Pore-scale visualization and quantitative analysis of the spontaneous imbibition based on experiments and micro-CT technology in low-permeability mixed-wettability rock

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Abstract
The pore-scale imbibition mechanism in fractured water-wetted reservoirs has been acknowledged as an efficient tool to enhance oil recovery. Most rock in a natural reservoir is mixed-wetted, but most studies on the pore-scale imbibition process only focused on the water-wetted rock. This paper adopts two mixed-wetted core samples for microspontaneous imbibition experiment, micro-CT scanning, and nuclear magnetic resonance (NMR) test. The results indicate that the pore radius distribution of the segmented CT images is consistent with that of the NMR test. The microimbibition recovery ratio of core No. 34-1 and core No. 64 in the spontaneous imbibition experiment are 27.7% and 58.2%, which agrees well with that computed by the micro-CT scanning image (27.63% and 56.09%). Based on the segmented images, the influences of the Jamin's effect, pore size distribution, and wettability on the microspontaneous imbibition are visualized and quantitatively studied. The Jamin's effect is the important factor that hinders the microimbibition process. The main pore size of imbibition distributes in the range of 1–25 μm. Furthermore, the pore-scale spontaneous imbibition process in a single pore with mixed wettability is investigated and analyzed. The relationships among the contact angle, capillary force, recovery ratio, wettability, and the microimbibition recovery are revealed.

KEYWORDS
low-permeability reservoir, micro-CT, mixed wettability, pore size distribution, pore-scale spontaneous imbibition, recovery ratio
Oil and natural gas still account for 63.5% proportion (Source: IEA) of the international energy market. Imbibition action has been widely considered an efficient method to enhance the oil recovery of low-permeability reservoirs and promote the utilization of the injected fluids, especially for fractured reservoirs. Browsoncombe and Dyer observed that the water spontaneously invaded the crude oil in experiments. In 1958, Amott measured the functional relationship between the rock wettability and the displacement properties of the water/oil system. Then, Moor and Graham proposed that the main driving force of the imbibition process was the capillary force and gravity. Based on these studies, Iffly explored the relationship among the gravity, capillary force, and boundary conditions and indicated that the imbibition process was mainly classified as countercurrent imbibition and cocurrent imbibition. Xie, Babadagli, and You et al. found that changing the wettability by surfactant could greatly improve the imbibition efficiency. Hou et al. analyzed the mechanisms of wettability alteration of oil-wetted sandstone by the cationic/nonionic surfactant. In addition, the influence of the Jamin’s effect plays a crucial role in the imbibition process. Nevertheless, traditional experiments have difficulty to visualize the imbibition process in the pore. In recent years, the highly developed computer has provided a platform to study imbibition. Al-Huthali performed a streamline simulation of the countercurrent imbibition. Wang et al. simulated the static and dynamic imbibition processes in a regular 2D model to study the effects of the crude oil viscosity, matrix permeability, core size, interfacial tension, and displacement rate on the imbibition. Sedaghat et al. simulated imbibition using the Finite Element-Centered Finite Volume Method and found the ultimate recovery in the water-wetted case was 2-3 times higher than that in the oil-wetted case. However, the regularly adopted (2D/3D) models are too simple to reproduce the natural pore structure. With the development of the microimaging technology, nuclear magnetic resonance (NMR) and X-ray computed tomography (CT) methods are quantitatively studied. Furthermore, the pore-scale spontaneous imbibition process in a single pore with mixed wettability is investigated and analyzed. The relationships among the contact angle, capillary force, recovery ratio, wettability, and the microimbibition recovery are revealed.

