

## Research Article

# Experimental Investigation on the Effect of Pore Size on Spontaneous Imbibition Recovery in Oil-Wet Reservoirs

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Spontaneous imbibition has been considered as a significant method to enhanced oil recovery (EOR) in unconventional reservoirs. The main impediment to comprehending the variation characteristics of spontaneous imbibition at different pore scales was the reservoir's opacity. To this end, a series of spontaneous imbibition experiments of visualized oil-wet microtubes with six different diameters ( $10\ \mu\text{m}$ ~ $60\ \mu\text{m}$ ) were performed, as were the corresponding macroscopic core imbibition tests with six different permeabilities under the same conditions. The results showed that formation brine mostly advanced along the wall surface of a capillary during the imbibition process, and some were even isolated within a capillary. The imbibition recovery in the capillary with a diameter of  $10\ \mu\text{m}$  was the highest (27.81%), which was more than three times that of the capillary with a diameter of  $60\ \mu\text{m}$  (8.3%). There was a good power function decline relationship between capillary diameter and imbibition recovery, and  $30\ \mu\text{m}$  appeared to be a critical inflection point in both capillary tube and macroscopic core imbibition tests. In addition, the majority of the detained residual oil clusters not only cut off the continuity of formation brine but also increased the imbibition flow resistance, accelerating the imbibition to balance. This research provides a new perspective for comprehending the imbibition characteristics at different pore scales.

## 1. Introduction

Spontaneous imbibition is a capillary-dominated displacement process in which a nonwetting fluid is displaced from a porous medium by the inflow of a more-wetting fluid [1]. As the primary source of energy in unconventional reservoirs, spontaneous imbibition has made significant contributions to the recovery of matrix area and fracture area during the process of fracturing fluid reverse drainage and well soaking, and thus, it is regarded as an important tool to EOR [2–4]. However, the opacity of porous media and the complexity of the pore structure may be the two main barriers to comprehending spontaneous imbibition characteristics [5, 6]. Therefore, it is considerably significant to reveal the spontaneous imbibition characteristics of formation brine in petroleum reservoirs.

At present, NMR and CT techniques were widely used in the study of spontaneous imbibition in petroleum reservoirs [2, 4]. The first one was the research of imbibition via NMR technology, the flow analysis of tight sandstone at pore scale, and the effects of boundary conditions, wettability, and temperature and oil viscosity on spontaneous imbibition were studied by the combination of spontaneous imbibition experiment and NMR technology [7]. Nuclear magnetic resonance was implemented to determine the oil distributions before and after spontaneous imbibition and forced imbibition for six cores [8]. Through the combination of mercury intrusion curve and nuclear magnetic resonance technology to study the pore structure of tight cores at different stages of imbibition and the distribution characteristics of oil in different pore diameters, submicropores were the main type of oil production, and nanopores had greater capillary force

and their imbibition efficiency [9–11]. When NMR technology was used for imbibition research, it was not only time-consuming but also unable to observe the imbibition flow in real-time, which was also inconvenient to reveal the mechanism of spontaneous imbibition. The second one was the study of imbibition through CT technology. CT scanning of carbonate rocks with different permeability under strong water and humidity conditions was carried out to study the variation of water saturation plane with distance and time [12]. Using high-resolution CT scanning technology and three-dimensional image reconstruction technology, the changes of fluid distribution in the process of spontaneous infiltration and the remaining oil image in three-dimensional pore space were observed, and the occurrence state of the remaining oil in pore space after spontaneous infiltration was studied [13]. The physical parameters of the core were studied by CT technology, the imbibition experiments of different solutions were performed on the core, and the complex wettability behavior of the reservoir core on the pore scale was studied [14–16]. On the whole, the cost of recording the dynamic imbibition process was extremely high, and the variations of pores with different scales could not be obtained through CT technology. In addition to the above two technologies, the visual micromodel was also adopted to observe the oil morphologies after two surfactant imbibition, while the rheometer and the emulsion stability analyzer were applied to analyze formation mechanisms [17, 18]. Nevertheless, there have been few visual studies on the variation characteristics of formation brine imbibition in pores with different scales when they first contact. Furthermore, the application of capillary imbibition to core scale imbibition was also extremely scarce.

