Impact of Wettability and Gravity on Fluid Displacement and Trapping in Representative 2D Micromodels of Porous Media (2D Sand Analogs)

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Abstract The combined effects of gravitational forces and wettability on displacement and trapping have not previously been investigated experimentally in realistic 2D porous media. Here we report the first use of representative 2D micromodels that accurately reproduce characteristic geometric, morphological, and topological properties of 3D sand packs. We use these representative micromodels to compare the wettability dependence of displacement front morphology and trapping processes using selected fluid pairs at capillary numbers between $10^{-6}$ and $10^{-5}$. The contact angles studied ranged from 30° (normal imbibition) to 150° (strong drainage). The impact of gravity was dramatic: In the horizontal flow case, only one finger developed, that is longitudinal displacement was always favored, whereas gravity created a more compact displacement front with broad lateral extension, and this lateral movement created loops, leading to by-pass trapping. As a result, the trapping efficiency rose from 7% to 29%. We observed universal scaling of the cluster size distribution demonstrating that our micromodels are statistically representative. The universal 2D-scaling exponent of 2.05 was obtained with an acceptable error smaller than 5%. Our experimental results obtained by fluorescence microscopy showed (a) precursor corner flow for weak imbibition at $\theta = 51°$, that is above the critical contact angle of $\alpha_c = 45°$ (Zhao et al., 2016, https://doi.org/10.1073/pnas.1603387113), and (b) a new wetting transition from corner flow to full duct flow under neutral wettability conditions, that is at $\theta_c \approx 90°$; and (c) above this second critical contact angle, that is under strong drainage conditions, we again observed corner flow (specifically, core-annular flow).

1. Introduction

Multiphase flows through porous media such as sand are important in natural and industrial processes including underground gas storage and enhanced oil recovery. The morphology and connectivity of the pore space, and the wettability and roughness of the pore–solid interface determine (a) the fluid-fluid patterns formed during multiphase flow, (b) the geometry of the displacement front, and (c) the capillary trapping efficiency of the defending fluid by the invading fluid. The displacement patterns formed by such flows can vary widely, ranging from stable compact displacement fronts to unstable highly ramified displacement with preferential flow paths (fingers).

The geometric characteristics of the displacement front, notably its compactness and fractality, are important in industrial processes. For example, a compact displacement front is required for oil recovery, whereas a fractal displacement front with a high trapping efficiency is required for underground gas storage.

Underground gas storage (CO₂, CH₄, H₂) is a vertical displacement process and is therefore affected by gravitational forces. However, most published studies have focused on the horizontal displacement process, neglecting the influence of gravity (e.g., Hu et al., 2017; Lenormand et al., 1988; Rabbani et al., 2017; Wang et al., 2020; Zhang et al., 2011; Zhao et al., 2016). Therefore, one objective of our study is to investigate the influence of gravity on front stabilization and trapping efficiency using a single fluid pair (water and air) while varying the wettability of the porous media by silanization (Geistlinger & Zulfiqar, 2020).

Recent research has partially explained the crossover from imbibition to drainage as the contact angle ($\theta$) increases (Ferer et al., 2004): increasing $\theta$ was shown to enhance the contribution of so-called overlaps resulting from nonlocal cooperative pore filling mechanisms that produce a compact displacement pattern.
This was demonstrated both for stable displacement (Jung et al., 2016; Singh et al., 2017) and unstable displacement (Trojer et al., 2015).

Intensive theoretical efforts were made to explain this complex fluid invasion behavior, focusing particularly on 2D horizontal displacement processes. Pore network models have been used to study quasi-static displacement (Cieplak & Robbins, 1988) and dynamic displacement (e.g., Holtzman & Segre, 2015), the Lattice Boltzmann method has been used to study stable displacement (e.g., Wang et al., 2019, 2020), and the direct Navier-Stokes solver OpenFoam was used to investigate unstable displacement (Rabbani et al., 2017).

Efforts to study this problem empirically have mainly relied on µ-CT experiments and microfluidic visualization experiments (the focus of this work). Real 3D-porous media such as glass beads, sands, and sandstone can be studied in µ-CT experiments. In principle, µ-CT experiments can only analyze the static fluid-fluid distribution or only quasi-static displacement and quasi-static trapping, because the process time scale must be much smaller than the time for a 360° scan (see Berg et al., 2013). This always means a compromise between process time scale and resolution. The advantage of micromodel experiments is that they allow direct visualization of the dynamics of displacement process. However, they are limited by their reliance on artificial 2D porous media such as circular posts with smooth surfaces (e.g., Hu et al., 2017; Jung et al., 2016; Zhao et al., 2016).

The same limitation holds for most numerical studies. Approaches in which 2D porous media are modeled as triangular networks with randomized ideal circular posts at nodes yield unrealistically high porosities (0.6–0.9), as exemplified by the works of Cieplak and Robbins (1990) (porosity = 0.6), Holtzman and Segre (2015) (porosity = 0.7), Jung et al. (2016) (porosity = 0.7–0.9), Rabbani et al. (2017) (porosity = 0.6), and Wang et al. (2019) (porosity = 0.7).

