



Wetting transition and fluid trapping in a microfluidic fracture

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Immiscible fluid–fluid displacement in confined geometries is a fundamental process occurring in many natural phenomena and technological applications, from geological CO₂ sequestration to microfluidics. Due to the interactions between the fluids and the solid walls, fluid invasion undergoes a wetting transition from complete displacement at low displacement rates to leaving a film of the defending fluid on the confining surfaces at high displacement rates. While most real surfaces are rough, fundamental questions remain about the type of fluid–fluid displacement that can emerge in a confined, rough geometry. Here, we study immiscible displacement in a microfluidic device with a precisely controlled structured surface as an analogue for a rough fracture. We analyze the influence of the degree of surface roughness on the wetting transition and the formation of thin films of the defending liquid. We show experimentally, and rationalize theoretically, that roughness affects both the stability and dewetting dynamics of thin films, leading to distinct late-time morphologies of the undisplaced (trapped) fluid. Finally, we discuss the implications of our observations for geologic and technological applications.

wetting transition | microfluidics | roughness | fracture | pattern formation

Immiscible fluid–fluid displacement in geometrical confinement occurs in many natural and engineering processes, including water infiltration into soil (1), oil recovery (2), fuel cell water management (3), and microfluidics (4, 5).

Many experimental and computational studies have shown that fluid invasion during these processes can range from a compact pattern to a fractal pattern including viscous fingering and capillary fingering on the macroscopic scales (6–14). These coarse-grained descriptions of displacement often discard the details of the interactions between the liquids and the solid walls and assume complete fluid displacement on the microscale. In the presence of moving contact lines, however, the dynamics on the microscale are rate dependent, with complete displacement at low displacement rates and incomplete displacement at sufficiently high displacement rates (15). The transition between these two regimes is accompanied by the formation of thin films of the defending liquid on the solid surface, which later dewet from the substrate and lead to pinch-off and the formation of disconnected bubbles and drops (15, 16).

The consequences of this wetting transition on macroscopic patterns remain poorly understood. Previous studies focused on smooth surfaces (17–19), while, in fact, most real surfaces and porous media are rough. Here, we investigate the influence of roughness on the wetting transition on both microscopic and macroscopic length scales. We show that roughness leads to the emergence of regimes that are absent on smooth surfaces.

We demonstrate that roughness significantly reduces the flow rate threshold for the onset of the wetting transition. Above this threshold, we observe two distinct regimes: a) the thin-film regime, where the displaced fluid is trapped in the crevices of the rough surface and b) the thick-film regime, where the displaced fluid fully covers the rough surface. The thin films may be discontinuous or remain energetically stable depending on the degree of roughness. We delineate these distinct regimes—complete displacement, thin-film (unstable, dewetting, stable), and thick-film—in the form of a phase diagram and explain theoretically the cross-overs among the regimes. We then show that the different hydrodynamic regimes lead to different late-time morphologies of the undisplaced (trapped) defending fluid.

Experimental Setup

We conduct our experiments in radial Hele-Shaw cells (Fig. 1A), consisting of two parallel circular plates, separated by spacers of thickness B . The radial geometry has been used extensively in the study of nonlinear pattern formation (7, 8, 13, 14, 20–24). While it introduces some arbitrariness in the definition of the capillary number, it

Significance

Fluid flow along fractures is critical in subsurface technologies like geological CO₂ sequestration, geothermal energy extraction, and hydrogen geostorage. Geologic fractures are rough, and the role of roughness on two-phase fluid displacement is not well understood. Here, we conduct laboratory experiments on patterned microfluidic cells with a precisely controlled structured surface to elucidate the fundamental role of surface roughness on two-phase displacement in confined geometries. In contrast to the better-understood case of smooth confinement, we observe two types of liquid films entrained on the rough surfaces, which lead to distinct late-time morphologies of trapped fluid. Our observations shed light on the microscale physics and macroscopic displacement patterns in rough fractures, which control long-term mixing and reactivity in geological systems and lab-on-a-chip applications.

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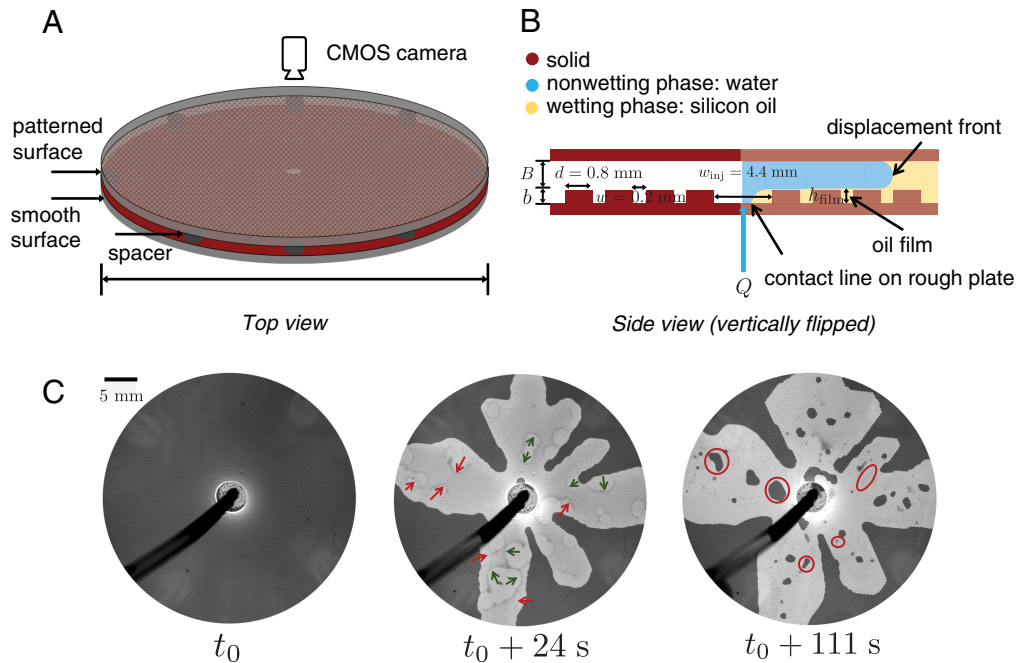