A series of visualized spontaneous imbibition experiments of formation brine in the capillary tubes with six different diameters ( $10\ \mu\text{m}\sim 60\ \mu\text{m}$ ) were carried out to comprehend the variation characteristics of formation brine imbibition in pores with different scales. To recreate the scene of the first contact between oil and water, an oil-saturated capillary with one end open and the other closed was chosen. In the meantime, the reason for reaching a stable equilibrium of spontaneous imbibition in a capillary was further analyzed. Based on the empirical relationship between core permeability and pore diameter, the corresponding six permeability core imbibition experiments were carried out under the same conditions. Finally, the empirical relationship between pore size and imbibition recovery could be obtained from the combination of capillary imbibition and core imbibition.

## 2. Experimental Section

**2.1. Experimental Material.** The target fracturing fluid in this experiment was Shengli Oilfield formation brine, and the crude oil used had a viscosity of  $8.26\ \text{mPa}\cdot\text{s}$  and a density of  $0.818\ \text{g}/\text{cm}^3$  ( $50^\circ\text{C}$ ). Furthermore, six capillary tubes with a length of  $6.35\ \text{cm}$  were etched on the chromium plate using a photolithography machine, and they were immersed in a mixed solution of hydrofluoric acid and nitric acid to deter-

mine the depth. The diameters of the capillary tubes were controlled in the range from  $10\ \mu\text{m}$  to  $60\ \mu\text{m}$ . More importantly, the etching depth of each capillary tube was proportional to its diameter. The etched chromium plate was then sintered together with another intact polished sheet in a high-temperature muffle furnace. Finally, the prepared microscopic model was vacuumed to age the saturated crude oil at high temperature ( $105^\circ\text{C}$ ), and the wettability was converted into a lipophilic capillary. In addition, the average contact angle of a water droplet on the lipophilicity glass was approximately  $66^\circ$  through the optical contact angle meter (OCA20, Germany). The oil-saturated micromodel was displayed in Figure 1.

**2.2. Experimental Apparatus.** The schematic diagram of the visualized capillary spontaneous imbibition experimental apparatus was shown in Figure 2. It could be mainly divided into four parts. The first one was the saturated part of the capillary tube, which included a high-precision injection pump, an intermediate container containing crude oil, and a vacuum pump. The second one was the spontaneous imbibition part of the capillary model, which contained a high-precision syringe pump, an intermediate container with fracturing fluid, and a container. The third one was the control part, which was composed of a micromodel holder and a microscope holder and a temperature control equipment. The last component was the observation section, which included a microscope, a camera, and a computer. The outlet end of the saturated microscopic model was closed, and the inlet end was placed in a petri dish containing fracturing fluid. Furthermore, each component pipeline was connected to complete the visualized spontaneous imbibition experiment device.

**2.3. Experimental Procedure.** To effectively calculate the imbibition recovery in capillaries, the mature drawing image processing technology was adopted in this experiment [19]. It was noted that the whole experiment process was kept at  $50^\circ\text{C}$ . The specific experimental procedures were as follows.

- (a) *Wettability Alteration.* After the microscopic capillary was evacuated, 3 pore volume (PV) crude oil was injected by a high-precision injection pump in constant flow mode of  $0.003\ \text{ml}/\text{min}$  for displacement saturation, and then 5 PV crude oil was continuously injected. Put it into a muffle furnace for sintering at  $80^\circ\text{C}$  for 3 days, and the capillary wettability was converted from hydrophilicity to lipophilicity.
- (b) *Tightness Inspection.* The micromodel was cooled to ambient temperature, and the experimental device was connected in turn to check the air tightness.
- (c) *Imbibition Environment Formation.* The formation brine was slowly injected into the petri dish at a constant flow mode of  $0.08\ \text{ml}/\text{min}$  until the micromodel was surpassed. It was noted that the inlet end of the micromodel was open so that oil and brine could contact each other. The outlet end of the