Jung et al. (2016) studied the Cieplak-Robbins transition in PDMS micromodels (porosity = 0.7–0.9, see Supporting Information S1) by using combinations of different fluids to vary the contact angle. By monitoring the evolution of the interface length and the final saturation of the defending fluid, it was shown that the crossover from compact to fractal flow occurred at a contact angle (θc) of roughly 90°. This is consistent with the results of our µ-CT study using glass beads (Geistlinger & Zulfiqar, 2020). A statistical analysis of the local displacement processes revealed that the compact displacement observed when θ < θc was due to cooperative filling events (overlaps, see Figure S13, Supporting Information S8). Interestingly, no precursor corner flow was observed for contact angles below 45° (Jung, 2015; see Figure S15d, Supporting Information S10).

Zhao et al. (2016) used large representative PDMS micromodels (porosity = 0.45, see Supporting Information S1). Their experiments demonstrated physical mechanisms including film formation and corner flow during the multiphase flow in porous media. Unexpectedly, they also found that the trend of increasingly compact displacement with decreasing θ is reversed at very low θ values because the system undergoes a wetting transition from mixed-wet imbibition (θ = 60°) to strong imbibition (θ = 7°). This dramatic change occurs at a critical contact angle, θc = 45° and is caused by precursor corner flow, which allows the invading fluid to propagate without filling the pore bodies (see Figure 1b, yellow arrows). There are thus two critical
contact angles: $\theta_c$ for the transition from compact to fractal displacement and $\alpha_c$ for the transition from duct to corner flow.

Figure 1 shows the static fluid-fluid pattern caused by corner flow (green rings) in (a) Si micromodels with irregular grain shapes (porosity $= 0.4$) and (b) precursor corner flow in glass-ceramic micromodels with circular grains (porosity $= 0.6$) under water-wet conditions ($\theta \approx 30^\circ$). In Figure 1a the static trapped gas phase (gray area) is surrounded by corner flow areas (green rings). This fractal corner flow leads to efficient trapping, as discussed by Zulfiqar et al. (2020). Figure 1b shows the complex interplay between duct flow (white areas), thick-film flow (TFF, indicated by white shadows between grains) and bridging between nearest-neighbor grains. The yellow arrows indicate the flow paths of fractal and precursor corner flow (see also Movie S1).

Intriguingly, Jung (2015) did not observe precursor corner flow at $\theta < 45^\circ$ (see Figure S15d, Supporting Information S10) despite using the same PDMS micromodels as Zhao et al. (2016).

Rabbani et al. (2017) studied the impact of wettability on fluid-fluid pattern formation in uniform-wet 2D micromodels (porosity $= 0.6$) whose microstructure was generated by taking a 2D cut of a 3D $\mu$-CT image of a sand pack (see Supporting Information S1). Their experiments are particularly interesting because the highly anisotropic and very accurate inductively coupled plasma-deep reactive ion etching (ICP-DRIE) technology they used for micromodel production is also used in this work. Their results indicate that irregular grain shapes strongly impact displacement patterns at the pore scale (see also the recent Lattice Boltzmann simulations of Wang et al., 2020). This was confirmed experimentally by Zulfiqar et al. (2020) using $\mu$-CT and micromodel experiments.

Rabbani et al. (2017) conducted 2D simulations using the open source solver OpenFoam and compared these theoretical results to those of micromodel experiments. To validate the numerical model, the numerical simulation used microstructures identical to those of the micromodel. However, this simulation leaves a key question unanswered: how realistic are numerical simulations that do not account for corner flow? Rabbani et al. (2017) demonstrated the details of the pore-scale dynamics during two phase immiscible fluid flow under intermediate-wet condition. Therefore, while compared the 2D numerical simulation results and microfluidic experiments, the reliability of their results has not been tested by comparison to data from representative micromodel experiments. In this context, the term “representative” implies two requirements:

1. The 2D microstructure should be representative of a 3D porous medium, that is important morphological and topological properties should be conserved.
2. The micromodel must be statistically representative, that is the area must be large enough for the obtained ensemble averages to be sufficiently representative for large-scale prognosis. In particular, different sections must give the same ensemble average and the influence of boundary effects must be negligible.

The precursor flow type, precursor corner flow and/or thick-film flow (see Figure 1), determines the trapping efficiency of the defending fluid (Zulfiqar et al., 2020). The occurrence of both flow types is strongly controlled by the wettability and the surface roughness. A second objective of the present study is to investigate the interplay between wettability, surface roughness and trapping efficiency using representative Si-micromodels. In particularly, we are interested at universal scaling behavior of the trapped defending fluid, because this determines macroscopic trapping behavior.

