Effect of Water Distribution on Spontaneous Imbibition of Tight Rocks: A Fractal Approach

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Received: 12 October 2020; Accepted: 6 December 2020; Published: 9 December 2020

Abstract: The original water distribution characteristic plays an important role in the fracturing liquid retention in actual tight reservoirs. In this paper, an analytical model was proposed to characterize the water distribution and its effect on the spontaneous imbibition, based on the capillary tube model and fractal theory. Furthermore, the effect of the water film and the non-piston-like front related to the pore size are included in our model. The proposed model was successfully validated with the experimental results of core imbibition tests. Our work demonstrates that water distribution is influenced by displacement pressure and pore structure. For a small differential pressure, the porous media with richer large pores usually possesses a lower water saturation, and this difference will decrease with the increase of differential pressure. Moreover, compared with previous studies, the proposed imbibition model can not only distinguish the valid pores and invalid pores for imbibition but it can also predict the initial imbibition rate and equilibrium time of tight porous media with different water saturation. The results show that the equilibrium time is controlled by the minimum effective pore radius while the initial imbibition rate is mainly controlled by the large pores. Both of these two parameters will decrease with an increase of water saturation; the former is more sensitive to a low water saturation, while the latter decreases more quickly for a middle-high water saturation.

Keywords: fractal theory; spontaneous imbibition; water distribution; pore size; non-piston-like front

1. Introduction

Hydraulic fracturing is the key technology for achieving the production of tight gas reservoirs in a useful and economical way [1,2]. During the fracturing process, large volumes of water are pumped into the reservoirs while the flowback efficiency is generally lower than 30% [3]. Spontaneous imbibition induced by the capillary force is widely acknowledged to be responsible for this phenomenon [3,4]. Meanwhile, in the original reservoirs, water is considered ubiquitous because of the strong capillary effect of small pores and adsorbed effects of the rock surface [5,6]. Therefore, characterizing the spontaneous imbibition behaviors of tight rocks with further consideration of the water distribution characteristics in the original reservoirs is important for understanding the fate of the fracturing fluids in actual tight reservoirs.

Since Lucas (1918) and Washburn (1921) obtained the classical scaling imbibition law that the imbibition length \( l \) versus the time \( t \) meets the relation of \( l \sim t^{1/2} \) for the horizontal conditions [7,8], some other effects, such as the gravity effect, water saturation and dynamic contact angle, etc. are added into the model to analyze the spontaneous imbibition characteristic under more complicated conditions [9,10]. Handy (1960) proposed a model to characterize the imbibition process with...
consideration of the piston-like displacement front [11]. Li and Horne (2001) thought that the effect of gravity should depend on the ratio of the gravity to the capillary force, and an approach with further consideration of the gravity effect, as well as the initial water saturation, was developed [12]. These models worked well in conventional reservoirs because of the good pore connectivity of porous media and the slight difference of capillary force in different size of micropores [13,14]. However, it may face challenges in unconventional reservoirs because of the non-piston-like imbibition front induced by the poor pore connectivity and nanoscale pores, as was validated with visualized experimental results by neutron imaging and X-ray CT scanning [15,16].

The pore space is usually oversimplified with a series of capillary bundles. Due to the highly random nature of the pore space, the size distribution is extremely complex to describe. Fortunately, fractal geometry proposed by Mandelbrot et al. (1982) can be used to describe the pore system in rocks [17]. Since this theory was introduced, it has been widely used to characterize the pore network and to simulate the fluid flow in porous media. Based on fractal theory, Li et al. (2012) proposed a model to characterize the relationship between the spontaneous imbibition rate and time [18]. Cai et al. (2012) analyzed the spontaneous co-current imbibition process of wetting fluid into gas-saturated porous media [19]. Nevertheless, the piston-like imbibition front is still adopted in these studies. Recently, recognizing the limited pore connectivity of tight porous media, Shi et al. (2018) extended the previous fractal model with further consideration of the non-piston-like imbibition in the elongated pore, with the assumption that the spontaneous imbibition can be considered as a laminar flow in different size pores by following the Lucas–Washburn (L–W) equation [20]. Nevertheless, these works only focus on the dry samples rather than the unsaturated tight porous media. The water distribution characteristic related to the pore size may have significant influence on the imbibition process of unsaturated tight porous media, but the correlational researches are rarely reported in published works.

