Analysis of Imbibition in Mixed-Wet Rocks Using Pore-Scale Modeling
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Summary
Countercurrent imbibition is an important recovery mechanism during waterflooding in fractured reservoirs. This may be a rapid and efficient recovery process in strongly water-wet systems, but if the reservoir is mixed-wet, while it is possible for some water to imbibe spontaneously, the ultimate recovery is lower and the imbibition rate may be several orders of magnitude slower than for strongly water-wet rock.

We use quasistatic pore-scale network modeling as a tool to study the behavior of mixed-wet rocks and to predict relative permeability and capillary pressure. The model uses a topologically disordered network that represents the pore space of Berea sandstone. We adjust the distribution of contact angles at the pore scale to match previously published experimental countercurrent waterflood recoveries and wettability indices on Berea. We then input the relative permeabilities and capillary pressures into a conventional grid-based code and simulate countercurrent imbibition in 1D. We make predictions, with no matching parameters, of the recovery as a function of time and compare the results with the experimental measurements. We are able to reproduce the observed dramatic increase in imbibition time as the system changes from being water-wet to mixed-wet. In a mixed-wet system, spontaneous imbibition, where the capillary pressure is positive, is limited to a narrow saturation range where the water saturation is small. At these low saturations, the water is poorly connected through the network in wetting layers and the water relative permeability is extremely low, leading to recovery rates tens to thousands of times slower than for water-wet media.

We present a semiempirical equation to correlate imbibition recovery in mixed-wet rocks of different wettability and viscosity ratio. The recovery rate is proportional to the water mobility at the end of imbibition.

Introduction
Fractured reservoirs are important oil and gas resources. The fracture network contains a small amount of oil in place compared to the lower-permeability matrix to which it is connected. High well productivity and relatively low ultimate recovery are typical characteristics of these reservoirs. Waterflooding is one of the most important mechanisms of oil production from fractured reservoirs. Imbibition is the displacement of nonwetting phase by wetting phase. In a strongly water-wet rock, water rapidly imbibes into the rock and displaces the nonwetting phase, oil. However, the majority of reservoir rocks are not strongly water-wet.

Salathiel introduced the term “mixed wettability” for rock that contains both water-wet and oil-wet fractions. After primary oil recovery, those larger pores occupied by oil may change their wettability, while smaller water-filled regions of the pore space remain water-wet. The adsorption of surface-active agents in the oil, such as asphaltene, to the pore surface in direct contact with the oil causes wettability alteration.

One of the important characteristics of mixed-wet rock is its ability to imbibe both water and oil. Zhou et al. performed 23 spontaneous imbibition and 27 waterflood experiments on Berea cores with different wettabilities and initial water saturations. The samples were aged in Prudhoe Bay crude oil at a temperature of 88°C for between 0 and 240 hours to alter the wettability of the samples from water-wet to mixed-wet. The results of the imbibition and waterflood tests are shown in Fig. 1 for an initial water saturation $S_{wi}=0.15$. As the aging time increases, the waterflood recovery improves, because the residual oil saturation is reduced.

However, the recovery rate from imbibition (defined in this paper as displacement where the oil/water capillary pressure $P_c(0)>0$) is several orders of magnitude slower than for water-wet conditions, where the core is not aged at all. Similar behavior has also been observed in chalk. In some cases, there is an apparent induction time before imbibition starts. As the aging time increases, corresponding to more of the pore space being oil-wet, the induction time increases. The induction time is interpreted as the time needed for water to make a connected flow path through water-wet surfaces through the rock.

In this paper, we will attempt to explain why the waterflood recovery increases with aging time, while imbibition recovery becomes both lower and slower. Several authors have predicted the rate of imbibition recovery for water-wet systems. Ma et al. modified the Mattax and Kyte hydration scaling law to define a dimensionless time for countercurrent imbibition in which gravity has no effect and the viscosities are finite:

$$t_D = t \left( \frac{K}{\phi} \frac{\sigma}{\mu_w \mu_o} \frac{1}{L_c^2} \right) \quad \text{.......... (1)}$$

where $t$ is time, $K$ is permeability, $\phi$ is porosity, $\sigma$ is interfacial tension, $\mu_w$ and $\mu_o$ are water and oil viscosities, and $L_c$ is the characteristic length determined by the size, shape, and boundary conditions of the sample defined by Ma et al. as:

$$F_c = \frac{1}{V} \sum A_i \quad \text{.......... (2)}$$

$$L_c = \sqrt{\frac{1}{F_c}} \quad \text{.......... (3)}$$

where $V$ is the volume of the sample, $A$ is the area open to flow, $l$ is the distance from the open surface to the no-flow boundary and the sum is over all open surfaces of the block. Ma et al. found that plots of the fraction of recoverable oil produced as a function of the dimensionless time, Eq. 1, for cores of different size with different fluid viscosities, boundary conditions, and interfacial tensions all lay approximately on the same universal curve. They studied experimental data presented by Mattax and Kyte for Alumnum samples and Weiler sandstones, Hamon and Vidal’s results for synthetic materials, and Zhang et al.’s results on Berea sandstone.

Zhou et al. used another equation to scale imbibition experiments in water-wet diatomite where the viscosity ratio was varied by four orders of magnitude:

$$t_D = t \left( \frac{K}{\phi} \frac{\sigma}{\mu_w \mu_o} \frac{1}{\sqrt{M^w} + \sqrt{M^o}} \right) \quad \text{.......... (4)}$$

where $\lambda^w(=k^w/\mu)$ is a characteristic mobility and $M^o(=\lambda^o/\lambda^w)$ is a mobility ratio. They used endpoint relative permeabilities to calculate $\lambda^w$ and $M^o$.
Eqs. 1 and 4 were developed for strongly water-wet rocks, where it is assumed that the oil/water contact angle $\theta$ is close to zero. To account for larger oil/water contact angles in mixed-wet media, Gupta and Civan\textsuperscript{19} and Cil et al.\textsuperscript{20} proposed the following expression based on Eq. 1:

$$t_D = \frac{1}{K \sqrt{\frac{\phi}{\mu_w \mu_o} L^2_w}}$$

where $t_D = t_{D,0}(0.5)$ is defined as the dimensionless time, Eq. 1, for one half of the total recovery. This gives a dimensionless time given by Eq. 5 with $\theta = \theta_{aD}$.

Xie and Morrow\textsuperscript{13} tested Eq. 1 on 32 weakly water-wet Berea sandstone samples. They suggested that when capillary forces are sufficiently small, gravity segregation will make a significant contribution to oil recovery; therefore, this force must be included in scaling laws for weakly water-wet rocks:

$$t_D = \frac{1}{K \sqrt{\frac{\phi}{\mu_w \mu_o} L^2_w}} \left( P, f(\theta) + \frac{\Delta \rho g L^2_w}{L_H} \right)$$

where $P$ is a representative imbibition capillary pressure proportional to $\sigma \sqrt{K/\phi}$, $f(\theta)$ is a wettability factor, and $L_H$ is the vertical height of the sample.

While these approaches are appealing, they lack a sound theoretical basis. If the cores are mixed-wet, there are water-wet and oil-wet regions of the pore space, and the assignment of a single effective contact angle is an empirical fit to the data that does not represent a typical contact angle in the porous medium. Furthermore, the thousand-fold decrease in imbibition rate is unlikely to be accounted for by contact-angle effects alone.

A more fundamental approach to understanding imbibition in mixed-wet systems is the use of pore-scale network modeling.\textsuperscript{21,22} In this paper, pore-scale modeling will be used to predict relative permeability and capillary pressure for different wettabilities. The contact angles will be chosen to match the results of cocurrent waterflooding experiments and the measured wettability indices. Then the relative permeabilities and capillary pressure will be input into a conventional grid-based simulator to predict countercurrent imbibition in one dimension, and the results will be compared to the experiments shown in Fig. 1.

**Network Modeling**

The void space of a rock is represented at the microscopic scale by a lattice of pores connected by throats. Rules are developed to determine the multiphase fluid configurations and transport in these pores and throats. The appropriate pore-scale physics combined with a geologically representative description of the pore space gives a model that can predict average behavior, such as capillary pressure and relative permeability. The model we use accounts for wetting layers in crevices of the pore space, cooperative pore filling, and different contact angles.\textsuperscript{21-23} The model simulates primary drainage, wettability alteration, and any subsequent cycles of waterflooding and secondary drainage. The model is described in more detail elsewhere.\textsuperscript{22,24} Here, we simply use it to compute relative permeabilities and capillary pressures for different wettabilities.