2. Material and Methods

The experimental methodology, the contact angle measurement procedure, and the method for producing Si micromodels used in this work are described in detail by Zulfiqar et al. (2020). We therefore review them only briefly here. The development of a new 3D-2D-mapping algorithm for generating representative micromodels is described in detail.
2.1. Experimental Methodology

In a first series of invasion experiments (Exp.0–Exp.5; see Table 1) the contact angle was varied over a range corresponding to normal inhibition at one extreme to strong drainage at the other by using different fluid-fluid pairs (Singh et al., 2017). In a second series (Exp.6–Exp.9), we used a single fluid pair (water and air) and varied the wettability through silanization.

It should be noted that 2 contact angles determine the wettability of the micromodel: the contact angle on the Pyrex-cover glass, denoted $\theta_{\text{glass}}$, and the contact angle on silicon, denoted $\theta_{\text{Si}}$. The values of these angles are given in brackets ($\theta_{\text{glass}}$, $\theta_{\text{Si}}$). If only one contact angle is given, it is $\theta_{\text{glass}}$. The corresponding contact angles for the invading fluid surrounded by the defending fluid were measured on flat-plate Pyrex and silicon samples (see Supporting Information S3).

For Exp.6 to Exp.9 we used silanization (Geistlinger & Zulfiqar, 2020) to achieve a neutral contact angle on silicon. The silanization reactants were injected at very low capillary numbers ($<10^{-6}$) to guarantee sufficient reaction time. The surface roughness of the silicon walls is in the sub-micrometer range (see Figure 10) and is thus orders of magnitude greater than that of the smooth Pyrex surface. Consequently, there are many more reaction sites on the walls than on the Pyrex, leading to asymmetry in the wettability of the Si-matrix and the Pyrex cover glass. As a result, there are always some thin-film water fingers in front of the invading fluid (Figure S5, Supporting Information S3). We measured the contact angle on silicon by the graphical method, which gave an average contact angle of 95.2° ± 8.9°. Because spontaneous thin film flow was observed, the contact angle of the silanized Pyrex glass must be in a range that permits such flow behavior. Therefore, we estimated $\theta_{\text{glass}} = 30^\circ$.

All experiments were conducted at capillary numbers between $Ca = 10^{-6}$ and $10^{-5}$. We assume that the flow at $Ca = 10^{-6}$ is dominated by capillary forces and that viscous forces start influencing fluid pattern formation at $Ca = 10^{-5}$. To maintain a constant flow rate during imbibition experiments, a high-precision bidirectional syringe pump with a step resolution of 0.046 microns (Fusion 200, Chemyx) was used. Water was used as the wetting fluid and air as the nonwetting fluid.

For visualization, we used a Canon EOS7D SLR camera with a 100-mm Macro lens (Canon EF 100-mm F2.8 USM Macro lens) in both single-shot mode (spatial resolution: 5184 × 3456 pixel) and video mode at 24 frames/sec (spatial resolution: 1920 × 1080 pixel). The camera was used in combination with a fluorescence microscope (Leica Leitz DMRB microscope; LM digital SLR wide-field adapter, micro-tech-lab Ltd.). Uranine and Oil red dye were used as fluorescent tracers.

### Table 1

**Contact Angles and Capillary Numbers Used in the Conducted Experiments**

| Exp.0 ($Ca = 10^{-6}$) | Exp.0 ($Ca = 10^{-5}$) | Exp.1 | Exp.3 | Exp.4 ($Ca = 10^{-6}$) | Exp.4 ($Ca = 10^{-5}$) | Exp.5 |
|------------------------|------------------------|-------|-------|------------------------|------------------------|-------|
| Invading fluid         | Water                  | Water | Glycerin* | Air | Heptane* | Heptane* | Water |
| Defending fluid        | Air                    | Air   | 61    | 119 | 131[4] | 131[4] | Water |
| Contact angle on Si (*[b]) | 47          | 47    | 61    | 119 | 131[4] | 131[4] | 112  |
| Contact angle glass ([*]) | 28          | 28    | 51    | 129 | 150[4] | 150[4] | 84   |
| Capillary number inv. fluid | 10^{-6}  | 10^{-5} | 10^{-5} | 10^{-5} | 10^{-6} | 10^{-5} | 10^{-5} |
| Capillary number inv. fluid$^d$ | 1.5 × 10^{-6} | 1.5 × 10^{-5} | 1.8 × 10^{-5} | 1.8 × 10^{-5} | 1.5 × 10^{-6} | 1.3 × 10^{-5} | 1.8 × 10^{-6} |
| Displacement velocity (cm/s) | 0.72 × 10^{-2} | 7.2 × 10^{-2} | 1.3 × 10^{-2} | 1.3 × 10^{-2} | 1.3 × 10^{-2} | 1.1 × 10^{-1} | 2.1 × 10^{-2} |

Note. Contact angles were measured by the droplet method using droplets of the invading fluid (see Supporting Information S3).