In this work, an analytical model has been proposed to characterize the water distribution and its effect on the spontaneous imbibition based on the capillary tube model and fractal theory. In this model, the effect of water distribution and a non-piston-like front induced by the different pore sizes are included. Drainage–imbibition experiments were conducted to validate our model. Furthermore, water distribution characteristics and their impact on the spontaneous imbibition characteristics (initial imbibition rate and equilibrium time) are discussed and analyzed based on our experimental samples. Finally, some conclusions are made. Additionally, some assumptions are listed as follows:

1. The pore space is assumed to be a series of capillary bundles with different dimensions, which obey the statistical fractal theory. The diameter along the flow path is constant.
2. The spontaneous imbibition is driven by the capillary force, and the effect of other mechanisms (water adsorption, osmotic effect and electric double layer, etc.) are ignored.
3. The variation of interfacial tension and the variation of wettability are ignored.

2. Mathematical Modeling

2.1. Fractal Scaling Laws of Porous Media

Fractal geometry, proposed by Mandelbrot et al. (1982), has been widely used to describe the pore structure of reservoir rocks [17]. Yu et al. (2001, 2002) extended the previous research works and obtained the fractal scaling law of pore distribution, that is [21,22]

\[ N(\geq r) = \left( \frac{r_{\text{max}}}{r} \right)^{D_f} \] (1)

where \( r \) is the pore radius, \( N \) is the number of pores with a radius larger than \( r \), \( r_{\text{max}} \) is the maximum pore radius, and \( D_f \) is the fractal dimension.

The number of pores with a radius in the range of \( r \) and \( r + dr \) can then be obtained as [19]

\[ -dN = D_f r_{\text{max}}^{D_f} r^{-{(D_f + 1)}} dr \] (2)
The fractal dimension $D_f$ can be determined by the porosity $\phi$ and the ratio $r_{\text{min}}/r_{\text{max}}$ in the porous media [21]

$$D_f = 2 - \frac{\ln \phi}{\ln \left( \frac{r_{\text{min}}}{r_{\text{max}}} \right)}$$  \hspace{1cm} (3)

Usually, the capillaries are tortuous, and the average tortuosity of the flow path can be obtained with the following widely used empirical correlation [19]

$$\tau = 1 + 0.41 \ln(1 / \phi)$$  \hspace{1cm} (4)

where $\tau$ is the average tortuosity, $\phi$ is the porosity, and the constant 0.41 is obtained by fitting experimental data for spherical particles.

2.2. Water Saturation in Fractal Porous Media

A bundle of cylindrical capillaries with different radii $r$ is used to represent the porous media. As illustrated in Figure 1, the capillaries with pore radii smaller than $r_c$ are still filled with water because of an insufficient driving force, while the pores with a radius larger than $r_c$ are covered with a water film because of the surface effects of the pore wall. Based on the physical argument, the water saturation can be calculated as

$$S_w = \frac{\int_{r_{\text{min}}}^{r_{\text{max}}} \pi r^2 (-dN) - \int_{r_{c}}^{r_{\text{min}}} \pi (r - h_n)^2 (-dN)}{\int_{r_{\text{min}}}^{r_{\text{max}}} \pi r^2 (-dN)}$$  \hspace{1cm} (5)

where $S_w$ is the water saturation, $h_n$ is the thickness of the water film, $r_{\text{max}}$ is the maximum radius, $r_c$ is the critical radius, and $r_{\text{min}}$ is the minimum radius.

Substituting Equation (2) into Equation (5), the water saturation can be expressed as

$$S_w = \frac{\int_{r_{\text{min}}}^{r_{\text{max}}} r^{1-D_f} dr - \int_{r_{c}}^{r_{\text{min}}} (r - h_n)^2 r^{-(D_f+1)} dr}{\int_{r_{\text{min}}}^{r_{\text{max}}} r^{1-D_f} dr} = 1 - \frac{\int_{r_{c}}^{r_{\text{max}}} \left[ (r - h_n)^2 r^{-(D_f+1)} \right] dr}{\int_{r_{\text{min}}}^{r_{\text{max}}} r^{1-D_f} dr}$$  \hspace{1cm} (6)

The key parameters for water saturation of tight porous media are the critical capillary radius $r_c$ and water film thickness $h_n$. According to the traditional Young–Laplace theory, the relationship
between critical cylindrical capillary radius and displacement pressure can be expressed in following form [22].

\[ r_c = 2\sigma \cos \theta / \Delta P \]  

where \( \theta \) is the wetting angle (°), \( \sigma \) is the interfacial tension of the water–gas system, and the value is 0.072 N/m at room temperature.