EXPERIMENTAL SECTION

2.1 Experimental apparatus

Considering the continuous operation of micro-CT scanning and microspontaneous imbibition experiment, a core holder device is designed to ensure the sealing of the experimental process and the stability and integrity of the core samples in this paper, as shown in Figure 1A.
Other necessary instruments are included in the imbibition device (Figure 1B), which is mainly composed of a constant-temperature sealed box and a high-precision balance (0.0001 g). The weight changes can be recorded at set time intervals. The micro-XCT scanning equipment (Xradia) is model Microxct-400 with a 4× scanning lens. The core nuclear magnetic resonance analysis system (NIN MAG) is model Animr-150 with a magnetic field intensity of 0.23 ± 0.03 T, a maximum echo number of 8000, a minimum echo interval below 150 μS, and a minimum digital acquisition interval of 50 ns. The interface parameter integrated measurement system (model DSA30S, KRUSS) is used to test the wettability.

### 2.2 Rocks and fluids

The tight sandstone cores in this paper are taken from the Xinzhao district with core depths of 1393.5-1402.99 m and 1416.10-1423.39 m, respectively. All operations are performed at 25°C. The imbibition and micro-CT scanning experiments are performed at 1 atm.

The experimental oil is kerosene with a dynamic viscosity of 2.21 mPa s at 25°C and a density of 0.88 g/cm³. To more easily identify the oil-water phase in the CT scan image processing, 13% iodobutane is added into the oil. The experimental water is simulated formation brine with a dynamic viscosity of 0.89 mPa s and a density of 1.04 g/cm³ at 25°C. The ion composition of the brine is shown in Table 1.

### 2.3 Experimental procedure

1. The wettability of the origin core samples (2.5 cm in diameter and 2.5 cm in length) are measured after washing and drying. After that, the two core samples are vacuum-saturated with water for 12 hours, and NMR testing is immediately performed to obtain the $T_2$ spectra.
2. Cores No. 34-1 and No. 64 with a size of 6 mm in diameter and 100 mm in length are drilled form core plugs (2.5 cm in diameter and 2.5 cm in length). Then, the cores are weighed ($m_1$) and loaded into the core holder device after drying, as shown in Figure 2. The physical parameters test is conducted for the core samples, as shown in Table 2.
3. The core holder device is fixed on the CT scanning device; then, the CT scanning (the first scan) is performed on the two core samples that are dried to obtain the pore structure.
4. Afterwards, the core samples are vacuum-saturated with kerosene for 12 hours. After weighing ($m_2$), the core samples are loaded into the core holder device. Then, CT scanning (second scan) is performed on two core samples to obtain the distribution of oil in the pores.
5. The saturated oil core samples are put into the imbibition device, and the full-immersion microspontaneous imbibition experiment is conducted. When the weight collected by the computer no longer changes, the core samples are removed, and we immediately put them into the core holder device.
6. Finally, the core samples are scanned (the third scan) to obtain the distribution of oil and water after the imbibition.

### 3 IMAGE PROCESSING AND ANALYSIS

#### 3.1 Image preprocessing

The micro-CT images is processed using the Avizo™. The basic parameters of the micro-CT images are shown in Table 3. In order to make the image location keep consistent before and after the imbibition, the center of the core images and some random feature points are selected and calibrated.

![FIGURE 1](image)

### Table 1

**The ion composition of the experimental water**

|          | $\text{Na}^+$/mg/L | $\text{Ca}^{2+}$/mg/L | $\text{Mg}^{2+}$/g/L | $\text{Cl}^{-}$/mg/L |
|----------|---------------------|------------------------|----------------------|---------------------|
| Salinity | 80 000              | 27 550                 | 2168                 | 1021                | 49 261             |
The nonlocal mean filtering algorithm is used to reduce noise.\textsuperscript{54} The rationale is to compare the neighborhood of all voxels with the neighbors of the voxel at the current location and replace the gray value of the current position with the gray values of other positions that are assigned with some suitable weight factors.\textsuperscript{55,56} In addition, in order to reduce the partial volume effects of the phase to phase, the Laplace operator edge enhancement method is used to obtain a clear oil-water phase interface. The basic principle is to copy of the original image and blends it with the original, and the image is sharpened without increasing noise.\textsuperscript{57} Two sample slices before and after processing are shown in Figure 3.

