Spontaneous Imbibition of Capillaries under the End Effect and Wetting Hysteresis
Leilei Zhang, Keliang Wang,* Huiming An, Gen Li, Yu Su, Wei Zhang, and Xinyi Yang

ABSTRACT: The phenomenon of spontaneous imbibition is widely present in the development process of tight oil/gas reservoirs. To further explore the spontaneous imbibition behavior of capillary tubes to provide theoretical and methodological references for the study of microscopic porous media imbibition phenomena, the capillaries that can be observed with the naked eye on the order of 10−100 μm were selected as research objects. Based on the theory of interface chemistry, the capillary end effect, and wetting hysteresis, the influence of the additional pressures generated by the two-phase interface on the spontaneous absorption of the horizontal capillary was studied. Some of the capillaries were processed for wettability, and then the water wettability of different capillaries was measured by the introduced concept, which is the conversion height of the self-absorption phase in the capillary. The capillaries were horizontally placed in the liquid for a spontaneous imbibition experiment, and the air−liquid twophase menisci behavior was observed at the same time, and then the influence of water wettability, surfactant, and capillary diameter on spontaneous imbibition was discussed. It was found that in the equal diameter capillaries, the spontaneous air−liquid imbibition behavior of capillary tubes with different water wetting properties is different in sensitivity to surfactants and tube diameters; when surfactants are used to improve capillary water wettability to increase spontaneous imbibition efficiency, the initial water wettability of the capillary and the comprehensive changes in the capillary pressure caused by interfacial tension should be considered.

1. INTRODUCTION
Capillary spontaneous imbibition refers to the phenomenon that the wet phase displaces the nonwet phase only by capillary pressure without an external force. This phenomenon is widespread in industries such as construction, biochemistry, farmland irrigation, and mineral development.1−3 Waterproof and antiseepage of high-rise building roofs,6 soilless cultivation of plants by imbibition,7 water-saving irrigation using soil imbibition,8 development of coalbed methane and tight oil/gas reservoirs,9 etc. are all closely related to spontaneous imbibition.

With the development of the human society, the continuous expansion of the economic system, and the continuous increase of energy consumption, oil and natural gas, as important fossil energy sources, play a vital role in the progress of science and technology. Shale gas and tight oil represented by North America have changed the world energy pattern.10−12 The newly discovered oil/gas burial conditions in the Asia−Pacific region are becoming increasingly complex and demanding, especially low permeability and tightness.13−15 Spontaneous imbibition, as an important mechanism for the development of low-permeability oil/gas reservoirs, has been increasingly attracting attention from petroleum workers.

The behavior of the two-phase menisci in the capillary under different conditions is the basis for studying the influence on spontaneous imbibition, which is related to the matrix pore structure, the matrix opening surface, and reservoir/fluid physical properties in low-permeability oil/gas reservoirs.16 Washburn17 connected one side of a capillary with water and the other side with air. Under the action of the capillary pressure, water spontaneously enters the capillary and the air is discharged. He found that the displacement of the water−air meniscus is proportional to the square root of the interface tension and the capillary radius is proportional to the capillary water−air contact angle.20

Received: November 2, 2021
Accepted: January 21, 2022
Published: January 28, 2022

https://doi.org/10.1021/acsomega.1c06155
ACS Omega 2022, 7, 4363−4371
capillaries and believed that the fluid flow between two capillaries mainly occurs near the oil–water interface. Unsal et al.\textsuperscript{22,23} made irregular nonequal diameter capillary models with different degrees of lateral connectivity to study the curved surface behavior in small and large capillary diameters, and it was found that when the side is disconnected and the nonwet phase end is not closed, the curved surface migration velocity in the large capillary is higher, while when the side is connected or the nonwet phase end is closed, the curved surface migration velocity in the small capillary is higher. Hatiboglu et al.\textsuperscript{24,25} studied the relative flow between fracture and the matrix by a microscopic visualization model and a microscopic filling model. They found that the wet phase preferentially enters the small pore, and the nonwet phase is discharged through the larger pore. At present, the research on the interfacial motion behavior of the multiphase fluid in capillaries is mostly focused on the micrometer scale.\textsuperscript{26–28} During the experiment, a capillary is generally placed horizontally or vertically, the pressure regulating device is established at both ends of the capillary to control the entry of liquid in the capillary and pressure difference, and a microscope with a high-speed camera is used to observe the movement speed and displacement of the two-phase interface. However, the effects of the additional pressure generated by wetting hysteresis and the end effect on the spontaneous imbibition behavior are less studied.

