New Method for Quantitative Prediction of Liquid Flowback Based on the Capillary Bundle Model

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ABSTRACT: Liquid invasion damage plays a main role in the formation damage of low-permeability sandstone reservoirs. The long-term retention of working fluid in the formation will cause serious water blocking damage. In order to research the quantitative prediction of liquid flowback in low-permeability sandstone reservoirs, a new method for quantitative prediction of liquid flowback based on the capillary bundle model was proposed, which took into account the boundary layer effect. Besides, taking the low-permeability sandstone samples of Penglaizhen formation in the Sichuan basin as an example, the quantitative prediction of liquid flowback was carried out. Finally, a series of experiments including gas displacement experiment and nuclear magnetic resonance experiment were conducted to examine the validation of the new method for quantitative prediction of liquid flowback. The study indicates that the results of quantitative prediction were in good agreement with the experimental results. Liquid flowback is closely related to pore structure, boundary layer effect, and displacement pressure in low-permeability sandstone reservoirs. For the sandstone sample with a permeability of 5.5 mD, most of the liquid could be displaced out at 1 MPa, and liquid saturation in the sandstone sample tends to be stable at 32%; for the sandstone sample with a permeability of 0.0349 mD, liquid flowback process is more durable, thereby the cumulative mass of liquid flowback and liquid saturation change gradually, and liquid saturation tends to be stable at 55%. This study not only provides an insight into the microscope mechanisms of liquid flowback but also predicts the cumulative mass of liquid flowback and liquid saturation accurately.

1. INTRODUCTION

With the increase of global energy requirement, conventional oil and gas resources have been unable to meet peoples’ energy demand, and development focus has gradually turned to unconventional oil and gas resources. According to IEA data, the potential resources of tight sandstone gas reach 210 × 1012 m³, which plays an increasingly important role in the current energy supply.1,2 Tight sandstone gas exists in low-permeability tight sandstone reservoirs, which has the characteristics of low porosity and permeability, high capillary pressure, strong stress sensitivity, and ultralow water saturation. Moreover, the phenomenon of liquid invasion during drilling, completion, and stimulation operation is common in low-permeability tight sandstone reservoirs.3−6 The long-term retention of working fluid in the formation will not only lead to both technical and environmental issues but also reduce the productivity of gas wells seriously.7−9 The study of liquid flowback is of great significance for the efficient development of low-permeability sandstone reservoirs.

At present, the research on liquid flowback mainly includes a chemical method, physical method, and theoretical method. A chemical method is that altering the reservoir wettability and gas−liquid surface tension by injecting surfactants into the formation, aiming to reduce capillary pressure during liquid flowback.10−12 A physics method is that studying the factors such as displacement pressure, permeability, clay content, soak time, and new microcracks on liquid flowback rate by a series of experiments.13−16 The chemical method and physics method belong to qualitative research. A theoretical method is that establishing the mathematic model by taking into consideration many factors such as displacement pressure, interfacial tension, contact angle, and viscosity of liquid,17,18 which is very significant for quantitative prediction of liquid flowback in low-permeability sandstone reservoirs. For low-permeability sandstone reservoirs that are water-wet in nature, pore throat is so small that the boundary layer effect is very remarkable. The seepage no longer follows Darcy’s law.19
However, the impact of boundary layer on liquid flowback is not taken into account in the above study. In this work, a new method for quantitative prediction of liquid flowback based on the capillary bundle model was proposed, which took into account the effect of boundary layer on liquid flowback. Taking the low-permeability sandstone samples of Penglaizhen formation in the Sichuan basin as an example, the quantitative prediction of liquid flowback based on the capillary bundle model was carried out. Furthermore, gas displacement experiment and nuclear magnetic resonance (NMR) experiment were carried out. The experimental results are in good agreement with the predicted results, which well examine the validation of the new method for quantitative prediction of liquid flowback. This study not only provides an insight into microscopic mechanisms of liquid flowback but also predicts the cumulative mass of liquid flowback and liquid saturation accurately.