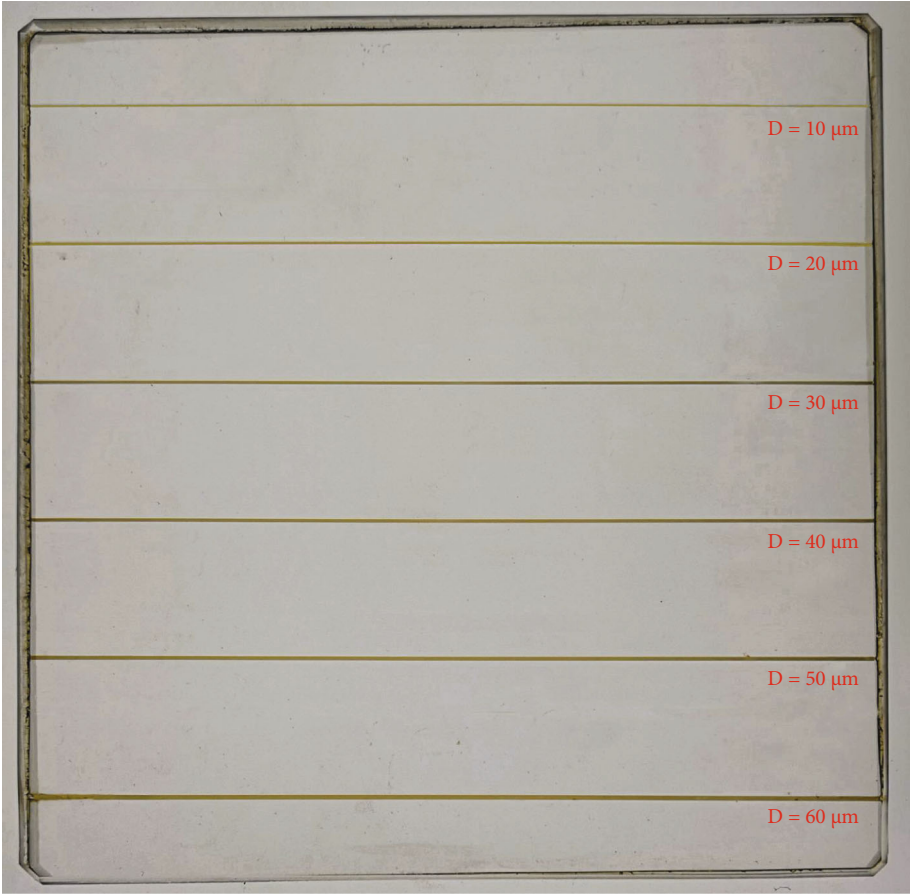


FIGURE 1: An overhead view of the oil-saturated visualized capillaries with six different diameters.

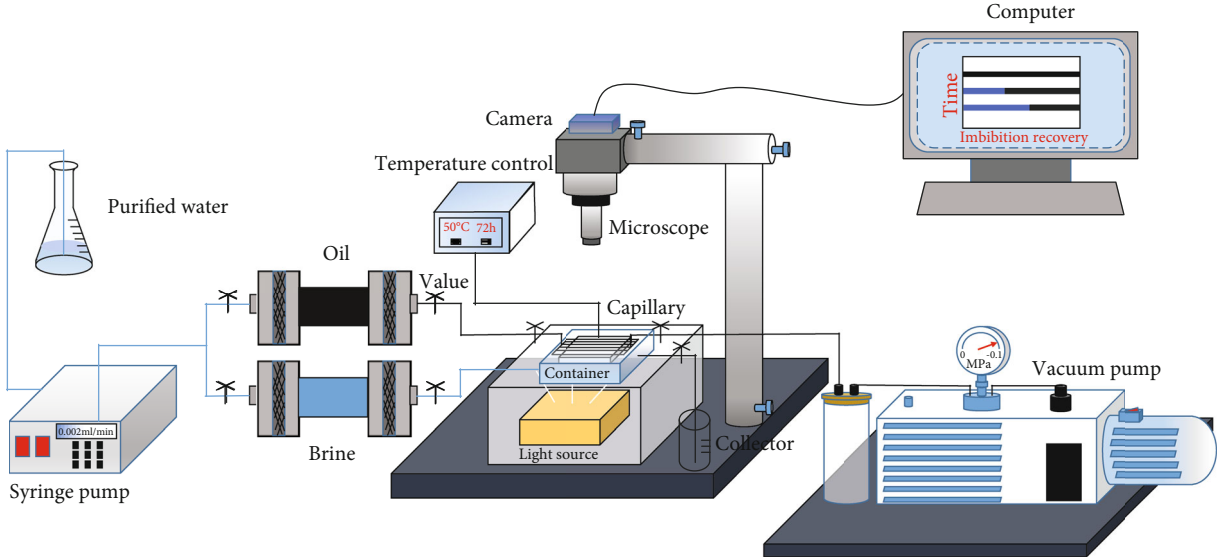


FIGURE 2: Schematic diagram of visualized spontaneous imbibition device.