In the larger pores \( (r > r_c) \), the increase of pressure difference will decrease the thickness of the water film. When the two interfaces (solid–liquid interface and liquid–gas interface) approach each other, repulsive force emerges to separate the two interfaces. This force is termed by Derjaguin et al. (1987) as the disjoining pressure \( \Pi(h_{\delta}) \), which is a function of the water film thickness [23]. As illustrated in Figure 2, the mechanical equilibrium can be expressed as [24]

\[ \Delta P = P_g - P_w = \sigma / (r - h_{\delta}) + \Pi(h_{\delta}) \]  

where \( P_g \) is the gas pressure, \( P_w \) is the water pressure, and the difference is the displacement pressure. The term \( \sigma / (r - h_{\delta}) \) is the capillary force of the cylindrical surface.

![Figure 2. Illustration of the water film and the mechanical characteristics, on the condition that \( r > r_c \).](image)

Usually, the disjoining pressure is a sum of the molecular component, electrostatic component and structural component in DLVO theory (Derjaguin-Landau-Verwey-Overbeek) [25]. For quartz-rich tight rocks, the molecular component dominates the other components, and the expression is proportional to \( h_{\delta}^{-3} \) [26,27]. Gee et al. (1990) investigated the film thickness as a function of disjoining pressure with ellipsometry measurements (Figure 3) [28]; a widely used relationship between disjoining pressure (MPa) and water film thickness (μm) can be expressed as [29]

\[ \Pi(h_{\delta}) = \frac{6.5 \times 10^{-8}}{h_{\delta}^3} \]  

Combining Equations (8) and (9), in the nanopores with pore radius larger than \( r_c \), the water film thickness can be expressed as

\[ h_{\delta} = \left[ \frac{6.5 \times 10^{-8} \Delta P - \sigma}{r} \right]^{1/3} \]
2.3. Spontaneous Imbibition in Fractal Tight Porous Media

In this section, an analytical spontaneous imbibition model that accounts for the water distribution characteristics and non-piston-like front is developed. Based on the fractal scaling laws of porous media, the total pore area \( A_p \) in a unit cell can be expressed as

\[
A_p = \int_{r_{\text{min}}}^{r_{\text{max}}} 2\pi r^2 (-dN) = \frac{\pi D_f \rho_f^D_f}{2 - D_f} \left( r_{\text{max}}^{2-D_f} - r_{\text{min}}^{2-D_f} \right)
\]

(11)

Then, the number of pores with radius larger than \( r \) can be expressed as

\[
N_f(> r) = \frac{A_p \phi}{A_p} N(> r) = \frac{A_p \phi (2 - D_f)}{\pi \tau D_f \rho_f^D_f (r_{\text{max}}^{2-D_f} - r_{\text{min}}^{2-D_f})} \left( r_{\text{max}} / r \right)^{D_f}
\]

(12)

where \( A_i \) is the cross-section area of samples. The term \( \phi / \tau \) is the surface porosity, which is different than the traditional volume porosity because of the tortuous flow path. The details were described by Shi et al. (2018) [20]. The number of pores with a radius in the range of \( r \) and \( r + dr \) can be obtained as

\[
-dN_f = A_i \frac{\phi}{\tau} \frac{(2 - D_f)}{\pi \left( r_{\text{max}}^{2-D_f} - r_{\text{min}}^{2-D_f} \right)} r^{-(D_f+1)} dr
\]

(13)

The imbibition of a single pore follows the classical Lucas–Washburn law and can be expressed as

\[
\frac{dh_f}{dt} = r \sigma \cos \theta / \left( 4 \mu_w h_f \right)
\]

(14)

where \( \mu_w \) is the water viscosity, mpa·s, \( h_f \) is the tortuous height of the liquid front in the capillary, and \( r \) is the pore radius under dry conditions.

