Supplementary Information

Spontaneous Unilamellar Polymer Vesicle in Aqueous Solution

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- Sum model

When the interparticle interference is negligible, the scattering intensity of a sum of form factors can be described as

$$I(Q) = n_a P_a(Q) + n_b P_b(Q) + BKG$$

where n_a and $P_a(Q)$ is the number density and form factor of the particle *a*, n_b and $P_b(Q)$ is the number density and form factor of the particle *b*, and BKG is an incoherent background.

- SANS Intensities of P85-5mS mixtures at varying temperature

When the temperature increased, SANS intensities of P85-5mS mixtures with the 5mS concentration of 0.1 %, 0.125 %, and 0.175 % also showed a variety of scattering pattern, indicating that a phase transition occurred (Figure S1). For all the temperature ranges, the scattering intensities were successfully reproduced by a single form factor, core-shell sphere, core-shell cylinder or vesicle, or a sum of core-shell sphere and core-shell cylinder form factor or core-shell cylinder and vesicle form factor. The fitted core radius, shell thickness,

and length for sphere, cylinder and vesicle are shown in Figure S2. It should be noted that the phase transition temperature of P85-5mS mixtures (sphere to cylinder or cylinder to vesicle) is decreased with increasing the 5mS concentration.

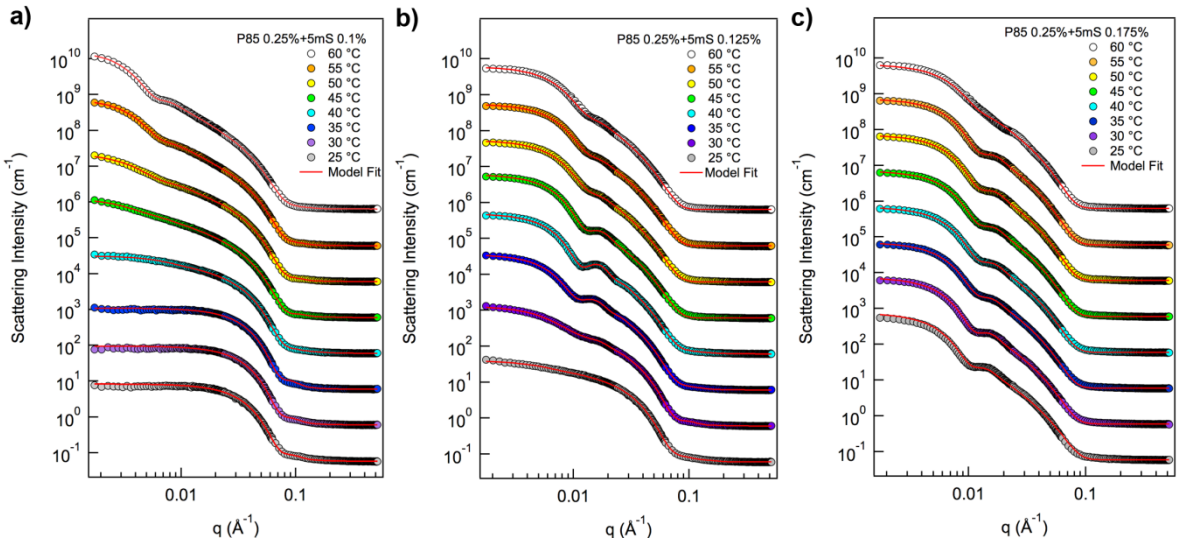

Figure S1. SANS intensities of P85-5mS mixtures with 5mS concentration of a) 0.1 %, b) 0.125 %, and c) 0.175 %.

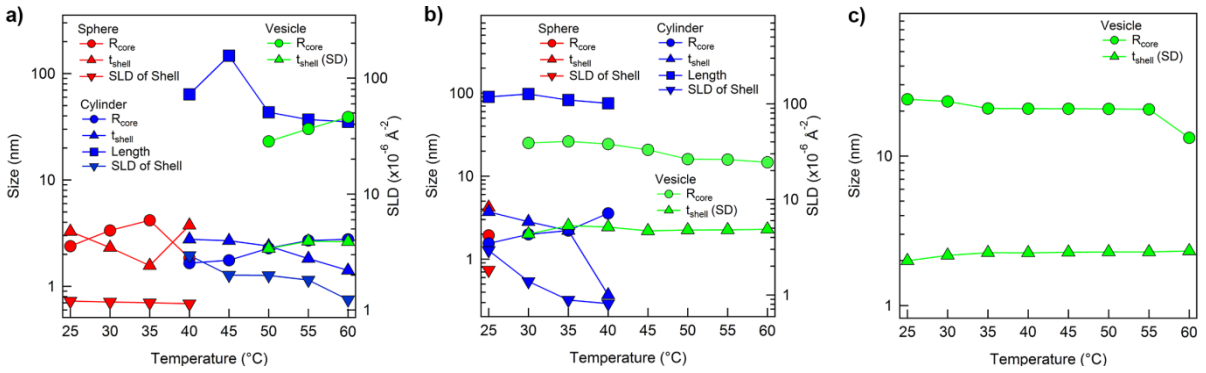

Figure S2. Fitting parameters obtained from SANS analyses of P85-5mS mixtures with 5mS concentration of a) 0.1 %, b) 0.125 %, and c) 0.175 %.