Fig. 1. We inject water into a rough Hele-Shaw cell filled with viscous silicone oil at constant injection rate Q . (A) Schematic figure of the experimental system. We take images of the experiment from above with a CMOS camera, measuring the gap-averaged oil saturation at high spatial and temporal resolution after calibration of the light intensity. (B) Cross-section of the rough Hele-Shaw cell. The rough plate is patterned with a regular honeycomb lattice of cylindrical posts with post diameter $d = 0.8$ mm and pore throat $w = 0.2$ mm. The fluid is injected at the center of the cell, within a post-free circular region of diameter $w_{\text{inj}} = 4.4$ mm. (C) Sequence of images from a typical experiment with $\text{Ca}_{\text{inj}} = 0.725$; $B = 0.5$ mm; $b = 0.08$ mm (SI Appendix, Movie S4). Less viscous water displacing more viscous silicone oil results in viscous fingering. A film of undisplaced residual oil is left behind the displacement front. This film dewets on the rough surface (red arrows at $t_0 + 24$ s) and at late times leads to clusters of trapped oil (red circles at $t_0 + 111$ s).

eliminates the boundary effects that plague rectilinear-cell experiments, both from the influence of lateral walls and the lack of uniformity in the initial displacement front. We make one plate's surface rough by patterning it with a regular honeycomb array of cylindrical posts of height b (25) (SI Appendix, section 1.A). The dimensions of the pore throat $w = 0.2$ mm and post diameter $d = 0.8$ mm are fixed in our study, which yields two constant quantities: 1) post density, defined as the areal fraction taken by the posts $\phi_s = \frac{\pi}{2\sqrt{3}} \left(\frac{d}{w+d}\right)^2 = 0.58$; and 2) post-pattern porosity $\phi = 1 - \phi_s = 0.42$. The injection port is at the center of the rough plate and is surrounded by a circular region of diameter $w_{\text{inj}} = 4.4$ mm without posts (Fig. 1B). We use silicone oil with viscosity $\mu_o = 500$ mPa s as the defending liquid and deionized water with viscosity $\mu_w = 0.99$ mPa s as the invading fluid. We measure the liquid-liquid interfacial tension to be $\gamma = 13$ mN/m. The static contact angle of water immersed in silicone oil is $\theta_E = 123 \pm 5^\circ$, indicating that water is less wetting to the solid surfaces. We add a light-absorbing dye to the oil phase for visualization and quantification of the film thickness using the light intensity (SI Appendix, section 1.B).

We investigate the roles of three variables: 1) the injection rate [Q in the range (2×10^{-2} , 2×10^2) mm³/s], 2) the aperture of the fracture [gap thickness B in the range (0.15, 0.76) mm], and 3) the amplitude of the surface roughness [post height b in the range (0, 0.135) mm]. The aperture B is the characteristic height of the flow channel, which determines the single-phase hydraulic conductivity (26) and capillary pressure gradient along the displacement direction (13). The post height b is chosen as the characteristic roughness height, a quantity that is known to impact the effective contact angle (27–29) and slip length (30–33).

In each experiment, the cell is first saturated with silicone oil. Then, water is injected into the cell at a constant injection rate Q .

We characterize the displacement dynamics of each experiment using the injection capillary number $\text{Ca}_{\text{inj}} = \frac{v_{\text{inj}} \mu_o}{\gamma}$, where $v_{\text{inj}} = \frac{Q}{\pi w_{\text{inj}} (B + b\phi)}$ is defined at the entrance of the post pattern. We vary Ca_{inj} from 10^{-3} to 4.2. Since water displaces oil radially outward and the front velocity decays away from the center, we also define a local capillary number, $\text{Ca} = \frac{v \mu_o}{\gamma}$, to represent the local competition between the viscous and capillary forces, where v is the measured local displacement front velocity (SI Appendix, section 1.C).

Experimental Results

We show images of a typical experiment (under conditions $\text{Ca}_{\text{inj}} = 0.725$, $B = 0.5$ mm, and $b = 0.08$ mm) before water injection, shortly after water breakthrough, and at late times, in Fig. 1C. The displacement front is subject to the viscous fingering instability due to the smaller viscosity of the invading fluid (7, 11). Films of the viscous defending liquid are left behind the displacement front and deposited on both the rough and smooth substrates (red and green arrows in Fig. 1C). At late times, these films dewet from the substrates, forming persistent clusters along the pathway of the invading fluid (circled regions in Fig. 1C). The formation and evolution of films on the smooth surface have been well studied (e.g., ref. 15), and we discuss them in the context of our hybrid smooth plate-rough plate system in SI Appendix, section 7. Here, we focus on analyzing the films on the rough surface of the cell.