To further explore the spontaneous imbibition behavior of capillary tubes and their influencing factors to provide theoretical and methodological references for the study of microscopic porous media imbibition phenomena, the capillaries that can be observed with the naked eye on the order of 10–100 \( \mu \text{m} \) were selected as research objects. The concept of the conversion height of the vertical self-absorption phase of a capillary tube was introduced. Through the measurement of the converted height, the water wetting performance of different capillaries was compared. The capillaries with different water wetting properties were horizontally placed in the liquid, and the initial state that the wetting phase is immersed into one end of the capillary with the help of vibration was created to perform a spontaneous imbibition experiment. By observing the behavior of the two-phase interface, the spontaneous imbibition of the capillary under the dual effects of wetting hysteresis and the capillary end effect was discussed; at the same time, the influence of the water wettability, surfactant, and capillary diameter on the spontaneous absorption of the capillary was analyzed.

## 2. RESULTS AND DISCUSSION

Experimental methods and experimental materials are given in Section 4. Schematic diagrams of experiments and the water quality analysis of the experimental water are shown in Section 4.

### 2.1. Capillary Water Wettability Measurement

The capillary pressure is affected by the dual effects of interfacial tension and wetting properties. According to the parameter relationship in the capillary pressure equation (eq 1), for the same diameter capillary, when the two-phase fluid is determined, the larger the value of \( \Delta \rho \times h \), the stronger the water wettability of the capillary. To determine the water wettability of the capillary by the rising height of the wet phase in the vertical capillary, the conversion height \( H = \Delta \rho \times h \) was defined.

\[
P_c = \frac{2\sigma \cos \theta}{r} = \Delta \rho g h = Hg
\]  

(1)

where \( \sigma \) is the surface tension, \( r \) is the capillary radius, \( \theta \) is the contact angle, \( \Delta \rho \) is the density difference between the two phases, \( h \) is the height of the meniscus, and \( H \) is the converted height of the meniscus.

As shown in Figure 1, under water–air interface conditions (WA), the \( H \) in the quartz capillary is a positive value, so relative to the air, water is the wet phase. However, under the surfactant solution–air interface conditions (SSA), the value of \( H \) is reduced and the capillary pressure decreases. As for how much water wettability may be changed after changing from WA to SSA, a comparative discussion will be made in the section that water wettability of quartz capillaries treated with mineral oil was measured.

As shown in Figure 2, under WA conditions, the \( H \) in the silicone capillary is a negative value, so relative to the air, water is the nonwet phase. After the surfactant was added, the \( H \) changes from a negative value to a positive value. Meanwhile, relative to the air, the water changes from a nonwet phase to a wet phase.

As shown in Figure 3, under WA conditions, the \( H \) in the quartz capillary treated with mineral oil is a positive value, so relative to the air, water is the wet phase. After the surfactant was added, the value of \( H \) and the capillary pressure increase. The addition of surfactants improves the water wettability of the capillary and reduces the interfacial tension. The former increases the capillary pressure and the latter reduces the capillary pressure. We believe that the former has a greater impact on the capillary pressure than the latter, so the capillary pressure increases. Comparing to Figure 1, we found that the effect of a surfactant on the water wettability of the capillary

\begin{figure}[h]
\centering
\includegraphics[width=0.8\textwidth]{quartz_capillary_water_wettability.png}
\caption{Quartz capillary water wettability.}
\end{figure}

\begin{figure}[h]
\centering
\includegraphics[width=0.8\textwidth]{silicone_capillary_water_wettability.png}
\caption{Silicone capillary water wettability.}
\end{figure}
Capillary action refers to the wetting of the solid surface by the liquid due to the difference in the cohesion of the liquid and the adhesion between the liquid and the solid when the liquid is in contact with the tube wall. The shape of the two-phase meniscus in the capillary, the radius of the curvature of the meniscus, and the size of the two-phase interfacial tension directly determine the size and the direction of the capillary pressure.