The major assumption of the model is that viscous effects are considered to be negligible at the pore scale; this means that the predicted relative permeabilities are the same for both countercurrent flow (imbibition) and cocurrent flow (waterflooding). Some authors have suggested that the countercurrent relative permeabilities may be up to 30% lower than cocurrent relative permeability.\textsuperscript{11,25} In countercurrent imbibition, by definition, the ratio of capillary to viscous forces across the whole sample is equal to 1 (it is capillary pressure that drives fluid flow). At the scale of a single pore, however, this ratio is approximately the inverse of the number of pores traveled by the invading water front. Because the cores studied were a few cm across, the water travels through thousands of pores, so the assumption of quasistatic flow at the pore scale is accurate. However, at early times when the invading water front has moved only a short distance, dynamic effects may be significant. This has been modeled theoretically using a relaxation time\textsuperscript{26} that slows the imbibition rate. Thus, we consider it appropriate to use a quasistatic network to simulate both co- and countercurrent flow using the same relative permeabilities. While this may tend to overestimate recovery at early time, we are confident that it accurately captures the late-time behavior.

**Analysis of Mixed-Wet Data**

The network model\textsuperscript{23,24} is used to match Zhou et al.’s\textsuperscript{10} experimental cocurrent waterflooding recoveries and wettability indices. Then the computed relative permeabilities and capillary pressures are used to simulate 1D countercurrent imbibition to predict the observed experimental behavior. A flow chart of the procedure is shown in Fig. 2.

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**Fig. 1**—Experimental recovery data on Berea cores aged in crude oil for different aging times, $t_a$ from Zhou et al.\textsuperscript{10} (a) Dimensionless oil recovery by imbibition. (b) Dimensionless oil recovery by waterflooding.
We use a network based on Berea sandstone, and hence the network geometry is representative of the Berea cores used in the experiments. Moreover, in previous work, we have accurately predicted primary drainage and waterflood relative permeabilities for water-wet Berea and waterflood recoveries for mixed-wet Berea.\textsuperscript{23,24} The prediction of mixed-wet data is a challenge because we need to distribute contact angles to each pore and throat.\textsuperscript{27} These are effective contact angles that account for the rock/fluid interaction as well as roughness and the converging/diverging geometry of the pore space.\textsuperscript{27} Contact angles are only adjusted in pores and throats filled with oil after primary drainage; water-filled elements remain water-wet. We assigned two contact-angle distributions. The first distribution has values of less than 105° and represents the water-wet or neutrally-wet regions of the rock. Such pores and throats are filled with oil after primary drainage, but undergo only a modest wettability alteration. The second distribution includes larger contact angles to represent oil-wet regions. We assume that contact angles are distributed at random uniformly between some upper and lower bounds. In order to provide good connectivity of oil-wet and water-wet regions, the oil-wet contact angles are distributed in clusters.\textsuperscript{23} A number of pores and throats are randomly selected. This pore or throat is assigned a contact angle from the oil-wet distribution, as are all its nearest neighbor connected pores or throats. Then, all the nearest neighbors of these oil-wet elements are made oil-wet. The process continues until a target oil-wet volume fraction is reached. In this way, there will be patches of oil-wet pores and throats distributed among water-wet pores and throats. In all cases, 50 clusters were used except for the case of an aging time equal to 240 hours where number of clusters was 10 (see Table 1 for the parameters used to match the data). The network contains 12,349 pores and 26,146 throats; 50 clusters correspond approximately to a correlation length of 6 pores, while 10 clusters is a correlation length of 11 pores.

Zhou \textit{et al}.,\textsuperscript{10} defined a wettability index as the ratio of separately determined imbibition and waterflooding recoveries:

\[
I' = \frac{R_{wim}}{R_{wf}} . \hspace{1cm} (8)
\]

This definition is slightly different from the traditionally measured Amott wettability index. The samples in the imbibition and waterflooding tests with the same aging time had slightly different initial water saturations. In the network model, we followed ex-

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**Fig. 2**—Flow chart showing the steps used to tune the network model to waterflood data and then predict imbibition recovery.