Regimes of Fluid-Fluid Displacement. We demonstrate the different regimes of fluid-fluid displacement with three representative experiments conducted on the same geometry ($B = 0.5$ mm; $b = 0.08$ mm).

At a low capillary number (local $Ca \approx 0.0008$ in Fig. 2A), we observe complete displacement, with only very small droplets of oil trapped behind the posts (34) (Fig. 2B and C, Movie S1).

At an intermediate capillary number (local $Ca \approx 0.02$ in Fig. 2D), we observe a thin film of undisplaced oil of thickness roughly equal to the post height b (see the measured thickness in the circled red dot in Fig. 3): The top surface of the posts is exposed to the invading fluid. This film dewets with the residual viscous fluid retracting as a film of uniform thickness and confined within the post pattern (Fig. 2E and F and Movie S2).

In contrast with classical dewetting on smooth surfaces, this film dewets with sublinear dynamics (SI Appendix, Fig. S3A). We call this regime the thin-film regime.

At a sufficiently high capillary number (local $Ca \approx 0.05$ in Fig. 2G), the thickness of the film of undisplaced oil is larger than the post height b (see the measured thickness in the circled red dot in Fig. 3), and the undisplaced oil completely covers the solid surface (SI Appendix, section 1.D). This film is not stable and dewets on the rough surface, similarly to how it dewets on a smooth surface (15, 18): The residual liquid retracts at a constant velocity (SI Appendix, Fig. S3B), and a rim is formed next to the

