

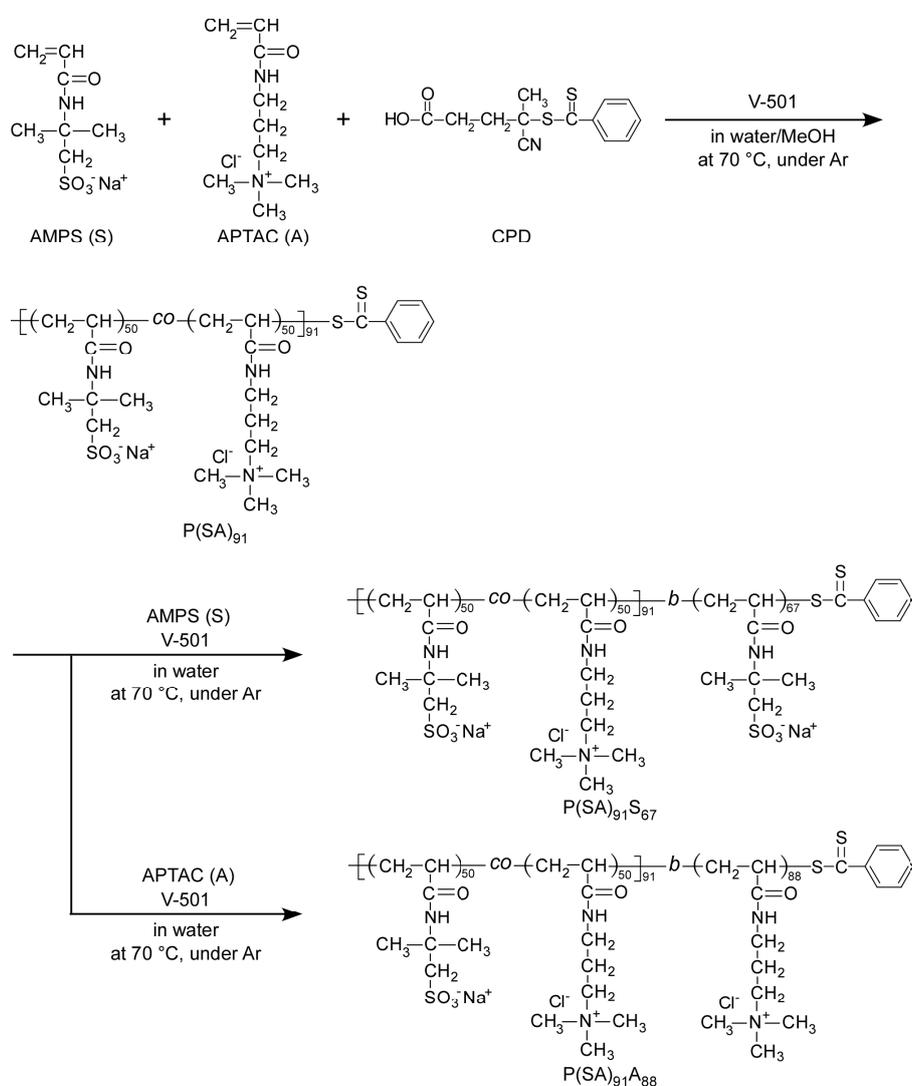
Electronic supplementary information (ESI)

# Preparation of Water-soluble Polyion Complex (PIC) Micelles Covered with Amphoteric Random Copolymer Shells with Pendant Sulfonate and Quaternary Amino Groups