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**TABLE 1**—CONTACT ANGLE DISTRIBUTIONS USED IN THE NETWORK MODEL TO MATCH EXPERIMENTAL\textsuperscript{10} WATERFLOODING RECOVERIES AND WETTABILITY INDICES SHOWN IN TABLE 2

<table>
<thead>
<tr>
<th>Aging Time (hours)</th>
<th>Water-Wet Contact Angle Distribution</th>
<th>Neutral and Oil-Wet Contact Angle Distribution</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Degrees</td>
<td>Min</td>
</tr>
<tr>
<td>Water-Wet</td>
<td>S\textsubscript{sw}</td>
<td>25</td>
</tr>
<tr>
<td>0</td>
<td>.155</td>
<td>50</td>
</tr>
<tr>
<td>4</td>
<td>.158</td>
<td>67</td>
</tr>
<tr>
<td>48</td>
<td>.162</td>
<td>74</td>
</tr>
<tr>
<td>72</td>
<td>.152</td>
<td>72</td>
</tr>
<tr>
<td>240</td>
<td>.157</td>
<td>79</td>
</tr>
</tbody>
</table>

The water-wet data are from a reference case that predicts water-wet relative permeabilities.\textsuperscript{23,24}
exactly the same sequence of saturation changes as observed in the experiments.

Matching Cocurrent Waterflood Recovery and Wettability Indices. The flooding sequence in the network model, given in the following, is identical to that in the experiments. We assign an immobile water volume to the network. This is adjusted to match the lowest water saturation reached in any of the experiments. This is the $S_{wc}$ of the sample. For Zhou et al.’s experiments, $10\% S_{wc}$. 1. Simulate primary drainage to $S_{wi}$ for each aging time. 2. Simulate wettability alteration by assigning the contact angle distributions. 3. Waterflood the sample until the water/oil ratio is 99. 4. Calculate the waterflood recovery using the Buckley-Leverett approach assuming no capillary pressure. Compute the wettability index and compare with experimental data. Repeat the procedure, adjusting the contact angle distributions in Step 3 until the experimental and predicted recoveries and wettability indices match (see the flow chart in Fig. 2).

The results of matching waterflooding recovery and wettability indices are presented in Fig. 3 and Table 2. To match the waterflood recovery and wettability index, five parameters were adjusted: the upper and lower bounds of the oil-wet and water-wet distributions and the oil-wet volume fraction. These parameters are shown in Table 1. The key parameter is the recovery at the end of imbibition (or the wettability index), because this determines the fraction of pores that are oil-wet.

The ranges of contact angle are plausible; note that for waterflooding, the water-wet regions are in fact weakly to neutrally wet. As the aging time increases, the oil-wet fraction increases, as do the contact angles in the oil-wet regions. The waterflood recoveries and wettability indices are accurately matched, which indicates that the model is able to capture the correct displacement physics with the right parameters. For zero aging time, the quality of waterflood and wettability index match is not as good as the others. It was not possible to match simultaneously waterflood recovery and the very high residual oil saturation for this case.

The capillary pressure and relative permeabilities calculated by the network model for all cases are presented in Figs. 4 and 5. Also shown are results for a water-wet reference case (labeled strongly water-wet) with an initial water saturation of 25% that has already been shown to predict steady-state relative permeabilities accurately. While we present the results in terms of macroscopic parameters—recovery, relative permeability, and capillary pressure—their values are a function of complex pore-scale interactions between the solid and fluids. As the aging time increases, the capillary pressure becomes lower and an increasing fraction of

<table>
<thead>
<tr>
<th>Aging Time (hours)</th>
<th>$S_{wc}$ at $P_i=0$</th>
<th>$S_{wc}$ at $t_o=99$</th>
<th>Wettability Index</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>EXP 0.414</td>
<td>SIM 0.465</td>
<td>EXP 0.507</td>
</tr>
<tr>
<td>4</td>
<td>EXP 0.438</td>
<td>SIM 0.426</td>
<td>EXP 0.557</td>
</tr>
<tr>
<td>48</td>
<td>EXP 0.405</td>
<td>SIM 0.395</td>
<td>EXP 0.648</td>
</tr>
<tr>
<td>72</td>
<td>EXP 0.387</td>
<td>SIM 0.370</td>
<td>EXP 0.683</td>
</tr>
<tr>
<td>240</td>
<td>EXP 0.254</td>
<td>SIM 0.260</td>
<td>EXP 0.687</td>
</tr>
</tbody>
</table>

FIG. 3—Comparison of experimental (points) waterflood recovery for samples with different aging times, $t_a$ (after Zhou et al.,10), and predicted results (lines) using network model relative permeabilities. A good match to the data is achieved using plausible distributions of contact angles (see Table 1).

