Supporting Information

For

Topography-regulated disorder-to-order transition of condensation droplets

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1. Water condensation dynamics inside cavities

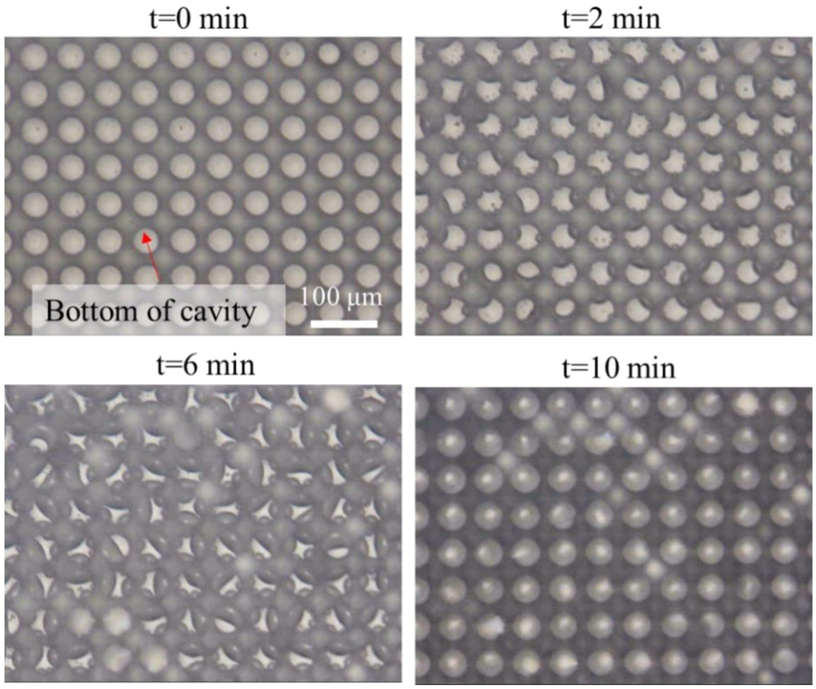

Figure S1. Time-lapse optical images of water condensation process inside cavities. Before the condensation, we first focused on the bottom surface of the cavity. As shown in the first image (t=0 min), the bottom surface of these cavities, indicated by white circles, stands in striking contrast to other areas. Initially, small droplets start to nucleate at the edge of the cavities, these droplets gradually grow up to coalesce with neighboring droplets, and finally cover the entire bottom area of cavities.

2. Droplets patterning on surfaces with cavities of different shapes

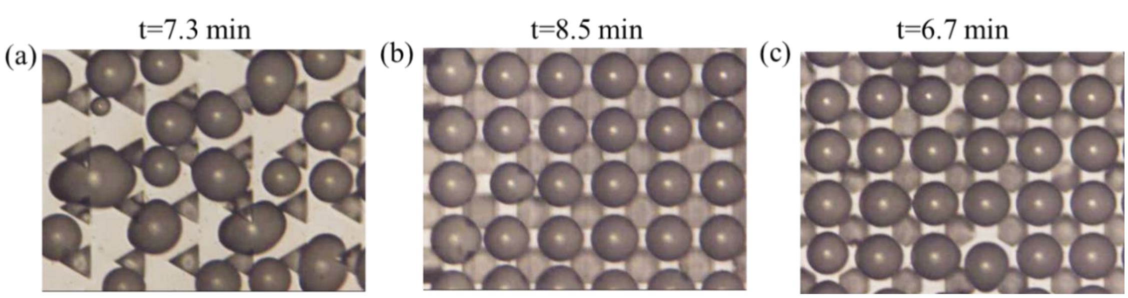

Figure S2. (a-c) Snapshots of droplets patterning on surfaces with triangular, rectangular and hexagonal cavities, respectively. Droplets nucleated on the triangular cavities surface are randomly distributed whereas on the other two surfaces, there is a pronounced disorder-to-order transition.