The relationship between the tortuous height \( h_f \) and the straight height \( h_s \) can be expressed as

\[
h_f = \tau h_s
\]

(15)
For unsaturated tight porous media, the water distribution plays an important role in the spontaneous imbibition. The pores with radius smaller than \( r_c \) are useless space for liquid flow because they are already filled with water, while for the pores with radius larger than \( r_c \), the effective radius for water flow is \( r - h_0 \). Combining Equations (14) and (15) and further considering the thickness of the adsorbed water film, the imbibed length in capillaries with radius larger than \( r_c \) can be expressed as

\[
h_{\text{im}} = \left[ \frac{(r - h_0) \sigma \cos \theta}{2 \tau \mu_w} \right] t^{1/2}
\]

(16)

Then, the volume of imbibed water in a single pore can be given as

\[
V = \pi (r - h_0)^2 \left[ \frac{(r - h_0) \sigma \cos \theta}{2 \mu_w} \right] t^{1/2}
\]

(17)

The volume of imbibed water for the pores with radius \( r \) can be calculated as

\[
V(r) = V \cdot dN = \frac{A_0 \varphi (2 - D_f)}{\tau (r_{max}^{2-2D_f} - r_{min}^{2-2D_f})} \left[ \frac{\sigma \cos \theta}{2 \mu_w} \right] t^{1/2} (r - h_0)^{2-2D_f} r^{-(D_f+1)} dr
\]

(18)

In our work, a bundle of cylindrical capillaries without any interconnection are used to represent the tight porous media. Before the water imbibition, the small pores are filled with water while the large pores are covered with a water film (Figure 4A). During the imbibition process, Equation (14) demonstrates that the larger the pore radius is, the faster will be the imbibition velocity. Therefore, the imbibition of unsaturated tight porous media can be divided into three stages. At the initial stage (Figure 4A,B), all pores are exposed to the liquid at the bottom, and the imbibition process in pores with \( r > r_c \) occurs simultaneously. Subsequently, since the imbibition rate is higher in larger pores, liquid will preferentially reach the top of large capillaries while the small pores can still imbibe the water (Figure 4C). At the last period, all the pores are filled with the imbibed water (Figure 4D), and the samples stop imbibing water.

Figure 4. Illustration of liquid imbibition into the bundle of capillaries with different diameters with further consideration of the water distribution: (A) original water distribution, (B) imbibition occurs in all effective pores \( (r > r_c) \), (C) part of the pores are filled with imbibed water, and (D) the equilibrium status.
At the initial stage, the pores with \( r > r_c \) can imbibe water, and the volume can be calculated as

\[
V_i = \int_{r_c}^{r_{\text{max}}} V(r) dr = \frac{A \phi(2-D_f)}{\tau(r_{\text{max}}^{2-D_f} - r_{\text{min}}^{2-D_f})} \left[ \frac{\sigma \cos \theta}{2\mu_w} \right]^{1/2} \int_{r_c}^{r_{\text{max}}} t^{1/2} (r - h_\delta)^{2.5} r^{-(D_f+1)} dr
\]  

(19)

This period ends when the largest pores are completely filled with water. Therefore, the limitation is

\[
t \leq \frac{2\tau^2 \mu_w h_\delta^2}{\sigma \cos \theta(r_{\text{max}} - h_\delta)}
\]  

(20)

Subsequently, the largest pores are filled with water while other pores continue imbibing water. The total volume of imbibed water can be given as

\[
V_f = \int_{r_c}^{r_{\text{max}}} V(r) dr + \int_{r_c}^{r_{\text{max}}} V_e(r) dr
\]  

(21)

where \( n \) is the critical radius of pores that are just filled with water at time \( t \), \( V_i(r) \) is the equilibrated volume for the pores for which water has reached the top.

The critical radius \( r_k \) can be calculated as

\[
r_k = \frac{2\tau^2 \mu_w h_\delta^2}{\sigma n \cos \theta} + h_\delta
\]  

(22)

The equilibrated volume \( V_e(r) \) is given as

\[
V_e(r) = \pi (r - h_\delta)^2 \tau H \cdot dN_f = \frac{HA \phi(2-D_f)}{(r_{\text{max}}^{2-D_f} - r_{\text{min}}^{2-D_f})} (r - h_\delta)^2 r^{-(D_f+1)} dr
\]  

(23)