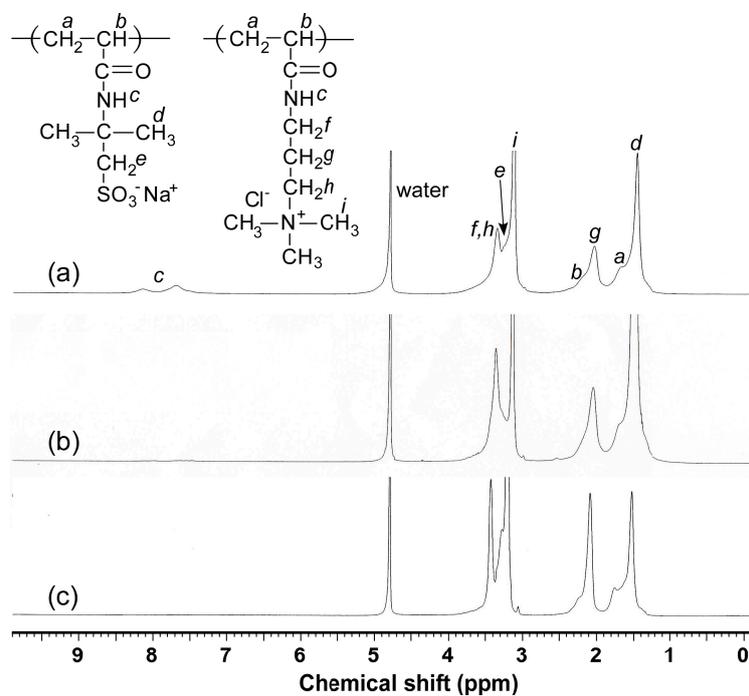
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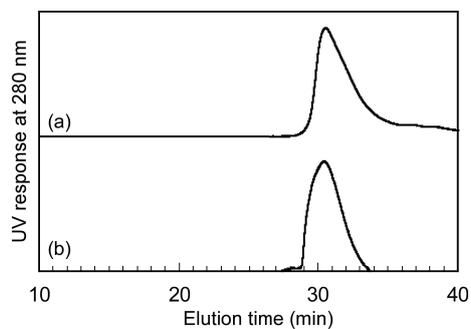
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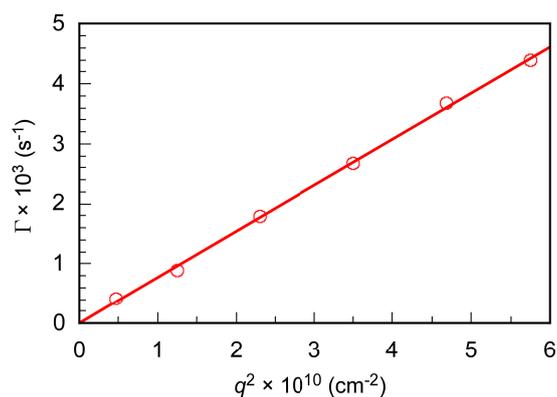
**Figure S1.** Synthesis routes of  $\text{P}(\text{SA})_{91}\text{S}_{67}$  and  $\text{P}(\text{SA})_{91}\text{A}_{88}$ .



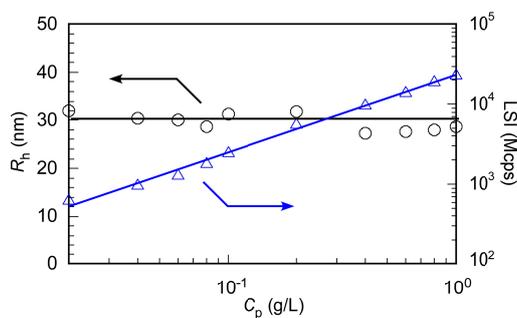
**Figure S2.**  $^1\text{H}$  NMR spectra for (a) P(SA)<sub>91</sub>, (b) P(SA)<sub>91</sub>S<sub>67</sub>, and (c) P(SA)<sub>91</sub>A<sub>88</sub> in  $\text{D}_2\text{O}$ .



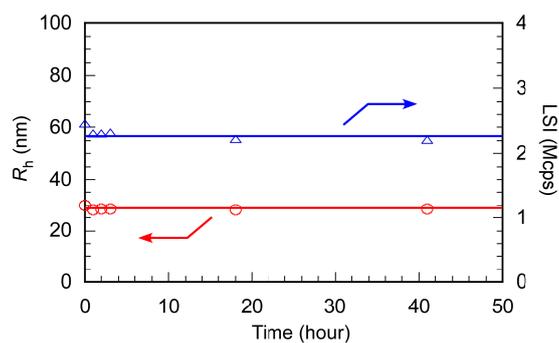
**Figure S3.** GPC elution curves for (a) P(SA)<sub>91</sub>S<sub>67</sub> and (b) P(SA)<sub>91</sub>A<sub>88</sub> using an acetic acid (0.5 M) solution containing sodium sulfate (0.3 M) as an eluent.



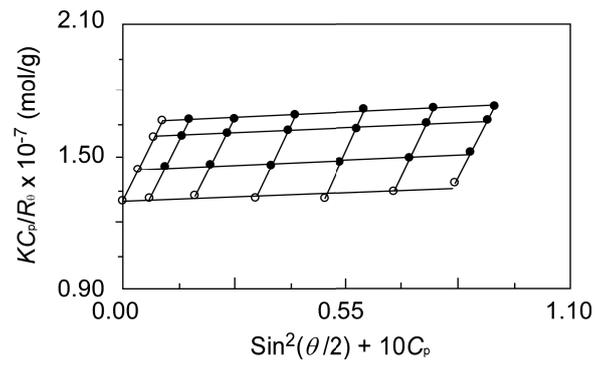
**Figure S4.** Relationship between the relaxation rate ( $\Gamma$ ) and the square of the magnitude of the scattering intensity vector ( $q^2$ ) for PIC micelles at  $C_p = 1 \text{ g/L}$  in  $0.1 \text{ M}$  aqueous NaCl at  $25 \text{ }^\circ\text{C}$ .