micromodel was closed to prevent the brine from being injected into the capillary

- (d) *Observation Record Data.* The imbibition process was observed at two-day intervals, and the oil-water interface variations in each capillary were recorded through a high-resolution microscope
- (e) *Experiment Closure.* After the experiment, the pipeline and micromodel of the whole experimental device were cleaned through petroleum ether, ethanol, deionized water, and high pressure nitrogen in sequence

### 3. Results and Discussions

*3.1. Variation Characteristics of Spontaneous Imbibition in the Oil-Wet Capillary with Different Pore Sizes.* The relationships between imbibition time and imbibition distance in the oil-wet capillary with six different diameters were shown in Figure 3(a). It was clear that formation brine intruded preferentially into the capillary with a diameter of  $10\ \mu\text{m}$  to replace crude oil, and the imbibition effect was far superior to that in the other five capillaries. Meanwhile, the increase in the diameter of a capillary caused the imbibition effect to deteriorate [20, 21]. During the imbibition process, formation brine was clearly observed to mostly advance along the wall surface of a capillary, and some were even isolated within a capillary. In other words, a significant amount of crude oil was displaced along the internal wall surface during the imbibition process.

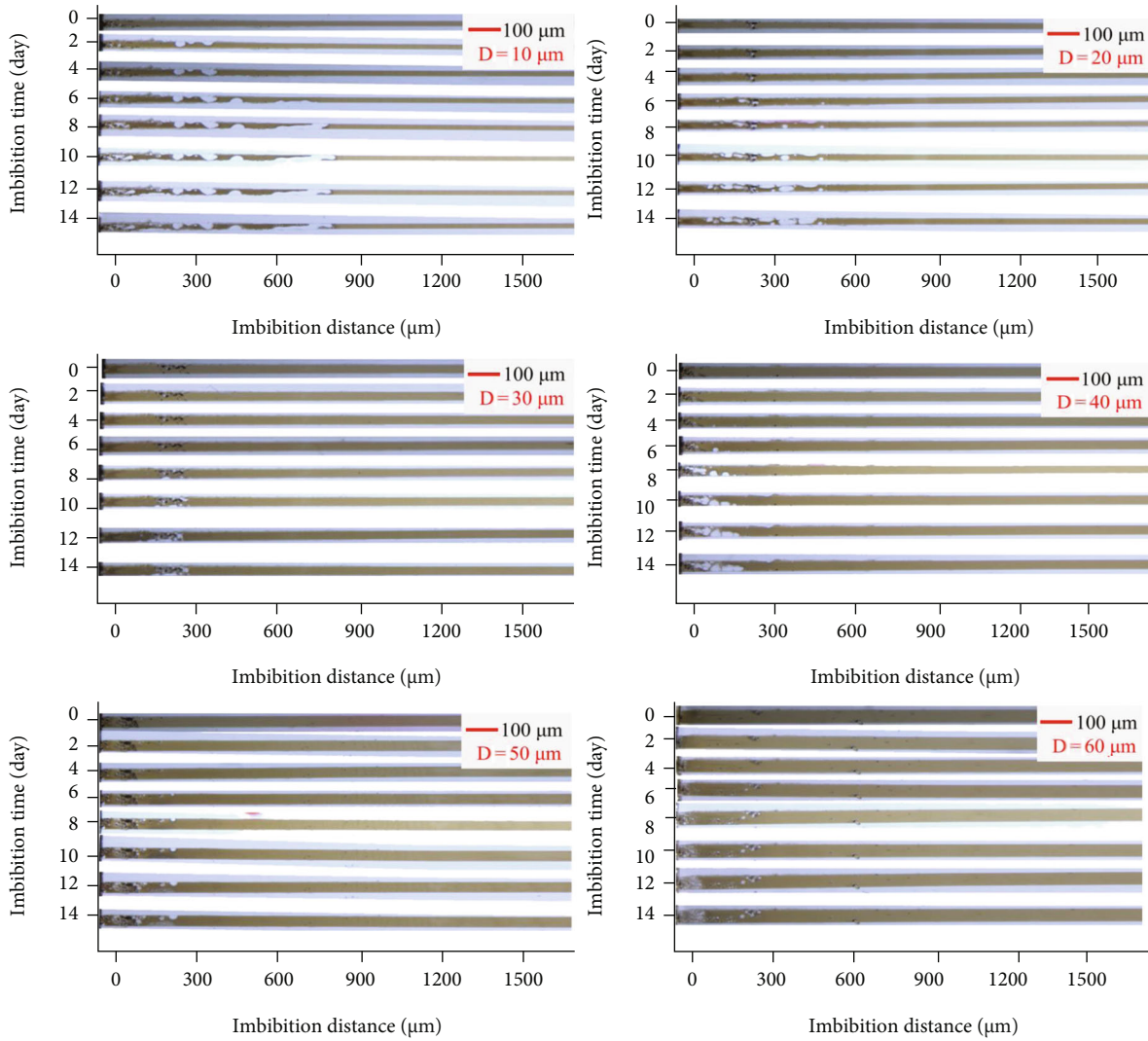
To quantitatively evaluate the effect of pore size on spontaneous imbibition recovery, the relationship between imbibition time and imbibition recovery in six different diameters capillaries was displayed in Figure 3(b), as was the relationship between microtube diameter and imbibition recovery in Figure 3(c). The spontaneous imbibition in capillaries of six different diameters was concentrated primarily in the first eight days or so. Imbibition recovery was 27.81% in the capillary with a diameter of  $10\ \mu\text{m}$ , which was more than three times that of the capillary with a diameter of  $60\ \mu\text{m}$  (8.3%). Meanwhile, in the diameters ranging from  $30\ \mu\text{m}$  to  $60\ \mu\text{m}$ , the effect of pore size on the imbibition recovery appeared to be negligible. In addition, a good power function decline relationship between capillary diameter and imbibition recovery was found, with  $30\ \mu\text{m}$  appearing to be a critical inflection point. This meant that the contribution of spontaneous imbibition to recovery with larger pore size was negligible.