Substituting Equation (18) and Equation (23) into Equation (21), the total volume of imbibed water at this stage can be expressed as

\[
V_f = \frac{A \phi(2-D_f)}{\tau(r_{\text{max}}^{2-D_f} - r_{\text{min}}^{2-D_f})} \left[ \frac{\sigma \cos \theta}{2\mu_w} \right]^{1/2} \int_{r_c}^{r_{\text{max}}} t^{1/2} (r - h_\delta)^{2.5} r^{-(D_f+1)} dr + H \int_{r_c}^{r_{\text{max}}} (r - h_\delta)^2 r^{-(D_f+1)} dr
\]  

(24)

At the last period, all pores are filled with water and the imbibition process has stopped. The imbibed water volume is

\[
V_f = \int_{r_c}^{r_{\text{max}}} V_f(r) dr = \frac{A \phi(2-D_f)}{(r_{\text{max}}^{2-D_f} - r_{\text{min}}^{2-D_f})} \left[ H \int_{r_c}^{r_{\text{max}}} (r - h_\delta)^2 r^{-(D_f+1)} dr \right]
\]  

(25)

The time \( t \) should satisfy the condition that

\[
t \geq \frac{2\tau^2 \mu_w h_\delta^2}{\sigma \cos \theta(r_{\text{min}} - h_\delta)}
\]  

(26)

Therefore, based on the capillary tube model and fractal scaling laws, an analytical model was proposed to characterize the effect of water distribution on spontaneous imbibition. In our model, two features are worth noting. Firstly, the water distribution characteristics are considered, and the
useless space and effective radius for water imbibition are distinguished. Secondly, unlike the piston-like imbibition front, the effect of pore size is demonstrated when we model the spontaneous imbibition process.

3. Experiments

3.1. Sample Description

In order to validate our model, two tight sandstones collected from the Yanchang field were used to conduct the experiments. The measured porosities of samples S1 and S2 were 9.32% and 9.5%, respectively, while the measured permeabilities of samples S1 and S2 were 0.093 and 0.059 mD, respectively. In order to obtain the pore information, high-pressure mercury intrusion capillary pressure (MICP) was conducted on an AutoPore IV mercury-injection analyzer 9320. The measured mercury intrusion capillary pressure curves and pore size distribution curves are shown in Figure 5. Usually, the maximum pore size is obtained with the minimum discharge pressure $P_{cmin}$, which can be obtained by extending the flat section to the Y-axis to get the intersection (Figure 5a). The results show that the $P_{cmins}$ of S1 and S2 are 1.7 MPa and 3.4 Mpa, respectively, and the corresponding maximum pore sizes are 0.433 and 0.216 um, respectively. Meanwhile, the maximum capillary pressure of the two samples is 136 Mpa, and the determined minimum pore size is 0.005 um. In addition, the water contact angles of the two samples are measured at room temperature and atmospheric pressure by the sessile drop method. As shown in Figure 6, the results show that the contact angles of samples S1 and S2 are 12.9° and 14.6°, respectively. Therefore, it is appropriate that the contact angle is assumed to be zero when we model the spontaneous imbibition. Based on the pore structure parameters, the fractal parameters are calculated based on Equations (3) and (4). The detailed rock information of our samples is shown in Table 1.

![Figure 5. Mercury injected pressure curve and pore size distribution of the samples.](image)

![Figure 6. The measured water contact angle of the two samples.](image)
Table 1. Properties of the samples used in the experiments.

| Sample | Length (cm) | Diameter (cm) | Porosity (%) | Permeability (mD) | $r_{\max}$ (μm) | $r_{\min}$ (μm) | $r_{av}$ (μm) | Df | τ |
|--------|-------------|---------------|--------------|-------------------|-----------------|-----------------|---------------|----|---|
| S1     | 2.33        | 2.52          | 9.32         | 0.093             | 0.433           | 0.005           | 0.1345        | 1.46| 1.97 |
| S2     | 2.31        | 2.52          | 9.50         | 0.059             | 0.216           | 0.005           | 0.0760        | 1.37| 1.96 |

3.2. Displacement Experiments

Figure 7 demonstrates the schematic of gas–water displacement experiments. The pressure was perceived by the precision pressure sensor, and the results were directly recorded on the computer. The metering component is made up of a precise electronic balance and a container with desiccant, and the water weight could be immediately measured. The primary experimental steps were as follows:

(1) Samples were cleaned using the solution immersion method. The solvent is a mixture of methylene chloride and ethyl alcohol (3:1) and we kept the core immersed in the solvent for seven days.

