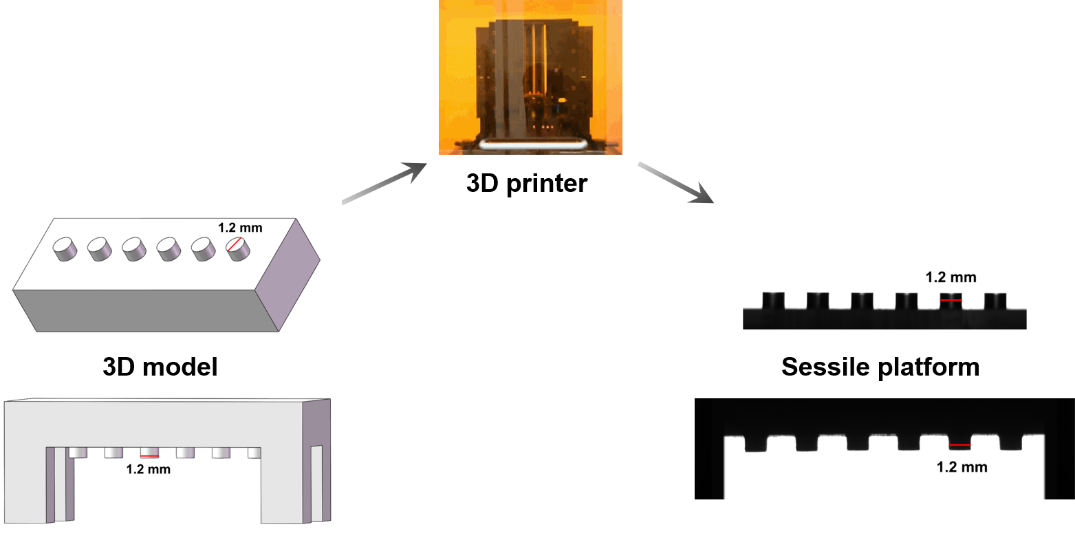
**Supplementary Information**

**Precise crystal regulation and harvest by constructing confined flexible droplet crystallizer on 3D-printed sessile platform**

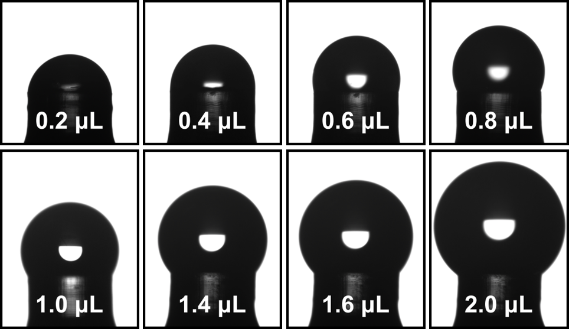
Zhijie Yuan1, Xiaobin Jiang1\*, Yuchao Niu1, Mengyuan Wu1, Yingshuang Meng1, Zhengtao Li1, Xuehua Ruan2, Xiangcun Li1, Wu Xiao1, Xuemei Wu1 and Gaohong He1,2\*

1 State Key Laboratory of Fine Chemicals, School of Chemical Engineering, Dalian University of Technology, Dalian, Liaoning, 116024, China

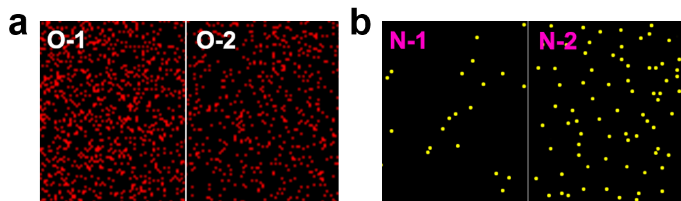
2 School of Chemical Engineering at Panjin, Dalian University of Technology, Panjin 124221, China