*3.2. Microscopic Mechanism of Spontaneous Imbibition in the Oil-Wet Capillary with Different Pore Sizes.* It was well known that capillary force and viscosity force were two principal decision parameters of spontaneous imbibition recovery. Furthermore, the capillary force, which was the motive force of spontaneous imbibition, was inversely proportional to the pore size. On the contrary, the viscosity resistance was found to be closely associated with the velocity of the fluid during the imbibition process [22–24]. The spontaneous imbibition results of six different pore sizes capillaries were

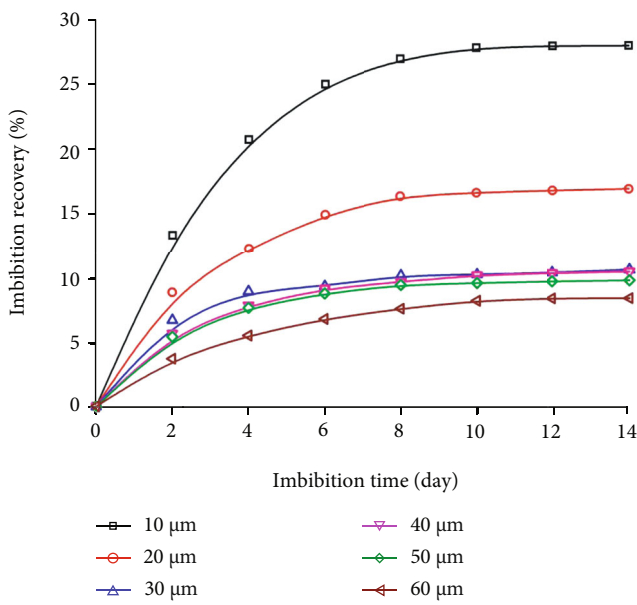
also demonstrated, as exhibited in Figure 4. It was clear that the motive power and resistance of spontaneous imbibition tended to balance after a period of time, rendering the oil was incapable of being expelled from a capillary.