### Image segmentation

In previous studies, most researchers extracted a cubic part of the original images for the segmentation to increase the calculation speed. In this paper, to quantitatively study the imbibition recovery ratio, only the outer edge cutting treatment is applied to the image. As shown in Figure 4, the watershed algorithm is adopted to segment the gray image, which is widely used in pore-scale multiphase image segmentation. The basic principle is that the unclassified zones are filled from different ends using the intensity gradient, and lines of highest gradient are used to demarcate class borders locally.\textsuperscript{55} Based on the segmented images, the rock grain and pore distribution of the dry core are obtained.

The dry sample pores are used as the reference pores to calibrate the areas of interest after the saturated oil and imbibition process, as shown in Figure 5. In the saturated oil process, iodobutane is added into the oil phase. The reagent is soluble in oil but not in water. Thus, the density of oil phase and the contrast of oil and water images are enhanced. After the imbibition process, the water phase in the pores appears black, while the oil phase appears light gray (as shown in Figure 5A2,B2). Further, oil-water segmentation results after imbibition are obtained, as shown in Figure 6.

### 3.3 Image 3-D reconstruction

Based on the segmentation process, the three-dimensional structure of the core is reconstructed from 857 2D slices of core No. 34-1 and 677 2D slices of core No. 64. Then, the pores, water phase, and residual oil phase are extracted, as shown in Figure 7. The rock porosity based on image segmentation is obtained by calculating the pore volume. Compared with the experimental porosity, the feasibility of image segmentation can be verified.

### 4 Quantitative Analysis Method

#### 4.1 Micro-occurrence of the water phase in the pore space

The shape factor $G$, that is the sphericity degree of the water phase, is used to classify the occurrence of the microscopic

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**TABLE 3** Micro-CT scanning parameters

| ID of samples | Number of slices | Pixel (diameter)/μm | Resolution ratio/μm |
|---------------|------------------|---------------------|---------------------|
| 34-1          | 677              | 835                 | 5.59154             |
| 64            | 858              | 879                 | 4.90747             |

**TABLE 2** Core sample physical parameters

| No. of samples | Diameter/mm | Length/mm | Porosity/% | Permeability/mD |
|----------------|-------------|-----------|------------|-----------------|
| 34-1           | 6           | 101       | 12.75      | 30              |
| 64             | 6.18        | 104       | 9.18       | 6.72            |
water phase after imbibition. The shape factor can be determined using Equation (1).\textsuperscript{58,59}

\begin{equation}
G = \frac{6\sqrt{\pi} V_0}{S_0^{1.5}}
\end{equation}

where $G$ is a dimensionless number, which represents the shape of the given water; $V_0$ is a given water volume, μm$^3$; $S_0$ is the pore surface area, μm$^2$.

\subsection{4.2 | Capillary force}

The capillary pressure $P_c$ for fluid to enter the pores should obey the Young-Laplace equation:

\begin{equation}
P_c = \frac{2\sigma \cos \theta}{r}
\end{equation}

where $\sigma$ is the surface tension, $\theta$ is the contact angle, and $r$ is the pore radius. The $\kappa = 2 \cos \theta/r$ is defined as surface curvature, and Equation (2) becomes:

\begin{equation}
P_c = \sigma \kappa
\end{equation}

\subsection{4.3 | Contact angles and interfacial curvature}

A novel algorithm had been developed by AlRatrout\textsuperscript{47} is adopted to measure contact angles and fluid-fluid interface curvature at the pore scale. First, a generalized marching cube algorithm is adopted to generate the mesh $M$.\textsuperscript{60} As shown in Figure 8, the mesh $M$ is divided into three face zones: $Z_1$ is the oil/rock interface, $Z_2$ is the oil/brine interface, and $Z_3$ is the brine/rock interface in an oil-brine system. A surface mesh ($M = \{Z_1, Z_2, Z_3\}$) from a segmented three-phase images (rock grain, oil, and water) is extracted; and then, the extracted mesh is smoothed using volume-preserving Gaussian smoothing algorithm; and finally, a local uniform volume-preserving curvature smoothing is conducted.\textsuperscript{47} The sensitivity analysis of reconstruction accuracy can be referred to.\textsuperscript{48}