*95 wt.% glycerin in water. $^b$See Table S1. "$^\text{n}$-heptane. $^c$Silicon oil 100 cP. $^d$Including \cos{\theta} for capillary forces (Ferer et al., 2004). $^e$The 180°-complement angle was taken from the measured contact angle of a droplet water in heptane. $^f$The capillary number and displacement velocity are calculated based on draining the defending fluid.
2.2. Production of Silicon Micromodels

A microfabrication method involving photolithography, ICP-DRIE and anodic bonding was used to fabricate the micromodel in a silicon wafer. Anisotropic edging was performed using an interval-based ICP-DRIE-technology (Küchler et al., 2003; Zue et al., 2013) that delivered high edge steepness and a true mapping of the lattice structure with depth (Figure 2b). Under-etching was minimal, with a deviation of 1.2° from the vertical line and a maximal deviation of 10 μm from the horizontal bottom line. This 1.2° deviation was achieved for a microstructure 9 times deeper than that prepared by Zue et al. (2013). The production process is described in detail in an earlier publication (Geistlinger et al., 2019).

2.3. Microstructures: A 3D-2D Mapping Algorithm

The experiments were conducted in a 2D stochastic pore space structure with similar morphological, topological, and geometric properties to 3D natural sand packs. We therefore call this microstructure a 2D sand analog. Details of this mapping algorithm are described in Zulfiqar et al. (2020). Figures 2a and 2b show sections of the developed MM4 micromodel in which its irregular pore space structure can be seen. The dimensions of the micromodel were 80 × 80 × 0.3 mm.

3. Results and Discussion

3.1. Impact of Wettability on Fluid Pattern: Transition From Corner Flow to Duct Flow

Figure 3 shows the wettability dependence of the horizontal fluid pattern at contact angles ranging from weak imbibition (a): \( \theta = 51^\circ \) to strong drainage (e): \( \theta = 150^\circ \). Under the drainage conditions imposed in Exp.3 (c) and Exp.4 (e), the fluid pattern clearly changes from viscous fingering (fractal dimension \( D_f = 1.61 \pm 0.02 \), accepted value is 1.62; Maloy et al., 1985) to capillary fingering (\( D_f = 1.80 \pm 0.02 \), accepted value is 1.82; Wilkinson & Willemsen, 1983). This is consistent with the classical \( M-Ca \) phase diagram.
shown in Figure 4 \( M = \mu_{inv}/\mu_{def}, \) Lenormand et al., 1988). The neutral case, represented by Exp.5 (b), is also consistent with the classical phase diagram, showing some transitional behavior between viscous and capillary fingering with a fractal dimension of 1.73 ± 0.02, which is between the two limiting values. In the weak imbibition case (a) there is a stable displacement front with some trapping of the defending phase. Physically, one would expect stable displacement when a more viscous fluid (e.g., glycerin) displaces a less viscous one (e.g., air). Note that under all other wetting conditions (Figures 3b–3e), the less viscous fluid displaces the more viscous one, leading to unstable displacement.

Figure 5 shows the wettability dependence of the fluid pattern at the pore scale. The trapped ganglia-like clusters of the defending fluid, for example, the green-colored glycerin clusters for Exp.3 (Figure 5c), can be seen clearly. Additionally, inspection of the trapped air clusters (black pores) in the weak imbibition case \( (\theta = 51°) \) shown in Figure 5a shows that they are surrounded by green glycerin films, which may be due to precursor corner flow (for a detailed discussion of this flow type and trapping mechanism see Zulfiqar et al., 2020). This is interesting because it contradicts the accepted picture that corner flow can only occur for contact angles below 45°.

Zhao et al. (2016) observed this interesting wetting transition from corner flow to duct flow in 2D PDMS micromodels exhibiting unstable displacement of oil by water (340 cP). At capillary numbers on the order of \( 3 \times 10^{-3} \) (using the \( \cos\theta \) definition; see the bottom row of their Figure 2), increasing the contact angle in these micromodels from \( \theta = 7° \) to \( \theta = 60° \) (corresponding to strong and weak imbibition, respectively; see Figures
20 and 21) causes a wetting transition from fractal corner flow with capillary fingering to full duct flow with compact displacement. Increasing the contact angle further results in a more fractal displacement front with no corner flow.

Our fluorescence microscopy observations (Figure 6) deviate from this well-accepted picture in several respects:

1. We observed corner flow under weak imbibition conditions ($\theta = 51^\circ$, Figure 6b) when the contact angle was above the critical value of $\alpha_c = 45^\circ$.

Figure 6. Wetting transitions from fractal corner flow ($\theta_{\text{glass}} = 28^\circ$, $2 \times 10^{-6}$), to more compact corner flow ($51^\circ$, $10^{-5}$), to duct flow ($84^\circ$), and to core-annular flow ($150^\circ$). The white arrows show the flow direction of the pore-scale fluid fingers.
2. We observed a new wetting transition under neutral wettability conditions, that is at $\theta = 84^\circ$ (Figure 6c), from corner flow to full duct flow (compare to Figure 2i of Zhao et al., 2016).

3. Above this second critical contact angle ($\theta_c$) of about $90^\circ$, that is for weak and strong drainage (weak and strong oil wet), we again observed corner flow, or more precisely, core-annular flow. In this case, the fingers of the invading nonwetting fluid flow in the middle of the pore channel, that is within the core (indicated by white arrows in Figure 6d), while the more viscous wetting fluid flows (or remains) at the pore corners/walls, that is in the annular ring of the pore channel.