It would be considerably meaningful to know how to make interpretation for the imbibition equilibrium in a capillary tube. As displayed by the red circular dashed line in Figure 4, there was plenty of residual oil clusters trapped in the spontaneous imbibition process, especially in capillaries with the diameters of  $10\ \mu\text{m}$  and  $20\ \mu\text{m}$ . In other words, spontaneous imbibition could not be piston displacement, but rather advanced along the dominant passageway. The majority of the residual oil clusters were linked together and closed to the wall surface, which not only cut off the continuity of formation brine but also increased the imbibition flow resistance. As a result, the spontaneous imbibition in the capillary tube was accelerated to reach equilibrium. For a capillary with a diameter of  $30\ \mu\text{m}$ , the retained residual oil blocked the flow passageway of spontaneous imbibition. The oil phase of imbibition replacement was most visible at the entrance of the other three capillaries. This was principally attributed to the relatively small imbibition motive force. As a result, disrupting the connectivity of replacement residual oil could improve spontaneous imbibition recovery to some extent. It was suggested that increasing the fluctuation of the nonwetting phase on an irregular basis helped to improve the imbibition recovery in reservoirs.

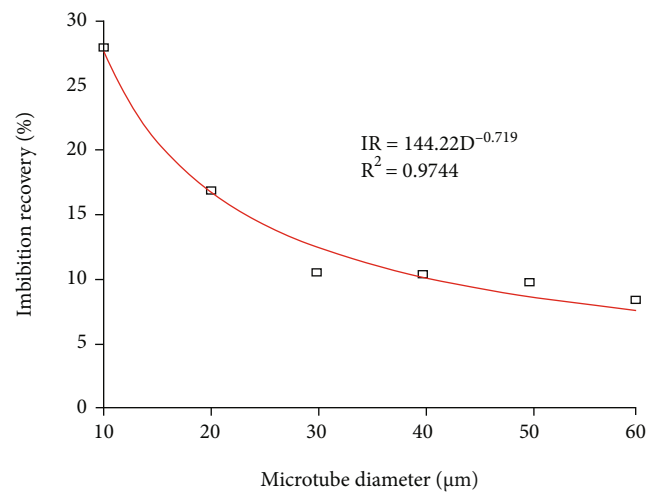
*3.3. Variation Characteristics of Spontaneous Imbibition in the Oil-Wet Macroscopic Cores with Different Permeabilities.* The idealized capillary model was upgraded to a core scale in order to make use of the above cognition derived from visual imbibition tests. Based on the classical Kozeny-Carman equation described relationship between permeability and pore radius, the permeability of rock samples corresponding to capillaries ranging from  $10\ \mu\text{m}$  to  $60\ \mu\text{m}$  could be estimated to be 80 mD, 300 mD, 700 mD, 1000 mD, 2000 mD, and 3000 mD [25, 26]. The oil-saturated artificial cores were aged for three days prior to the imbibition experiment. Then, in the same experimental environment, cores with six permeabilities were placed in the corresponding imbibition cells surrounded by formation brine. The spontaneous imbibition results of cores with six different permeabilities were exhibited in Figure 5(a). The imbibition effect was most noticeable during the first 40 hours. In comparison to the capillary results, the imbibition velocity of the core was fast, whereas the imbibition recovery was slightly low. Meanwhile, oil droplets were visible on all surfaces of the core, as shown in the upper left corner of Figure 5(a). All of this was caused by the emergence of buoyancy and gravity during the core imbibition process [27, 28]. Additionally, the relationship between permeability and imbibition recovery of core was displayed in Figure 5(b). The two parameters also showed a power function decreasing relationship, which was similar to that seen in the capillary imbibition. The core with a permeability of 700 mD ( $D = 30\ \mu\text{m}$ ) was used to assess the contribution of permeability to imbibition recovery, and the imbibition recovery



(a)



(b)



(c)

FIGURE 3: (a) Spontaneous imbibition results in the oil capillaries with six different diameters. (b) Results of imbibition time versus with imbibition recovery in six different diameter capillaries. (c) Relationship between microtube diameter and imbibition recovery.

