**Supporting Information** 

## Control of Crystal Size Distribution in Continuous Cooling Crystallization Using Non-isothermal Taylor Vortex

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**Fig. S1** Simulated temperature profiles of bulks solution and heating-cooling surface with different directions of medium flow: (a) Direction-I (Counter-Counter), (b) Direction-II (Co-Count), (c) Direction-III (Co-Co), (d) Direction-IV (Count-Co).



Fig. S2 Simulated temperature profiles of bulk solution and heating-cooling surface with varying temperature differences ( $\Delta$ T) between inner and outer cylinders: (a) 0 °C, (b) 3.6 °C, (c) 8.9 °C, (d) 16.4 °C, (e) 18.1 °C. (f) Deviation of simulated bulk solution temperature along the axial direction of the crystallizer with varying  $\Delta$ T.



Fig. S3 Optical microscope images of crystals at different axial ports of the non-isothermal CT crystallizer with varying  $\Delta T$ : (a) 0 °C, (b) 3.6 °C, (c) 8.9 °C, (d) 16.4 °C, (e) 18.1 °C (1) Port-I, (2) Port-II, (3) Port-III, (4) Port-IV). Crystallization conditions in non-isothermal Mode-I are constant at  $T_b = 28^{\circ}$ C, mean residence time of 5 min, inner cylinder rotation speed of 500 rpm, feeding concentration of 900 g/L, and medium flow direction of Direction-I.



Fig. S4 Optical microscope images of crystals obtained with varying rotation speeds in the non-isothermal CT crystallizer: (a) 200 rpm, (b) 500 rpm, (c) 700 rpm, (d) 900 rpm (1) Port-I, 2) Port-II, (3) Port-III, (4) Port-IV). Conditions in non-isothermal Mode-I:  $T_b = 28 \text{ °C}$ ,  $\Delta T = 16.4 \pm 0.2 \text{ °C}$ , mean residence time of 5 min, feeding concentration of 900 g/L, and medium flow direction of Direction-I.



**Fig. S5** Optical microscope images of crystals obtained with rotation speeds in the isothermal CT crystallizer: (a) 200 rpm, (b) 500 rpm, (c) 700 rpm, (d) 900 rpm (1) Port-I, (2) Port-II, (3) Port-III, (4) Port-IV). Conditions:  $T_b = 28$  °C, mean residence time of 5 min, feeding concentration of 900 g/L, and medium flow direction of Direction-I.



**Fig. S6** (a) Simulated temperature profiles of bulk solution in non-isothermal (solid line) and isothermal (dashed line) CT crystallizer. (b) Heat transfer coefficients at inner and outer cylinder surfaces with varying rotation speeds.



**Fig. S7** Optical microscope images of crystals obtained with different bulk solution temperatures in the isothermal CT crystallizer: (a)  $T_b = 20^{\circ}$ C, (b) 24°C, (c) 28°C, (d) 32 °C. (e) Supersaturation ratio for initial nucleation at Port-I with increasing bulk solution temperature. Conditions:  $T_b = 28^{\circ}$ C, mean residence time of 5 min, rotation speed of inner cylinder of 500 rpm, feeding concentration of 900 g/L, and medium flow direction of Direction-I.



Fig. S8 Heat transfer coefficients at the inner and outer cylinder surfaces with varying mean residence times in the CT crystallizer.



**Fig. S9** Optical microscope images of crystals obtained at different mean residence times along axial ports in non-isothermal CT crystallizer: (a) 2.5 min, (b) 5 min, (c) 10 min, (d) 15 min, (1) Port-I, (2) Port-II, (3) Port-III, (4) Port-IV). Conditions:  $T_b = 28^{\circ}C$ ,  $\Delta T = 16.4\pm0.2^{\circ}C$ , rotation speed of inner cylinder of 500 rpm, feeding concentration of 900 g/L, and medium flow direction of Direction-I. And all the yields at different mean residence times are the same.



**Fig. S10** Optical microscope images of crystals obtained at different mean residence times along axial ports in isothermal CT crystallizer: (a) 2.5 min, (b) 5 min, (c) 10 min, (d) 15 min, (1) Port-I, (2) Port-II, (3) Port-III, (4) Port-IV). Conditions:  $T_b = 28$  °C, rotation speed of inner cylinder of 500 rpm, feeding concentration of 900 g/L, and medium flow direction of Direction-I.