(2) Then, the clean samples were dried at a temperature of 105 °C. The weight of each sample ($W_{dry}$) was measured with a precise electronic balance. When there was no further mass change for two consecutive measurements, the dry samples were obtained.

(3) Samples are placed in a vacuum for 4 h and then saturated with distilled water.

(4) The sample was put into the core holder and distilled water was driven through the sample to ensure that the samples were fully saturated. Then, the weight of the saturated samples ($W_{sat}$) was measured. The difference between $W_{sat}$ and $W_{dry}$ was used to estimate the pore volume (PV), and the results showed that the PV of Sample S1 and S2 were 1.065 cm$^3$ and 1.070 cm$^3$.

(5) The water was displaced with gas under a small pressure difference. If there was no bubble, the pressure difference was enhanced. The pressure and the liquid production were recorded until the core reached the stable state. Then, the weight of the wet sample ($W_{wet}$) was measured.

(6) The water saturation of the sample was calculated by the following expression:

$$S_w = \frac{W_{wet} - W_{dry}}{W_{sat} - W_{dry}} \times 100$$  \hspace{1cm} (27)
holder, (7) precision pressure sensor, (8) gas–water separator, (9) precise electronic balance, (10) gas flowmeter, (11) confining pressure pump, (12) numerical recorder, and (13) the computer.

3.3. Spontaneous Imbibition Experiments

Because of the low permeability and porosity of tight rocks, the imbibition rate is very low and the accuracy of measurement should be strictly controlled. A picture of spontaneous imbibition experiments is shown in Figure 8. In our work, the imbibition fluid is distilled water, whose viscosity is 1 mPa·s at room temperature T = 298 K. The length of our sample is small to ensure that the influence of gravity can be neglected. The main procedure of the experiments is as follows:

1. Firstly, the spontaneous imbibition experiments of dry samples were conducted. Before the experiments were performed, the samples were dried at the temperature of 105 °C until no further mass change occurred.
2. The electronic balance was placed in a constant temperature chamber, in which the temperature was kept at 298 K;
3. The sample was suspended by nonelastic and impermeable string. Once the sample contacts the surface of the distilled water, the mass of imbibed water was recorded in time by the electronic balance (JJ632BC) with an accuracy of 0.0001 g and a range from 0 to 620 g.
4. The imbibition experiments were over if the mass of imbibed water remained constant within two hours.
5. After the experiments, the displacement process following the procedure in Section 3.2 is conducted to prepare the samples with different water saturation. Then, steps (2) through steps (5) were repeated.

![Figure 8. Picture of the co-current spontaneous imbibition experiments.](image)

4. Model Verification and Discussion

4.1. Model Verification

4.1.1. Validation of Fractal Water Saturation Model

The comparison of the fractal water saturation model and experimental results is shown in Figure 9. The figure shows that the simulated results have the same variation trend with the experimental data, proving that the water saturation calculated by the proposed fractal model with consideration of the water distribution is reliable. Meanwhile, the comparison of experimental results shows that the pore structure also influences the displacement process. The water saturation of Sample S1 is always smaller than that of Sample S2, and the difference decreases with an increase of displacement pressure. This is because of the fact that Sample S1 is relatively richer of large pores than that of Sample S2, and more wetting phase (e.g., water) can be displaced with a smaller
differential pressure. When the displacement pressure reaches 5 Mpa, the critical capillary radius decreases to 28 nm. Under this condition, most of the water-saturated pores in the two samples have been displaced by the nonwetting phase.

Meanwhile, the discrepancy between the fractal model and experimental results with the weighting method should be noted. The discrepancy at the beginning can be understood from the following point: the maximum pore radius for the two samples is 0.433 um (S1) and 0.216 um (S2), respectively, and the corresponding critical minimum capillary pressure is 1.7 and 3.4 MPa, respectively. When the displacement pressure is smaller than this threshold drainage pressure, all pores are still saturated with wetting fluid. Therefore, the calculated water saturation is still 100% at the early stage. However, in the experimental process, before the critical minimum capillary pressure, very little nonwetting fluid (e.g., mercury) has already entered the pit of the sample surface or the big pores (e.g., micro-fracture) [30]. In our work, the maximal mercury injection pressure is 136 MPa (the corresponding pore radius is 0.005 um), the remaining volume contributed by the pores with radius smaller than 5 nm cannot be measured with the mercury injection method. Therefore, the water saturation with the fractal model will be a little lower than that of the weighting method, especially in the case of a low water saturation.