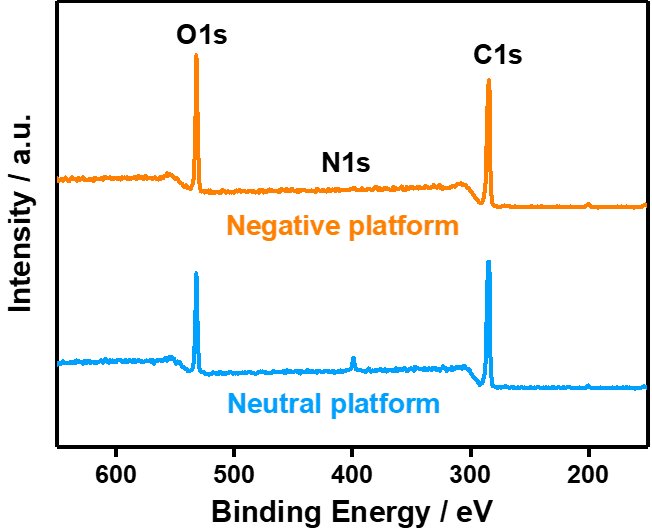
**Supplementary Figure 1.** Preparation of micro-chip with numerous of matrix-type and regular sessile platforms by high precise 3D printer.



**Supplementary Figure 2.** The construction of confined flexible droplet crystallizer on sessile platform.



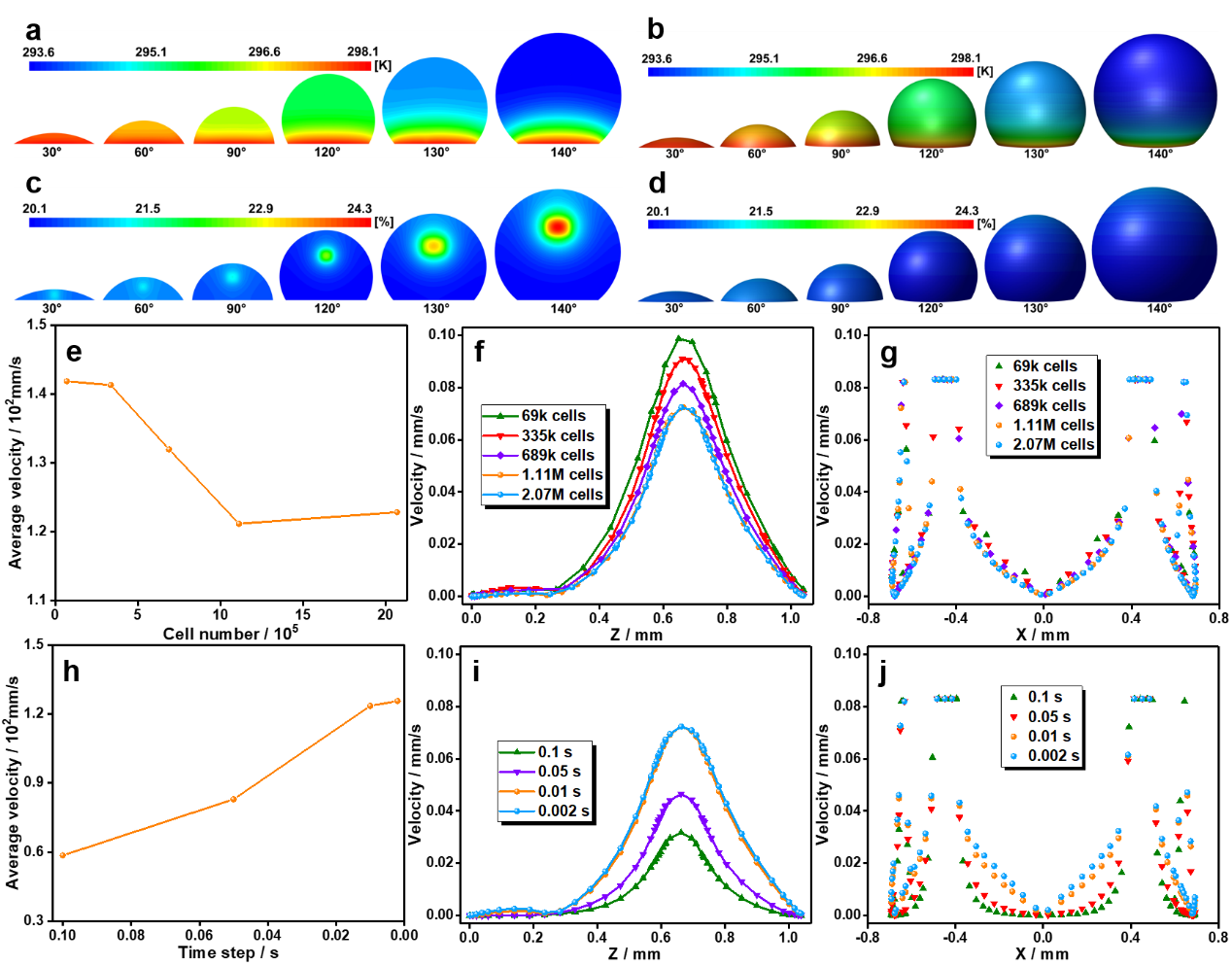
**Supplementary Figure 3.** The O elemental (**a**) and N elemental (**b**) maps of negative (O-1 and N-1) and neutral (O-2 and N-2) sessile platform surface.



