

# Bundling Process of Citrulline Polypeptides upon UCST-type Phase Separation

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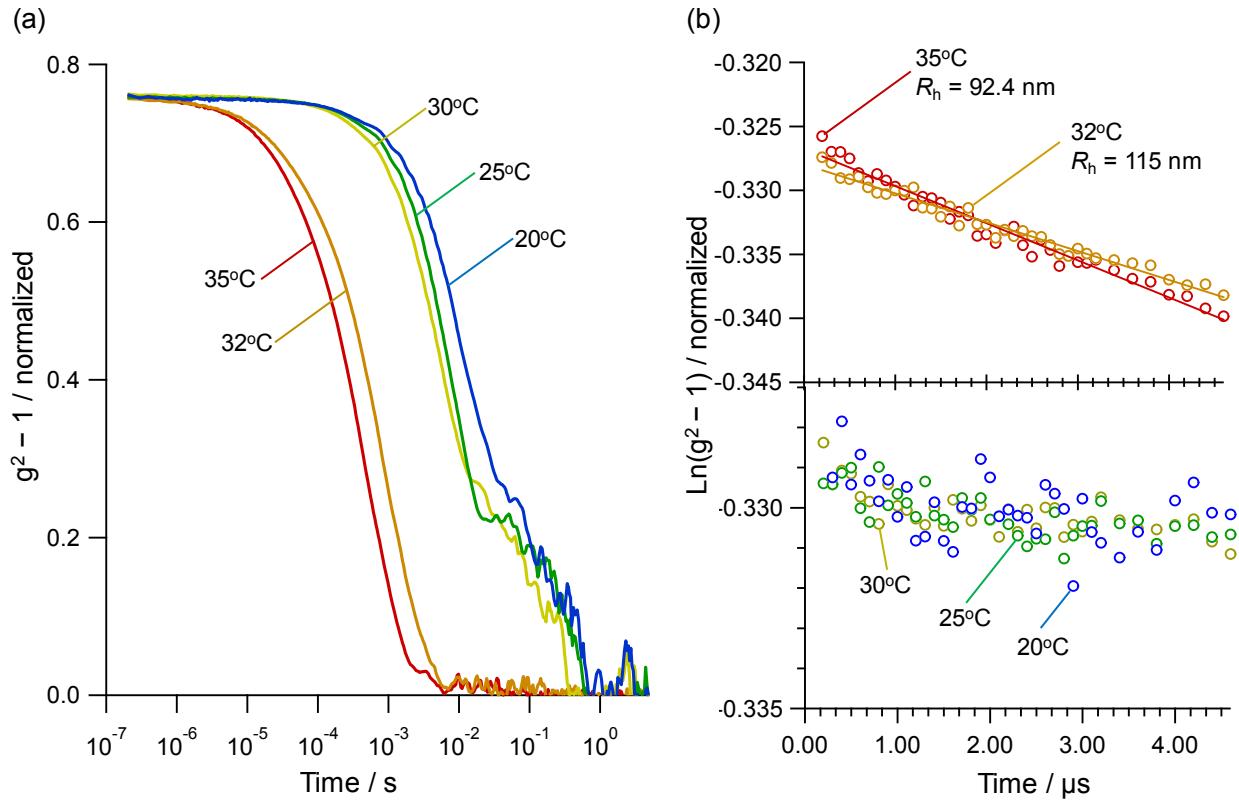
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Figure S1



**Figure S1.** Autocorrelation function [(a)  $g^2 - 1$  or (b)  $\ln(g^2 - 1)$ ] of  $P_L$ OC in 10 mM HEPES-NaOH (pH 7.5) containing 150 mM NaCl at different temperatures.