As shown in Table 1, under the condition of WA and an equal pipe diameter, the menisci do not move in the capillaries with radii of 50 and 25, while the menisci in the capillaries with radii of 15 and 5 move and bubbles are continuously expelled. Under the condition of an equal pipe diameter and SSA, all menisci move and bubbles are continuously expelled. Under the condition of a nonequal pipe diameter, all menisci move from the small pipe diameter end to the large pipe diameter end, and bubbles are continuously discharged, which is consistent with the results of previous studies.

Some researchers found that the wetting angle at the meniscus of the entry end was not constant using different liquids to test the spontaneous imbibition process of the capillary. During the movement of the meniscus, the dynamic contact angle is greater than the static contact angle and is related to the self-absorption speed of the fluid. The capillary end effect refers to an additional resistance phenomenon caused by the deformation of the meniscus when the capillary suddenly loses continuity, and the nonwet phase leaves the capillary port to enter the wet phase. As shown in Figure 4, the capillary pressure (PC) and the additional pressure (P) are generated by the menisci at both ends of the air phase in the capillary tube, and the magnitude and the direction of the pressures are the key factors for the movement of the meniscus and expulsion of bubbles. For a horizontally placed capillary, the hydrostatic pressures at two ports are equal and opposite, and if the fluid viscosity is neglected, the fluid system in the capillary is only affected by the curved surface pressure difference generated by the menisci at both ends of the air phase.

For the process of moving the meniscus in the equal diameter capillary, we believe that the wetting hysteresis will cause the dynamic contact angle of the meniscus at the inlet end to increase first and then fluctuate around a certain value, which means that the capillary pressure PC is reduced to a certain value and then levels out. However, the radius of the curvature of the meniscus at the bubble discharge end of the capillary decreases first and then increases, which causes the additional pressure P to increase first and then decrease, as shown in Figure 4. During the entire period from the bubble generation to the bubble leaving the capillary, the additional pressures of the meniscus at both ends of the nonwet phase change with the displacement of the meniscus at the liquid inlet end, as shown in Figure 5. When the pressure difference value is always positive, as shown in Figure 5a, the meniscus keeps moving and bubbles are continuously discharged. When the pressure difference appears negative, as shown in Figure 5b, the wetting hysteresis phenomenon is observed, and the meniscus may stop moving or the bubble may not be expelled.

### Table 1. Behavior of the Meniscus in Horizontal Quartz Capillaries

| radius/10 μm | WA | SSA |
|--------------|----|-----|
|              | left | right | meniscus behavior | left | right | meniscus behavior |
| 50           | 50    | static     | 50 | 50    | move and bubbles are expelled |
| 25           | 25    | static     | 25 | 25    | move and bubbles are expelled |
| 15           | 15    | move and bubbles are expelled | 15 | 15    | move and bubbles are expelled |
| 5            | 5     | move and bubbles are expelled | 5  | 5     | move and bubbles are expelled |
| 50           | 25    | move from right to left and bubbles are expelled | 50 | 25    | move from right to left and bubbles are expelled |
| 15           | 5     | move from right to left and bubbles are expelled | 50 | 15    | move from right to left and bubbles are expelled |
| 50           | 15    | move from right to left and bubbles are expelled | 50 | 5     | move from right to left and bubbles are expelled |

*Note: The static of the meniscus means that there is no meniscus in the capillary, and the air fills the entire capillary, or the meniscus has a certain displacement in the capillary due to the instant high-frequency vibration of the capillary, but there is no continuous bubble discharge behavior.*
and if the meniscus moves under the action of inertia and crosses the negative interval, the meniscus can keep moving and bubbles are continuously discharged, but if the movement speed of the meniscus is reduced to 0 in the negative interval, the fluid system inside the capillary reaches a force balance under the dual adjustment of the end effect and the wetting hysteresis, and the meniscus remains stationary and the capillary stops expelling bubbles.

$$p = \sigma \left( \frac{1}{R_1} + \frac{1}{R_2} \right) = A\sigma$$  \hspace{1cm} (2)

where $R_1$ and $R_2$ are the parameters describing the curvature radius of the meniscus, $\sigma$ is the interfacial tension of the two phases, and $A$ is the parameter related to the curvature radius. The larger the radius of the curvature, the smaller the value of $A$.