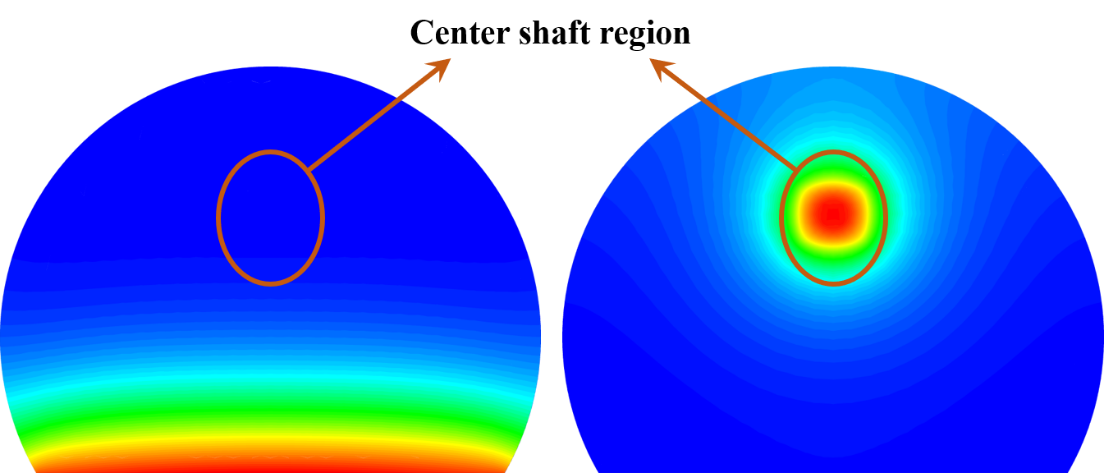
**Supplementary Figure 4.** The XPS spectra for C 1s, N 1s, and O 1s of negative and neutral sessile platform surfaces.



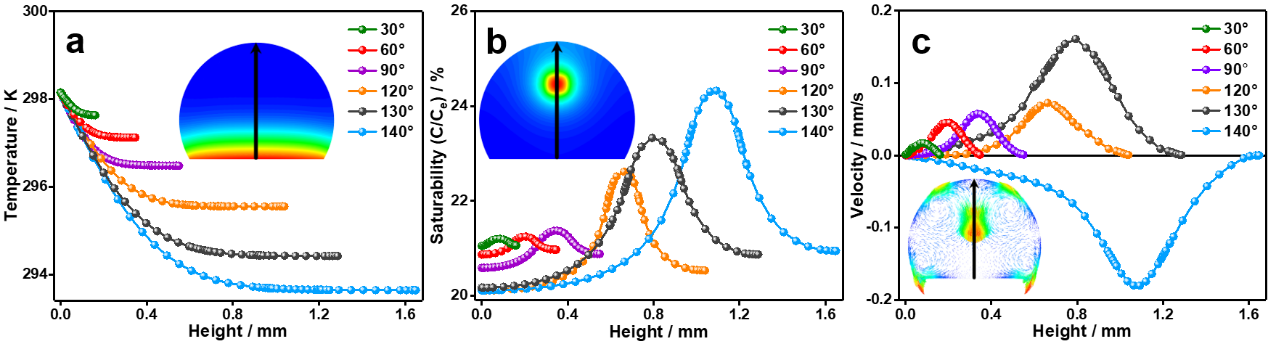
**Supplementary Figure 5.** The trajectories of particles inside CFDCs with initial contact angles of 90° (**a**), 120° (**b**), and 140° (**c**).



