Supporting Information

Imaging and Characterizing Fluid Invasion in Micro-3D Printed Porous Device with Variable Surface Wettability

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S1. Micromodel Printing and Flow Imaging

S1.1 Printing with Rock Micro-CT Image

A rock pattern is printed based on the CT image of natural carbonate. Figure S1 shows the rock pattern from the CT image, which has 671×560 pixels in total and each pixel is 8 µm. The right-top image in Figure S1 shows the printed patterns under the SEM. It can be found that the printed patterns can capture the rock features vividly. The printing resolution of 2µm is a good match for a µ-CT scan image of 8µm. In the image, some very fine features, e.g. solid grain, pore and throat (~50 µm) are highlighted in the red box. The shape of the grain/throat and smooth surfaces are clearly observed in the SEM images, which evidences the printing accuracy of our micro-3D printer. Together with the observations in Figure 1 in the manuscript, we have shown that the µ-SL printing is an effective way for the fabrication of microfluidic device fabrication owing to its capability of quick prototyping complicated 3D structures at the micrometer scale.



Figure S1. Surface morphology of 3D printed micromodel. The rock pattern is from micro-CT image of natural carbonate. The CT image resolution is 8 μ m and the printing resolution is 2 μ m. The SEM image in the right-top corner is assembled from four images because of the limited size of field of view of each single image.

S1.2 Repeatability of The Pore-Scale Flow Experimental Setup

To evaluate the repeatability of the experimental setup, we first conduct a series of test-retest experiments. In these experiments, the inlet pressure is set at 800 mbar and then decreased gradually to 0 mbar (marked as Test 1). Meanwhile, we recorded the pressure value (the black curve) and the flow rate (the red curve) from the sensors, as shown in **Figure S2**. Then, the whole process is reversed, by increasing the pressure from 0 mbar to 800 mbar (marked as Test 2). Finally, the pressure again decreased to 0 mbar (Test 3). By replotting the measurements with pressure as x-axis and flow rate as y-axis in **Figure S2**b, we can find a good consistency between these repeating experiments. Most importantly, the linear relationship between the pressure drop and flow rate confirms the laminar flow inside the micromodels, which is resonable for slow fluid flow in subsurface applications.⁴²⁴³⁴⁴



Figure S2. The test-retest reliability of the experimental setup. (a) the experiment procedure with the recorded pressure drop and the corresponding flow rate; (b) repeatability verification of the experimental setup through the pressure vs. flow rate curve.

S2. Effect of Channel Depth on Imaging and Fluid Invasion Behavior

In this work, the confocal microscope is operating at reflection mode. In this way, we avoided using fluorescence because it can change the fluid property, i.e., surface tension and viscosity. The confocality is limited by the light source and the intrinsically small numerical aperture (NA) of low magnification lens (5x magnification lens with 0.15 NA is used in this work). As illustrated in **Figure** S3, the focal plane is with a certain thickness (the purple area), so the collected image actually contains a stack of out-of-focus images. When the pinhole size is 64 μ m, the thickness of focal plane is about 200 μ m. As a result, in a shallow channel, a wide interfacial region is captured where the greyscale indicates the fluid thickness or concentration; however, with the increase of channel depth, the interfacial region narrows down in the range of focal plane, as illustrated at the right side of **Figure** S3. Therefore, in order to get a clear image of the fluid interface, we have carefully considered the effect of channel depth on the imaging quality. In this work, the channel depth is 1 mm and that is why we can clearly observe the lateral interface between two fluids.

There is another benefit of increasing the channel depth. As mentioned in the manuscript, the capillary pressure is calculated as $p_c = \gamma \cos \theta (1/R_1 + 1/R_2)$, where R1 and R2 are the channel width and depth. So only when the channel depth is much larger than the channel width, the capillary pressure value is dominated by the channel width. The large depth/width ratio allows us to accurately study the lateral interfacial meniscus in parallel channels and in porous patterns during fluid invasion.