To elucidate the physical reasons for these unexpected observations, we discuss the pore-scale displacement processes occurring under the different wetting conditions shown in Figure 6: i) normal imbibition (Exp.0), ii) weak imbibition (Exp.1), iii) neutral wettability (Exp.5), and iv) strong drainage (Exp.4). In the following we discuss the three interesting cases: the weak-imbibition case, the neutral-wettability case, and the strong-drainage case. The normal imbibition case is discussed in the Supporting Information S11.

### 3.1.1. Weak Imbibition

The intermediate wet case ($\theta_{\text{glass}} = 51^\circ$, $\theta_{\text{Si}} = 61^\circ$) with glycerin displacing air (Exp.1)

Upon inspecting Figure 6a, we suspected that the green rings around the trapped air cluster were due to corner flow (CF) during invasion. This is proven by Figure 6b. As shown in the time series $t_1$ to $t_4$ in Figure 7, the precursor corner flow invades the porous media first and is immediately followed by duct flow (see also Movie S3).

In contrast to the normal wet case, both contact angles are larger than $45^\circ$, giving rise to an unstable meniscus (Figure 8a). Therefore, CF is physically not possible even though the experimental data imply the occurrence of precursor CF. These findings are reconciled by the results presented in Figure 9: the anisotropic ICP-DRIE-etching process (for details see Geistlinger et al., 2019) creates vertical and horizontal grooves capillaries (Constantinides & Payatakes, 2000), giving rise to wall roughness in the sub-micrometer range. According to Wenzel's argument (Wenzel, 1936), surface roughness amplifies both hydrophilicity and hydrophobicity. Consequently, a high degree of surface roughness can lead to complete wetting for contact angles smaller than $90^\circ$. This was theoretically and experimentally demonstrated by Zulfiqar et al. (2020).

According to a realistic wetting model derived by modifying Wenzel's model, the complete-wetting condition (Zulfiqar et al., 2020, Section 4.1.1.) is given by:

$$\theta_0 = \theta_{\text{Si}} < \theta_c, \quad \text{with} \quad \theta_c = \arccos \left( \frac{1 - \phi_s}{r - \phi_s} \right),$$

(1)

where $\theta_0$ is the equilibrium contact angle of the liquid on an ideal flat Si surface of the same chemical composition (i.e., the intrinsic contact angle) and $\phi_s$ is the dry part of the surface area. If water flows only along the valleys of the grooves, the hills define $\phi_s$. The physical meaning of Equation 1 is that for all $\theta_0 < \theta_c$, the TFF advances as long as the entire surface is covered by a water film. The range of the critical contact angle is determined by the degree of surface roughness $r$ (the ratio of rough surface area to flat surface area). Assuming that the cross sections of the grooves can be approximated by equilateral triangles, the rough surface area is twice the flat area. Inserting $r = 2$ into Equation 1 and assuming that the grooves are completely covered by a thick film flow ($\phi_s = 0$), one obtains $\theta_c = 60^\circ$. This is a striking result because it means that complete wetting occurs even at the relatively large intrinsic contact angle of $60^\circ$. This can be explained physically by recalling the Young-Laplace Force...
Free Energy diagram and noting that the force along the glycerin-air interface is twice that along the glycerin-Si interface. In mechanical equilibrium, this requires that \( \cos \theta_0 = \frac{1}{2} \). However, if the effective interface area of the glycerin-Si interface is doubled due to surface roughness, mechanical equilibrium is only possible if \( \cos \theta = 1 \) (\( \theta = 0^\circ \)), corresponding to complete wetting. Consequently, corner flow can be observed when \( \theta_{\text{glass}} = 51^\circ \) as shown in Figure 8b.

### 3.1.2. Neutral Wettability

The neutral wet case (\( \theta_{\text{glass}} = 84^\circ, \theta_{\text{Si}} = 112^\circ \)) with water displacing silicon oil (Exp.5)

The most interesting feature of the neutral wet case is that CF is absent and only duct flow occurs (Figure 6c). The tips of the green fingers always end with convex menisci, indicating contact angles above 90°. Naively, one would expect surface roughness to cause complete wetting and that CF would occur for \( \theta_{\text{glass}} = 84^\circ \) as shown in Figure 8b. However, now Wenzel’s argument works in the other direction: when \( \theta_{\text{Si}} > 90^\circ \), surface roughness amplifies hydrophobicity. In the Young-Laplace force diagram, the force along the water-Si interface that acts to draw the 3-phase boundary inwards becomes stronger as the surface roughness increases. This leads that to an apparent contact angle \( \theta_{\text{Si,app}} \) that is larger than the intrinsic one (\( \theta_{\text{Si,0}} = 112^\circ \)). Because \( \theta_{\text{glass}} \) is \( \sim 90^\circ \) and TFF and CF do not occur, \( \theta_{\text{Si,app}} \) can be observed directly (Figure 8b).