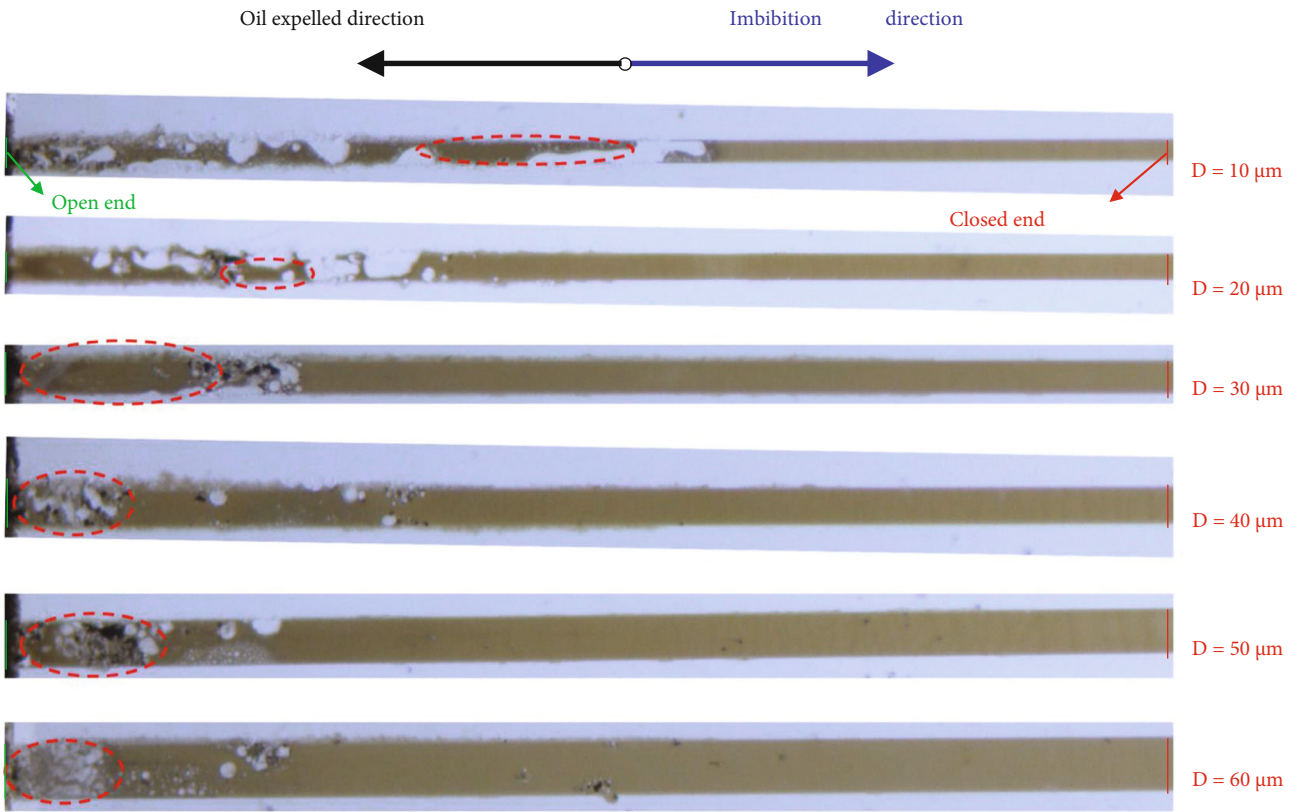


FIGURE 4: Final results of spontaneous imbibition in the left open-end location of capillaries with six different diameters.