2. CAPILLARY BUNDLE MODEL

Porous media are widely used in many fields, such as resource development engineering, irrigation and water conservancy engineering, chemical filtration, biological organization, and so on. The complex porous media are simplified as a pipe network model, a spherical pore model, a parallel plate model, and a capillary bundle model. In most cases, a capillary bundle model is widely used in many porous media. Purcell\textsuperscript{20} first proposed the capillary bundle model and obtained the absolute permeability model of rock based on the Darcy formula and Poiseuille equation, which is of great significance for the calculation of rock porosity and permeability. Therefore, this model is called “classical capillary bundle model”. Later, Kozeny and Carmen\textsuperscript{21} pointed out that the actual flow distance in porous media is far greater than the length of rock; thus, tortuosity was introduced to modify the capillary bundle model. More recently, Ruth et al.\textsuperscript{22} derived the formation factor model based on the capillary bundle model. The calculation of porosity and permeability of rock is performed theoretically and the relationship between the physical properties is studied based on the capillary bundle model. However, it is rarely reported that some parameters of capillary bundle model including the capillary radius, distribution frequency, capillary volume, and the number of capillary tube were calculated. Additionally, the permeability contribution of capillary tube with a different radius is not accounted for.

In this paper, the pore of low-permeability sandstone is regarded as the capillary bundle model (Figure 1). To make some simplifications, we assume that each capillary tube is a smooth cylinder, and the length of each tube is equal, while the radius of each tube is different. The hypothesis for establishing the capillary bundle model is as follows:\textsuperscript{23} (1) The pore volume of sandstone sample is equal to the sum of capillary tube volume. (2) The distribution frequency of initial capillary radius is equal to that of throat radius. (3) The permeability contribution rate of initial capillary radius is equal to that of throat radius.

Some parameters are needed to describe this capillary bundle model. \( r_i \) represents the type of capillary tube (e.g., the first-type capillary tube means \( i = 1 \). Here, \( i = 1, 2, 3, \ldots, m \)). The tortuosity of the capillary bundle model is \( \tau \). The length of the sandstone sample is \( L \). A typical capillary tube has length \( L_i \), initial capillary radius \( R_{ini(i)} \), number of capillary tube \( N_{i(i)} \), distribution frequency \( f_{(i)} \) and capillary volume \( V_{(i)} \). What’s more, \( R_{ini(i)} > R_{ini(i+1)} \). The capillary bundle model was established based on the mercury injection data, and the basic parameters are calculated as follows:

According to the Laplace equation, throat radius can be obtained accurately.

\[
\eta_i = \frac{2\sigma \cos \theta}{P_{ini(i)}} \tag{1}
\]

The initial capillary radius was calculated as follows

\[
R_{ini(i)} = \frac{\eta_i + \eta_{(i+1)}}{2} \tag{2}
\]

The distribution frequency was calculated as follows

\[
f_{(i)} = S_{(i)} - S_{(i-1)} \tag{3}
\]

The capillary volume was calculated as follows

\[
V_{(i)} = f_{(i)} V_p \tag{4}
\]

The tortuosity of capillary bundle model was calculated as follows

\[
\tau = \sqrt{\frac{(\sigma \cos \theta)^2}{2K} \int_0^{\text{Hgmax}} \phi \frac{dS}{[P_{ini(i)}]^2}} \tag{5}
\]

The single capillary volume was calculated as follows

\[
v_{(i)} = \pi R_{ini(i)}^2 L_i \tag{6}
\]

The number of capillary tube was calculated as follows

\[
N_{(i)} = \frac{V_{(i)}}{v_{(i)}} \tag{7}
\]

3. QUANTITATIVE PREDICTION OF LIQUID FLOWBACK

For the hydrophilic capillary bundle model, the capillary pressure is the resistance during gas displacement. When displacement pressure is strong enough to overcome the capillary pressure, the liquid in the capillary tube will be displaced out. According to eq 1, capillary pressure varies with the initial capillary radius. As shown in Figure 2, the liquid in the large capillary tube is displaced out at low displacement pressure, while the liquid in the small capillary tube is displaced out at high displacement pressure. Because of high capillary pressure in the smallest capillary tube, displacement pressure is not strong enough to overcome the capillary pressure. Therefore, the liquid will not be displaced out and remains in the smallest capillary tube.\textsuperscript{24}

The hypothesis for the quantitative prediction of liquid flowback is as follows

(1) The porous media of the low-permeability sandstone sample are simplified as the capillary bundle model. (2) The capillary bundle model is saturated with liquid.
(3) The process of flowback is one-dimensional flow in the capillary tube.