The additional pressure $P$ produced by the end effect of a single capillary in the process of imbibition is opposite to the capillary force $PC$, and $P$ is the resistance in the process of imbibition. The $P$ on the curved surface can be described by the Laplace equation (eq 2). The addition of surfactants reduces the interfacial tension $\sigma$, so from the Laplace equation (eq 2), it can be seen that the additional pressure $P$ and the change range of $P$ at the bubble discharge end reduce compared to WA conditions. Moreover, as shown in Figure 1 above, the addition of surfactants also reduces the capillary pressure $PC$ at the liquid inlet end. As shown in Table 1, under WA conditions, the meniscus moves, and bubbles are continuously discharged after the capillary diameter decreases. We analyze that with the decrease of the capillary diameter, the increase of the capillary pressure $PC$ at the liquid inlet end is higher than the increase of the additional pressure $P$ at the bubble discharge end, which also increases the pressure difference between the two ends, as shown in Figure 9. Therefore, the reduction of the capillary

![Figure 5](image1)  
**Figure 5.** Variation of meniscus pressures at both ends of the nonwet phase in the equal diameter capillary. (a) Pressure difference is always positive and (b) there is a negative pressure difference interval.

![Figure 6](image2)  
**Figure 6.** Influence of the surfactant on meniscus pressures in the strong water wetting capillary: $r = 25 \times 10^\mu m^2$.

![Figure 7](image3)  
**Figure 7.** Bubbles are continuously discharged: (a) phenomenon of the first experiment, (b) front view of the phenomenon in the second experiment, and (c) top view of the phenomenon in the second experiment.

![Figure 8](image4)  
**Figure 8.** Influence of tube diameter reduction on the meniscus curvature radius at the capillary bubble discharge end.
pressure to decrease at the liquid inlet end, and the additional pressure due to the capillary water wettability causes the capillary pressure $P_C$ to decrease at the bubble discharge end remains unchanged, so the pressure difference between the two ends is limited, and it is not enough to move the meniscus. Therefore, improving the water wettability by surfactants may not achieve favorable spontaneous imbibition, and the initial water wet strength of the capillary is the key to the occurrence of spontaneous imbibition. However, pieces of literature related to oil production by imbibition show that a surfactant solution can significantly improve the effect of spontaneous imbibition under suitable interfacial tension conditions (mostly $10^{-3}$ mN/m), which makes the efficiency of spontaneous imbibition reach a higher level.\(^\text{37,38}\) According to analysis, different surfactants have different abilities to improve the spontaneous imbibition of capillaries with different water wettabilities. The addition of the surfactant can reduce the wetting angle $\theta$ and interfacial tension $\sigma$ at the same time, but the change of the capillary pressure is opposite. The initial water wet strength of the capillary and the properties of the surfactant directly determine the change of the capillary pressure, which, in turn, affects the effect of spontaneous imbibition.

2.4. Spontaneous Imbibition of the Quartz Capillary with One End Treated. In the process of mineral oil processing the quartz capillary, one end of the quartz capillary is immersed in the mineral oil, and the oil is instantly adsorbed on the inner wall surface of the whole capillary in the form of a film.\(^\text{39-41}\) Due to the internal viscous force of the liquid, even if the oil is blown out from the other end of the capillary, the thickness of the oil film adhering to the inner wall of the capillary at the immersion end is still greater than that at the other end, as shown in Figure 11.