We measured the geometrical contact angles at 45 locations (see Figure S6, Supporting Information S3) in the dynamic case (during invasion) and the static case after stopping injection, obtaining values of \( \theta_{\text{Si,app}} \) (dynamic) = 136° ± 16° and \( \theta_{\text{Si,app}} \) (static) = 137° ± 14°. These results support Wenzel’s argument. Furthermore, the dynamic and static contact angles are almost identical, indicating that the convex curvature of the menisci is a geometric phenomenon resulting from pore angularity (compare with Rabbani et al., 2017) rather than viscous forces (Hagen-Poiseuille velocity profile).

### 3.1.3. Strong Drainage

The oil wet case (\( \theta_{\text{glass}} = 150^\circ, \theta_{\text{Si}} = 131^\circ \)) with heptane displacing water (Exp.4)
Increasing the contact angle of the invading fluid into the strong drainage regime (150°/131°) restored corner flow (Figure 6d). However, in this case the less viscous invading fluid (heptane) flowed in the center of the pore channels to minimize flow resistance, while the more viscous fluid (water) flowed along the walls and corners because of the lower water pressure (or equivalently, the higher capillary pressure) there. This type of flow is known as core-annular flow in fluid mechanics (Joseph & Renardy, 1993).

Based on these experimental observations, we have to comment on the experiments reported by Rabbani et al. (2017), Zhao et al. (2016), and Jung et al., (2016):

Rabbani et al. (2017) conducted experiments using Si-micromodels that were generated using the ICP-DRIE-technology also used in this work and covered with Pyrex glass. They thus closely resemble the micromodels used in our experiments and would be expected to exhibit the same flow types (i.e., CF and TFF) resulting from surface roughness. However, the only flow type considered in the corresponding 2D numerical simulations was duct flow; CF and TFF were neglected. Therefore, the results of these experiments cannot be used to validate the 2D numerical simulations other than for the neutral wet case, where only duct flow occurs (not shown in their Figure 2a, Rabbani et al., 2017).

The displacement and trapping behavior observed in artificial porous media with ideal circular grains and smooth surface (soft PDMS micromodels) differs significantly from that in realistic porous media with irregular and rough grains (Zulfiqar et al., 2020). Zhao et al. (2016) studied soft polymer (PDMS) micromodels, for which the 45° constraint for CF is valid. TFF and complete wetting do not occur in such micromodels; instead, DF is preceded by CF because of the relatively high capillary pressure. Grains are connected via CF, which “searches” for the nearest neighbor (i.e., the grain to which bridging is easiest). The actual bridging occurs via thin film flow in PDMS micromodels and thick film flow in Si and glass ceramic micromodels (see Movie S2). To snap-off the bridge and achieve complete pore throat filling, the geometrical snap-off condition must be satisfied (see Figure 10 in Zulfiqar et al., 2020). This condition is only satisfied if the precursor corner flow (Figure 2o in Zhao et al., 2016) follows the path of lowest grain-grain distances. We demonstrate this by matching the fluid pattern to the spatial grain distribution (see Figure S12, Supporting Information S7). If the throat width is excessive, snap-off (and thus bridging) becomes impossible. This was demonstrated by Zulfiqar et al. (2020) for a stochastic quadratic micromodel in which the front advance is initiated by a Haines jump at a specific position that is followed by horizontal front advance (see also

![Figure 10](image-url). Gravity causes a transition from unstable horizontal to stable vertical water-air displacement at different capillary numbers ($10^{-6}, 10^{-5}$). Upper row (a–d): neutral wet case; lower row (e–h): normal wet case ($D_1$ – fractal dimension, $S_{trap}$ – trapped saturation of defending phase). The insets in Figures (a and e) show the displacement types occurring at the pore scale: duct flow fingering and fractal corner flow. The spatial distribution of trapped clusters for the normal wet case is shown in Figure S7b of the Supporting Information S4.
Because of this geometric constraint, high porosity microstructures (0.65–0.85) such as those studied by Jung et al. (2016) and Jung (2015; see Figure S15, Supporting Information S10), will hinder bridging and precursor corner flow. This explains why Zhao et al. (2016) observed CF but Jung et al. (2016) did not even though both groups used soft PDMS micromodels: the geometric snap-off condition was satisfied in the first case but not in the second. In this context, Jung et al. (2016) made the statement that they discarded the experimental analysis of fluids with small contact angles $\theta < 45^\circ$, because of spreading the invading fluid along the edges of the cell. It is interesting because it strongly suggests that corner flow did in fact occur along the edges of the PDMS cover glass in their micromodel.

3.2. Impact of Gravity on Fluid Pattern

In this section we discuss the influence of gravity on front stabilization and trapping efficiency using a single fluid pair (water and air) while varying the wettability of the Si micromodel by silanization (Geistlinger & Zulfiqar, 2020). This creates contact angle asymmetry because silanization occurs more extensively on the rough Si walls than on the smooth Pyrex cover glass ($\theta_{\text{glass}} = 30^\circ, \theta_{\text{Si}} = 95^\circ$; Exp.6).