(4) Liquid is viscous incompressible.

(5) Boundary layer effect should be taken into account.

(6) The loss mass of liquid due to evaporation is not considered.

The procedures for quantitative prediction of liquid flowback are as follows

(1) Calculate the boundary layer thickness. It can be seen from Figure 3 that the liquid in the capillary tube consists of a capillary liquid and a boundary layer. The capillary liquid is far away from the wall and could flow, while the boundary layer is close to the wall and could not flow. The adjacent liquid molecules during liquid flow and the boundary layer thickness will become thinner. At present, the boundary layer thickness can be obtained by two methods, one is displacement experiment of deionized water in the microtube and the other is dissipative particle dynamics (DPD) method. It is noteworthy that the minimum radius of the microtube used for displacement experiment is more than 1 μm, while the minimum radius of channel for simulation is 1 nm. Most of pore throat of low-permeability sandstone reservoirs are micro–nanopores. Therefore, compared to the displacement experiment of deionized water in the microtube, the boundary layer thickness model obtained by the DPD method is more suitable for the calculation of liquid flowback in this work. The boundary layer thickness model can be obtained as

$$\delta_{(i)} = 0.6931R_{\text{init}(i)}^{-0.2822} R_{\text{ef}(i)}^{-0.491} \left( \frac{\Delta P_{j}}{L_{j}} \right) \mu$$

(2) From the perspective of microscope seepage, the flowback process is two-phase flow of gas and liquid in capillary tube. There is a single-phase flow on both sides of the hemispherical meniscus. The direction of the capillary pressure is toward the nonwetting phase (Figure 4). The effective seepage radius with an initial capillary radius of $R_{\text{init}(i)}$ at $\Delta P_{j}$ was denoted as

$$R_{\text{ef}(i)} = R_{\text{init}(i)} - \delta_{(i)}$$

It can be seen from eq (9) that if the initial capillary radius is equal to or less than the boundary layer thickness, the capillary tube will become an invalid channel.

(3) According to the Laplace equation, the capillary pressure with an effective seepage radius of $R_{\text{ef}(i)}$ is

$$P_{c(i)} = \frac{2\sigma \cos \theta}{R_{\text{ef}(i)}}$$
(4) The capillary liquid is forced by displacement pressure and capillary pressure. When

$$P_{ci}(i) \leq \Delta P_j < P_{ci(i+1)}$$

(11)

where \(i = s, \ldots, m\), it indicates that the liquid in the capillary tube with the initial capillary radius greater than \(R_{ini(i)}\) will be displaced out, and the capillary liquid will be replaced by gas. However, the liquid in the capillary tube with the initial capillary radius less than \(R_{ini(i)}\) will not be displaced out, and this kind of capillary tube will be full of liquid. Namely, the minimum initial capillary radius of the gas flow channel is \(R_{ini(i)}\).

When

$$\Delta P_j \leq P_{ci(i)}$$

(12)

where \(i = 1, 2, \ldots, m\) it indicates that the liquid of the whole capillary bundle model will not be displaced out.

(5) Displacement pressure was set to \(\Delta P_{ji}\), repeat steps (1–4). The calculation continues until the displacement pressure increases to a maximum value.

The flow efficiency is defined as the ratio of the effective seepage radius to the initial capillary radius on the cross section of a single capillary tube, which was calculated as follows

$$\gamma(i) = \frac{R_{eff}(i)}{R_{ini(i)} - \delta(i)} = 1 - \beta(i)$$

(13)

where \(\beta(i) = \delta(i)/R_{ini(i)}\) which represents retention rate.

