin qr}{qr}dr \tag{S5}$$

After mixing with AUC, scattering contrast of the PAA corona was increased due to the complexation of UC species. Since the X-ray scattering length density is proportional to the electron number density, the number of UC species (m) per PS-*b*-PAA micelle can be estimated by:

$$\frac{\Delta\rho_{\text{PAA}}}{\Delta\rho_{\text{PAA-UC}}}=\frac{\text{X}_{\text{PAA}}n^{e}_{\text{PAA}}-N^{e}_{\text{water}}}{\text{X}_{\text{PAA}}n^{e}_{\text{PAA}}-N^{e}_{\text{water}}+mn^{e}_{\text{UC}}/N_{\text{agg}}} \tag{S6}$$

$$N^{e}_{\text{water}}=\frac{\text{X}_{\text{PAA}}V_{\text{PAA}}\rho_{\text{water}}n^{e}_{\text{water}}N_{\text{A}}}{M_{\text{water}}} \tag{S7}$$

where, X_{PAA} (69) is the degree of polymerization of PAA, N_{agg} the aggregation number of the PS-*b*-PAA polymers, $n^{\text{e}}_{\text{PAA}}$ the electron numbers per PAA segment, $n^{\text{e}}_{\text{water}}$ the electron numbers per water molecule, n^{e}_{UC} the electron numbers per $UO_2(CO_3)_2^{2-}$ species, which is the main speciation of UC, V_{PAA} the volume per PAA segment, M_{water} the molar mass of water, N_{A} the Avogadro constant, ρ_{water} the density of water, and m the number of UC species ($UO_2(CO_3)_2^{2-}$) adsorbed by one micelle.

4. Fitted parameters of SAXS data

Table S1. Structural parameter obtained from SAXS data on PS-*b*-PAA-*n*AUC micelles by curve fitting using a micelle model.

Samples	R_0 (nm)	σ (nm)	t (nm)	$\Delta\rho_{core}$ (a. u.)	$\Delta\rho_{chain}$ (a. u.)	b (nm)[a]
PS-*b*-PAA	12.3	3.4	6.7	0.029	0.5	0.85
PS-*b*-PAA-0.5AUC	12.5	3.5	4.8	0.029	0.71	0.85
PS-*b*-PAA-1.5AUC	12.3	2	3.7	0.029	1.04	0.72
PS-*b*-PAA-2.5AUC	12.6	1.5	3.5	0.029	1.74	0.85
PS-*b*-PAA-3.5AUC	12.5	1.5	3.2	0.029	2.10	0.75
PS-*b*- PAA-5AUC	12.7	1.8	2.8	0.029	2.49	0.72

[a] R_0 is the mean radius; σ is the standard deviation; t is the thickness of the corona; $\Delta\rho_{core}$ and $\Delta\rho_{chain}$ are the excess SLD of the PS and PAA blocks to the surrounding solution; b is the Kuhn length of the chain.

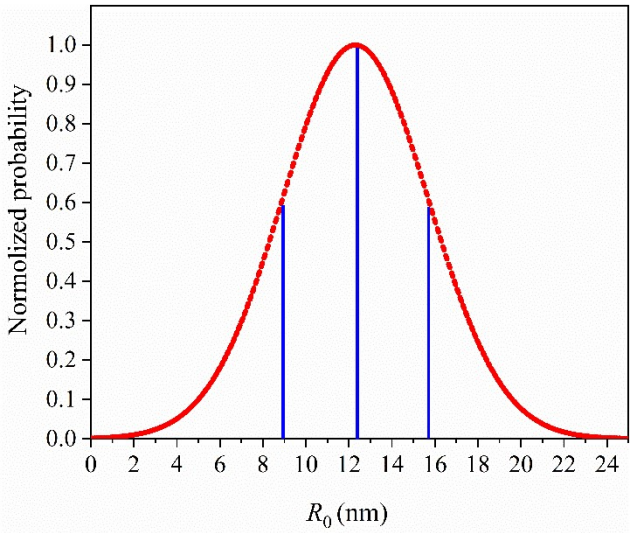

Fig. S4 Normal size distribution of the PS cores (R_0 = 12.3 nm, σ = 3.4 nm).