Figure S2

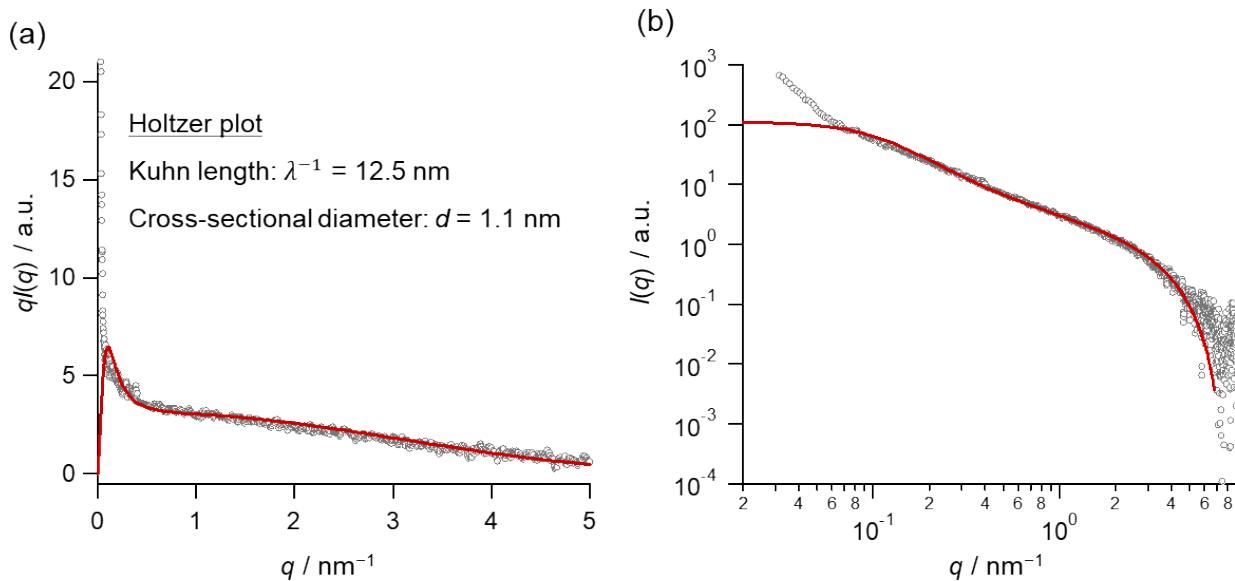
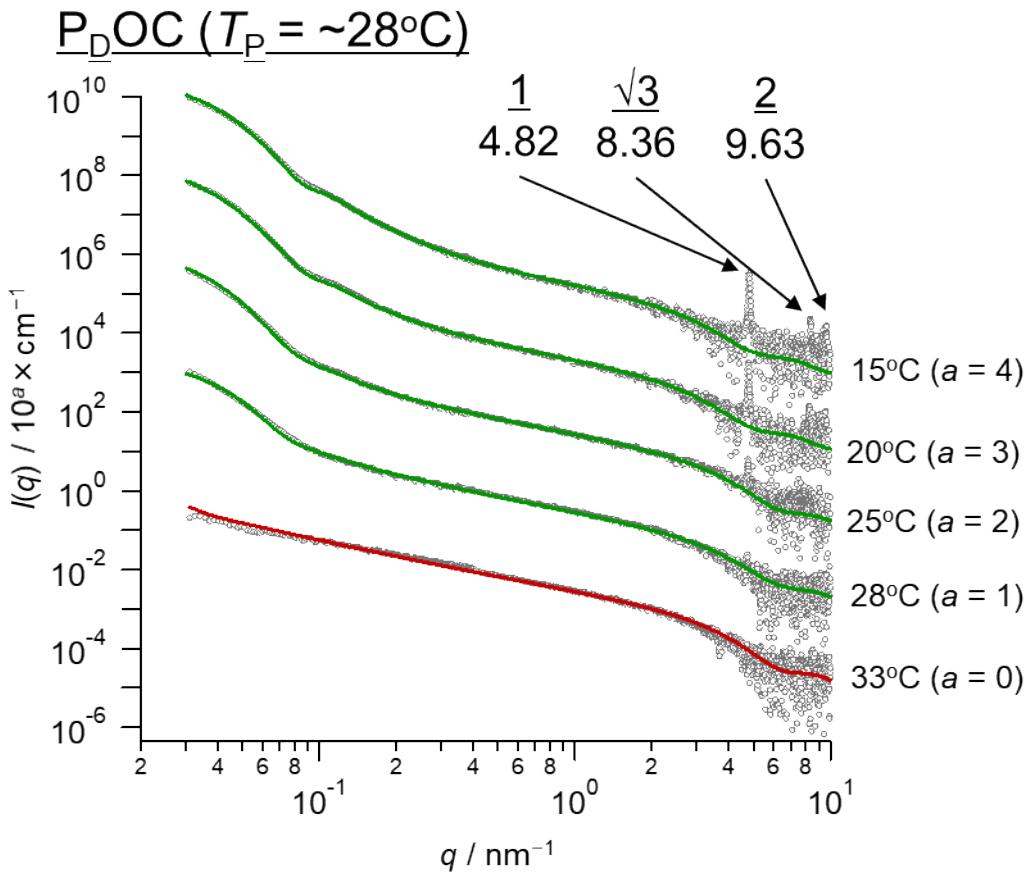


Figure S2. (a) The Holtzer plot for the SAXS profile of P<sub>L</sub>OC at 35°C and comparison with the Norisuye and Nakamura theory<sup>1</sup> with the indicated Kuhn length ( $\lambda^{-1}$ ) values and the cross-sectional diameter ( $d$ ). (b) Comparing SAXS profile of P<sub>L</sub>OC at 35°C with the calculated fitting curve.

## References

1. Nakamura, Y.; Norisuye, T., Scattering function for wormlike chains with finite thickness. *Journal of Polymer Science Part B: Polymer Physics* **2004**, *42*(8), 1398-1407.