The capillary volume with the initial capillary radius of \(R_{ini(i)}\) was calculated as follows

$$V(i) = \pi (R_{ini(i)}^2 \Delta t N(i))$$

(14)

The boundary layer has a negative effect on liquid flowback, which is the liquid that does not flow at a certain displacement pressure. Therefore, subtracting the boundary layer volume from the capillary volume gives the liquid volume displaced. The liquid volume displaced in the capillary tube of type \(i\) at \(\Delta P_j\) was calculated as follows

$$V_{bf(i)} = \pi (R_{ini(i)}^2 \Delta t N(i))$$

(15)

By combining eq 14 with 15

$$\frac{V_{bf(i)}}{V(i)} = \frac{\pi (R_{ini(i)}^2 \Delta t N(i))}{\pi (R_{ini(i)}^2 \Delta t N(i))}$$

(16)

Then, eq 16 can be simplified as

$$V_{bf(i)} = (\gamma(i))^2 V(i)$$

(17)

The cumulative volume of liquid flowback at \(\Delta P_j\) turns out as follows

$$V_j = \sum_{i=1}^{s} V_{bf(i)} = \sum_{i=1}^{s} (\gamma(i))^2 V(i)$$

(18)

The cumulative mass of liquid flowback at \(\Delta P_j\) is

$$M_j = \sum_{i=1}^{s} (\gamma(i))^2 V(i) \rho_{\text{water}}$$

(19)

The liquid saturation at \(\Delta P_j\) was calculated as follows

$$S_{wi(j)} = \left(1 - \frac{V_i}{V_p}\right) \times 100\%$$

(20)

4. CALCULATION EXAMPLE

4.1. Calculation of Capillary Bundle Model. The capillary bundle model was established based on mercury injection data. The basic parameters of the capillary bundle model can be obtained by eqs 1–7. The tortuosities of the capillary bundle model corresponding to sandstone samples with permeabilities of 5.5 and 0.0349 mD are 3.8 and 5.52, respectively. The basic parameters of the capillary bundle model are shown in Tables 1 and 2. The distribution frequency of initial capillary radius and permeability contribution rate are shown in Figure 5.

| Table 1. Basic Parameters of Capillary Bundle Model of No s-7# |
|---------------------------------|
| no. | initial capillary radius (μm) | distribution frequency (%) | permeability contribution rate (%) | capillary volume (cm³) | number of capillary tube (cm³) |
| 1   | 7.993                        | 1.648                      | 14.0                               | 0.048                  | 1.873 × 10⁷                    |
| 2   | 6.545                        | 2.119                      | 19.7                               | 0.062                  | 3.215 × 10⁶                    |
| 3   | 5.665                        | 8.617                      | 9.6                                | 0.250                  | 1.990 × 10⁶                    |
| 4   | 4.591                        | 25.137                     | 32.036                             | 0.730                  | 1.512 × 10⁷                    |
| 5   | 2.845                        | 12.982                     | 20.78                              | 0.377                  | 3.125 × 10⁵                    |
| 6   | 1.422                        | 7.258                      | 3.346                              | 0.211                  | 7.233 × 10⁵                    |
| 7   | 0.699                        | 6.133                      | 0.234                              | 0.178                  | 4.853 × 10⁵                    |
| 8   | 0.248                        | 13                         | 0.145                              | 0.378                  | 1.037 × 10⁶                    |
| 9   | 0.078                        | 1.776                      | 0.079                              | 0.052                  | 8.277 × 10⁵                    |
| 10  | 0.032                        | 7.667                      | 0.05                               | 0.222                  | 1.015 × 10⁶                    |
| 11  | 0.019                        | 7.553                      | 0.02                               | 0.219                  | 2.074 × 10⁵                    |
| 12  | 0.013                        | 2.467                      | 0.007                              | 0.072                  | 1.196 × 10⁵                    |
| 13  | 0.010                        | 3.643                      | 0.002                              | 0.106                  | 2.754 × 10⁵                    |