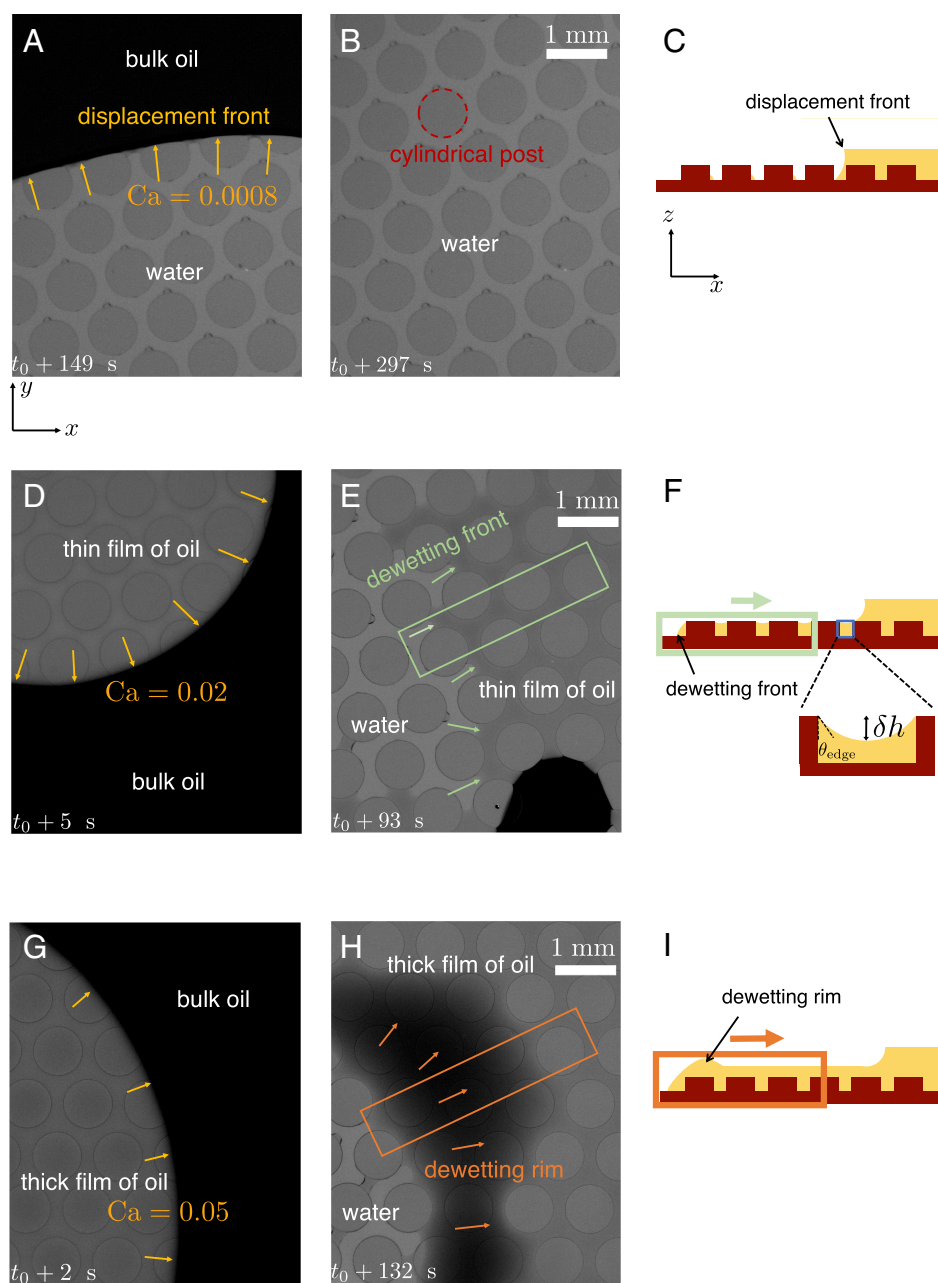


Fig. 2. Experimental demonstration of different fluid-fluid displacement regimes on the same geometry, with $B = 0.5$ mm and $b = 0.08$ mm. (A) and (B) show the complete displacement regime (SI Appendix, Movie S1), here for local capillary number $Ca \approx 0.0008$. They show the moments when water is displacing oil and when oil has been completely displaced. (C) is the side-view schematic of complete displacement. (D) and (E) show the thin-film regime (SI Appendix, Movie S2), here for local capillary number $Ca \approx 0.02$. They show the moments when water is displacing oil leaving the thin film trapped behind the fluid-fluid front and during dewetting of the residual thin film. (F) is the side-view schematic of thin-film dewetting. (G) and (H) show the thick-film regime (SI Appendix, Movie S3), here for local capillary number $Ca \approx 0.05$. They show the moments when water is displacing oil with the thick film trapped behind the front and during dewetting of the residual thick film. (I) is the side-view schematic of thick-film dewetting.

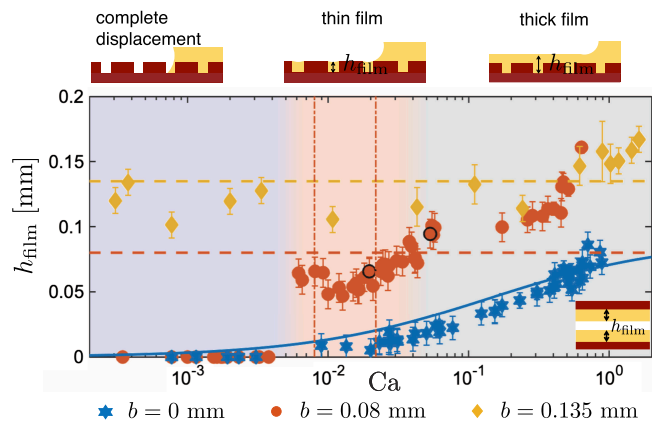


Fig. 3. Experimentally measured local capillary number of the displacement front Ca vs. film thickness h_{film} , for gap height $B = 0.5$ mm. We show data for a smooth cell ($b = 0$, blue symbols) and two different post heights ($b = 0.08$ mm, red symbols; and $b = 0.135$ mm, yellow symbols). Circled red dots are the measurements for Fig. 2 D, Left and Fig. 2 G, Right. The blue solid line is the extended Bretherton law (36): $\frac{h_{\text{film}}}{B/2} = \frac{0.643(3Ca)^{2/3}}{1 + 2.79 \times 0.643 \times (3Ca)^{2/3}}$. The vertical dashed lines, from left to right, represent the value $Ca_{c,\text{tf}}$ that sets the transition from complete displacement to the thin film and the value $Ca_{c,\text{tr}}$ that sets the transition to thick film for the $b = 0.08$ mm case. The critical threshold for transitions for the smooth case $b = 0$ and the rough case $b = 0.135$ mm are shown in SI Appendix, Fig. S9.

dewetting front (Fig. 2 H and I and Movie S3). We call this regime the thick-film regime.

Fluid-Film Thickness and Transitions among Regimes. In order to characterize the transitions among the three displacement regimes, we measure the local velocity v of the displacement front and employ the local capillary number Ca (SI Appendix, section 1.C) as well as the film thickness of the residual fluid h_{film} (SI Appendix, section 1.B).

In the absence of posts, i.e., for a smooth surface with $b = 0$, and at low capillary numbers, the displacement is complete and no film is left behind. As the capillary number increases beyond a critical value set by the onset of forced wetting transition (15) (blue dashed line in SI Appendix, Fig. S9), a thin film of the defending fluid becomes entrained on the walls. The thickness of this film increases monotonically with the capillary number, as predicted by the extended Bretherton model (blue stars and the blue solid line in Fig. 3).

In the presence of roughness, the dependence of film thickness on the capillary number changes qualitatively. For the case with $B = 0.5$ mm and $b = 0.08$ mm (red dots in Fig. 3), at low capillary numbers, we observe complete displacement, similar to that on a smooth surface. Beyond a critical capillary number $Ca_{c,\text{tf}}$, however, the entrained film thickness jumps to a value slightly smaller than the post height ($h_{\text{film}} \approx b$). As the capillary number increases further beyond a second critical capillary number $Ca_{c,\text{tr}}$, the film height becomes larger than the post height, and thick films of the defending fluid coat the rough surface. This transition from the thin-film to the thick-film regime has also been observed in the context of the Landau–Levich–Derjaguin problem, where unconfined rough solid surfaces were withdrawn from a liquid bath (35).

As we increase the roughness further, i.e., for the case with $B = 0.5$ mm and $b = 0.135$ mm (yellow diamonds in Fig. 3), we observe yet another qualitatively distinct coating behavior. In this case, the defending liquid becomes entrained in between the posts even at vanishingly small capillary numbers ($Ca \rightarrow 10^{-4}$);

that is, the complete displacement regime is not observed. As the capillary number increases beyond $Ca_{c,\text{TF}}$, the entrained film becomes thicker than the post height, and the thick-film coating regime is observed.

Analysis and Discussion

Here, we distill our experimental observations and provide theoretical arguments that rationalize the different coating regimes as well as the late-time fluid trapping. We delineate them in the form of a phase diagram in terms of three dimensionless numbers: 1) the injection capillary number Ca_{inj} , 2) the roughness-to-aperture-height ratio b/B , and 3) the roughness factor $r = 1 + \frac{4(1-\phi)b}{d}$, which is defined as the ratio of the real area of the rough surface over its projected area. The experimental parameters and the dimensionless numbers of the data used in the phase diagrams (Fig. 4) are listed in SI Appendix, Table 1.