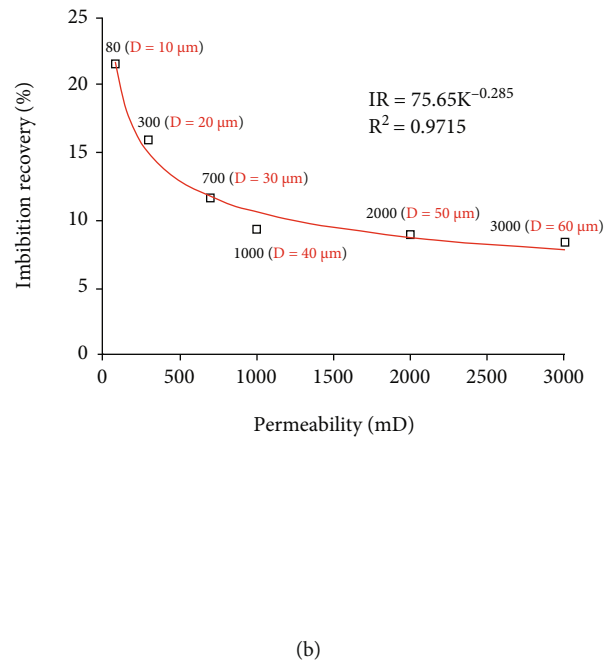
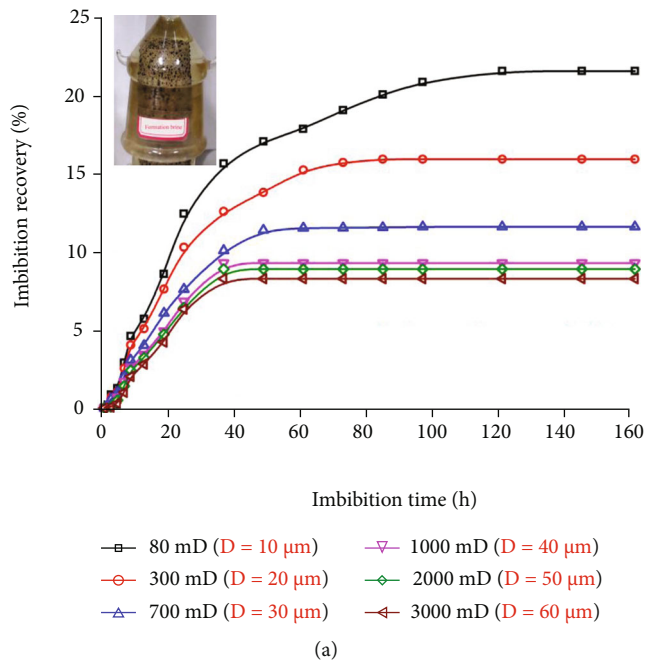


FIGURE 5: (a) Results of imbibition time versus with imbibition recovery in six different permeabilities cores. (b) Relationship between core permeability and imbibition recovery.

of cores beyond the critical point was nearly identical. When combined with the capillary imbibition results, spontaneous imbibition was ineffective for enhanced oil recovery in high permeability reservoirs.

#### 4. Conclusions

In this paper, a series of visualized spontaneous imbibition experiments of formation brine were performed in oil-wet microtubes with diameters ranging from 10  $\mu\text{m}$  to 60  $\mu\text{m}$  under formation conditions (50°C). The relationship between imbibition distance and imbibition time in six different microtubes was studied, as was the relationship between imbibition recovery and microtube diameter. Furthermore, the corresponding core imbibition experiments with six different permeabilities derived from the upgraded capillary diameters were carried out under the same conditions.

During the imbibition process, formation brine mostly advanced along the wall surface of a capillary, and some were even isolated within a capillary. Imbibition recovery was 27.81% in the capillary with a diameter of 10  $\mu\text{m}$ , which was more than three times that of the capillary with a diameter of 60  $\mu\text{m}$  (8.3%). In diameters ranging from 30  $\mu\text{m}$  to 60  $\mu\text{m}$ , the effect of microtube diameter on the imbibition recovery appeared to be negligible. Meanwhile, there was a good power function decline relationship between capillary diameter and imbibition recovery, with 30  $\mu\text{m}$  appearing to be a critical inflection point. The majority of the detained residual oil clusters not only interrupted the continuity of formation brine but also increased imbibition flow resistance. The imbibition velocity of core was rapid in the presence of buoyancy and gravity, whereas the imbibition recovery was slightly low. Imbibition recovery and core permeability also demonstrated a good power function decline relationship, indicating that the contribution of imbibition to enhanced oil recovery was limited in scope. It was suggested that increasing the fluctuation of the nonwetting phase on an irregular basis helped to improve the imbibition recovery in actual reservoirs.

#### Data Availability

The data used to support the findings of this study are included within the manuscript.

#### Conflicts of Interest

The authors declare that they have no conflicts of interest.

#### Acknowledgments

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