We report an approach to fully visualize the flow of two immiscible fluids through a model three-dimensional (3-D) porous medium at pore-scale resolution. Using confocal microscopy, we directly image the drainage of the medium by the nonwetting oil and subsequent imbibition by the wetting fluid. During imbibition, the wetting fluid pinches off threads of oil in the narrow crevices of the medium, forming disconnected oil ganglia. Some of these ganglia remain trapped within the medium. By resolving the full 3-D structure of the trapped ganglia, we show that the typical ganglion size, as well as the total amount of residual oil, decreases as the capillary number Ca increases; this behavior reflects the competition between the viscous pressure in the wetting fluid and the capillary pressure required to force oil through the pores of the medium. This work thus shows how pore-scale fluid dynamics influence the trapped fluid configurations in multiphase flow through 3-D porous media.

Keywords: fluid mechanics and transport phenomena, porous media, multiphase flow, permeability, capillarity, wetting
densely packed hydrophilic glass beads, with polydispersity \( \approx 4\% \), in thin-walled rectangular quartz capillaries (Figure 1a); these have cross-sectional areas \( A \approx 1 \times 1 \) mm\(^2\) or \( 1 \times 3 \) mm\(^2\). The beads have average radius \( a=75 \) or 32 \( \mu \)m; the media thus have lateral dimensions spanning from approximately 7 to 50 beads. Scattering of light from the interfaces between the wetting and nonwetting fluids, as well as from the interfaces between the fluids and the beads, typically precludes direct observation of the multiphase flow in 3-D. We overcome these limitations by matching the refractive indices of the wetting fluid, the nonwetting oil, and the beads, enabling full visualization of the multiphase flow in 3-D. We formulate a wetting fluid comprised of a mixture of dimethyl sulfoxide and water at 91.4 and 8.6% by weight, respectively; to visualize this fluid using confocal microscopy, we add 0.01 vol % fluorescein dye buffered at pH 7.2. Additionally, we formulate another nonwetting fluid comprised of a mixture of aromatic and aliphatic hydrocarbon oils (Cargille). These mixtures are designed to closely match the refractive indices of the wetting fluid and the nonwetting fluid to each other and to the refractive index of the glass beads. The wetting and nonwetting fluids have densities \( \rho_w=1.1 \) g/cm\(^3\) and \( \rho_{nw}=0.83 \) g/cm\(^3\), respectively; the flow through the porous medium is horizontal. The interfacial tension between the fluids is \( \gamma=13.0 \) mN/m, as measured using a du Noüy ring; this value is similar to the interfacial tension between crude oil and water.26 The viscosities of the wetting and nonwetting fluids are \( \mu_w=2.7 \) mPa s and \( \mu_{nw}=16.8 \) mPa s, respectively, as measured using a strain-controlled rheometer; thus, our experiments are characterized by a viscosity ratio \( M \equiv \mu_w/\mu_{nw} \approx 0.2 \). We use confocal microscopy to estimate the three-phase contact angle made between the wetting fluid and a clean glass slide in the presence of the nonwetting fluid, \( \theta \approx 5^\circ \). For some of the results reported here, we use a wetting fluid comprised of a mixture of dimethyl sulfoxide, benzyl alcohol, ethanol, and water at 55.8, 21.4, 10.4, and 12.3% by weight, respectively.

\[ a \approx 75, \quad 32 \mu m \]

Because of the limited lateral size of the porous media, we note that boundary effects may influence the results. Future work is required to elucidate the role played by boundaries. However, we observe similar behavior to that reported here for pores near the boundaries.