After that, for the same vertices, the curvature $\kappa_{iCL}$ is calculated using the Equation (4):

\begin{equation}
\kappa_{iCL} = \sum_{j \in \text{adj}(i)} \left| (s_i \cdot n_i) - (s_j \cdot n_j) \right| / \left| \vec{\partial} \partial r \right|, \quad \{n_i, n_j \in e, i, j \in V(4)\}
\end{equation}

Also, for vertices belonging to the oil-brine interface, the curvature $\kappa_{iOB}$ is calculated using the Equation (5):
And then, a smoothed curvature is calculated for the contact line $\langle \kappa_i \rangle_{CL}$:

$$\langle \kappa_i \rangle_{CL} = 0.5 \left( \frac{\sum_{j \in \text{adj}(i)} a_j K_j}{\sum_{j \in \text{adj}(i)} a_j} + \kappa_{i,CL} \right), \{i,j \in V_{CL} \} \tag{6}$$

Also, for vertices belonging to the oil-water interface, the smoothed curvature $\langle \kappa_i \rangle_{OB}$ is calculated by:

$$\langle \kappa_i \rangle_{OB} = 0.5 \left( \frac{\sum_{j \in \text{adj}(i)} a_j K_j}{\sum_{j \in \text{adj}(i)} a_j} + \kappa_{i,OB} \right), \{i,j \in V_{OB} \} \tag{7}$$

where $i$ is a label of vertices, $j$ represents at each vertex $i$, the adjacent vertices are recorded, $\text{adj}(i)$ is the total number of adjacent vertices to each vertex $i$, $s_i$ is the interface tangent vector, $n_i$ is vertex normal, $o_i$ is smoothing displacement of vertices, $\alpha_j$ is a weight factor for the adjacent points, $e_j$ is a set of four connected edges, the set of vertices shared by all face zones represents the three-phase contact line, $V_{CL} = i \in \{Z_1, Z_2, Z_3\}$, and the $V_{OB}$ is the oil/brine interface.

Further, the contact angle $\theta_i$ is calculated by:

$$\theta_i = \pi - \cos^{-1} \left( n_i \cdot n_{Z_2} \mid n_i \cdot n_{Z_3} \right), i \in V_{CL} \tag{8}$$

where is $n_{Z_2}$ a vector normal to the oil/brine interface and $n_{Z_3}$ is a vector normal to the brine/rock interface.

5 | RESULTS AND DISCUSSION

5.1 | Imbibition recovery ratio

In this paper, the mass method is adopted to monitor oil production in the microimbibition process. Due to the density difference of oil and water, the weight of the core will increase theoretically in the imbibition process. However, the experimental core weight (Figure 9B) which is recorded by the high-precision balance decrease until it reaches equilibrium. By observing the experimental phenomenon (Figure 9A), we find that displaced oil accumulates and eventually
forms droplets that are adsorbed on the core surface. The oil droplets attach the core cause the overall volume become larger, and the buoyancy increases. Thus, the weight of the core decreases in the initial stage of imbibition. After that, the imbibition process is completed and the core weight no longer changes.

The cumulative oil production curve is obtained by calculating the weight change data, as shown in Figure 9C.
According to the curve, for No. 34-1 and No. 64 core samples, cumulative oil production increases almost linearly before 150 and 200 minutes. And then, the imbibition process slows down in 150-450 minutes and 200-600 minutes, respectively. Finally, the imbibition process reaches equilibrium. In addition, the core weight is shown in Table 4.

According to Table 4, the recovery ratio can be calculated by Equation (9), as shown in Table 5.

\[
Q_e = \frac{m_c}{m_2 - m_1} \times 100\% \quad (9)
\]

where \(Q_e\) is the ratio of imbibition recovery based on the experimental, \(m_1\) is the dry weight of the core sample, \(m_2\) is the saturated oil weight of the core sample, and \(m_c\) is the cumulative oil production.