FIG. 4—Capillary pressures for different wettability states (represented by different aging times, $t_a$) simulated by the network model. Strongly water-wet data from Ref. 23.

FIG. 5—Relative permeabilities for different wettability states simulated by the network model to match experimental waterflooding recoveries (Fig. 3). Strongly water-wet data from Ref. 23.
the curve lies below zero, indicating oil-wet properties. Recovery by imbibition is governed by the regions of the capillary pressure curve above zero. Clearly the amount of oil recovered will decrease as more of the pores become oil-wet. The rate of recovery will be governed by the magnitude of the capillary pressure. While the capillary pressure is certainly lower than for a water-wet sample, it is typically less than an order of magnitude lower for the aged samples in the region where the capillary pressure is positive. This indicates that capillary pressure alone cannot explain the long imbibition times seen experimentally.

The relative permeabilities shown in Fig. 5 show that the residual oil saturations for the longest aging times are considerably lower than for a water-wet sample. This has been observed and explained by Salathiel²; in the oil-wet regions, oil layers maintain connectivity of the oil phase down to very low saturation.

The most remarkable feature of the relative-permeability curves is that the water relative permeability for the aged samples is around one to two orders of magnitude lower than for a water-wet system at low and intermediate oil saturations (where the capillary pressure is positive). This phenomenon has already been observed using pore-scale modeling.³,⁴,24 During waterflooding in a mixed-wet system, water-wet pores and throats are preferentially filled first. However, if the oil-wet fraction is large, these pores and throats fail to make a connected path of filled elements across the network. This means that the water relative permeability is controlled by thin wetting layers in the corners of the pore space that have a very low conductance. During forced displacement, the larger oil-wet pores are filled by water. This can lead to a rapid change in saturation, but until the water forms a connected pathway across the system, the water relative permeability remains very low.

For waterflood recovery, the combination of a low water relative permeability and low residual oil saturation leads to a high recovery as a function of pore volumes injected; the oil can escape readily from the system, while the low water relative permeability holds the water back. This can be seen in the predicted and experimental recovery curves (Fig. 3) where waterflood recovery increases with increasing aging time. However, for imbibition, this leads to very slow recovery, as the recovery rate is governed by a combination of the capillary pressure and the water relative permeability. This is the physical origin of the very long imbibition times observed experimentally. We will now test this hypothesis through simulation.

### Prediction of Experimental Countercurrent Imbibition Recovery

The relative permeabilities and capillary pressures derived from pore-scale modeling were used in a conventional simulation (using Eclipse®-100) of countercurrent imbibition in 1D. In the 1D model, illustrated in Fig. 6, a reservoir of water was connected to the rock with no flow boundaries on the other faces. We used the same fluid properties as in the experiments, and the porosity and permeability were those calculated by the network model (Table 3). Gravitational effects were neglected. No injection or production wells were included in the model; hence, no viscous forces were present. This means that the mechanism of recovery was counter-current imbibition as a result of capillary forces only. In Zhou et al.’s¹⁰ experiments, the cylindrical core samples had diameters and lengths in the range of 3.8 to 3.82 cm and 6.4 to 7.8 cm, respectively. In the simulations, the block width and length were set to 4 and 8 cm, respectively. 42 gridblocks were used in the simulation; we performed grid-refinement studies to ensure that we used sufficient gridblocks to obtain converged results. While for the waterflooding results the contact angles are varied to match the experiments, the comparison of imbibition recoveries shown in Fig. 7 are genuine predictions; no adjustment to the network model properties was made. While the agreement between predictions and experiment is not perfect, we are able to reproduce the trend in recovery with aging time and demonstrate that for mixed-wet systems the imbibition times can indeed be orders of magnitude longer than for a water-wet system. The principal reason for long imbibition times is the very low water relative permeabilities observed in the mixed-wet samples and is not solely the consequence of reduced capillary forces, although they are also low. Fig. 7 plots the results in terms of dimensionless time (Eq. 1) and recovery. However, we also accurately predict the absolute recovery that decreases with aging time (this is governed by the water saturation when the capillary pressure is zero); see Table 2.