As shown in Table 3, the menisci in the equal diameter capillary do not move under WA conditions. After the surfactant was added, the menisci in the capillaries move from the end with the thicker oil film to the end with the thinner oil film, and bubbles are continuously discharged. It is

### Table 2. Meniscus Behavior of the Treated Quartz Capillary at the Horizontal Level

| radius $\mu$m | meniscus behavior in the condition of WA | meniscus behavior in the condition of SSA |
|---------------|---------------------------------|---------------------------------|
| 5             | static                          | static                          |
| 15            | static                          | static                          |
| 25            | move from right to left and bubbles are discharged | move from right to left and bubbles are discharged |
| 50            | move from right to left and bubbles are discharged | move from right to left and bubbles are discharged |
| 50            | move from right to left and bubbles are discharged | move from right to left and bubbles are discharged |

*Figure 9. Influence of pipe diameter reduction on the meniscus pressures on both sides of the nonwet phase: (a) $r = 5 \times 10^{-3} \mu$m and (b) $r = 25 \times 10^{-3} \mu$m.*

*Figure 10. Influence of the decrease in water wetness on the meniscus pressures in the capillary: $r = 5 \times 10^{-3} \mu$m.*

*Figure 11. Schematic diagram of spontaneous imbibition of the quartz capillary treated at one end.*
analyzed that under the condition of WA, due to the reduced water wettability of the capillary, the capillary pressure PC at the inlet end is small, and the pressure difference between the two ends is not enough to push the meniscus to move. After the surfactant is added, the active agent molecules diffuse and adsorb on the inner wall of the capillary, which improves the water wettability of the capillary and increases the capillary pressure PC at the inlet end. In addition, the actual flow cross-sectional radius of the fluid at the thicker oil film end is smaller than that at the other end, which further increases the capillary pressure PC. But the actual flow cross-sectional radius of the fluid at the end of the thinner oil film is larger than that at the other end, which makes the additional pressure P at the end of the capillary outlet smaller. The pressure difference between the two ends is increased to the extent that it is sufficient to push the meniscus to move, so the meniscus moves from the thick oil film end to the thin oil film end, and bubbles are continuously discharged. The thickness of the nonwet phase on the inner wall of the equal diameter capillary affects the direction in which the wet phase is immersed.

### 2.5. Spontaneous Imbibition of Capillaries with Different Water Wettabilities at Both Ends

As shown in Table 4, under SSA conditions, all of the menisci move from the quartz end to the silicone end, and bubbles are continuously discharged. Although the diameter of the silicone end is smaller than that of the quartz end, there is no phenomenon that the meniscus moves from the silicone end to the quartz end.

As shown in Figures 1 and 2 above, although the addition of a surfactant reduces the capillary pressure of the quartz capillary and increases the capillary pressure of the silicone capillary, comparing the value of \( H \) in the same diameter of the quartz capillary and the silicone capillary under SSA conditions, it is found that the range of reduction and increase is limited, and the capillary force of the former is significantly greater than that of the latter. It is analyzed that even if the water wettability of the silicone capillary is improved by surfactants, only when the quartz end is used as the entry end, the pressure difference is sufficient to move the meniscus, and bubbles are discharged. It is further illustrated that the initial water wet strength of the capillary is the key to the occurrence of spontaneous imbibition.

### 2.6. Comment on the Limitations of the Experiments and Plans for Future Work

Spontaneous imbibition is an effective way to develop tight oil/gas reservoirs. Liquid can penetrate into the reservoir through self-absorption. Therefore, an in-depth study of the capillary imbibition behavior is of great significance for understanding the spontaneous imbibition mechanism of displacement fluid in the bedrock. In actual low-permeability tight oil/gas layers, the pores and fractures are mostly nanometric in size,16,26 and the physical and chemical properties are complex and changeable. The wetting hysteresis and end effects are influenced by many factors, such as the roughness of the pore throat surface, the cross-sectional shape of the pore throat, the composition of the adsorption layer on the pore throat surface,27 and nonuniform wetting of the pore throat. To improve the application effect of imbibition oil recovery in the development of tight oil/gas reservoirs, we need to combine a large number of actual oil reservoir imbibition oil production data, statistically distinguish the main macrocontrol factors that affect imbibition oil production, and further subdivide oil and gas reservoir categories. In addition, the effect of the increase of the dynamic contact angle and its fluctuation range due to the wetting lag in the spontaneous imbibition of the capillary needs to be further explored.