3. Droplets patterning on surfaces with cavities of different depths

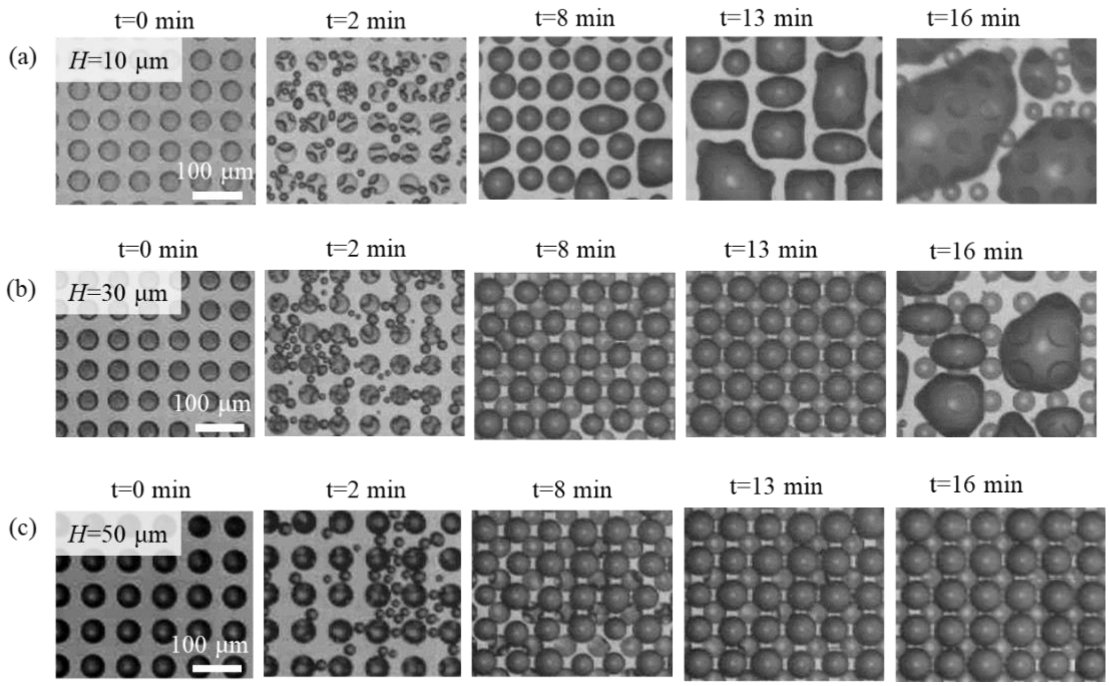

Figure S3. (a-c) Time-lapse optical images of water condensation process on surface with cavities of identical diameter (40 μm) and spacing (60 μm), but different depths of 10, 30, and 50 μm, respectively. On the surface with a cavity depth of 10 μm, droplets initially randomly nucleate on all the surfaces, with time progression, droplets that located on the outer edge of cavities grow up to coalesce with droplets inside the cavities, and eventually be dragged into the cavities. As a result, each droplet is confined in one cavity, the existing droplets continuously grow to touch each other and coalesce. Finally, the surface is covered by randomly distributed droplets. Note that on the surface with a small cavity depth of 10 μm, there is no ordered droplets pattern forming on the top surface during the entire condensation process, while on the surfaces with a cavity depth of 30 μm and 50 μm, we found similar disorder-to-order transition of droplets pattern, and the ordered droplets pattern sustain for longer time on the latter surface, which implies that surfaces with a larger depth of cavities are favorable for the formation of stable and ordered droplets pattern.

4. Derivation of interfacial energy of droplets

The surface energy of a droplet pinned at the edge of two cavities with advancing contact angle θ_a can be expressed as

$$E_a = \gamma_{LG}(A_a - \cos\theta_Y S_a) \tag{S1}$$

where A_a is the area of water cap that contact with gas, S_a is the base area of droplet that contact with flat solid surface. The parameters satisfy the geometry relationship: $A_a = \frac{2\pi R_1^2}{1+\cos\theta_a}$, $S_a = \pi R_1^2$, R_1 is the base radius of droplet, which is given as $R_1 = \frac{L-D}{2}$.

Similarly, the surface free energy of a droplet spreads across the cavities with equilibrium contact angle θ_Y can be formulated as

$$E_b = \gamma_{LG}\left(A_b - \cos\theta_Y S_b + S_c\left(1+\cos\theta_Y\right)\right) \tag{S2}$$

where A_b is the area of water cap that contact with gas, S_b is the base area that contact with flat solid surface. $A_b = \frac{2\pi R_2^2}{1+\cos\theta_Y}$, $S_b = \pi R_2^2$, R_2 is base radius of droplet. S_c is the portion of droplet base area that suspend on cavities, which is approximated as $S_c \sim \frac{\pi D^2}{4}$. Assume droplet volume V is identical in the two scenarios, which is given as $V = \frac{\pi R_1^3\left(1-\cos\theta_a\right)^2\left(2+\cos\theta_a\right)}{3\sin^2\theta_a} = \frac{\pi R_2^3\left(1-\cos\theta_Y\right)^2\left(2+\cos\theta_Y\right)}{3\sin^2\theta_Y}$.

Making the appropriate substitutions and non-dimensionalizing the energy as $E_a^* = \frac{E_a}{\gamma_{LG}L^2}$ and $E_b^* = \frac{E_b}{\gamma_{LG}L^2}$, we find

$$E_a^* - E_b^* = k_1\left(1-\frac{D}{L}\right)^2 - k_2\left(\frac{D}{L}\right)^2. \tag{S3}$$

$$k_1 = \frac{\pi}{4}\left\{\frac{2}{1+\cos\theta_a} - \cos\theta_{\mathrm{Y}} - \left(\frac{2}{1+\cos\theta_{\mathrm{Y}}} - \cos\theta_{\mathrm{Y}}\right)\left[\frac{\sin^2\theta_{\mathrm{Y}}\left(1-\cos\theta_a\right)^2\left(2+\cos\theta_a\right)}{\sin^2\theta_a\left(1-\cos\theta_{\mathrm{Y}}\right)^2\left(2+\cos\theta_{\mathrm{Y}}\right)}\right]^{2/3}\right\}$$

$$k_2 = \frac{\pi}{4}\left(1+\cos\theta_{\mathrm{Y}}\right)$$

For the calculation, we set θ_{a} and θ_{Y} as the advancing contact angle and Young's contact angle of droplet on flat PDMS surface, respectively, which is given as 125 $^{\mathrm{o}}$ and 105 $^{\mathrm{o}}$, respectively.