| Table 2. Basic Parameters of Capillary Bundle Model of No 3# |
|---------------------------------|
| no. | initial capillary radius (μm) | distribution frequency (%) | permeability contribution rate (%) | capillary volume (cm³) | number of capillary tube (cm³) |
| 1   | 0.826                        | 1.467                      | 9.636                              | 0.047                  | 7.896 × 10⁸                    |
| 2   | 0.522                        | 7.388                      | 33.381                             | 0.236                  | 9.958 × 10⁸                    |
| 3   | 0.339                        | 21.338                     | 40.576                             | 0.680                  | 6.835 × 10⁸                    |
| 4   | 0.213                        | 15.836                     | 11.915                             | 0.505                  | 1.282 × 10⁸                    |
| 5   | 0.112                        | 9.214                      | 2.677                              | 0.294                  | 1.931 × 10⁸                    |
| 6   | 0.082                        | 9.452                      | 1.054                              | 0.301                  | 5.161 × 10⁷                    |
| 7   | 0.052                        | 11.338                     | 0.505                              | 0.361                  | 1.551 × 10⁷                    |
| 8   | 0.034                        | 9.452                      | 0.178                              | 0.301                  | 3.055 × 10⁷                    |
| 9   | 0.021                        | 9.103                      | 0.069                              | 0.290                  | 7.292 × 10⁶                    |
| 10  | 0.013                        | 1.197                      | 0.003                              | 0.038                  | 2.505 × 10⁶                    |
| 11  | 0.008                        | 4.215                      | 0.005                              | 0.134                  | 2.304 × 10⁷                    |

The distribution frequency of no s-7# (Table 1) with capillary radius more than 1 μm is 57.761%, and the cumulative permeability contribution is more than 99%. By comparison, the capillary tube distribution of no 3# (Table 2) is relatively scattered, and the permeability contribution rate in the small capillary tube is very small.

4.2. Quantitative Prediction of Liquid Flowback.

4.2.1. Retention Rate. The relationship between retention
The retention rate and displacement pressure is shown in Figure 6. The displacement pressure varied from 0.5 to 7 MPa, and the step was 0.1 MPa. With the increase of displacement pressure and initial capillary radius, the retention rate decreases. The maximum and minimum values of initial capillary radius of no s-7# are 7.993 and 0.010 μm, respectively. The retention rates are 5.36 and 51.00% at 0.5 MPa, respectively, which decrease to 1.77 and 16.88% at 7 MPa, respectively. The maximum and minimum values of initial capillary radius of no 3# are 0.826 and 0.008 μm, respectively. The retention rates are 47.37 and 59.67% at 0.5 MPa, respectively, which decrease to 15.68 and 19.75% at 7 MPa, respectively.

Figure 5. Distribution frequency of initial capillary radius with (a) no s-7# and (b) no 3#.

Figure 6. Relationship between retention rate and displacement pressure with (a) no s-7# and (b) no 3#.

Figure 7. Results of quantitative prediction of liquid flowback. (a) Cumulative mass of liquid flowback and (b) liquid saturation of capillary bundle model.
From the molecular point of view, the larger the displacement pressure, the more liquid molecules flow, the thinner the boundary layer thickness, and the smaller the retention rate. When the displacement pressure increases to a certain value, the boundary layer will tend to the solid, and the retention rate will tend to constant.\(^3\)

4.2.2. Cumulative Mass of Liquid Flowback and Liquid Saturation. The curves of cumulative mass of liquid flowback and liquid saturation are shown in Figure 7. It can be seen that with the increase of displacement pressure, the cumulative mass of liquid flowback increases, while the liquid saturation decreases. In addition, there is an obvious difference on the cumulative mass and liquid saturation between sandstone samples of higher permeability and that of lower permeability. For the sandstone sample with a permeability of 5.5 mD, the cumulative mass of liquid flowback and liquid saturation change quickly between 0.5 and 1 MPa. It indicates that most of the liquid could be displaced out at 1 MPa. When the displacement pressure increases from 1 to 7 MPa, the cumulative mass of liquid flowback and liquid saturation change slowly. Liquid saturation tends to be stable at 32%. For the sandstone sample with a permeability of 0.0349 mD, the cumulative mass of liquid flowback and liquid saturation change uniformly, and the process of liquid flowback needs more time to reach a stable state. Liquid saturation tends to be stable at 55%. The differences among liquid flowback curves are mainly influenced by pore structure, boundary layer effect, and capillary pressure.\(^4\)

The boundary layer effect has a great impact on the liquid flowback in a small capillary tube. As a result, it is hard to overcome the capillary pressure of liquid in a small capillary tube. Besides, the distribution frequency of a small capillary tube is relatively scattered, and the permeability contribution rate in a small capillary tube is very small. Therefore, it is relatively difficult to displace the liquid in a small capillary tube, and the cumulative mass of liquid flowback increases gradually.