### 3. CONCLUSIONS

Through the abovementioned experiments and discussion, the following conclusions can be drawn within the capillary size range of the order of 10–100 \( \mu \text{m} \).

1. When the capillary diameter and two-phase fluid are unchanged, it is good that the method of measuring the converted height of the self-absorbent phase is used to determine the strength of the capillary water wettability.
2. In strong water wetting equal diameter capillaries, the addition of surfactants or the reduction of the capillary diameter is conducive to the occurrence of spontaneous imbibition.
3. In weak water wetting equal diameter capillaries, the addition of a surfactant or the change of the capillary diameter has little effect on spontaneous imbibition.
4. In the equal diameter capillary, the addition of a surfactant is easy to make the end with the thicker hydrophobic adhesion layer on the inner wall of the capillary become the entrance end of the meniscus.
5. In terms of surfactants to improve wettability, weakly wetted materials are better than strongly wetted materials. However, the strong initial water wettability of the capillary plays a more important role in spontaneous imbibition.
6. When surfactants are used to change the water wettability of materials to improve spontaneous imbibition efficiency, the initial water wet strength of
the capillary and the comprehensive changes in the capillary pressure caused by interfacial tension need to be considered.

4. MATERIALS AND METHODS

4.1. Materials. The main component of mineral oil is \( \text{C}_{16}^{-\text{C}_{31}} \) normal alkanes and isoparaffins, the relative molecular mass is in the range of 300–400, the surface tension is 23 mN/m, and the apparent viscosity is 20 MPa·s at atmospheric pressure and 25 °C. The water quality analysis of experimental water is shown in Table 5. The surface tension of the water is 72.1 mN/m at 25 °C and atmospheric pressure. A BHS-01A anionic surfactant has good interfacial activity, it can reduce the oil–water interfacial tension to \( 10^{-3} \) mN/m and change the wettability of quartz and silicone. The material of the quartz capillary is uniform, and the circulation radii are 50, 150, 250, and 500 μm, respectively. The material of the silicone capillary is uniform too, and the flow radii are 250, 400, and 500 μm, respectively. The silicone capillary is an organic silica capillary, which is like a rubber capillary, and the quartz capillary is a glass capillary. The wettability of the two is quite different.

4.2. Experimental Methods. 4.2.1. Capillary Water Wettability Measurement. Quartz capillary tubes and silicone capillary tubes were individually placed on the capillary bracket and then vertically placed on the water–air interface (WA) and the surfactant solution air interface (SSA). The interface behavior in the capillary was observed through an optical microscope for a long time until the interface state was stable, and the height of the meniscus was recorded, as shown in Figure 12a. The measurements were repeated three times and the average value was recorded.

4.2.2. Mineral Oil-Treated Quartz Capillary. Some quartz capillaries with different diameters were totally immersed in mineral oil to discharge the bubbles. The capillaries were soaked for 48 h and then taken out. A rubber suction ball was used to blow out the oil in the capillaries, and then the capillaries were placed in a fume hood to dry for later use.

4.2.3. One End of the Quartz Capillary Treated with Mineral Oil. One end of quartz capillaries with different diameters was put in mineral oil upright for 48 h and then taken out. A rubber suction ball was used to blow out the oil in the capillaries from the nonoil-immersed end, and then the capillaries were placed in a fume hood to dry for later use.

4.2.4. Spontaneous Imbibition of Equal Diameter Capillaries. Different quartz capillaries and silicone capillaries were put on the capillary bracket individually. The capillary tubes were quickly placed horizontally in clean water or a 0.3% (wt) BHS-01A solution through the bracket. The instantaneous high-frequency vibration of the bracket was controlled so that the wetting phase was immersed into one end of the capillary, which is the initial state of spontaneous imbibition. The bubble discharge behavior of the capillary was observed through an optical microscope, and the additional pressure values at different displacements were calculated using the shape of the meniscus, as shown in Figure 12b. The experiment was repeated three consecutive times until the behavior was the same.