The segmentation data in three states of core No. 34-1 and No. 64 can be calculated: dry samples, saturated oil samples, and after imbibition, as shown in Table 6.

According to the data in Tables 2 and 4, the oil saturations of the two cores are 96.0% and 94.36%. In the segmentation process of CT image after saturated oil (Table 6), the oil saturation can be calculated as 95.6% and 94.0%. The porosity measured by experiments and calculated by images is 12.75% and 9.18% (Table 2), 12.72% and 9.10% (Table 6), respectively. It indicates that the image segmentation is reasonable. Finally, using Equation (10), the imbibition recovery ratio based on the micro-CT scan image is obtained, as shown in Table 7.

\[
Q_{CT} = \frac{V_w}{V_w - V_{ro}} \times 100\% \quad (10)
\]

where \(Q_{CT}\) is the imbibition recovery ratio based on the CT image, \(V_w\) is the volume of the water phase, and \(V_{ro}\) is the volume of residual oil.

The imbibition recovery ratios obtained based on the spontaneous imbibition experiment and micro-CT scanning image are basically consistent with each other. Core No. 64 has a much higher imbibition recovery ratio than core No. 34-1.

5.2 Influence of Jamin’s effect

As shown in Figure 10A,B, when water invades into the narrow throat, the oil phase would be discharged from wide channels. But the capillary force is sharply decreasing when the water phase transfers from the narrow throat to the wide channels, see the Equation (2). As a result, the interface between oil and water would hardly be moved that caused the water phase cannot be continuously imbibed, as shown in Figure 10C,D.

The algorithm developed by AlRatrout et al.\(^{47,48}\) is adopted to measure the contact angle and the fluid-fluid interface curvature at the pore scale after imbibition. The OpenFOAM solver is used to implement the mathematical model (Equations 6-8). The histograms of the measured oil-water interface curvature are shown in Figure 11A. Furthermore, we calculate capillary pressure by oil/brine interface curvature model with the Equation (3), \(\sigma\) is 0.05 N/m for cores No. 34-1 and No. 64. Figure 11B shows the histograms of the capillary pressure for two typical Jamin’s effects in the pore-throat chain. The capillary pressure can be positive and negative, which indicates that the pore-throat chain has mixed wettability.

The ratio of the pore size to the connected throat size is the most important factor to quantificate Jamin’s effect. In addition, with Figures 10E,F and 11, we find that the mean curvature (capillary force) is negative at the pore-throat, which prevents the water phase to continue to flow, and Jamin’s effect appears. Finally, the residual oil block is trapped in the large pore, and this phenomenon will terminate the spontaneous imbibition process.

5.3 Occurrence state of the microscopic water phase after imbibition

According to the Equation (1), the water phase of two cores is classified as three types: network water phase (0 < \(G\) < 0.3), cluster water phase (0.3 < \(G\) < 0.7), and isolated water phase (\(G\) > 0.7), as shown in Figure 12. The volume fraction of each type of core is calculated, as shown in Table 8.

The typical micro-occurrence of water phases is extracted, as shown in Figure 13. The network and cluster water phases are distributed in regular pores, which are also the main sites of imbibition. When the water phase flows along the narrow throat, under the comprehensive action of Jamin’s effect and capillary force, it begins to thin, break, and separate; the network water phase begins to separate into a cluster water phase and an isolated water phase; the cluster water phase begins to separate into the isolated water phase. The isolated water phase is basically distributed in unconnected small pores.

5.4 Effect of the pore size distribution

As shown in Figure 14, the \(T_s\) spectra are obtained in the experimental test from the large core of 2.5 cm in diameter and 2.5 cm in length, while the pore size distribution is calculated based on micro-CT images from the small core with size of 6 mm in diameter and 100 mm in length drilled on the large core. There is a certain difference between the experimental data and the calculated result, but the overall distribution rule was consistent.