Fluid Configuration in the Microfluidic Roughness. We first discuss the influence of geometry (b/B and r) on the stability of the entrained thin films.

Geometric criterion for the formation of thin films. In the thin-film configuration (Fig. 2F), the oil bridges neighboring posts and the oil–water interface is pinned at the posts’ edges with a local angle θ_{edge} and bowing into the pattern with a penetration length δh due to the Laplace pressure (37, 38). We assume pressure equilibrium between the oil meniscus in the post and the defending fluid ahead of the displacement front, such that the Laplace pressure is $P_{\text{Lap}} = 2\gamma \cos \theta/B$. Larger Laplace pressure or gap thickness would result in a smaller local angle at the edge and deeper penetration that may destabilize the thin film. On the one hand, the interface depins when the local angle is smaller than the equilibrium contact angle. On the other hand, when the penetration depth δh is larger than the post height b , the interface touches the bottom surface and the meniscus between the posts cannot form. Thus, two conditions must be simultaneously satisfied for the formation of thin films: $\delta h < b$ and $\theta_{\text{edge}} > 180^\circ - \theta_E$. To compute δh and θ_{edge} , we simulate the 3D shape of the meniscus in the post pattern via energy minimization using Surface Evolver (39). By setting $\theta_{\text{edge}} = 180^\circ - \theta_E$ and $\delta h = b$, we arrive at the following geometric criterion (SI Appendix, section 2):

$$r_{c,1} = \max \left\{ \beta_1 \frac{b}{B} + 1, \quad \beta_2 \left(\frac{1}{\cos \theta_E} \frac{B}{b} - 1 \right)^{-\frac{1}{2}} + 1 \right\}, \quad [1]$$

in which $\beta_1 = 0.87$ and $\beta_2 = 0.55$. It indicates that thin films of the defending fluid can form in the post pattern only if the roughness factor is larger than this critical value, $r > r_{c,1}$ (Fig. 4A). Otherwise, the oil meniscus between posts quickly retracts from the center of each pore and moves along the pore throats, and thus, no continuous thin film can be left behind the displacement front. We present the boundaries on the formation of thin films controlled by dynamic conditions in Fig. 4 B and C, where the region corresponding to $r < r_{c,1}$ is not shown.

Energetic stability of the entrained thin films. Whether the entrained thin films of the defending liquid dewet from the rough surfaces is determined by comparing the free energy of the coated and dewetted states. The coated state is akin to the Cassie state (40), where a water drop only touches the top of the posts and does not fill the space within the post pattern. The dewetted state is akin to the Wenzel state (41), where the drop fills all the spaces within the post pattern. Therefore, the coated state is stable if the