4.2.5. Spontaneous Imbibition of Unequal Diameter Capillaries. Capillary ports of different pipe diameters were connected, and the capillaries were made in the same straight line. The joint was sealed with petroleum jelly to make an unequal diameter capillary. The experiment method of spontaneous imbibition is the same as the one described above (Section 4.2.4).

4.3. Analysis of the Clear Water Quality

| Parameter | \( \text{HCO}_3^- \) (mg/L) | \( \text{Ca}^{2+} \) (mg/L) | \( \text{Mg}^{2+} \) (mg/L) | \( \text{Cl}^- \) (mg/L) | \( \text{SO}_4^{2-} \) (mg/L) | \( \text{K}^+ + \text{Na}^+ \) (mg/L) | Total Salinity (mg/L) |
|-----------|----------------|----------------|----------------|----------------|----------------|----------------|-----------------|
| Salinity  | 101.1          | 75.6           | 4.2            | 147.1          | 0.7            | 123.1          | 452.0           |

Figure 12. Schematic diagrams of experiments: (a) schematic diagram of the capillary water wettability performance test and (b) schematic diagram of the capillary spontaneous imbibition experiment.

### AUTHOR INFORMATION

**Corresponding Author**

Keliang Wang — Key Laboratory of Enhanced Oil and Gas Recovery, Ministry of Education, Northeast Petroleum University, Daqing 163318, China; orcid.org/0000-0003-3039-8470; Email: wangkeliang0608@126.com

**Authors**

Leilei Zhang — Key Laboratory of Enhanced Oil and Gas Recovery, Ministry of Education, Northeast Petroleum University, Daqing 163318, China; Baili College of Petroleum Engineering, Lanzhou City University, Lanzhou 730070, China

Huiming An — Baili College of Petroleum Engineering, Lanzhou City University, Lanzhou 730070, China

Gen Li — Key Laboratory of Enhanced Oil and Gas Recovery, Ministry of Education, Northeast Petroleum University, Daqing 163318, China
Yu Su — Key Laboratory of Enhanced Oil and Gas Recovery, Ministry of Education, Northeast Petroleum University, Daqing 163318, China
Wei Zhang — Key Laboratory of Enhanced Oil and Gas Recovery, Ministry of Education, Northeast Petroleum University, Daqing 163318, China
Xinyi Yang — Baili College of Petroleum Engineering, Lanzhou City University, Lanzhou 730070, China

Complete contact information is available at: https://pubs.acs.org/10.1021/acsomega.1c06155

Notes
The authors declare no competing financial interest.

■ ACKNOWLEDGMENTS
The authors are grateful for the financial support from the National Natural Science Foundation of China (No. 51974088) and the National College Students Innovation and Entrepreneurship Training Program (CN) (Nos. 202010737008 and 202110737005).