5. TEM image of PS-*b*-PAA-*n*AUC

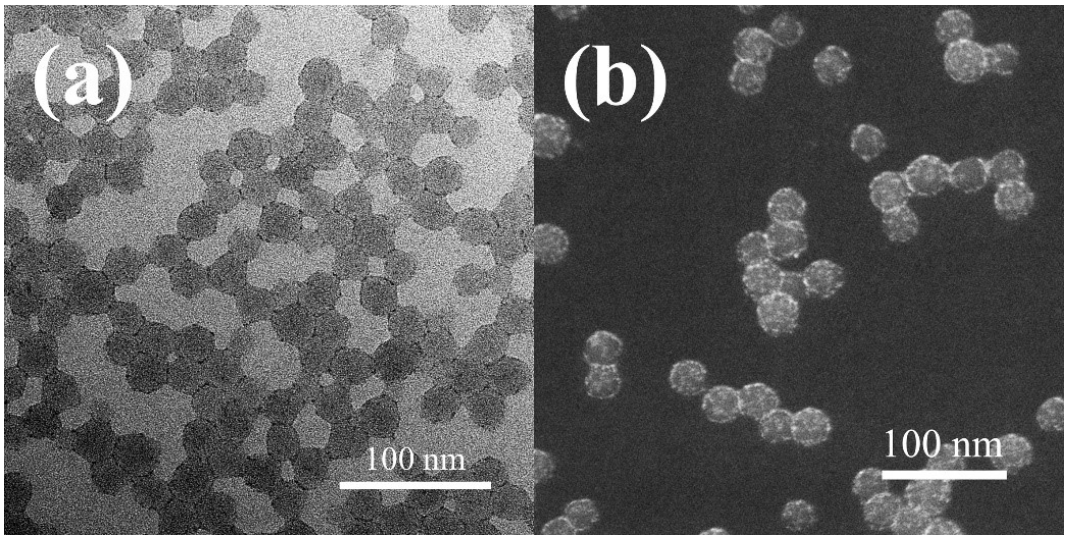

Fig. S5 TEM bright-field image (a) and HAADF-STEM image (b) obtained from PS-*b*-PAA-5AUC.

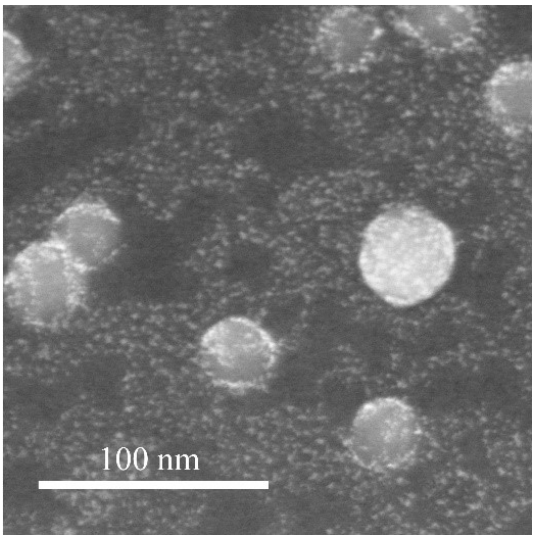

Fig. S6 HAADF-STEM image of PS-*b*-PAA-2.5AUC effluent through the quartz sand.

6. Hydrodynamic diameters and ζ potentials of colloidal FA-*n*AUC and PS-*b*-PAA-*m*NaCl

Table S2. Hydrodynamic diameter and zeta potential of PS-*b*-PAA-*m*NaCl dispersions at 25°C.

Samples	NaCl concentration (mg/mL)	pH	Hydrodynamic diameter (nm)	Zeta potential (mV)
PS-*b*-PAA	0	6.5	102(1)	-37(2)
PS-*b*-PAA-0.6NaCl	0.6	6.5	100(1)	-41(1)
PS-*b*-PAA-1.7NaCl	1.7	6.5	98(3)	-38(4)
PS-*b*-PAA-2.8NaCl	2.8	6.5	99(4)	-44(3)
PS-*b*-PAA-3.9NaCl	3.9	6.6	103(1)	-40(1)
PS-*b*-PAA-5.6NaCl	5.6	6.6	102(2)	-43(4)

Table S3. Hydrodynamic diameter and zeta potential of FA-*n*AUC dispersions at 25°C.