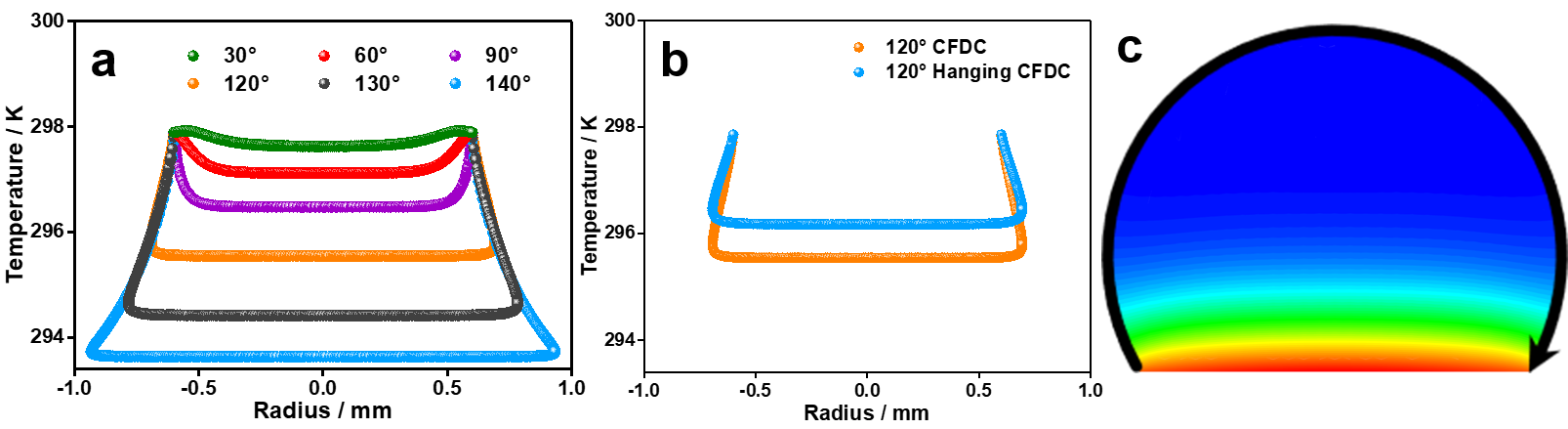
**Supplementary Figure 6.** The 2D (**a**) and 3D (**b**) temperature and the 2D (**c**) and 3D (**d**) concentration contours of CFDCs with initial contact angles of 30°, 60°, 90°, 120°, 130°, and 140°. The independence verifications of mesh (**e-g**) and step (**h-j**).



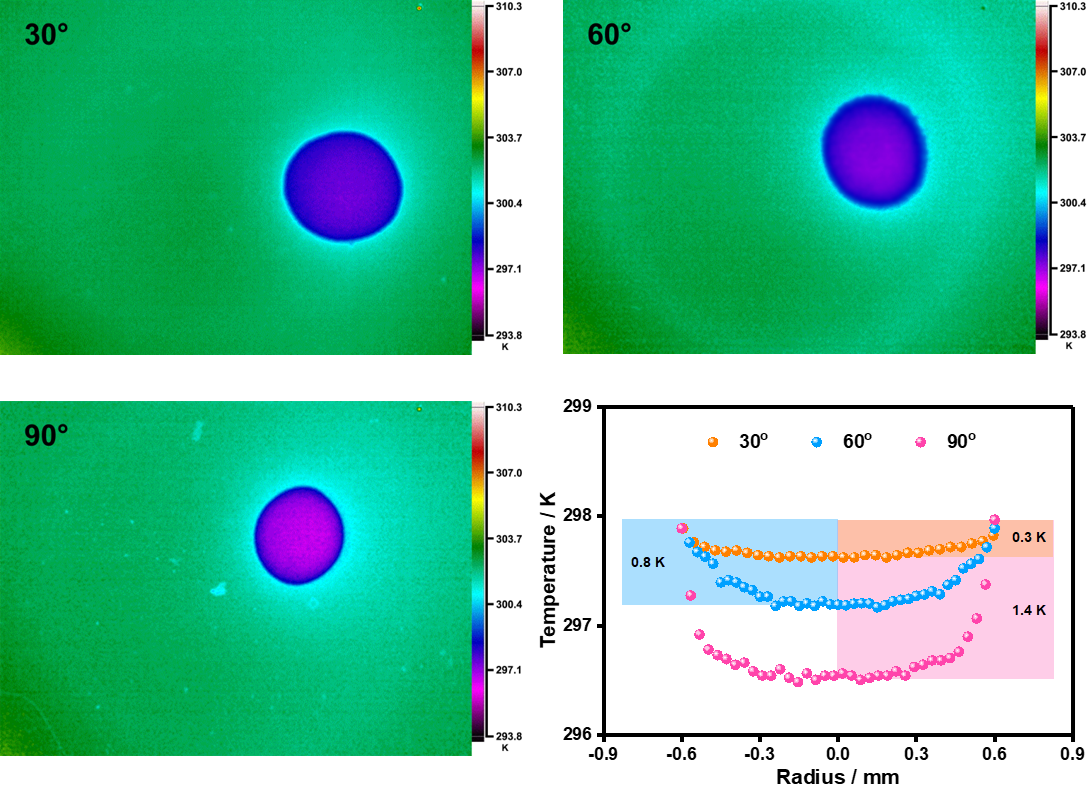
**Supplementary Figure 7.** Illustration of center shaft region with the lowest temperature and highest concentration inside CFDC.