Figure 7 shows the stepped curve because the capillary tube radius distribution is discontinuous. Additionally, the liquid in the capillary tube consists of a capillary liquid and a boundary layer, the former is much greater than the latter. When the displacement pressure overcomes the capillary pressure of liquid in the capillary tube, the capillary liquid in the capillary tube will be displaced out immediately and a new gas flow channel will be produced. Thus, the cumulative mass of liquid flowback and liquid saturation change suddenly.

4.2.3. Minimum Initial Capillary Radius of a Gas Flow Channel. The minimum initial capillary radius of a gas flow channel at different displacement pressures is shown in Figure 8. For the sandstone sample no s-7#, the minimum initial capillary radius of the gas flow channel is 0.248 \(\mu\)m at 1 MPa. In other words, a gas flow channel with an initial capillary radius of 0.248 \(\mu\)m or above (7.993, 6.545, 5.545, 4.591, 2.845, 1.422, 0.699, and 0.248 \(\mu\)m) has been produced. The minimum initial capillary radius of the gas flow channel is 0.032 \(\mu\)m at 7 MPa. That is to say, a new gas flow channel with initial capillary radii of 0.078 and 0.032 \(\mu\)m has been produced. The change trends of no 3# are similar to that of the sandstone sample of s-7#. The minimum initial capillary radius of the gas channel is 0.034 \(\mu\)m at 7 MPa, namely, a gas flow channel with an initial capillary radius of 0.034 \(\mu\)m or above (0.826, 0.522, 0.339, 0.213, 0.132, 0.082, 0.052, and 0.034 \(\mu\)m) has been produced gradually.

5. EXPERIMENTAL METHOD AND PROCEDURES

In order to verify the validation of a new method for quantitative prediction of liquid flowback, a series of experiments including gas displacement experiment and NMR experiment were carried out on a low-permeability sandstone sample.

5.1. Samples. Low-permeability sandstone samples were taken from the Penglaizhen formation in the Sichuan basin. The sandstone samples were dried in an air oven at 70 ± 3 °C for at least 48 h until the mass of sandstone samples remains unchanged. The basic parameters of sandstone samples are listed in Table 3. Porosity was measured by Boyle’s law. The permeability was measured by a high-temperature and high-pressure measurement system (SCMS-E type). The confining pressure was 5 MPa.

5.2. Experimental Procedures. In this paper, the NMR apparatus (type of MacroMR23-060V-VT) was produced by Suzhou Niumai Corporation. The waiting time was 3000 ms, the number of echo was 15,000, the temperature was 32 °C, and the echo spacing was 0.15 ms.

The displacement experiment was conducted to simulate the low-permeability sandstone reservoir condition during flowback. Analysis balance (precision 0.0001 g) was used in this process. The experimental steps are as follows

1. The sandstone sample was dried in an air oven at 70 ± 3 °C for at least 48 h until the weight of the sandstone sample was constant, whose weight was recorded as \(m_1\).

2. Measure the physical parameters of the sandstone sample, including the length, diameter, porosity, and gas permeability.

3. The sandstone sample was saturated completely and then taken out to weigh, whose weight was recorded as \(m_2\). In order to avoid the effects of clay swelling on the results of liquid flowback, 2.00 wt % KCl brine is used as a saturated liquid. The mechanism of 2.00 wt % KCl brine inhibiting clay swelling is that the lower the hydration energy of ion

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Table 3. Basic Parameters of Cylindrical Sandstone Samples

| sample no. | length (mm) | diameter (mm) | dry weight (g) | porosity (%) | gas permeability (mD) | pore volume (cm³) |
|------------|-------------|---------------|----------------|--------------|----------------------|-----------------|
| s-7#       | 50.00       | 25.48         | 57.71          | 11.39        | 5.5                  | 2.905           |
| 3#         | 50.03       | 24.8          | 54.65          | 13.18        | 0.0349               | 3.186           |

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Figure 8. Minimum initial capillary radius of the gas flow channel.
is, the easier it is to be adsorbed. Compared with Ca\(^{2+}\) and Na\(^{+}\), K\(^{+}\) has lower hydration energy, which are embedded in the crystal layer after being adsorbed by montmorillonite and may discharge the water molecules between crystal layers. Furthermore, K\(^{+}\) has a function of inhibiting the expansion or separation between adjacent layers.\(^{34,35}\) The basic parameters of KCl brine are as follows: the density is 1.046 g/cm\(^3\), the viscosity is 1.12 mPa-s, the interfacial tension is 0.079 N/m, and the contact angle is 37.5°.