■ REFERENCES

(1) Liang, X.; Sheng, J. J. Comparison of Chemical-Induced Fracturing by Na2S2O8, NaClO, and H2O2 in Marcellus Shale. Energy Fuels 2020, 34, 15905–15919.
(2) Meng, M.; Ge, H.; Shen, Y.; Li, L.; Tian, T.; Chao, J. The effect of clay-swelling induced cracks on shale permeability during liquid imbibition and diffusion. J. Nat. Gas Sci. Eng. 2020, 83, No. 103514.
(3) Vinogradova, I. S.; Falaleev, O. V. Free water in bean seeds: the imbibition process observed by micro-magnetic resonance imaging. J. Struct. Chem. 2016, 57, 812–814.
(4) Zhao, J.; He, Y.; Li, X.; Weng, X.; Feng, D.; Ying, J.; Wang, Z. An integrated RNA-Seq and physiological study reveals gene responses involving in the initial imbibition of seed germination in rice. Plant Growth Regul. 2020, 90, 249–263.
(5) Parada, M.; Vontobel, P.; Rossi, R. M.; Derome, D.; Carmeliet, J. Dynamic wicking process in textiles. Transp. Porous Media 2017, 119, 611–632.
(6) Muhammad, N. Z.; Keyvanfar, A.; Majid, M. Z. A.; Shafaghat, A.; Mirza, J. Waterproof performance of concrete: A critical review on factors of Longmaxi Shale: Implications from Water Physisorption and Imbibition Measurements. Energy Fuels 2021, 35, 11958–11975.
(7) Savvas, D.; Gruda, N. Application of soilless culture technologies in the modern greenhouse industry—A review. Eur. J. Hortic. Sci. 2018, 83, 280–293.
(8) Yang, J.; Liu, K.; Wang, Z.; Du, Y.; Zhang, J. Water-saving and high-yielding irrigation for lowland rice by controlling limiting values of soil water potential. J. Integr. Plant Biol. 2007, 49, 1445–1454.
(9) Habibi, A.; Esparza, Y.; Boluk, Y.; Dehghanpour, H. Enhancing Imbibition Oil Recovery from Tight Rocks by Mixing Nonionic Surfactants. Energy Fuels 2020, 34, 12301–12313.
(10) Zhang, S.; Pu, H.; Zhao, J. X. Experimental and numerical studies of spontaneous imbibition with different boundary conditions: case studies of middle bakken and berea cores. Energy Fuels 2019, 33, 5135–5146.
(11) Wang, Q.; Zhou, W.; Hu, Q.; Xu, H.; Meensdén, F.; Shu, Y.; Qiao, H. Pore Geometry Characteristics and Fluid–Rock Interaction in the Haynesville Shale, East Texas, United States. Energy Fuels 2021, 35, 237–250.
(12) Chenghua, O.; Rui, R.; Chaochn, L.; Haiyan, Y. Multi-index and two-level evaluation of shale gas reserve quality. J. Nat. Gas Sci. Eng. 2016, 35, 1139–1145.
(13) Hou, C.; Yu, B.; Liu, L.; Xu, Y.; Zhang, Y.; Zhang, L.; Zhao, J.; Zhao, X.; Zuo, Q.; Sun, M. Water Uptake Behavior and Influence Factors of Longmaxi Implications: From Water Physiosorption and Imbibition Measurements. Energy Fuels 2021, 35, 11958–11975.
(35) Reynolds, C. A.; Blunt, M. J.; Krevor, S. Multiphase flow characteristics of heterogeneous rocks from CO₂ storage reservoirs in the United Kingdom. *Water Resour. Res.* **2018**, *54*, 729–745.

(36) Moghaddam, R. N.; Jamiolahmady, M. Steady-state relative permeability measurements of tight and shale rocks considering capillary end effect. *Transp. Porous Media* **2019**, *128*, 75–96.

(37) Tangirala, S.; Sheng, J. J. Roles of surfactants during soaking and post leak-off production stages of hydraulic fracturing operation in tight oil-wet rocks. *Energy Fuels* **2019**, *33*, 8363–8373.

(38) Saputra, I. W. R.; Park, K. H.; Zhang, F.; Adel, I. A.; Schechter, D. S. Surfactant-assisted spontaneous imbibition to improve oil recovery on the Eagle Ford and Wolfcamp shale oil reservoir: Laboratory to field analysis. *Energy Fuels* **2019**, *33*, 6904–6920.

(39) Puntervold, T.; Mamonov, A.; Piñerez Torrijos, I. D.; Strand, S. Adsorption of Crude Oil Components onto Carbonate and Sandstone Outcrop Rocks and Its Effect on Wettability. *Energy Fuels* **2021**, *35*, 5738–5747.

(40) Yu, L.; Wardlaw, N. C. The influence of wettability and critical pore-throat size ratio on snap—off. *J. Colloid Interface Sci.* **1986**, *109*, 461–472.

(41) Firincioglu, T.; Blunt, M. J.; Zhou, D. Three-phase flow and wettability effects in triangular capillaries. *Colloids Surf., A* **1999**, *155*, 259–276.