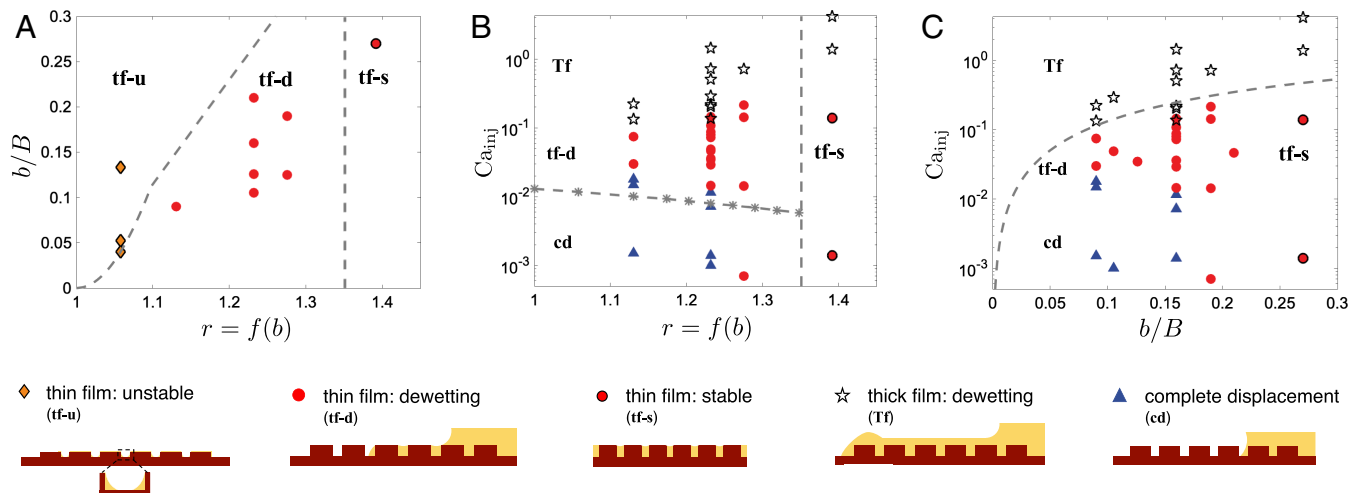


Fig. 4. Phase diagrams of the different displacement regimes observed in the experiments. The diagrams demonstrate how the interplay between the degree of surface roughness (r), the vertical aspect ratio (b/B), and the injection capillary number (Ca_{inj}) impact the wetting transition and fluid trapping. The plotting ranges are $r \in [1, 1.45]$, $b/B \in [0, 0.3]$, and $Ca_{inj} \in [0.0005, 4.3]$. (A) Geometric criteria of the stability of thin films. The left dashed line represents $r_{c,1}$ (Eq. 1). When $r < r_{c,1}$, the thin film is unstable and cannot form under any flow conditions. The vertical dashed line represents $r_{c,2}$ (Eq. 2), which is determined by the roughness factor only. When $r > r_{c,2}$, the thin film is stable and does not dewet. (B) Transition from complete displacement to thin film as $Ca_{inj} > Ca_{inj,tf} = Ca_{c,tf}$ (Eq. 3). To the right of the vertical dashed line ($r = r_{c,2}$), the transition occurs at $Ca_{inj} \rightarrow 0$. (C) Transition from the thin film to the thick film as $Ca_{inj} > Ca_{inj,Tf} = \frac{L}{4w_{inj}} Ca_{c,Tf}$ (Eq. 4).

free energy of the Cassie-like state is lower than the free energy of the Wenzel-like state (SI Appendix, section 3). Equating the free energy of these two states, we arrive at the following critical amplitude of roughness:

$$r_{c,2} = 1 - \frac{\phi(1 + \cos \theta_E)}{\cos \theta_E}, \quad [2]$$

which suggests that if $r > r_{c,2}$, the entrained thin films are stable and will not dewet from the post pattern (Fig. 4A).

Forced Wetting Transition. Even when the geometric criterion (Eq. 1) is satisfied, a thin film is observed only when the capillary number is above a critical value (red symbols in Fig. 3). We rationalize the transition from complete displacement to the thin film by calculating the critical threshold $Ca_{c,tf}$ for the onsets of the wetting transition (15). At $Ca_{c,tf}$, the meniscus of the fluid–fluid displacement front is deformed, and the apparent contact angle of the nonwetting phase reaches 180° . Beyond this critical value, the defending oil is forced to wet the rough plate as a film.

To obtain the critical capillary number at the onset of the wetting transition, we solve the Stokes equations using the generalized lubrication approximation and balance the viscous stress and Laplace pressure jump across the interface to arrive at (15, 42, 43):

$$\frac{d^2\theta}{ds^2} = \frac{3Caf(\theta, R)}{z(z + 3\lambda_s)}, \quad [3]$$

where θ is the local interface slope, s is the arc length along the interface, $M = \mu_w/\mu_o$ is the viscosity ratio between water and oil, and λ_s is the slip length (on the order of nanometers) introduced to resolve the contact-line singularity (44). The function $f(\theta, R)$ is from Cox's solution (42, 45). The differential equation is solved with the boundary conditions $\theta|_{s=0} = \theta^*$, $z|_{s=0} = 0$, $\theta|_{s=1} = \pi/2$, and $z|_{s=1} = h^*$. Here, we account for the change in the apparent contact angle on a rough surface, which we denote as θ^* (SI Appendix, section 3; Fig. S6D). The height h^* represents the location across the gap where the

interface slope becomes $\pi/2$. Since only one of the plates in our cell is rough, the apparent contact angles at the top and bottom plates are different, leading to an asymmetric meniscus profile across the gap; h^* is no longer equal to the half-cell height. Instead, we determine h^* by presolving the meniscus shape across the whole channel at equilibrium (SI Appendix, section 4A). We assume that h^* is unchanged even if the capillary number changes.

The critical capillary number $Ca_{c,tf}$ predicted by Eq. 3 for $b = 0.08$ mm is shown in Fig. 3 (SI Appendix, section 4B). We note that the thin film generated near the injection port, where the front velocity is above the threshold $Ca_{c,tf}$, would not catch up and merge with the advancing fluid–fluid front in a radial cell even though the front velocity decays with distance from the center. The viscous residual oil inside the thin film drains away slowly due to the high viscosity and low interfacial driving force (see the calculations in SI Appendix, section 5). Thus, we predict the occurrence of a thin film in a radial cell experiment if the capillary number at injection is larger than the critical threshold, i.e., $Ca_{inj} > Ca_{c,tf}$ (Fig. 4B).

For very rough surfaces, i.e., when $r > r_{c,2}$ (Eq. 2), the defending fluid cannot be removed from the post pattern, as these films become energetically stable. Therefore, the complete displacement regime cannot be observed on very rough surfaces even at vanishingly small capillary numbers, as shown in Fig. 3 and Fig. 4B.

Transition to a Thick Film. As the capillary number increases, the entrained film height becomes larger than the post height, indicating a transition from the thin-film to the thick-film regime. We calculate the film thickness in this transition using the lubrication approximation—assuming that the flow in between the posts is horizontal—and that it is governed by Darcy's law (see the derivation in SI Appendix, section 6). Under this assumption, the flux through the film is still linearly proportional to the pressure gradient and hence the presence of a rough surface only modifies the coefficient of the classical Bretherton law (46). Equating the film thickness to the post height, we obtain the

critical capillary number for the onset of the thick-film regime: $Ca_{c,Tf} = \frac{\phi}{\tau} \left[\frac{b}{0.67(b+B)} \right]^{3/2}$, where $\tau = 1.8$ is the flow path tortuosity that corresponds to porosity $\phi = 0.42$ in our case (47), which remains constant for different post heights as b does not change the porosity. The result for the case $b = 0.08$ mm is plotted in Fig. 3.