In Figure 10 we compare the horizontal and vertical water-air displacement processes for two capillary numbers ($10^{-6}$ and $10^{-5}$) and two wetting cases: the normal wet case (Exp.0: $\theta_{\text{glass}} = 28^\circ, \theta_{\text{Si}} = 47^\circ$) and the neutral wet case (Exp.6). These experiments show how the interplay of capillary, viscous and gravitational forces affects the displacement process and the resulting fluid patterns. The displacement is stable because the more viscous water displaces the less viscous fluid air. The position of Exp.6 in the $M$-$Ca$-phase diagram (Figure 4) is the same as for Exp.0. The corresponding fluid pattern of Exp.0 at breakthrough are shown in Figure S7a (Supporting Information S4).

When the capillary number was low (Ca = $10^{-6}$) under normal wet conditions, the impact of gravity was dramatic (compare the first column of Figures 10a and 10e to the third column (c and g)). In the horizontal flow case, only one finger developed, that is longitudinal displacement was always favored. The positive values of modified Bond number (Blunt & Scher, 1995), $1.8 \times 10^{-2}$ for vertical and 0 for horizontal cases, are consistent with the classical instability analysis (Chuoke et al., 1959; Ferer et al., 2004; Lenormand et al., 1988): gravity created a more compact displacement front with broad lateral extension, and this lateral movement created loops, leading to by-pass trapping.

As a result, the trapping efficiency ($S_{\text{trap}}$, i.e., the saturation of the trapped defending fluid) rose from 7% to 29%.

Increasing the capillary number to $10^{-5}$ (compare the second column of Figures 10b and 10f to the fourth column (d and h)), that is strengthening the viscous forces, caused more fingering (resulting in two fingers instead of one) in the horizontal case and less compact displacement in the vertical case (there was insufficient time for lateral movement). However, the fluid patterns in the two cases still differ significantly.

Wettability also had a significant impact, as demonstrated by considering the horizontal displacement at Ca = $10^{-5}$: by comparing the bottom and top row of images, it can clearly be seen that the fluid patterns were more compact in the normal wet case. This is indicated by the transition of the fractal dimension from values near 1.7 (viscous fingering) to values of 1.8 (corresponding to more compact capillary fingering). These findings show that the results of visualization studies on horizontal displacement pattern cannot be used to draw conclusions about vertical flow processes if gravitational forces are comparable in strength to capillary forces.

3.3. Representative Micromodels and Universal Scaling

Almost all micromodel studies and numerical studies on the impact of wettability on fluid displacement claim that their results are of high practical relevance for geo-sequestration, enhanced oil recovery, and ground water remediation. However, a key question is whether the domains of such models are statistically representative. To answer this question, we consider as an instructive example the displacement process at low capillary numbers (Ca = $10^{-6}$), where invasion percolation governs the advancement of the front. The basis of our analysis is the spatial air-cluster distribution (Exp.0, normal wet case) shown at breakthrough in Figure 11 and at an earlier time in Figure 10g.
It is well-known that the size of the representative elementary volume (REV) or window (REW) is process-dependent. For static quantities such as the porosity, a relatively small window size is sufficient (a few grain diameters in the case of glass bead packs). Process characteristics such as the permeability, the fractal dimensions of fluid patterns, and universal scaling necessitate the use of large REWs to capture fluid patterns and relevant large trapped clusters (see Stauffer & Aharony, 1994). In other words, the REW must be large enough to make boundary effects insignificant and prevent artificial disruption of fluid patterns.

We begin by analyzing the saturation using three different window sizes (w1, w2, and w3; see the red windows in Figure 11). Even with the smallest window, w3, the saturation was approximated with reasonable accuracy; the obtained value of 34% differs by only 8% from that obtained with the largest window, w1 (37%). The mean and standard deviation (SDV) of the saturation over all three windows was 35% ± 1%.

We next considered four smaller windows (R1-R4) with the same dimensions as the micromodel and modeling domain used by Rabbani et al. (2017) (see Supporting Information S1). The resulting saturation values, which are shown in the top left corner of Figure 11, deviate significantly (by around 20%) from the correct value of 37%. This micromodel is thus too small to capture the REV saturation of the vertical water-air displacement process.

Finally, we studied the characteristics of the invasion process. At the percolation threshold $p_c$ (i.e., the terminal point at which the non-wetting/defending fluid becomes disconnected; Blunt & Scher, 1995) and for large clusters, the normalized cluster number decayed in accordance with a universal power law with a Fisher exponent of $\tau = 2.05$ in 2D (Fisher, 1967). For a detailed discussion of this behavior, see Geistlinger et al. (2015). MSE fits (MSE = Mean Square Error estimator) give almost identical scaling exponents for all 3 windows (2.05) with acceptable errors smaller than 5% (see Figure S7c, Supporting Information S4).

