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Electronic supplementary information

Tunable fabrication of concave microlens arrays by initiative cooling based water droplets condensation

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1. Numerical model of three phase flow based on the phase filed method

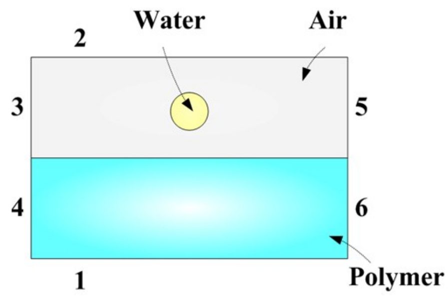

Fig. S1. Schematic diagram of three phase flow simulation model.

The water droplets imprinting process is the key for the microlens fabrication. Thus, we build a three phase flow based on phase field method to simulate the morphology evolving process of water droplet dispensing on the polymer surface.[S1] In simulations, the phase filed formulation was introduced to track the moving interface among air, water, and polymer. In the phase field formulation of three-phase flow, the interface of each two of the three phases is considered to be physically diffused and has a tiny thickness. The commercial finite element method software COMSOL Multiphysics was applied to analyze the microlens formation process of water droplet imprinting. The geometry of the model was presented in schematic diagram in Fig. S1. The water droplet with initial diameter of 2 μm locating in air was dispensed to polymer surface. The boundary conditions for the fluid flow are no slip at boundaries 1, 2, 3, 4, 5 and 6.

2. Morphology evolving process

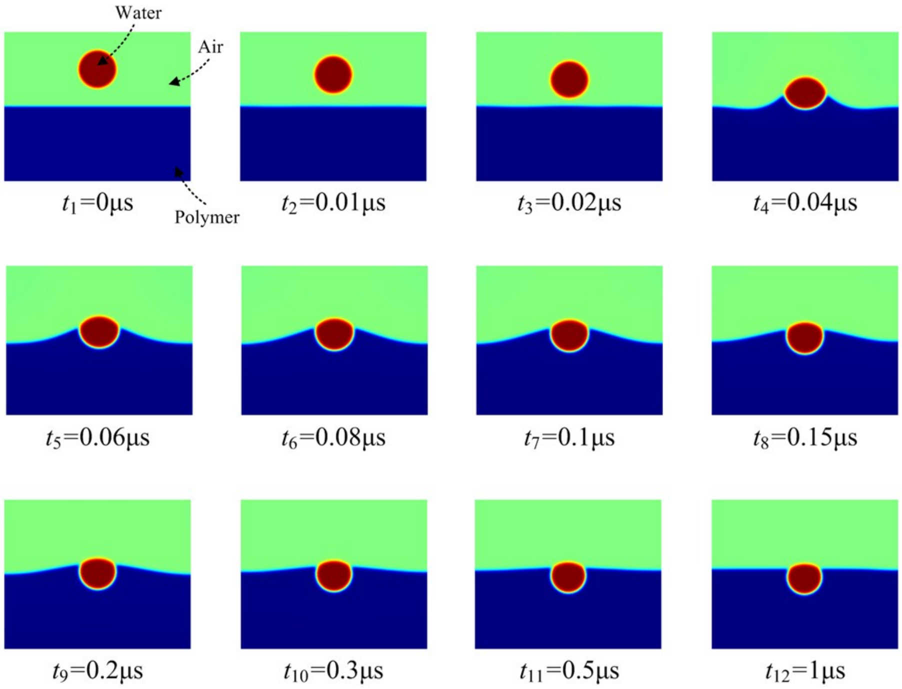

Fig. S2. Morphology evolving process of water droplet at different times.

Fig. S2 demonstrated the morphology evolving process of water droplet. The red, green, and blue regions represent water, air, and polymer, respectively. Due to the interfacial tension of the water droplet and the liquid polymer, the droplet penetrated into the polymer in part gradually. The equilibrium state of air-water-polymer three-phase structure of partial spreading was obtained after a certain period of time. When the polymer was cured, the droplet evaporated and escaped from the polymer surface, a concave microlens was formed on the polymer surface finally. Therefore, the condensed water droplets can be used as the template to imprint concave microlens arrays on the polymer surface.