Samples	AUC concentration (mg/mL)	pH	Hydrodynamic diameter (nm)	Zeta potential (mV)
FA	0	7.1	146(4)	-20(2)
FA-0.5AUC	0.5	7.4	142(1)	-17(1)
FA-1.5AUC	1.5	7.4	135(1)	-23(2)
FA-2.5AUC	2.5	7.7	140(2)	-25(3)
FA-3.5AUC	3.5	7.8	138(1)	-47(1)
FA-5AUC	5	8	135(2)	-28(3)

7. SAXS data and TEM image of FA-*n*AUC

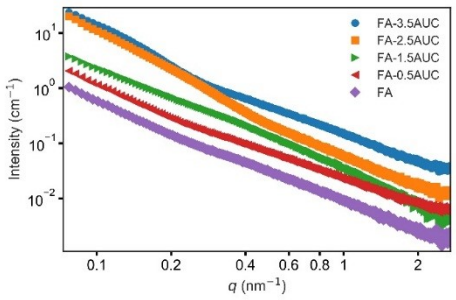

Fig. S7 SAXS curves obtained from the FA-*n*AUC dispersions. The curves are shifted vertically by factors of 2, 4, 5 and 10 for clarity.

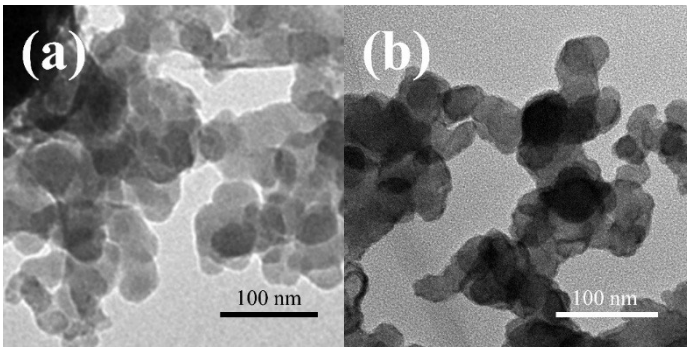

Fig. S8 TEM images obtained from the FA (a) FA-2.5AUC dispersions (b).

8. The multimodal size distributions of hydrodynamic sizes of micellar dispersions

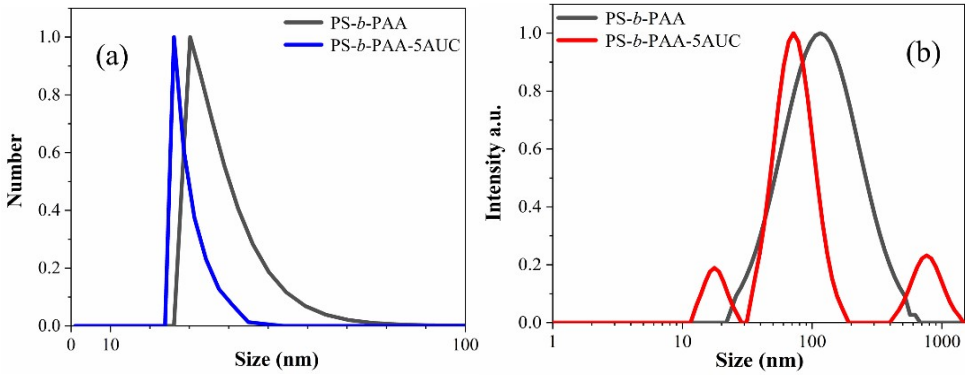

Fig. S9 The number-weighted (a) and intensity-weighted (b) multimodal size distributions of hydrodynamic size. All the samples were filtered with 0.45 um polyethersulfone syringe filters before the characterization.

References:

1. L. Rayleigh, Form factor of a homogenous sphere, *Proc. R. Soc. London, Ser. A*, 1911, **84**, 25-38.
2. P. Sharp and V. A. Bloomfield, Light scattering from wormlike chains with excluded volume effects, *Biopolymers: Original Research on Biomolecules*, 1968, **6**, 1201-1211.
3. J. S. Pedersen and M. C. Gerstenberg, Scattering form factor of block copolymer micelles, *Macromolecules*, 1996, **29**, 1363-1365.
4. C. Wijmans and E. B. Zhulina, Polymer brushes at curved surfaces, *Macromolecules*, 1993, **26**, 7214-7224.