Figure S3



**Figure S3.** SAXS profiles of 1.0 mg/mL  $P_D$ OC ( $M_w = 8.3 \times 10^4$  g/mol, citrulline content = 91.0%,  $T_p = 28^\circ\text{C}$ ) in 10 mM HEPES-NaOH (pH 7.5) containing 150 mM NaCl at different temperatures. The red and green curves were calculated using the modified UF model described in eq. (1) or (2), respectively.

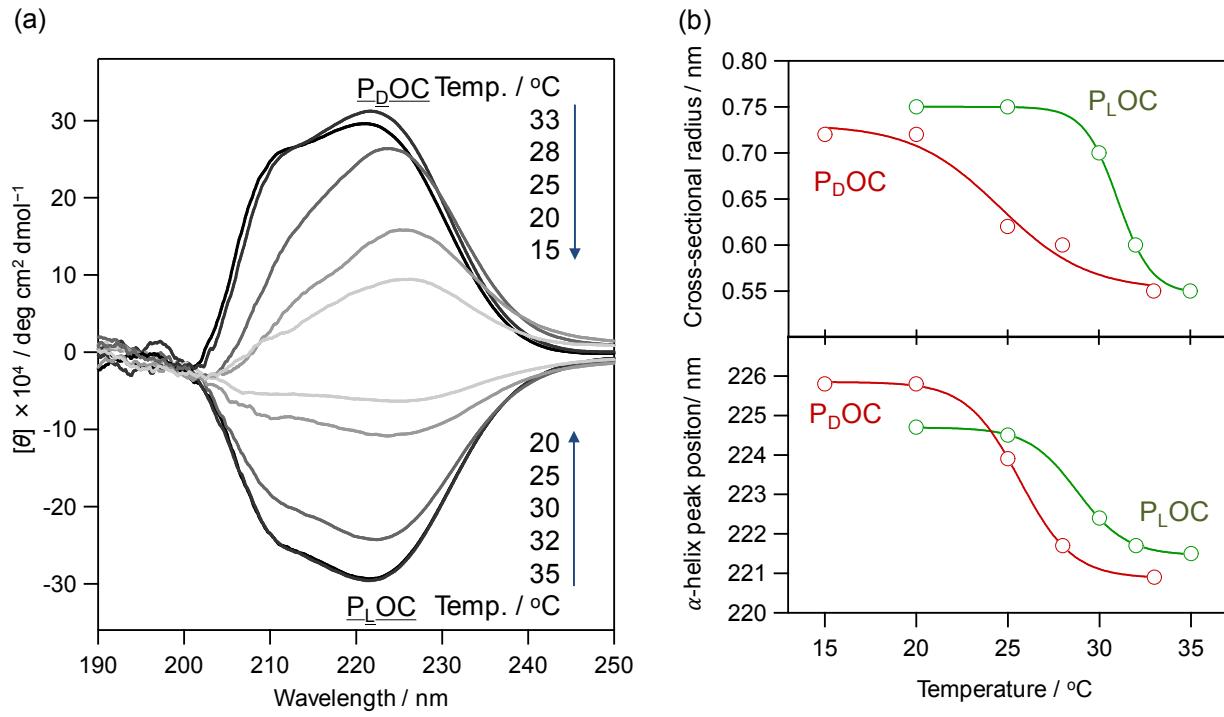
**Table S1**

**Table S1.** Fitting Parameters for the SAXS Profiles of P<sub>D</sub>OC in 10 mM HEPES-NaOH (pH 7.5) Containing 150 mM NaCl at Different Temperatures Using the Modified UF Models

Temp. [°C]	$R_c$ [nm]	$R_g$ [nm]	$D_M$	$R_s$ [nm]	$\sigma^*/R_s$
33	0.55	100	1.7	-	-
28	0.60	68	1.7	40	0.38
25	0.62	68	1.7	42	0.29
20	0.72	68	1.7	42	0.24
15	0.72	68	1.7	45	0.22

\*We assumed that the size distribution can be described by Gaussian function and  $\sigma$  is the standard deviation of the spherical aggregation.

Figure S4



**Figure S4.** (a) CD spectra of  $P_{\text{L}}\text{OC}$  ( $M_w = 1.02 \times 10^5$  g/mol, citrulline content = 92.5%,  $T_p = 30^\circ\text{C}$ ) and  $P_{\text{D}}\text{OC}$  ( $M_w = 8.3 \times 10^4$  g/mol, citrulline content = 91.0%,  $T_p = 28^\circ\text{C}$ ) in 10 mM HEPES-NaOH (pH 7.5) containing 150 mM NaCl at different temperatures. (b) Temperature dependence of the cross-sectional radius of  $\alpha$ -helix determined by SAXS analysis and peak top position relating to the  $\alpha$ -helix structure.