Experimental Methodology

We prepare rigid 3-D porous media by lightly sintering\(^{16}\) densely packed hydrophilic glass beads, with polydispersity \( \approx 4\% \), in thin-walled rectangular quartz capillaries (Figure 1a); these have cross-sectional areas \( A \approx 1 \times 1 \) mm\(^2\) or \( 1 \times 3 \) mm\(^2\). The beads have average radius \( a=75 \) or 32 \( \mu \)m; the media thus have lateral dimensions spanning from approximately 7 to 50 beads. Scattering of light from the interfaces between the wetting and nonwetting fluids, as well as from the interfaces between the fluids and the beads, typically precludes direct observation of the multiphase flow in 3-D. We overcome these limitations by matching the refractive indices of the wetting fluid, the nonwetting oil, and the beads, enabling full visualization of the multiphase flow in 3-D. We formulate a wetting fluid comprised of a mixture of dimethyl sulfoxide and water at 91.4 and 8.6% by weight, respectively; to visualize this fluid using confocal microscopy, we add 0.01 vol % fluorescein dye buffered at pH 7.2. Additionally, we formulate another nonwetting fluid comprised of a mixture of aromatic and aliphatic hydrocarbon oils (Cargille). These mixtures are designed to closely match the refractive indices of the wetting fluid and the nonwetting fluid to each other and to the refractive index of the glass beads. The wetting and nonwetting fluids have densities \( \rho_w=1.1 \) g/cm\(^3\) and \( \rho_{nw}=0.83 \) g/cm\(^3\), respectively; the flow through the porous medium is horizontal. The interfacial tension between the fluids is \( \gamma=13.0 \) mN/m, as measured using a du Noüy ring; this value is similar to the interfacial tension between crude oil and water.26 The viscosities of the wetting and nonwetting fluids are \( \mu_w=2.7 \) mPa s and \( \mu_{nw}=16.8 \) mPa s, respectively, as measured using a strain-controlled rheometer; thus, our experiments are characterized by a viscosity ratio \( M \equiv \mu_w/\mu_{nw} \approx 0.2 \). We use confocal microscopy to estimate the three-phase contact angle made between the wetting fluid and a clean glass slide in the presence of the nonwetting fluid, \( \theta \approx 5^\circ \). For some of the results reported here, we use a wetting fluid comprised of a mixture of dimethyl sulfoxide, benzyl alcohol, ethanol, and water at 55.8, 21.4, 10.4, and 12.3% by weight, respectively.

Unlike other imaging approaches like MRI or X-ray \( \mu \)CT, our approach does not enable full 3-D imaging of optically opaque systems in which the refractive indices of the fluids are not matched to that of the glass beads.

![Figure 1. Overview of the experimental approach.](image-url)
and a nonwetting fluid comprised a different mixture of the aromatic and aliphatic hydrocarbon oils (Cargille); these proportions closely match the refractive index of the glass beads and the nonwetting fluid, and are characterized by $\rho_w = 1.05$ g/cm$^3$, $\rho_{nw} = 0.82$ g/cm$^3$, $\mu_w = 2.2$ mPa s, $\mu_{nw} = 7.4$ mPa s, and $\gamma = 27.3$ mN/m.

We instrument the porous media to enable measurement of the bulk transport properties simultaneously with flow visualization. We use a differential pressure sensor to measure the pressure drop $\Delta P$ across a porous medium prior to and during visualization of the flow using confocal microscopy. We vary the volumetric flow rate $Q$ and measure the proportionate variation in $\Delta P$; this enables us to determine the absolute permeability of a medium with length $L$ and cross-sectional area $A$, $k \equiv \mu_w (QL/A)/\Delta P$. The permeability of a disordered packing of spheres can be estimated using the Kozeny–Carman relation, $k = a^2 \varphi^4/(5(1-\varphi)^2)$, where $\varphi$ is the porosity of the packing and $a$ is the average sphere radius. The dependence of the measured permeability on bead size is consistent with $k \sim a^2$, in agreement with this prediction. Moreover, for a medium with bead radius $a = 75$ $\mu$m and $\varphi = 0.41\%$, we expect $k = 25$ $\mu$m$^2$; we find $k \approx 75$–95 $\mu$m$^2$, in reasonable agreement with our expectation. The discrepancy between the measured permeability and the theoretical prediction is unclear; it may, for example, arise from the effect of the capillary walls confining our porous media.$^4$