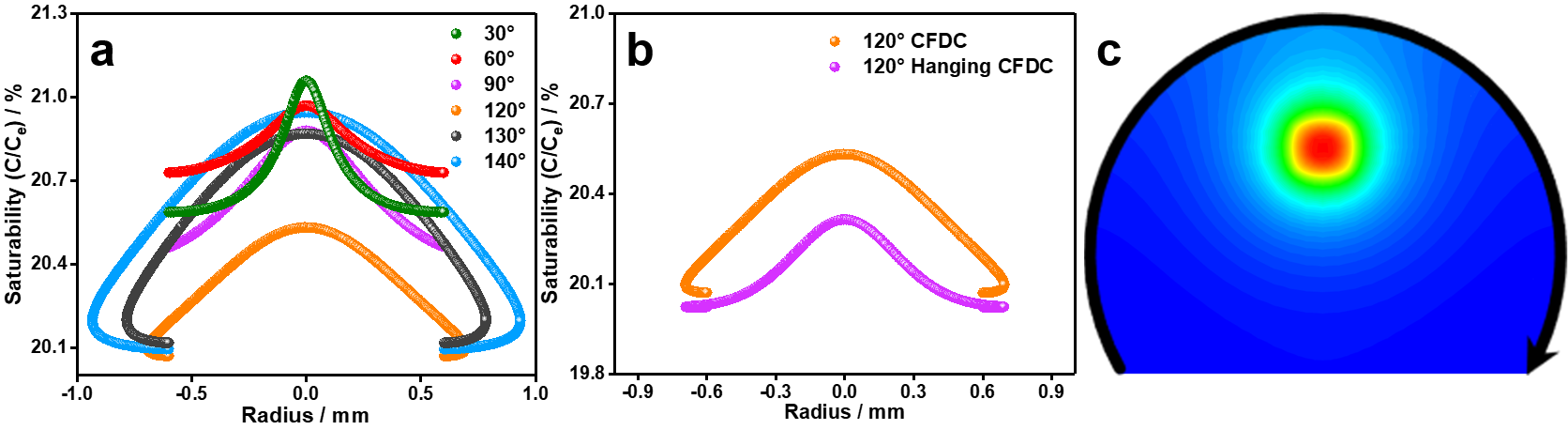
**Supplementary Figure 8.** The temperature (**a**), concentration (**b**), and velocity (**c**) distributions along the axes of CFDCs with initial contact angles of 30°, 60°, 90°, 120°, 130°, and 140°.



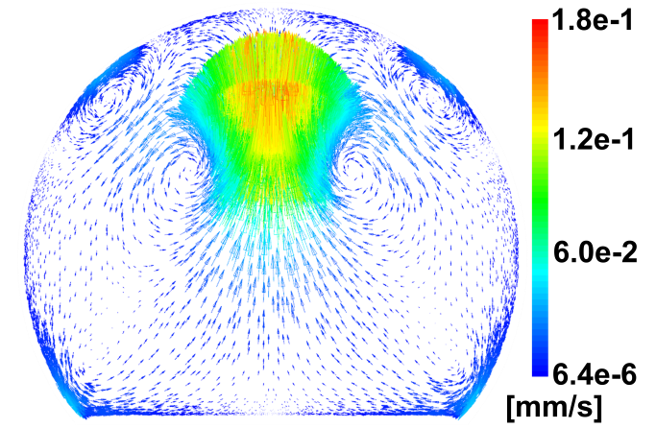
**Supplementary Figure 9.** The temperature distributions along the gas-liquid interface of CFDCs obtained by CFD simulations with initial contact angles of 30°, 60°, 90°, 120°, 130°, and 140° (**a**), the sitting and hanging 120° CFDCs (**b**), and the illustration of temperature distribution along the gas-liquid interface (**c**).



