Support Information for

Supramolecular aggregates from polyacrylates and Gd(III)-containing

cationic surfactant as high-relaxivity MRI contrast agents

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Figure S1. GPC curves of PtBA of different molecular weights



Figure S2. ¹H NMR spectra of PtBA (a) in CDCl₃ and PAA (b) in DMSO-d₆.



Figure S3. DLS spectra of the PAAS-MS mixtures, (A) P3-MS, (B) P4-MS, (C) P5-MS.

z value	0.2	0.3	0.4	0.5	0.6	0.7	0.8	0.9	1.0	1.1
$R_{h}(nm)$	169.3	182.0	176.3	170.1	207.9	182.3	190.5	197.6	207.9	178.6
PDI	0.162	0.236	0.060	0.069	0.080	0.050	0.058	0.065	0.091	0.054

Table S1. $R_{\rm h}$ and PDI values of P1-MS PSCs in Figure 2B

z value	0.2	0.3	0.4	0.5	0.6	0.7
$R_{h}(nm)$	292.8	273.2	267.3	259.3	236.6	224.6
PDI	0.013	0.045	0.036	0.053	0.093	0.114

Table S2. $R_{\rm h}$ and PDI values of P2-MS PSCs in Figure 2C

Table S3. $R_{\rm h}$ and PDI values of P3-MS PSCs in Figure S3A

z value	0.3	0.4	0.5	0.6	0.7
$R_{h}(nm)$	252.7	229.8	208.5	215.7	175.0
PDI	0.066	0.089	0.041	0.154	0.104

Table S4. R_h and PDI values of P4-MS PSCs in Figure S3B

z value	0.2	0.3	0.4	0.5	0.6
$R_{h}(nm)$	238.0	226.4	190.8	181.7	176.8
PDI	0.162	0.141	0.092	0.124	0.126

Table S5. $R_{\rm h}$ and PDI values of P5-MS PSCs in Figure S4C

z value	0.2	0.3	0.4	0.5
$R_{h}(nm)$	184.8	134.5	113.1	102.9
PDI	0.114	0.146	0.142	0.133



Figure S4. TEM images of PAAS-surfactant mixtures at different charge ratio, (A) P3-0.3 (B) P3-0.5 (C) P4-0.3 (D) P4-0.5 (E) P5-0.3 (F) P5-0.5.



Figure S5. SEM element mapping of P1-MS (A), P2-MS (B), P3-MS (C), P4-MS (D) and P5-MS (E) at z = 0.3.



Figure S6. Relative cell viability (%) of Hela cell after treated by pure metallosurfactant and PAAS.