We exploit the close match between the refractive indices of the fluorescently dyed wetting fluid and the glass beads to visualize the structure of the porous medium in 3-D. Prior to each experiment, the porous medium is evacuated under vacuum and saturated with CO$_2$ gas; this gas is soluble in the wetting fluid, preventing the formation of any trapped gas bubbles. We then fill the medium with the fluorescently dyed wetting fluid by imbibition; a similar approach is used to saturate a rock core prior to core-flood experiments. We then flow dyed wetting fluid at the same flow rate; this process is often referred to as primary drainage. We then flow dyed wetting fluid at the same flow rate; this process is referred to as secondary imbibition. Our experiments are performed at controlled flow rates to investigate the influence of the capillary number $Ca \equiv \mu Q/\gamma$ on the flow; this represents the ratio of viscous and capillary forces, with $\mu = \mu_{nw}$ during drainage and $\mu = \mu_w$ otherwise. Important differences may arise in pressure-controlled flow. Our experiments span the range $Ca \sim 10^{-6}$–$10^{-7}$.

### Results and Discussion

To investigate the pore-scale dynamics of primary drainage, we use confocal microscopy to visualize oil invasion at a single 11-$\mu$m-thick optical slice within a porous medium of width 3 mm and height 1 mm, acquiring a new image every 35 ms. Because the oil is undyed, we identify it by its contrast with the dyed wetting fluid in the measured pore space. At low $Ca \sim 10^{-6}$–$10^{-7}$, the oil menisci displace the wetting fluid through a series of abrupt bursts into the pores (Figure 3a); this indicates that a threshold pressure must build up in the oil at a pore entrance before it can invade the pore.$^{26–31}$ This pressure is given by the pore-scale capillary pressure, $P_c = \alpha/(\alpha + \rho_w g l)$, where $\alpha \approx 0.18$ $a$ is the radius of a pore throat.$^{32–34}$ The bursts are typically only one pore wide$^{35}$ but can span many pores in length along the direction of the local flow (third frame of Figure 3a); moreover, the oil remains continually connected during flow. We find that bursts can proceed along directions other than the bulk flow direction; as a result, the interface between the invading oil and the wetting fluid is ramified. After oil invasion, we observe a $\sim 1$-$\mu$m-thick layer of the wetting fluid coating the bead surfaces,$^{28–31}$ as indicated in the rightmost panels of Figure 3; because we use optical imaging, we can resolve this layer to within hundreds of nanometers.

The speed of the fluid meniscus during a burst of oil into a pore, $v$, can be estimated by balancing the threshold capillary pressure, $2\alpha/a$, with the viscous pressure required to displace the wetting fluid over a length $l$, $\mu_w \varphi D l/k$. Our experimental approach enables us to directly visualize and quantify the speed of, individual bursts. For a porous medium with $a = 75$ $\mu$m, $k = 75$ $\mu$m$^2$, $\varphi = 0.41\%$, and cross-sectional width $w = 3$ mm, we measure a maximum burst speed $v \approx 10$ mm/s; this corresponds to wetting fluid flow over $l \sim 10$ mm. Although the details of this flow are complex, this simple scaling estimate suggests that the wetting fluid is displaced over a length scale comparable to the width of the porous medium,$^{36}$ spanning many pores in size$^1$; interestingly, this observation is consistent with previous measurements of the fluorescence intensity over all slices making up a stack. To probe the spatial dependence of $\varphi$, we also image stacks at multiple locations along the length of the medium. We find $\varphi = 41 \pm 3\%$ independent of position along the length of the medium, as shown in Figure 2; this is comparable to the porosity of highly porous sandstone.$^{27}$ Moreover, $\varphi$ is similar for different realizations of a porous medium, as shown by the different symbols in Figure 2; this illustrates the reproducibility of our protocol.