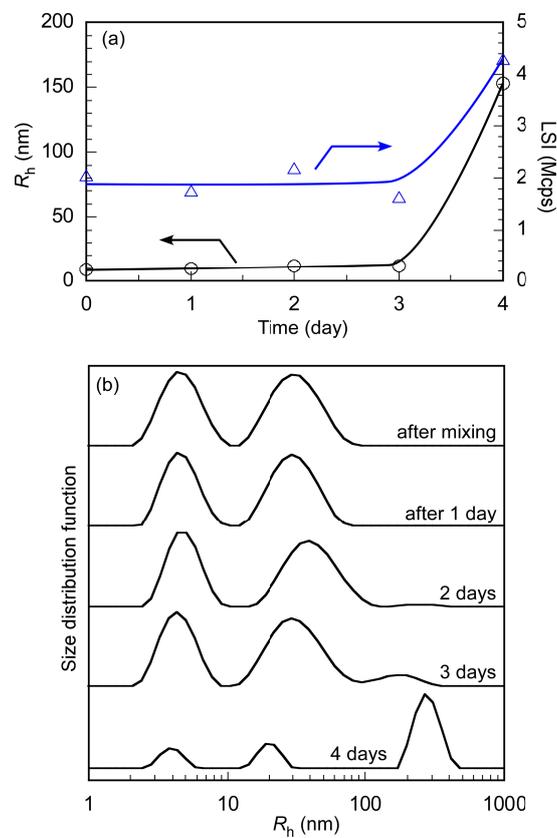
**Figure S5.** Hydrodynamic radius ( $R_h$ ,  $\circ$ ) and light scattering intensity (LSI,  $\Delta$ ) of PIC micelles as a function of polymer concentration ( $C_p$ ) in  $0.1 \text{ M}$  aqueous NaCl.



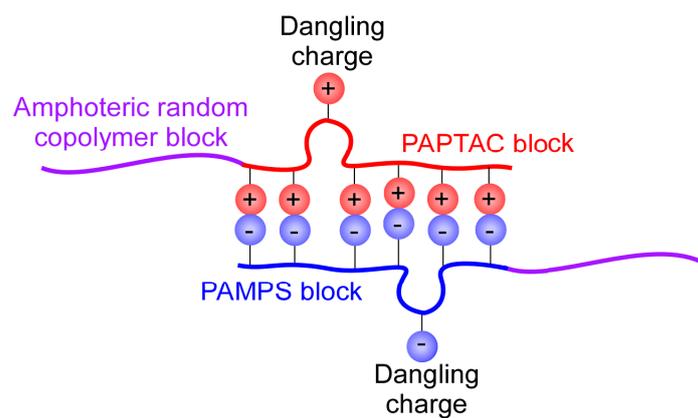
**Figure 6.** Relationship between  $R_h$  ( $\circ$ ) and light scattering intensity (LSI,  $\Delta$ ) as a function of time for PIC micelle with  $f^* = 0.5$  at  $C_p = 1.0 \text{ g/L}$  in  $0.1 \text{ M}$  NaCl aqueous solution.



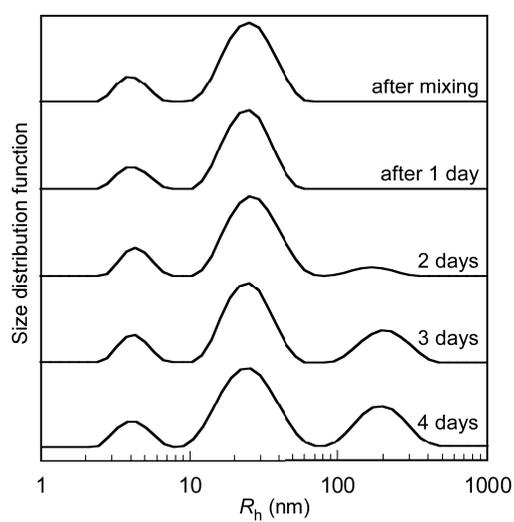
**Figure S7.** A typical Zimm plot for PIC micelles in 0.1 M aqueous NaCl at 25 °C.



**Figure S8.** (a) Relationship between  $R_h$  ( $\circ$ ) and light scattering intensity (LSI,  $\Delta$ ) as a function of time, and (b)  $R_h$  distributions for a mixture of PIC micelles/BSA at  $C_p = 0.1$  g/L and  $[BSA] = 5.0$  g/L in PBS at 25 °C.



**Figure S9.** Conceptual illustration of dangling charge groups in the unit PIC of  $P(SA)_{91}S_{67}/P(SA)_{91}A_{88}$ .



**Figure S10.**  $R_h$  distributions for mixture of PIC micelle/FBS at  $C_p = 0.1$  g/L and  $[FBS] = 40$  g/L in PBS at 25 °C.