### Discussion

The match to the waterflood data is not necessarily unique. However, in our simulations, we were unable to find significantly dif-

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**TABLE 3—MATRIX, ROCK, AND FLUID PROPERTIES USED IN THE SIMULATIONS (AFTER ZHOU ET AL.¹⁰)**

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Unit</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Porosity</td>
<td>fraction</td>
<td>0.207</td>
</tr>
<tr>
<td>Permeability</td>
<td>Darcy</td>
<td>3.131</td>
</tr>
<tr>
<td>Oil density</td>
<td>kg·m⁻³</td>
<td>895</td>
</tr>
<tr>
<td>Oil viscosity</td>
<td>Pa·s</td>
<td>0.03925</td>
</tr>
<tr>
<td>Water density</td>
<td>kg·m⁻³</td>
<td>1.012</td>
</tr>
<tr>
<td>Water viscosity</td>
<td>Pa·s</td>
<td>0.000967</td>
</tr>
<tr>
<td>Interfacial tension</td>
<td>N·m⁻¹</td>
<td>0.0242</td>
</tr>
</tbody>
</table>

*from network modeling

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**Fig. 6—Grid system used for 1D simulations of imbibition. A total of 42 gridblocks were used.**

**Fig. 7—Simulation (lines) of experimental imbibition recovery (points) for different aging times, tₐ (after Zhou et al.¹⁰), using network model data (Figs. 4 and 5). The dimensionless time is defined by Eq. 1. Imbibition data (diamonds) from Zhou et al.’s¹⁰ experiments with initial water saturations of 20 and 25% are also included in the zero aging time data (squares for Sₜₐ=15%).**
different contact-angle distributions that simultaneously matched the wettability index and waterflood recovery. In all cases, we predicted low water relative permeability at low saturation leading to much slower imbibition than for water-wet samples.

For the case of zero aging time, the experimental results\textsuperscript{10} show a water-wet behavior because we see very small difference in waterflood and imbibition recoveries. For this case, all data for zero aging time with different initial water saturations of 15, 20, and 25% are plotted in Fig. 7 because the results are expected to be less sensitive to initial water saturation \( (S_{wi}) \). Network model relative permeabilities for a strongly water-wet system and for zero aging time both give good predictions of the zero aging-time data, except for dimensionless times greater than 100, where recovery is overestimated.

We also ran other simulations in which we assumed smaller immobile water saturations down to 8%, because it is possible to reduce \( S_o \) to below 15% in Berea sandstone.\textsuperscript{10} In all cases, the results were similar. We still see a very low water relative permeability in the range where the capillary pressure is positive, because the water is poorly connected.

Scaling of Countercurrent Imbibition in Mixed-Wet Rocks

The scaling of viscosity in the scaling group (Eq. 1) has been confirmed experimentally for Berea systems that had been aged in crude oil.\textsuperscript{29} However, Eq. 1 cannot correlate systems with different wettability, because it does not account for the relative permeability. In contrast, Eq. 4 does account for the water relative permeability. However, we suggest that rather than use the endpoint water relative permeability (at the end of waterflooding), which increases with aging time (Fig. 5), we use the water relative permeability at the end of spontaneous imbibition (when \( P_c = 0 \)) that decreases with aging time. With this definition, in the cases we study, \( \lambda_{w0} > \lambda_{w} \); thus, \( M_{w} \ll 1 \), and Eq. 4 reduces to

\[
t_D = t \sqrt{\frac{K_0 \sigma}{\phi L_w}} \lambda_{w0}. \hspace{1cm} (9)
\]