Then, the water phase diameter distribution histogram (Figure 15) is obtained. The distribution of the water phase first increases and subsequently decreases with the increase in diameter, which indicates the following:
1. The two cores are in the 0-0.5 μm pore size interval, which basically consists of tiny pores that do not participate in the imbibition process.

2. Although the water phase begins to appear in the pore size interval of 0.5-1 μm, this part is basically the isolated water phase, which accounts for 1.497% and 1.994% of the volume of the entire imbibition water phase.

3. In the pore size interval of 1-5 μm, the imbibition water phase begins to change from isolated to cluster, which accounts for 11.81% and 15.47% of the total volume of the water phase.

4. In the pore size range of 5-12 μm, the cluster water phase begins to connect and forms the network water phase, which accounts for 47.01% and 48.54% of the volume of the entire imbibition water phase.

5. In the pore size interval of 12-25 μm, the network water phase gradually begins to disperse and changes into clusters due to the increase in pore size, which decreases the capillary force.

6. Finally, in the interval above 25 μm, the cluster water phase begins to separate under the action of the weak capillary force and forms isolated water droplets again, which account for 6.17% and 5.89% of the volume of the entire imbibition water phase.

Table 9 shows the proportion of water phase distribution in different pore sizes. From Table 9, the distribution evolution rule of the water phase in the pore is consistent with the occurrence state of the three-dimensional microscopic water phase in the micro-CT image, which is analyzed by using the shape factor.

The proportion of the pore size distribution is further quantified and classified into three categories: non-imbibition area, sub-main imbibition area, and main imbibition area. Specific classification results are shown in Table 10.
In the imbibition process of the fractured reservoir, the main imbibition area should be taken as the key area.

Table 6

| ID  | Sample status | Pore volume | Isolate pore volume | Water volume | Residual oil volume | Rock grain volume | Total volume | Porosity/% |
|-----|---------------|-------------|---------------------|--------------|---------------------|-------------------|--------------|------------|
| 34-1| Dry           | 0.008703    | –                   | –            | 0.059738            | 0.068441          | 12.72%       |
|     | Saturated oil | –           | 0.000383            | –            | –                   | –                | –            | –          |
|     | Imbibition    | –           | –                   | 0.002211     | 0.005792            | –                | –            | –          |
| 64  | Dry           | 0.005591    | –                   | –            | 0.055863            | 0.061454          | 9.10%        |
|     | Saturated oil | –           | 0.000335            | –            | –                   | –                | –            | –          |
|     | Imbibition    | –           | –                   | 0.003113     | 0.002437            | –                | –            | –          |

Table 7

| No. of samples | Imbibition recovery ratio |
|----------------|--------------------------|
| 34-1           | 27.63%                   |
| 64             | 56.09%                   |

5.5 Effect of the complex mixed wettability

The wettability of the core samples was tested using the sessile drop method. As shown in Figure 16, water droplet and oil droplet are adopted for the sessile drop method test. The tested contact angle of the core No. 34-1 is 21.09° for water droplet (Figure 16A), and the test contact angle is 56.59° for oil droplet (Figure 16B). This indicates that core No. 34-1 is mixed-wetted. Similarly, core No. 64 is also mixed-wetted.

Figure 10 Jamin’s effect: 2D slices of the oil and water distribution: (A) core No. 34-1; (B) core No. 64; two typical Jamin’s effects in the pore throat; (C) core No. 34-1; (D) core No. 64; oil-water interface curvature surface; (E) core No. 34-1; and (F) core No. 64. Note: blue represents water, red represents oil, and the arrow indicates the flow direction.

Figure 11 Histograms of the calculated distributions of the measured (A) oil/brine interface curvature and (B) capillary pressure.
As shown in Figure 17, there are water-wetted pores, oil-wetted pores, and mixed pores. More interestingly, the oil-wetted and water-wetted walls (mixed wettability at the pore scale) coexist in the same pore. The schematic diagram of the mechanism of spontaneous imbibition in low-permeability mixed-wettability rock is shown in Figure 18. The imbibition does not occur in strong oil-wetted pore (Figure 18A), while water is completely absorbed along strong water-wetted pore wall (Figure 18B) and oil is displaced. In mixed-wetted pore, the water is absorbed along the water-wetted wall and stops when it encounters the oil-wetted wall (Figure 18C).