Since the front velocity decays with distance from the cell center, the thickness of the entrained film also decreases in a radial cell experiment. It indicates that the thick film generated near the injection port may transition to a thin film as the fluid–fluid front advances. In our experiments, we identify the displacement regime based on the observation window, which is placed at approximately half the cell radius. In such a window, the velocity does not vary much, and we avoid observing two different displacement regimes. Thus, we require the capillary number at the radius $L/4$ to be larger than the critical value $Ca_{c,Tf}$, i.e., $Ca_{inj} \frac{4w_{inj}}{L} \geq Ca_{c,Tf}$. We then obtain the second critical injection capillary number for the observation of a thick film around the half-radius of the cell:

$$Ca_{inj,Tf} = K \left(\frac{b}{B+b} \right)^{\frac{3}{2}}. \quad [4]$$

The constant coefficient is $K = 0.67^{-3/2} \frac{\phi}{\tau} \frac{L}{4w_{inj}} \approx 4.8$. This critical Ca simultaneously depends on the varying experimental parameters b and B , in agreement with the experimental data in the phase diagram as a function of b/B and Ca_{inj} (Fig. 4C).

Phase Diagram. We plot Eqs. 1–4 in a 3-dimensional parameter space (Fig. 5) to demonstrate how the interplay between roughness factor r , vertical aspect ratio b/B , and injection capillary number Ca_{inj} govern the cross-overs between the different regimes: complete displacement, thin film (dewetting, stable, and unstable) and thick film.

The geometric parameters (b/B and r) determine the stability of the thin film: For $r < r_{c,1}$ (light blue plane), the thin film

is unstable and cannot form at any flow rate (tf-u); for $r > r_{c,2}$ (gray plane), the thin film is energetically stable and does not dewet (tf-s); and for $r_{c,2} > r > r_{c,1}$, the entrained thin film can dewet after it forms (tf-d).

The injection capillary number further controls the displacement regimes: Complete displacement (cd) transitions to the thin film (tf) when $Ca_{inj} > Ca_{inj,tf}$ (red plane) and then to the thick film (Tf) when $Ca_{inj} > Ca_{inj,Tf}$ (green plane). $Ca_{inj,tf}$ is vanishingly small in the tf-s region. In the tf-u region, $Ca_{inj,tf}$ indicates the transition from complete to incomplete displacement, where thin films do not form due to the geometric constraint $r < r_{c,1}$.

Residual Fluid Morphology at Late Times. The different regimes in the phase diagram (Fig. 5) lead to distinct trapped residual fluid morphologies at late times (Fig. 6).

Residual microdroplets and throat-bridges (Fig. 6A) and residual lubrication films (Fig. 6B) are observed in the thin-film dewetting regime. Residual microdroplets and throat-bridges form as a result of snap-off events on the edge at the base of the cylindrical posts when the dewetting front of the thin film retracts through the throats. A residual lubrication film is an isolated cluster that is confined within the post pattern, while the top of the post is exposed to the invading liquid. It is formed when the connection between the thin film and the bulk oil is cut off by dewetting and later the thin film retracts. When the thin film is stable (tf-s in Fig. 5), dewetting does not occur and the lubrication film remains within the roughness in the whole cell (Fig. 6C).

Residual liquid caps (Fig. 6D) and residual liquid bridges (Fig. 6E) are observed in the thick-film dewetting regime during which a rim is formed to collect the retracting liquid that coats the patterned surface. A liquid cap is an isolated droplet that covers an entire post. It forms when the connection between thick film and bulk oil is cut off during dewetting and the disconnected thick film then retracts. When the oil volume in such a disconnected film is sufficiently large, the rim in the dewetting front grows to touch the smooth plate and forms a liquid bridge between the two plates.

Conclusion

In this study, we show the essential role of roughness in controlling the fluid–fluid displacement and fluid trapping in fractured media. We do so by means of laboratory experiments on patterned microfluidic cells that permit precise control over the confinement and degree of roughness of the fracture analogue. We inject a nonwetting, low-viscosity fluid (water) to displace a wetting, high-viscosity fluid (silicone oil) and identify different displacement regimes.

We demonstrate that roughness significantly lowers the critical capillary number for the onset of the wetting transition and that for sufficiently strong roughness, this wetting transition occurs at a vanishingly small capillary number. Above this threshold capillary number, we uncover a heretofore unrecognized thin-film regime in which a film of undisplaced oil is left behind entirely within the posts that conform the roughness. This thin film may remain stable or dewet with uniform thickness, deviating from the behavior of classic viscous dewetting on smooth surfaces. The entrained thin-film regime transitions to a thick-film regime at a second critical capillary number, where the liquid fully covers the rough surface. Beyond this critical capillary number, the film height increases with the capillary number according to Bretherton's law. Our theoretical