Validity of the Scaling Functions. To check the validity of the scaling functions, Eqs. 1, 4, and 9, a series of 1D imbibition simulations for different oil-to-water viscosity ratios, \( M \), were performed using the network model derived data for an aging time of 72 hours. Our simulations covered a range of viscosity ratios from 0.01 to 200. The recoveries from the simulations are plotted as a function of the different dimensionless times in Figs. 8 and 9. For a given aging time, but different mobility ratios, all the recoveries lie on the same universal curve using Eq. 9 as the dimensionless time. While there is some scatter in the data, the simulation results for different aging times also, approximately, fall on the same curve. The water relative permeability is around four orders of magnitude lower than the oil relative permeability in the saturation range where imbibition occurs (Fig. 5). In order for the oil mobility to affect the imbibition rate significantly, the viscosity ratio would need to be around 10,000 or greater. The same analysis using Eq. 4 gives identical results, because \( M_{w} \ll 1 \), as long as the relative permeabilities at the end of imbibition are used. In contrast, using Eq. 1 gives poorer results, with a wider scatter in the recoveries for different aging times (Fig. 7) and for different mobility ratios (Fig. 10).
This analysis. Initial water saturations were different from those assumed in broadly consistent with the simulation results in Fig. 9, although a quantitative match is not possible, because the aging times and initial water saturations were different from those assumed in this analysis.

Many authors have matched imbibition data, albeit for water-wet systems, with an exponential recovery with time. This analysis has been limited to direct comparison with mixed-wet experimental data for one value of the initial water saturation, $S_{wi}$, and $t_i$ from Eqs. 4 or 9 and $\alpha = 0.2$ (see Fig. 10), although the correlation underpredicts recovery at early time. However, note that our simulations overpredict recovery at early time (Fig. 6), so the correlation may be a better match to experiment. Unfortunately, we cannot compare our correlation directly against experiment, because we do not know the experimental water relative permeability. However, we can compare experiment and theory by using the relative permeabilities predicted by the network model in Eq. 9. The dimensionless rate constant $\alpha$ is higher than that used to match water-wet data using Eq. 1 ($\alpha = 0.0517$) despite the much longer real imbibition times. The reason for this is that Eq. 1 does not account for the water relative permeability that is typically very low.

So far, this analysis has been limited to direct comparison with mixed-wet experimental data for one value of the initial water saturation, $S_{wi}$, and $t_i$, Zhou et al.35 studied systems with other values of $S_{wi}$ that could be analyzed using the same approach. Furthermore, we have neglected the effect of gravity. As mentioned by Xie and Morrow,31 in mixed-wet media it is likely that gravitational effects also impact recovery, in which case a dimensionless time including buoyancy effects (such as Eq. 7) needs to be tested and validated.

An analytic expression for the recovery (Eq. 10) can be used to derive a transfer function for field-scale dual-porosity simulation of flow in fractured reservoirs.32–37 This work suggests that the imbibition rate in mixed-wet systems is proportional to the water mobility.

In some cases, the quantitative match to experiment was poor. We tended to overestimate recovery at early time, which suggests that there might be an induction time before imbibition starts. All the imbibition recovery curves fell on approximately a universal curve if plotted as a function of a dimensionless time (Eq. 9) that was proportional to the water mobility at the end of imbibition. We proposed a correlation to match the simulation results (Eq. 10) that could be used to predict imbibition rates for mixed-wet media.

**Conclusions**

We used pore-scale network modeling to study waterflooding and countercurrent imbibition in mixed-wet Berea. We adjusted the contact angles in the network model to match experimental waterflood recovery and wettability index for samples aged in crude oil for different times. As the aging time increased, more of the pore space became oil-wet. We then used the computed relative permeabilities and capillary pressures in a conventional simulator to predict recovery from countercurrent imbibition with no further adjustment of any parameters. We were able to match the results using plausible ranges of contact angles. We were able to predict the orders-of-magnitude increase in imbibition time over water-wet media seen in mixed-wet samples. This was principally the result of extremely low water relative permeability caused by the low connectivity of water at intermediate saturation in a mixed-wet network.

In some cases, the quantitative match to experiment was poor. We tended to overestimate recovery at early time, which suggests that there might be an induction time before imbibition starts.

**Nomenclature**

\[ R = R_{eo}(1 - e^{-\alpha t}) \] \hspace{1cm} (10)

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