4.1.2. Validation of Spontaneous Imbibition Model

Figure 10 demonstrates the comparison of the spontaneous imbibition process between experimental data and calculated results. As shown in the figure, the fractal model can almost simulate the imbibition curves of tight porous media with different water saturation, implying that the presented model is reliable for analyzing the imbibition characteristics of unsaturated tight porous media. The imbibition curves show that the amounts of imbibed water increase rapidly in the initial period of time and then increase more slowly and approach the equilibrium. Different from the traditional explanation at the porous media scale, our model gives the explanation of this phenomenon at the pore scale. At the initial stage, all gas-saturated pores \((r > r_c)\) can imbibe the water and the imbibition rate is the fastest. Subsequently, with the increase of time, the larger pores will be preferentially filled with water, resulting in the decrease of the imbibition rate. Finally, only the smaller pores can imbibe the water. The effect of water saturation on spontaneous imbibition can also be observed in Figure 8. As the water saturation increases, both the amount of imbibed water and the equilibrium time decreases.

In addition, Figure 10 also shows that there are deviations between the model prediction and the experimental results, especially for the dry condition. The discrepancy may be understood from
the following points: (1) Regarding the displacement process, some corners and disconnected pores have no contribution to the displacement flow. Oppositely, for the imbibition process of dry samples, the wetting phase may spontaneously occupy some of these volumes. As the water content increases, the influence of these volumes will decrease. (2) In our work, the contact angle of the whole rock ($\theta = 0^\circ$) is used to represent the wettability of all pores. However, the contact angle strongly depends on the properties of the pore wall. The variation of the contact angle in different pores influences the imbibition process, especially for the dry conditions. Moreover, for unsaturated samples, the pore surface is covered with a water film, which means that the third and necessary phase (air) for the contact angle is missing. At such a condition, although the explanation is not perfect, the value $\theta = 0^\circ$ was validated to be useful to model the liquid flow in published works [31,32].

**Figure 10.** Comparison between the experimental results and the fractal spontaneous imbibition model of unsaturated tight porous media. (a) Spontaneous imbibition of sample S1; (b) Spontaneous imbibition of sample S2.

4.2. Imbibition Characteristic of Unsaturated Tight Porous Media

4.2.1. Effect of Water Saturation on Equilibrium Time

Based on the proposed model and the experimental samples, we investigated the effect of water saturation on equilibrium time of spontaneous imbibition. As shown in Figure 11, as the initial water saturation increases, the imbibition process takes less time to approach equilibrium. In our work, the length of the sample is near 2.30 cm, and the minimum pore radius is 0.005 um. The calculated equilibrium time of dry samples is 3.7 h. When the water saturation increases to more than 20%, the equilibrium time drops to less than 0.6 h. For the capillary filling in a single pore, the imbibition is faster in a larger pore. Therefore, with further consideration of the effect of water distribution, the equilibrium time of porous media strongly depends on the minimum effective pore radius. The smaller the minimum effective pore radius is, the longer is the equilibrium time. In our work, the initial water saturation is established by the drainage process, and the large capillaries are easily occupied by the nonwetting phase, while small pores are still filled with water because of the larger barrier of displacement. Therefore, with the increase of water saturation, the smaller pores will be preferentially filled with water, and the equilibrium time is more sensitive in the case of a low water saturation. Meanwhile, at the same water saturation, the equilibrium time of Sample S1 is lower than that of Sample S2 because the latter possesses richer small pores.
4.2.2. Effect of Water Saturation on the Imbibition Rate at the Initial Stage

At the initial stage, the imbibed water volume can be expressed as $V_t = K t^{1/2}$ based on Equation (19), where parameter $K$ represents the influence of pore structure and water distribution on the initial imbibition rate. Based on our model and the experimental samples, the effect of water saturation on the imbibition rate at the initial stage is illustrated in Figure 12. The calculation results show that parameter $K$ gradually decreases with the increase of water saturation. The slopes of the curves are gentle under a low water saturation condition, where the water mainly occupies the smaller pores. With the increase of water saturation, relatively larger pores will be occupied by water, and the decrease rate of the curves will increase, which implies that the large pores have a more significant effect on the initial imbibition rate. When water saturation is over 80%, the imbibition rate can be ignored because most of the pores have lost their imbibition capacity.