To mimic discontinuous core-flood experiments on reservoir rocks, we subsequently flow $> 30$ pore volumes of nonwetting oil at a prescribed volumetric flow rate $Q$ through the porous medium; this process is often referred to as primary drainage. We then flow dyed wetting fluid at the same flow rate; this process is referred to as secondary imbibition. Our experiments are performed at controlled flow rates to investigate the influence of the capillary number $Ca \equiv \mu Q/\gamma$ on the flow; this represents the ratio of viscous and capillary forces, with $\mu = \mu_{nw}$ during drainage and $\mu = \mu_w$ otherwise. Important differences may arise in pressure-controlled flow. Our experiments span the range $Ca \sim 10^{-6}$–$10^{-7}$.

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$^1$Consistent with this hypothesis, we find that the discrepancy between the measured permeability and the theoretical prediction is smaller for porous media with larger cross-sectional areas and comprised smaller beads.

$^2$The estimated $l$ is larger than the width of the medium, suggesting that the wetting fluid may not only flow across the width of the medium, but along it, as well.
pressure fluctuations during drainage, as well as imaging of drainage through a monolayer of glass beads.

To explore the dependence of the water displacement on flow conditions, we visualize primary drainage for varying Ca. Unlike the low Ca case, the oil bursts are not successive during primary drainage at higher Ca ~ 10^{-4} to 10^{-2}; instead, neighboring bursts occur simultaneously, typically in the bulk flow direction (Figure 3b). As a result, over the scale of multiple pores, the interface between the invading oil and the wetting fluid is more compact; this behavior reflects the increasing contribution of the viscous pressure in the invading oil at higher Ca. As in the low Ca case, we do not observe evidence for oil pinch-off or subsequent reconnection; interestingly, this behavior is in contrast to the prediction that the oil can be pinched off during drainage.

Similar to the low Ca case, we observe a ~1-μm-thick layer of the wetting fluid coating the bead surfaces after oil invasion. This behavior is indicated in the rightmost panels of Figure 3.

To mimic discontinuous core-flood experiments on reservoir rocks, we flow dyed wetting fluid immediately after oil invasion. During secondary imbibition at low Ca ~ 10^{-6} to 10^{-5}, the wetting fluid does not flow directly into the pores. Instead, it pinches off threads of oil at multiple nonadjacent pore constrictions; the resultant state is shown in the first frame of Figure 4a. This is in stark contrast to the case of drainage. Such behavior must require the wetting fluid to initially flow through the thin wetting layers coating the bead surfaces. This observation directly confirms predictions for water-wet 3-D porous media. As flow proceeds, the wetting fluid spreads from the filled constrictions, displacing oil from the surrounding pores. The flow is highly nonlocal. The wetting fluid eventually forms a tortuous, continuous network of filled pores through which it continues to flow, forming disconnected oil ganglia in the process. For sufficiently long times, we do not observe any additional oil displacement, and a significant amount of oil remains trapped in the medium (last frame in Figure 4a). The pressure drop across the porous medium does not appreciably change, further confirming that a steady state is reached.

To explore the dependence of oil displacement on flow conditions, we visualize secondary imbibition for varying Ca. Unlike the low Ca case, we do not observe oil pinch-off at higher Ca ~ 10^{-4} to 10^{-2}; instead, the wetting fluid displaces the oil from the pores, as shown in Figure 4b. This indicates that flow through the thin wetting layers becomes less significant as Ca is increased. This observation confirms the predictions of recent simulations. However, some of the oil is still bypassed by the wetting fluid, forming disconnected oil ganglia; in several cases, the ganglia break up into smaller ganglia. Many of these ganglia are mobilized from the medium; however, a few smaller ganglia remain trapped (last frame in Figure 4b). For sufficiently long times, these ganglia cease to move, and the pressure drop across the medium does not appreciably change, indicating that a steady state is reached. Our results highlight the important
role played by the wetting layers in influencing the flow behavior.