3. Effect of the interfacial tension

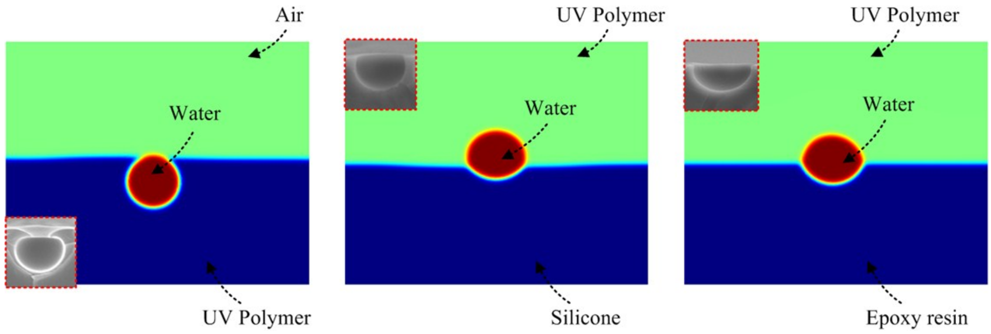

Fig. S3. Equilibrium states of water droplet condensation with different interfacial tension (insets are the corresponding cross-sectional profiles of the fabricated MLAs).

It can be noted that the interfacial tension is one of the factors that influences the morphology of concave microlens. Therefore, we analyzed the effect of the interfacial tension in the water imprinting process. The contact angle between different materials was measured by drop shape analyzer (DSA100, KRUSS, Germany). Since UV polymer, water, silicone, and epoxy resin are all transparent, it is difficult to directly get the three-phase contact angle. Therefore, we chose to measure the contact angle of the two-phase materials, and then calculated the interfacial tension of two-phase materials by solving the Young's relation.

Table S1. Surface tension of materials.

Materials	Water	UV polymer	Silicone	Expoxy resin
Surface tension(N/m)	0.072	0.4	0.31	0.46

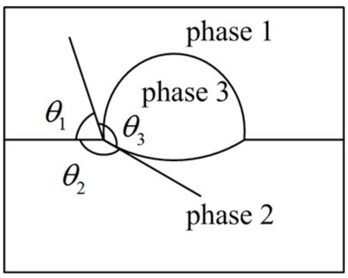

Fig. S4. Shape of the droplet at equilibrium.

Take phase 1 as air, phase 2 as UV polymer, and phase 3 as water for example. θ_1 = 170.7º was measured by the drop shape analyzer, and σ_{12} = 0.04 N/m， σ_{13} = 0.072 was known. The above three known parameters were induced into the Young's relation:

$$\frac{\sin\theta_1}{\sigma_{23}} = \frac{\sin\theta_2}{\sigma_{13}} = \frac{\sin\theta_3}{\sigma_{12}}$$

As a result, the interfacial tension of UV polymer/water was obtained as σ_{23} = 0.0332 N/m. Similarly, the interfacial tension between UV polymer/silicone, UV polymer/epoxy resin, water/silicone and water/epoxy resin can be obtained separately, which are 0.0180 N/m, 0.0181 N/m, 0.0432 N/m, and 0.036 N/m, respectively.

By substituting the calculated interfacial tension parameters into the three-phase flow numerical model, the three-phase cross-sectional profiles with no sacrificial layer and with different sacrificial layer materials can be simulated respectively. Fig. S3 shows the steady equilibrium state of the water droplet with different interfacial tension. With the change of the interfacial tension by using no sacrificial layer and different sacrificial layer materials, the cross-sectional profile of water droplet changes from sphere to hemisphere, which is consistent with the experiment results (Fig. S3 inset is the corresponding cross-sectional profiles of the fabricated MLAs). Thus, the concave MLAs with different morphology can be obtained by using different sacrificial layer materials. However, it should be noted that there may be deviations between the simulation and the experiment results, which may be caused by the following two reasons. The interfacial tension change with the variation of temperature, which make it not easy to accurately calculate the interfacial tension among the three-phase interfaces. Meanwhile, the UV polymer is not curing instantaneously, which may lead to physics properties slight changes in the experiment process.