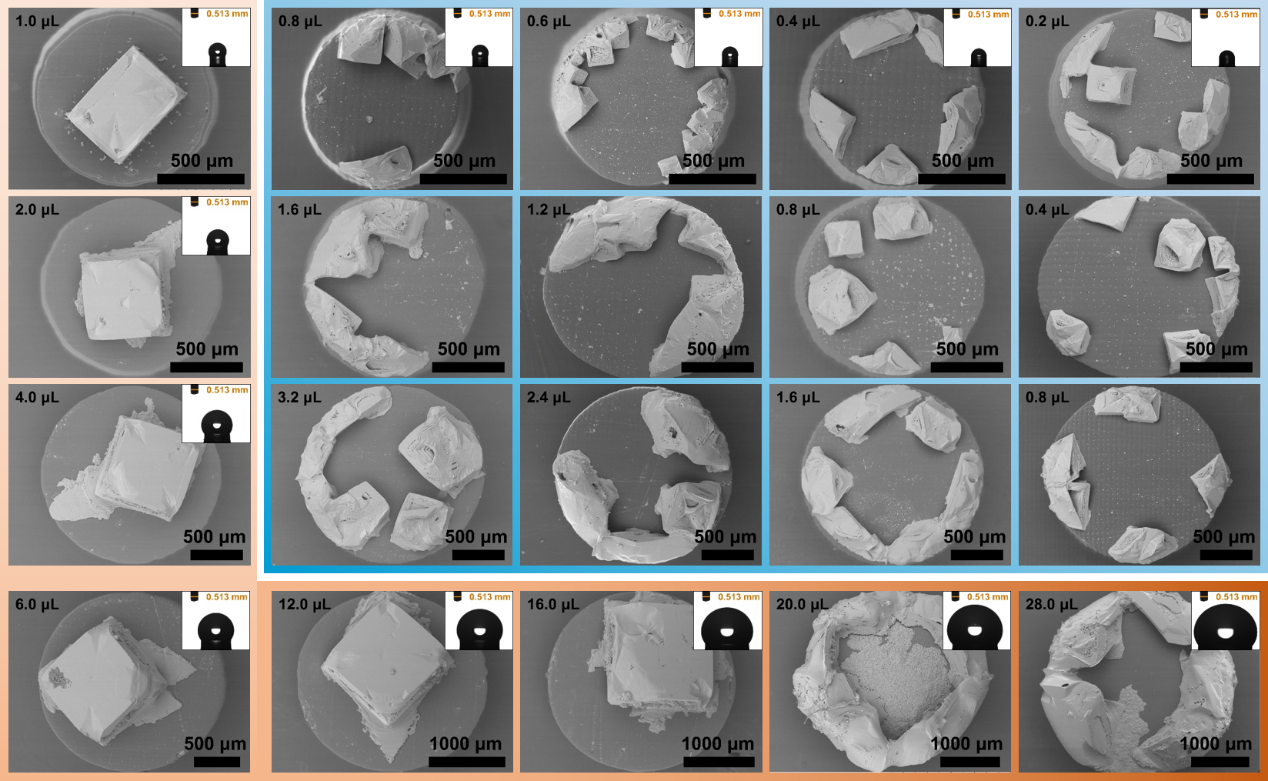
**Supplementary Figure 10.** The temperature contours of projections on the top and the temperature distributions along the gas-liquid interface of CFDCs obtained by infrared thermal imagers with initial contact angles of 30°, 60°, and 90°.