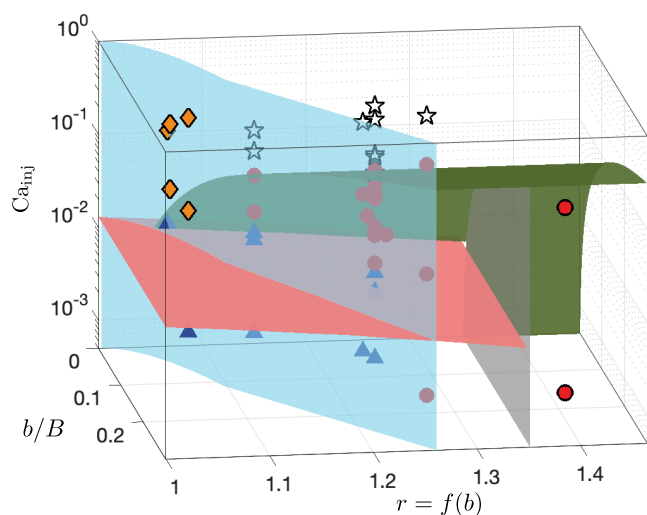


Fig. 5. Phase diagram of the different drainage regimes in a rough microfluidic cell. Shown are the transitions among the five regimes observed experimentally (cd: complete displacement with blue triangles; tf-u: thin-film unstable with yellow diamonds; tf-d: thin-film dewetting with solid red dots; tf-s: thin-film stable with circled red dots; Tf: thick film with star symbols) in the 3-dimensional parameter space defined by geometry (b/B), roughness (r) and injection capillary number (Ca_{inj}).

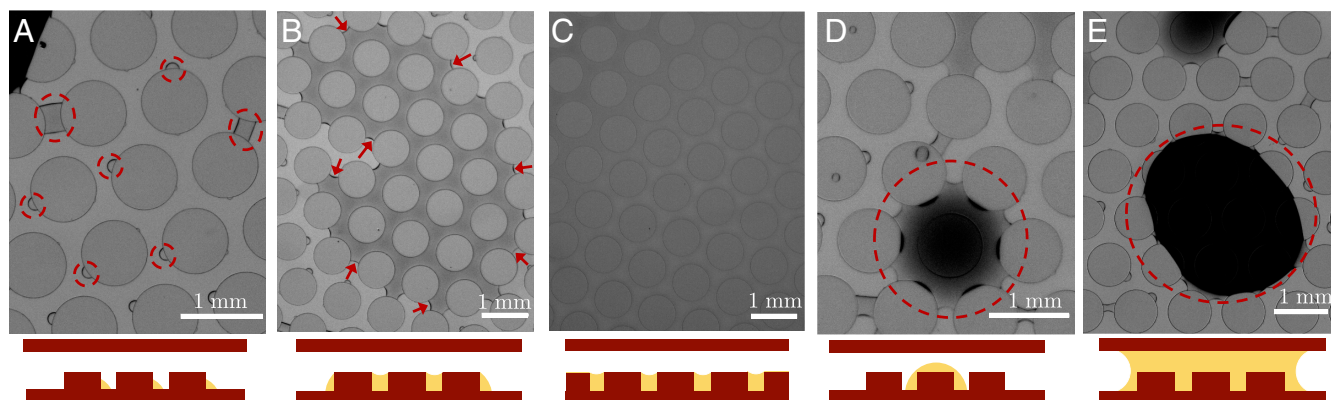


Fig. 6. Late-time morphologies of trapped defending fluid and corresponding side-view schematics. They are observed in the thin-film dewetting regime (A and B), thin-film stable regime (C), and thick-film regime (D and E), respectively. (A) Residual microdroplets attached to the corner of the post and throat-bridges connecting the neighboring posts. Experiment parameters for the image: $B = 0.5$ mm; $b = 0.08$ mm; $Ca_{inj} = 0.11$. (B) Residual lubrication film where the undisplaced oil fills a patch of pores without covering the top of the posts. Experiment parameters: $B = 0.5$ mm; $b = 0.08$ mm; $Ca_{inj} = 0.16$. (C) Residual lubrication film filling the pores of the whole cell. It forms when the thin film is energetically stable and does not dewet. Experiment parameters: $B = 0.5$ mm; $b = 0.08$ mm; $Ca_{inj} = 0.28$. (D) Residual liquid cap that covers an entire post. Experiment parameters: $B = 0.5$ mm; $b = 0.08$ mm; $Ca_{inj} = 0.28$. (E) Residual liquid bridge that joins the top and bottom surfaces and blocks flow paths locally. Experiment parameters: $B = 0.5$ mm; $b = 0.135$ mm; $Ca_{inj} = 0.14$.

arguments rationalize the cross-overs among the displacement regimes—complete displacement, thin film (unstable, dewetting, stable), and thick film—in good agreement with our experimental data.

We further show that the different displacement regimes lead to distinct residual fluid morphologies at late times: microdroplets, single-throat-bridges and lubrication films in the thin-film regime, and liquid caps and liquid bridges between plates in the thick-film regime. The distribution and morphology of the trapped fluid affect the availability of the nucleation sites and interfacial area, thus possibly controlling long-term biogeochemical reactions in porous and fracture media, which could play a profound role in natural and engineered subsurface flows like geothermal energy extraction, geological CO₂ sequestration, and hydrogen geostorage.

Materials and Methods

Materials and Methods are described in *SI Appendix*, which include 1) detailed description of the microfluidic cell fabrication process, 2) experimental method to determine the thickness of oil films via transmitted light intensity, 3) experimental characterization of the dewetting dynamics on smooth and

rough substrates, 4) experimental observation of the presence of thick oil films, 5) determination of the geometric criterion for the formation of thin films via simulations with *Surface Evolver*, 6) calculation of surface energy for different dewetting states and definition of static apparent contact angle on the rough surface, 7) determination of the meniscus shape and threshold for the wetting transition, 8) calculation of the thin-film draining velocity, 9) calculation of the transition from thin to thick film via an extension of Bretherton's law, and 10) estimation of the wetting transition and film dewetting on the smooth plate.

Data, Materials, and Software Availability. All study data are included in the article and/or [supporting information](#).

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