4. Effect of the cooling temperature

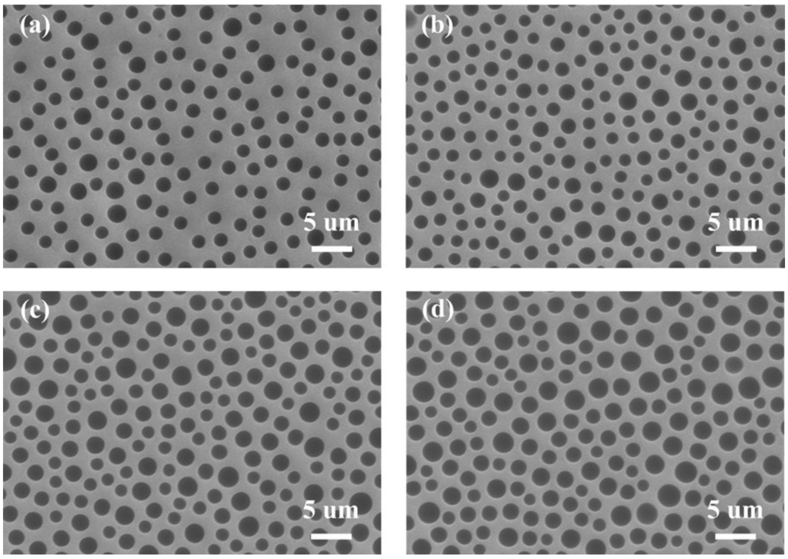

Fig. S5. SEM images of fabricated concave MLAs with different cooling temperature of (a) 14.0 °C, (b) 11.5 °C, (c) 9 °C, and (d) 7 °C at water condensation time of 5 s.

We investigate the effect of cooling temperature on the morphology of concave MLAs by using different cooling temperature changed from 14 to 7 °C. Fig. S5 illustrates the SEM images of fabricated concave MLAs with different cooling temperature at the condensation time of 5 s. The average diameter is gradually increased with the decrease of the cooling temperature, while the average packing distance shows a slightly decrease tendency. When the cooling temperature is 14, 11.5, 9, and 7 °C, the diameters are 1.30±0.19, 1.41±0.24, 1.61±0.33, and 1.67±0.36 μm, and the packing distances are 3.32±1.13, 3.05±0.85, 3.03±0.61, and 2.95±0.53 μm, respectively. The reason is that with the temperature of the sacrificial layer decreasing, more water vapor condenses on the polymer surface, then grows and self-assembles into water droplets with larger diameter.

5. Effect of the extra sacrificial layer

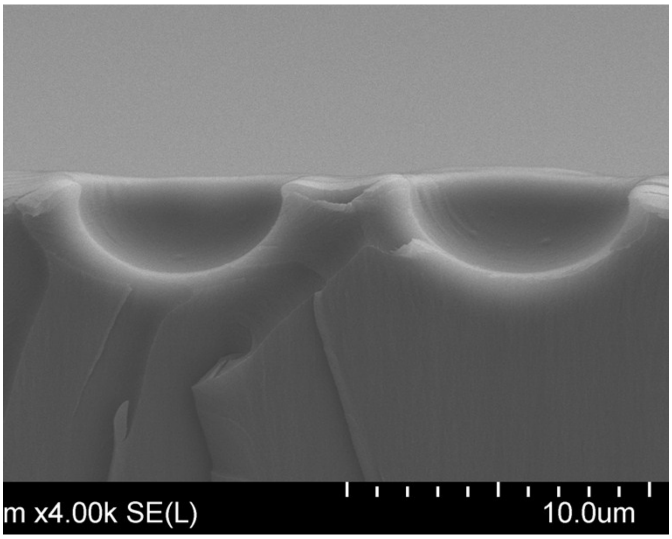

Fig. S6. Cross-sectional SEM image of fabricated concave MLAs at extra sacrificial layer polymer volume of 40 uL for water condensation time of 15 s and cooling temperature of 11.5 °C.

(S1) F. Boyer, C. Lapuerta, S. Minjeaud, B. Piar and M. Quintard, *Transport Porous Med.*, 2010, **82**, 463-483.