(4) Install the device for gas displacement experiment and put the sandstone sample into the core holder and then carry out the displacement experiment. The displacement medium for liquid flowback is nitrogen. The initial confining pressure and the initial displacement pressure were set as 2.5 and 0.5 MPa, respectively.

(5) When the displacement process reached a stable state for at least 1 h (when no liquid flowed out from the outlet and the value of the outlet gas flowmeter remained unchanged, the displacement process tends to be stable), take out the sandstone sample and weigh, whose weight was recorded as \(m_1\), and calculate the cumulative mass of liquid flowback and saturation by the formula of \((m_2 - m_1)/\rho_\text{fl}\) and \((m_2 - m_1)/\rho_\text{fl} \times V_\text{fl}\), respectively, where \(\rho_\text{fl}\) is the density of 2.00 wt % KCl solution. NMR testing was conducted.

(6) Change the confining pressure and displacement pressure and then repeat steps (4–5).

It is noteworthy that some errors might exist during the experiment. In order to ensure the accuracy of liquid saturation as much as possible, some points should be paid attention during the experiment: first, in order to reduce the error caused by weighing, the weight is measured three times with a balance and then the average value is used for calculation (including \(m_1\), \(m_2\), and \(m_3\)). Besides, evaporation of liquid might occur. In order to reduce the evaporation of liquid, the exposure time of sandstone sample in air should be as short as possible. What reduces the surface wear, sandstone sample should be handled with care during the experiment.

Figure 9 shows a schematic diagram of the displacement setup.

**Figure 9.** Schematic diagram of the displacement setup.

### 6. RESULTS AND DISCUSSION

**6.1. Analysis of Cumulative Mass of Liquid Flowback and Liquid Saturation.** By comparing the experimental results with the predicted results (Figure 10), it is found that the results of gas displacement experiment are in good agreement with the results of quantitative prediction, which examine the validation of the new method for quantitative prediction of liquid flowback. For the sandstone sample no s-7#, the cumulative mass of liquid flowback is 1.6161 g at 1 MPa, which accounts for 78.54% of the total cumulative mass of liquid flowback, and liquid saturation is 46.81%. The cumulative mass of liquid flowback increases to 2.0577 g at 7 MPa, and liquid saturation decreases to 32.28%.

For the sandstone sample no 3#, with the increase of displacement pressure, the cumulative mass of liquid flowback and liquid saturation change slowly, and the cumulative mass of liquid flowback and liquid saturation are 1.4852 g and 55.43% at 7 MPa, respectively.

**6.2. Distribution Characteristics of Liquid in the Porous Media.** NMR is a nondestructive technology and has been widely used in the exploration of oil/gas field. Some information such as total porosity, pore size distribution, and liquid content in different pores can be acquired by the NMR technique.\(^{36}\) The pore size is inversely proportional to the NMR \(T_2\) value, the liquid in macropore has a longer NMR \(T_2\) value, while the liquid in micropore has a shorter NMR \(T_2\) value. Additionally, the NMR \(T_2\) spectral area can reflect the liquid content in a certain range of pore.