To understand why some ganglia remain trapped, we analyze the distribution of pressures in the wetting fluid as secondary imbibition proceeds, before the oil is mobilized from the medium. Because the oil occludes some of the pore volume, the permeability of the medium to the wetting fluid is modified by a factor $\kappa \sim 0.1$; we note that $\kappa$ increases as the oil saturation decreases, as confirmed by our independent measurements. Thus, we estimate the viscous pressure gradient in the wetting fluid as $\mu \tau_0 (Q/A)/(\kappa h)$; for displacement at low $Ca$, this gradient is $\sim 10$ Pa/pore. Thus, for low $Ca$, the viscous pressure during oil displacement becomes comparable to the capillary pressure required to force oil through the medium, $2/ja_t \sim 10^3$ Pa, only on length scales larger than $\sim 10^3$ pores. As a result, oil ganglia smaller than $\sim 10^3$ pores cannot be mobilized; thus, we expect many large ganglia to remain trapped in the medium, consistent with our observations (last frame in Figure 4a). By contrast, the viscous pressure gradient in the wetting fluid can be as large as $\sim 10^3$ Pa/pore for the highest $Ca$ studied. Consequently, the viscous pressure during oil displacement becomes comparable to the capillary pressure on length scales larger than $\sim 10$ pores. As a result, all ganglia larger than $\sim 10$ pores can be mobilized; thus, we expect only a few smaller ganglia to remain trapped in the medium, consistent with our observations (last frame in Figure 4b).

To further test this picture, we exploit the close match between the refractive indices of the fluorescently dyed wetting fluid, the undyed non wetting fluid, and the glass beads to directly visualize the pore-scale configurations of the trapped oil. We image a second 3-D image stack of 2-μm-thick slices, spaced by 2 μm along the z-direction, within the porous medium, and we identify the undyed oil by its additional contrast with the dyed wetting fluid in the measured pore volume. By comparing the optical slices to slices taken at the same positions within the medium prior to primary drainage, we resolve the full 3-D structure of the trapped oil ganglia; two representative examples are shown in Figure 5.

The spatial resolution of this approach is on the order of hundreds of nanometers, significantly better than the typical limits of MRI and X-ray μCT. Interestingly, the ganglia sizes and shapes are highly dependent on $Ca$. At low $Ca \sim 10^{-6} - 10^{-5}$, the trapped ganglia are ramified and can span many pores, as shown in Figure 5a. By contrast, ganglia produced at higher $Ca \sim 10^{-4} - 10^{-3}$ are typically smaller and less ramified, as shown in Figure 5b.

To explore the variation of ganglia configurations with flow conditions, we use the 3-D reconstructions to measure the volume of each oil ganglion visualized. Moreover, we image additional stacks at multiple locations along the length of the medium, resolving a total of over 500 individual ganglia for each $Ca$ investigated. We summarize these data by calculating the cumulative probability distribution function of the ganglion volume for each $Ca$. At the lowest $Ca \sim 10^{-6} - 10^{-5}$, the ganglia typically occupy tens of pores. The largest ganglia occupy at most several hundred pores (rightmost curves in Figure 6a), consistent with our expectation.

Figure 4. Pore-scale dynamics of secondary imbibition depend strongly on $Ca$.