Figure S3. Effect of channel depth on the imaging of fluid interface

S3. Lattice Boltzmann Modeling on Fluid Invasion

The lattice Boltzmann modeling (LBM) is employed to simulate the oil/water/gas multiphase flow in the existence of the solid with different surface wettability. Here we implement the Shan-Chen multicomponent LBM³ into the two and three dimensions simulation. In the model, the distribution function f is introduced for each component in oil/water/gas system as shown in **Equation** S1.

$$f_a^{\sigma}(\mathbf{x} + \mathbf{e}_a \,\Delta \mathbf{t}, \mathbf{t} + \Delta \mathbf{t}) = f_a^{\sigma}(\mathbf{x}, \mathbf{t}) - \frac{\Delta \mathbf{t}}{\tau_{\sigma}} [f_a^{\sigma}(\mathbf{x}, \mathbf{t}) - f_a^{\sigma, \text{eq}}(\mathbf{x}, \mathbf{t})]$$
(S1)

where f_a^{σ} is the σ -th component density distribution in the *a*-th direction. τ is the relaxation time which is related to the kinematic viscosity. $f_a^{\sigma,eq}$ is the equilibrium distribution function, which can be calculated as

$$f_a^{\sigma, \text{eq}}(\mathbf{x}, \mathbf{t}) = w_a \rho_a \left[1 + \frac{e_a u_\sigma^{\text{eq}}}{c_s^2} + \frac{(e_a u_\sigma^{\text{eq}})^2}{2c_s^4} - \frac{(u_\sigma^{\text{eq}})^2}{2c_s^2}\right]$$
(S2)

$$u_{\sigma}^{\rm eq} = \left(\sum_{\sigma} \sum_{a} \frac{f_{a}^{\sigma} e_{a}}{\tau_{\sigma}}\right) / \left(\sum_{\sigma} \frac{\rho_{a}}{\tau_{\sigma}}\right) + \frac{\tau_{\sigma} F_{\sigma}}{\rho_{a}}$$
(S3)

In Equation S2, e_a is the discrete velocity in the *a*-th direction. w_a is the weight factor, and ρ_{σ} is the density Eof the σ -th component. Their values can be found in our previous paper^{4,5}. In Equation S3, F_{σ} is the total force acting on the σ -th component by the sum of the fluid-fluid cohesion force $F_{int,\sigma}$, the fluid-solid adhesion force $F_{ad,\sigma}$, and the external force F_{ext} such as the gravity force. The cohesive force and the adhesive force are defined as

$$F_{\text{int},\sigma}(\mathbf{x},\mathbf{t}) = -\varphi_{\sigma}(\mathbf{x},\mathbf{t}) \sum_{\sigma} G_{\sigma\sigma} \sum_{a} w_{a} \varphi_{\sigma}(\mathbf{x}+\mathbf{e}_{a} \Delta \mathbf{t},\mathbf{t}) \mathbf{e}_{a}$$
(S4)

$$F_{\rm ad,\sigma}(\mathbf{x},\mathbf{t}) = -\varphi_{\sigma}(\mathbf{x},\mathbf{t})G_{\rm ad,\sigma}\sum_{a} w_{a}\varphi(\rho_{\rm s})s(\mathbf{x}+\mathbf{e}_{\rm a}\,\Delta\,\mathbf{t},\mathbf{t})\mathbf{e}_{\rm a}$$
(S5)

where φ_{σ} and $\varphi_{\overline{\sigma}}$ are the effective densities in the interaction force of different fluid components. $G_{\sigma\overline{\sigma}}$ is the parameter controlling the strength of cohesive force, while $G_{ad,\sigma}$ controls the strength of adhesive force. By varying the values of $G_{\sigma\overline{\sigma}}$ and $G_{ad,\sigma}$, the desired surface tension and solid surface wettability can be achieved^{5,6}.