The NMR \(T_2\) spectrum of no s-7# at a saturated condition has double continuous peaks (right peak area > left peak area). The NMR \(T_2\) spectrum peaks are distributed in 0–7.56 and 7.56–657 ms corresponding to micropores and macropores, respectively (Figure 11a). A comparison of \(T_2\) spectrum curves of no s-7# at a saturated condition, 0.5 and 1 MPa, indicates that the peak area decreases remarkably in the \(T_2\) time range of 7.56–657 ms (Figure 11b). Because of the weak effect of the boundary layer and low capillary pressure, the liquid in macropores was displaced out quickly at low displacement pressure compared with the \(T_2\) spectrum curves of no s-7# at 1, 3, and 5 MPa, which show that the peak area decreases obviously in the \(T_2\) time range of 0–7.56 ms (Figure 11c). When the displacement pressure overcomes the capillary pressure of liquid, the liquid in the micropores will be displaced out and a new gas flow channel will be produced. A comparison of \(T_2\) spectrum curve of no s-7# at 5 and 7 MPa indicates that two \(T_2\) spectrum curves nearly coincide (Figure 11d). It is because that when the displacement pressure is less than the capillary pressure, the liquid will remain in the micropores.

The NMR \(T_2\) spectrum of no 3# at a saturated condition has three continuous peaks (left peak area > middle peak area > right peak area). The NMR \(T_2\) spectrum peaks are distributed in 0–2.98, 2.98–40.37, and 40.37–312 ms corresponding to micropores, mesopores, and macropores, respectively (Figure 12a). A comparison of \(T_2\) spectrum curves of no 3# at a saturation condition, 0.5 and 1 MPa, indicates that the peak area decreases uniformly in the \(T_2\) time range of 2.98–40.37 and 40.37–312 ms (Figure 12b). It is expected because that sandstone sample no 3# has lots of micro–nanopores and high capillary pressure. With the increase of displacement pressure, the liquid in macropores and mesopores will be displaced out, compared with the \(T_2\) spectrum curve of no 3# at 1, 3, and 5 MPa, which shows that the peak area decreases obviously in the \(T_2\) time range of 0–2.98 and 2.98–40.37 ms (Figure 12c). When
the displacement pressure overcomes the capillary pressure of liquid in the micropores, the capillary liquid in the micropores will be displaced out gradually. At the same time, the boundary layer effect is remarkable because of the low permeability. Part of liquid molecules in the boundary layer of mesopores could also flow with the increase of displacement pressure. It is found that the $T_2$ spectrum curve of no 3# at 5 MPa is similar to that of no 3# at 7 MPa (Figure 12d). It is because that when the displacement pressure is not strong enough to overcome the capillary pressure. The liquid will not be displaced out and remain in the micropores. In a word, the liquid in the macropores is preferentially displaced out, followed by mesopores and micropores; this change trend is similar to that of the liquid in the capillary tube.

7. CONCLUSIONS

This work proposed a new method for quantitative prediction of liquid flowback based on the capillary bundle model, which took
into account the effect of boundary layer on liquid flowback. Taking the low permeability sandstone samples of Penglaizhen formation in the Sichuan basin as an example, the quantitative prediction of liquid flowback based on the capillary bundle model was carried out. Furthermore, a series of experiments were conducted to compare with the predicted results. The conclusion below can be drawn in this work:

(1) The boundary layer thickness is related to the initial capillary radius, displacement pressure, and liquid properties. With the increase of displacement pressure and initial capillary radius, retention rate decreases. When displacement pressure increases to a certain value, the boundary layer will tend to solid, and the retention rate will tend to constant. The smaller the initial capillary radius is, the more remarkable the boundary layer effect is and the larger the retention rate is.

(2) A comparison of the experimental results with the predicted results reveals that both of them are in good agreement, which examines the validation of the new method for quantitative prediction of liquid flowback. It is proved that this method can predict the cumulative mass of liquid flowback and liquid saturation accurately.

(3) The liquid flowback is closely related to pore structure, boundary layer effect, and displacement pressure. With the increase of displacement pressure, the cumulative mass of liquid flowback increases, while the liquid saturation decreases. For the sandstone sample with a permeability of 5.5 mD, most of the liquid could be displaced out at 1 MPa, and liquid saturation tends to be stable at 32%; for the sandstone sample with a permeability of 0.0349 mD, the liquid flowback process is more durable because of lots of micro-nanopores and high capillary pressure, thereby the cumulative mass of liquid flowback and liquid saturation change uniformly, and liquid saturation tends to be stable at 55%. The liquid in the macropores is preferentially displaced out, followed by mesopores and micropores.

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