Images show multiple frames, taken at different times after drainage, of a single optical slice. The slice is 11-μm thick and is imaged within a porous medium comprised beads with average radius $a_t=75$ μm, with cross-sectional width 3 mm and height 1 mm. The bright areas show the dyed wetting fluid; the dark circles are the beads, whereas the additional dark areas are the undyed oil being displaced from the pore volume. Direction of bulk wetting fluid flow is from left to right. The final frame shows the unchanging steady state. Labels show time elapsed after first frame. (a) At low $Ca=7 \times 10^{-6}$, the wetting fluid pinches off the oil at multiple nonadjacent constrictions and then displaces the oil from the surrounding pores. The wetting fluid eventually flows through a tortuous, continuous network of filled pores, forming many trapped oil ganglia. (b) At high $Ca=6 \times 10^{-4}$, the occurrence of oil pinch-off is reduced, and the wetting fluid displaces the oil from the pores in a piston-like manner, leaving a few small oil ganglia trapped within the medium. Scale bars are 200 μm. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]
that ganglia larger than ~10^9 pores are mobilized from the medium. As \( \text{Ca} \) increases, the viscous pressure in the wetting fluid during oil displacement increases; consequently, even smaller ganglia are mobilized and removed from the medium. Consistent with this expectation, we find that the median ganglion volume decreases with increasing \( \text{Ca} \), in agreement with previous work.\(^{52,56-59}\) Indeed, in stark contrast to the low \( \text{Ca} \) case, ganglia formed at the highest \( \text{Ca} \) typically occupy only a few pores. The largest ganglia occupy at most 10 pores (leftmost curves in Figure 6a), in good agreement with our expectation that ganglia larger than ~10^9 pores are mobilized from the medium. These results support the idea that oil mobilization and trapping is determined by the interplay between the macroscopic viscous and capillary pressures.\(^{31}\)

To further quantify the properties of the trapped oil, we use the 3-D reconstructions to calculate the residual oil saturation \( S_{or}=V_{oil}/\phi V \), where \( V_{oil} \) is the total volume of oil trapped within the entire volume of porous medium imaged, \( V \), for each \( \text{Ca} \) investigated. We find that \( S_{or} \approx 15\% \) for the lowest \( \text{Ca} \sim 10^{-6}-10^{-5} \), in agreement with previous measurements.\(^{60}\) Interestingly, \( S_{or} \) decreases precipitously as \( \text{Ca} \) increases above a threshold ~2 \times 10^{-4}, reaching \( \approx 5\% \) at the highest \( \text{Ca} \sim 10^{-3} \) [Figure 6b]. This behavior reflects
the combined effect of the diminished ganglion formation and the enhanced mobilization of oil by the wetting fluid, as $Ca$ increases. In particular, as $Ca$ increases above this threshold, the viscous pressure in the wetting fluid balances the capillary pressure required to mobilize oil on length scales approaching the scale of an individual pore, consistent with theoretical predictions.\textsuperscript{62}

Conclusions

The experimental approach reported here provides a way to investigate both the pore-scale dynamics of ganglion formation and trapping, and the complex configurations of the trapped ganglia, within a 3-D porous medium. By matching the refractive indices of the wetting fluid, the nonwetting oil, and the porous medium, we fully visualize the multiphase flow in 3-D. We use confocal microscopy to directly visualize the drainage of the medium by a nonwetting oil and subsequent imbition by a wetting fluid. During imbibition, we find that the wetting fluid can flow through thin layers coating the solid surfaces of the medium, pinching off threads of oil in the narrow crevices. This nonlocal flow forms disconnected oil ganglia, some of which remain trapped within the medium. During oil displacement, the oil pinch-off is diminished for increasing capillary number $Ca$. Moreover, the viscous pressure due to wetting fluid flow can mobilize increasing amounts of oil at higher $Ca$. Consequently, both the typical ganglion size and the total amount of residual oil decrease, as $Ca$ increases. Thus, our observations highlight the critical role played by pore-scale fluid dynamics in determining the trapping and mobilizing of oil in 3-D porous media. These results may also be relevant to many other technologically important multiphase flows, such as groundwater contamination by nonaqueous pollutants\textsuperscript{63,64} and the storage of super-critical CO$_2$ within saline aquifers.\textsuperscript{65-68}

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