Two typical mixed-wetted pores are selected in each core, as shown in Figure 19. In Figure 20A, the contact angle calculated from the image is represented by the angle between the oil/brine and brine/rock interfaces, which is based on the water phase. Therefore, if the contact angle is less than 90°, it is water-wetted. When the contact angle is greater than 90°, it is oil-wetted. The core No. 34-1 is also proven to be mixed-wetted by image analysis. Similarly, core No. 64 is also mixed-wetted.

We define that the proportions of water phase surface area to the total surface area are used to characterize the water area ratio, see the Equation (11):

\[ R = \frac{A_w}{A} \times 100\% \]  (11)

where \( R \) is the water area ratio, \( A_w \) is the water phase surface area, and \( A \) is the total surface area; \( A_w \) and \( A \) are calculated through CT segmented images.

In addition, the proportion of contact angle below 90° to the total contact angle is used to characterize the water-wetted ratio. see the Equation (12):

\[ W = \frac{\theta_w}{\theta} \times 100\% \]  (12)

where \( W \) is the water-wetted ratio, \( \theta_w \) is the frequency of contact angle below 90°, and \( \theta \) is the frequency of total contact angle; \( \theta_w \) and \( \theta \) are calculated by AlRatrout.47

**TABLE 8** Different types of the water phase volume fraction based on the CT image

| ID of samples | Type   | Volume/cm³ | Volume fraction (%) |
|---------------|--------|------------|---------------------|
| 34-1          | Network| 0.00101051 | 45.73               |
|               | Cluster| 0.00103865 | 47.00               |
|               | Isolated| 0.000160761| 7.27                |
| 64            | Network| 0.000708035| 48.61               |
|               | Cluster| 0.00196568 | 43.25               |
|               | Isolated| 0.000342007| 8.14                |
Moreover, the ratio of the water phase volume to the total volume of the pore is used to characterize the imbibition recovery ratio at the pore scale, see the Equation (13):

$$Q_p = \frac{V_w}{V} \times 100\%$$

where $Q_p$ is the imbibition recovery ratio at the pore scale, $V_w$ is the water phase volume, and $V$ is the total volume of the pore, $V_w$ and $V$ are calculated through CT segmented images.

According to Figure 21A, the $R$ is linearly correlated with the $W$, which indicates that at the pore scale, greater contact

**Figure 13** Different types of water phase distributions: network water phase distributions of (A) core No. 34-1 and (B) core No. 64; cluster water phase distributions of (C) core No. 34-1 and (D) core No. 64; isolated water phase distributions of (E) core No. 34-1 and (F) core No. 64. Note: Blue represents water, and green represents the pores

**Figure 14** Measured pore size distribution: (A) Pore size distribution curve based on the NMR test; (B) histogram of the pore size distribution calculated based on the micro-CT scan image
between the water phase and the pore wall corresponds to the stronger water wettability of the entire pore. Furthermore, $Q_p$ is also linearly correlated with the $W$ (Figure 21B). Thus, a higher water wettability of the entire pore corresponds to a higher imbibition efficiency at the pore scale. In the imbibition process, a larger water-wetted wall corresponds to a higher imbibition recovery ratio.

Since the four typical mixed wet pores cannot represent generality, based on the above work, 20 mixed-wetted pores are randomly selected for each sample, and the correlation analysis is conducted, as shown in Figure 22A,B. These results also show a consistent correlation.

In addition, as shown in Figure 22C, the mean capillary pressure is linearly correlated with the $Q_p$. With the increase in mean capillary force, the $Q_p$ increases. The mean capillary force in typical pores are both positive and negative. It is well known that imbibition cannot occur with negative capillary force at the reservoir scale. However, in our work, a negative mean capillary force can also produce an imbibition effect because the positive capillary force on the water-wetted wall of a single pore is the driving force, and effective imbibition can occur at the pore scale. When the oil-wetted wall is encountered, the capillary force becomes negative and resists the imbibition effect, so the imbibition process stops.