In our work, an analytical model was proposed to characterize the water distribution and its effect on spontaneous imbibition based on the capillary tube model and fractal theory. Both of the
influence factors, i.e., water distribution and water saturation, and the imbibition characteristics, i.e., equilibrium time and the initial imbibition rate of the unsaturated porous media, can be analyzed with this approach, which could not be done with the previous models. This work provides a complementary viewpoint to understand the spontaneous imbibition characteristic of unsaturated tight porous media.

4.3. Limitations and Expectations

In this work, an analytical model was proposed to characterize the effect of water distribution on spontaneous imbibition based on the capillary tube model and fractal theory. Furthermore, the effects of a water film and a non-piston-like front induced by the pore size are also included. Although the model works well for our experiments, some special issues should be addressed and could be the purpose of future works:

(1) In our work, a series of circular pores without connection are assumed to represent the pore structure because of the poor pore connectivity of tight porous media. However, pore geometries vary along the flow direction, which may alter the observed classical L-W theory considerably [33]. These features may be further considered to improve the accuracy of our model.

(2) The pores in tight porous media are on the nanoscale, and the surface effects play an important role for the transport phenomenon [34]. Most remarkably, the surface force and fluid layering effects result in higher flow resistance compared with the bulk water [35], which implies that more time is needed to reach the imbibition equilibrium or that some smaller micropores cannot be invaded by the external fluid.

(3) The difference between the experimental conditions and the reservoir environment are worth noting. Our experiments were conducted in an atmospheric environment. The high temperature and pressure under geological conditions may result in a deviation if we evaluate the spontaneous imbibition characteristics in actual reservoirs, based on the traditional experiments and models. All imbibition parameters, such as interfacial tension, wettability and fluid properties are temperature- and pressure-dependent parameters [36,37]. Our future work could add these effects to capture the reservoir imbibition characteristics more accurately. Based on the issues mentioned above, combining the surface area of the fracture network predicted with micro-seismic detection, we can upscale the results of porous media to the applications on the field scale.

5. Conclusions

In this paper, the spontaneous imbibition of tight porous media with different water saturation is analyzed based on the capillary tube model and fractal theory. The effects of the water film and the non-piston-like front induced by the pore size are included in the model. Furthermore, displacement experiments and spontaneous experiments of two samples are conducted to validate our model. Both of the influence factors, i.e., water distribution and water saturation, and the imbibition characteristics, i.e., equilibrium time and initial imbibition rate, are analyzed and discussed. The following conclusions can be drawn:

(1) The proposed fractal model with consideration of the water distribution is valid for characterizing the water saturation of tight rocks. Both the displacement pressure and pore structure influence water saturation. For a small differential pressure, the porous media with a richer collection of large pores usually possesses a lower water saturation, and this difference will decrease with the increase of differential pressure. For our samples, when the displacement pressure reaches 5 Mpa, the water saturation is equal because most of the water-saturated pores have been displaced.

(2) The equilibrium time of unsaturated samples is controlled by the minimum effective pore radius. As the minimum effective pore radius decreases, the equilibrium time will increase. With the increase of water saturation, the smaller pores will be preferentially filled with water; the equilibrium time is more sensitive in the case of low water saturation.
The imbibition rate at the initial stage is mainly controlled by the large pores in tight porous media. A rich set of large pores results in a high initial imbibition rate. Due to the effect of water distribution characteristics, the initial imbibition rate will decrease with an increase of initial water saturation and will be more sensitive for a middle-high water saturation.

Author Contributions: Conceptualization, H.Y. and D.F.; methodology, Q.L.; software, D.F.; validation, H.Y.; formal analysis, X.L. All authors have read and agreed to the published version of the manuscript.

Funding: This research was funded by the National Natural Science Foundation Projects of China (50974128), Beijing Natural Science Foundation (2204093), the Science Foundation of China University of Petroleum, Beijing (No.2462018YJRC033) and financial support from the China Scholarship Council ((No. 201906440129).

Acknowledgments: We would like to thank Yanchang Oilfield Co., Ltd for providing experimental samples.

Conflicts of Interest: The authors declare no conflict of interest.

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