These results indicate two things: (a) the vertical invasion process at $Ca = 10^{-6}$ is controlled by capillary forces and gives rise to a fluid pattern characteristic of a 2D displacement process, and (b) the window w3 is large enough to ensure that the important large clusters are captured.

This argument and the preceding saturation analysis clearly show that the R-windows are too small for testing the universal scaling of large trapped air clusters.

We note that recent studies of cluster size distribution often use Maximum-Likelihood (ML) estimators (Clauset et al., 2009; Goldstein et al., 2004; Iglauer & Wülling, 2016) based on the argument that standard MSE fitting is significantly biased compared to ML estimators. We have shown that this bias is often due to the log-log-representation of data, which assigns high statistical weight to small clusters that occur more frequently than large ones. For a critical discussion of MSE and ML estimators, see Geistlinger and Zulfiqar (2020).

The main conclusion of this section is that no reliable conclusions about displacement mechanisms and pore level dynamics can be drawn from numerical and micromodel studies that (a) examine a non-representative pore structure, (b) consider only duct flow and not the important precursor corner flow, and (c) focus solely on horizontal displacement processes rather than their industrially relevant vertical counterparts.
4. Summary and Conclusions

Using Si micromodels that reproduce characteristic geometric, morphological, and topological properties of 3D sand packs, we conducted a comparative micromodel study on the impact of wettability and gravity on displacement front morphology and the trapping process at capillary numbers of $10^{-6}$ to $10^{-5}$. The contact angles were varied by using different fluid pairs ($30^\circ$–$150^\circ$) and by silanization of the micromodel ($30^\circ$ and $90^\circ$).

Our main conclusions are:

1. For industrial applications such as underground gas storage and enhanced oil recovery, it is important to study vertical rather than horizontal displacement processes. We demonstrated this through an illustrative study of a vertical water-air displacement process under normal wet conditions at a capillary number of $10^{-6}$. The impact of gravity was dramatic: In the horizontal flow case, only one finger developed, that is longitudinal displacement was always favored, whereas gravity created a more compact displacement front with broad lateral extension, and this lateral movement created loops, leading to by-pass trapping. As a result, the trapping efficiency rose from 7% to 29%.

2. For the vertical displacement process we observed universal scaling of the cluster size distribution demonstrating that our micromodels are statistically representative. The universal 2D-scaling exponent of 2.05 was obtained with an acceptable error smaller than 5%.

3. Precursor corner flow is an immanent flow characteristic of natural porous media consisting of irregular grains with rough surfaces. Irregularity of shape and surface roughness give rise to interesting fluid patterns on both the pore and REV scales as wettability changes. These phenomena are triggered by two physical conditions: (a) the complete wetting condition and (b) the geometrical (snap-off) instability condition. We showed that bridging occurs exclusively via thick-film flow (in rough-surfaced media) if the complete wetting condition is satisfied. Conversely, in smooth-surfaced PDMS models, bridging is initiated by thin-film flow over the distance between adjacent grains. This requires strong wettability of the smooth surface (e.g., $\theta < 45^\circ$). We emphasize that bridging (i.e., a Haines jump over two adjacent grains) also requires the geometrical snap-off condition to be satisfied. Both elementary front displacement processes lead to precursor fractal corner flow in Si micromodels produced using ICP-DRIE.

4. In contrast to the well-accepted picture that corner flow occurs at a critical contact angle of $45^\circ$, we observed for the first time a new wetting transition from corner flow to full duct flow under neutral wettability conditions (i.e., at $\theta_c \approx 90^\circ$). Corner flow (more precisely, core-annular flow) was restored above this second critical contact angle, that is under weak and strong drainage conditions. We showed that surface roughness in the sub-micrometer range is responsible for this new phenomenon. Wenzel's argument states that surface roughness amplifies both hydrophilicity and hydrophobicity. Therefore, a realistic surface area doubling due to roughness can reduce the apparent contact angle to zero, causing complete wetting. In the opposite case, that is for hydrophobic Si surfaces (neutral wet case, $\theta_{Si} = 112^\circ$), Wenzel's argument implies that the contact angle increases to $137^\circ$ (geometrical proof).

5. Wenzel's argument is not valid for soft PDMS micromodels with smooth surfaces. Therefore, the new phenomena mentioned above will not be observed in such systems. However, the geometric snap-off condition must still be satisfied for precursor fractal corner flow. A counterexample is provided by the experimental study of Jung et al. (2016), in which high porosity (0.7–0.9) hindered bridging of adjacent grains.

6. Reliable predictions of fluid pattern formation on the REV scale and related properties (e.g., recovery and trapping efficiency) can only be obtained on the basis of representative micromodel experiments or numerical simulations that include the important precursor corner flow as well as duct flow.

Future representative micromodel studies should (a) investigate other technologically relevant natural porous media such as sandstones and (b) evaluate the reliability of recent 2D Lattice Boltzmann simulations (see Wang et al., 2019, 2020) that neglect precursor corner flow.

Data Availability Statement

The data can be accessed at https://www.ufz.de/index.php?en=39791.
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