6 | CONCLUSIONS

1. We integrate two methods to quantitatively calculate the microimbibition recovery: base on the imbibition experiment, the microimbibition recovery is 27.7% and 58.2% for core No. 34-1 and core No. 64, respectively; base on micro-CT scanning images calculation, the microimbibition recovery is 27.63% and 56.09% for core No. 34-1 and core No. 64, respectively.
2. When the water phase moves to the narrow throat, the negative mean capillary force at the water-oil interface

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**TABLE 9** The proportion of different types of water phases in the pores

| ID of samples | Type   | Pore size (μm) | Proportion (%) |
|---------------|--------|----------------|----------------|
| 34-1          | Network| 5-12           | 47.01          |
|               | Cluster| 1-5, 12-25     | 45.33          |
|               | Isolated| 0.5-1, >25   | 7.66           |
| 64            | Network| 5-12           | 48.54          |
|               | Cluster| 1-5, 12-25     | 43.58          |
|               | Isolated| 0.5-1, >25   | 7.88           |

**TABLE 10** Pore size classification for imbibition

| Pore size classification   | Non-imbibition area/μm | Sub-main imbibition area/μm | Main imbibition area/μm |
|----------------------------|------------------------|----------------------------|-------------------------|
| Imbibition                 | 0-0.5 μm               | 0.5-1.0 μm                  | 1-25 μm                 |

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**FIGURE 15** Histogram of the water phase diameter distribution calculated based on the micro-CT scan image

**FIGURE 16** Wettability tested: oil and water drops on the core surface: (A, B) core No. 34-1; (C, D) core No. 64
FIGURE 17  Pore-scale wettability after imbibition: (A) core No. 34-1; (B) core No. 64

FIGURE 18  Schematic diagram of the mechanism of spontaneous imbibition in low-permeability mixed-wettability rock. (A) strong oil-wetted pore imbibition; (B) strong water-wetted pore imbibition; (C) mixed-wetted pore imbibition. Note: red represents oil, and blue represents water; the blue arrow indicates the water flow direction, and the red arrow indicates the oil flow direction

FIGURE 19  Typical mixed wetting at the pore scale: mixed wet pores of (A) and (B) No. 34-1 and (C) and (D) No. 64. Note: red represents oil, and blue represents water

FIGURE 20  Contact angle measurement at the pore scale: contact angle distribution of mixed-wetted pores of (A) No. 34-1 and (B) No. 64
directly causes Jamin’s effect, which will make the residual oil trapped in the large pores. Jamin’s effect will stop the imbibition process and reduce the imbibition recovery ratio.

3. The occurrence of the microscopic water phase is divided into three types: network water phase, cluster water phase, and isolated water phase. The calculated volume fractions of the three shapes of core No. 34-1 are 45.73%, 47%, and 7.27%, and those of core No. 64 are 48.61%, 43.25%, and 8.14%. These findings indicate that the network and cluster water were the main contribution areas.

4. The evolution of the water phase in different pore sizes is obtained by analyzing the pore size distribution and water phase diameter distribution. The pore size distribution is further quantified and classified into three categories: non-imbibition area, sub-main imbibition area, and main imbibition area.

5. The presence of water-wetted walls in a mixed wettability pore can also produce effective imbibition. The $R$ and $Q_p$ are almost linearly correlated with the $W$. A larger surface area of the water-wetted wall in the pore corresponds to stronger water wettability and higher imbibition recovery ratio. In addition, the mean capillary pressure is linearly correlated with the $Q_p$. A negative mean capillary force can also produce the imbibition effect at the pore scale.

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CONFLICT OF INTEREST

The authors declare that there is no competing financial interest with any other people or groups regarding the publication of this manuscript.

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