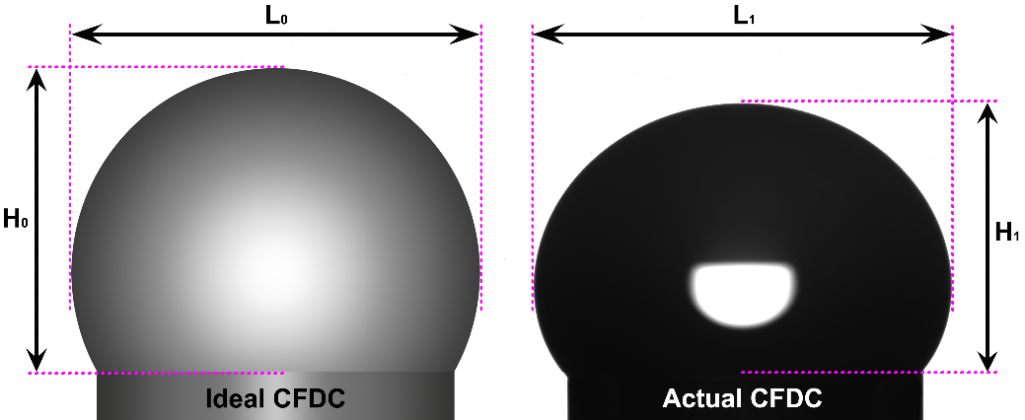
**Supplementary Figure 11.** The concentration distributions along the gas-liquid interface of CFDCs with initial contact angles of 30°, 60°, 90°, 120°, 130°, and 140° (**a**), the sitting and hanging 120° CFDCs (**b**), and the illustration of the concentration distribution along the gas-liquid interface (**c**).



**Supplementary Figure 12.** The velocity vector projection on the cross-section plane of CFDC with initial contact angle of 130°.

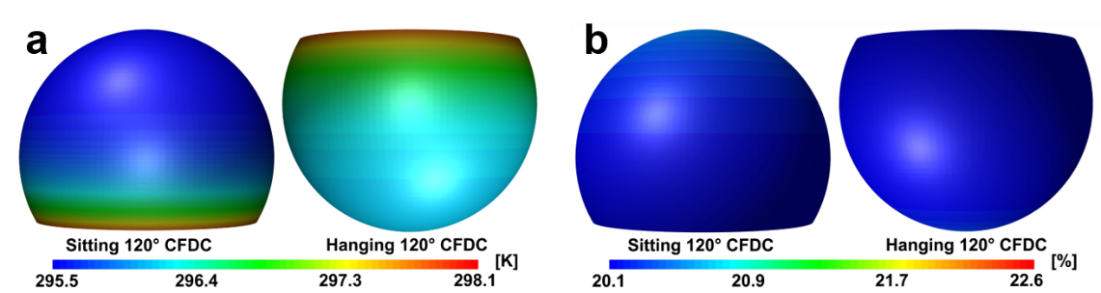


**Supplementary Figure 13.** The crystal morphologies and the corresponding CFDCs with the scale-up and stage-up.



**Supplementary Figure 14.** Illustration of the aspect ratio for ideal and actual CFDCs.

The aspect ratio (H/L) was calculated by height (H) and wide (L) of CFDC, where the error of aspect ratio was defined as the absolute value of .

**Supplementary Figure 15.** The 3D contours of temperature (**a**) and concentration (**b**) for the sitting and hanging CFDCs with initial contact angle 120°.

**Supplementary Table 1.** The Marangoni number (Ma) of the Marangoni flow inside CFDCs with initial contact angles of 30°, 60°, 90°, 120°, 130°, 140°, and the hanging CFDC (120°-H).

|  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- |
| Initial contact angle / ° | 30 | 60 | 90 | 120 | 130 | 140 | 120-H |
| Ma | 154.1 | 452.4 | 828.1 | 1373.7 | 2036.0 | 2493.1 | 1003.9 |

The Ma was utilized to assess the Marangoni flow intensity inside the CFDCs. In general, the Ma can be calculated written as1:

where R is the radius of the contact line of CFDC, γ is the surface tension, T is the temperature, ΔT is the temperature difference along the CFDC surface. μ is the viscosity, and the α is the thermal diffusivity.

**Supplementary Table 2.** The error of aspect ratio for the actual FCDCs compared with the ideal droplet (120°) with identical crystallization solution.

|  |  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- | --- |
| FCDCs | Ideal FCDC | Actual FCDCs | | | | | | | |
| Volume / μL | - | 1 | 2 | 4 | 6 | 12 | 16 | 20 | 28 |
| Aspect ratio | 0.75 | 0.766 | 0.78 | 0.763 | 0.719 | 0.699 | 0.694 | 0.67 | 0.643 |
| Error / % | 0 | 2.16 | 4 | 1.78 | 4.14 | 6.8 | 7.41 | 10.72 | 14.11 |

1. Basu, N. & Mukherjee, R. Evaporative Drying of Sodium Chloride Solution Droplet on a Thermally Controlled Substrate. *J. Phys. Chem. B* **124**, 1266–1274 (2020).