Effect of Shale Reservoir Characteristics on Shale Oil Movability in the Lower Third Member of the Shahejie Formation, Zhanhua Sag

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Abstract: To reveal the effect of shale reservoir characteristics on the movability of shale oil and its action mechanism in the lower third member of the Shahejie formation (Es3l), samples with different features were selected and analyzed using N\textsubscript{2} adsorption, high-pressure mercury injection capillary pressure (MICP), nuclear magnetic resonance (NMR), high-speed centrifugation, and displacement image techniques. The results showed that shale pore structure characteristics control shale oil movability directly. Movable oil saturation has a positive relationship with pore volume for radius > 2 μm, as larger pores often have higher movable oil saturation, indicating that movable oil is present in relatively larger pores. The main reasons for this are as follows. The relatively smaller pores often have oil-wetting properties because of organic matter, which has an unfavorable effect on the flow of oil, while the relatively larger pores are often wetted by water, which is helpful to shale oil movability. The rich surface provided by the relatively smaller pores is beneficial to the adsorption of immovable oil. Meanwhile, the relatively larger pores create significant pore volume for movable oil. Moreover, the larger pores often have good pore connectivity. Pores and fractures are interconnected to form a complex fracture network, which provides a good permeability channel for shale oil flow. The smaller pores are mostly distributed separately; thus, they are not conducive to the flow of shale oil. The mineral composition and fabric macroscopically affect the movability of shale oil. Calcite plays an active role in shale oil movability by increasing the brittleness of shale and is more likely to form micro-cracks under the same stress background. Clay does not utilize shale oil flow because of its large specific surface area and its block effect. The bedding structure increases the large-scale storage space and improves the connectivity of pores at different scales, which is conducive to the movability of shale oil.

Key words: shale oil; movability; shale reservoirs; pore structure characteristics; lower third member of the Shahejie formation (Es3l)

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1 Introduction

As an important unconventional oil and gas resource, shale oil is widely distributed in the continental basins of China (Jia et al., 2012; Zou et al., 2015; Ning, 2015a), Particularly, in the Paleogene shale of eastern China, oil and gas shows are common (Ning et al., 2015b), and even industrial oil and gas flows are obtained (Li et al., 2016; Lu et al., 2016). However, there is an urgent problem to be solved in actual shale oil exploration, that is, the oil-bearing shale interval does not produce high-yield industrial oil flow, because the shale oil is present in the micro-nano pores of the shale reservoir in the adsorbed state and free state (Su et al., 2018). The pore structure and mineral fabric characteristics of the shale reservoir determine the enrichment (Jiang et al., 2017) and movability of shale oil and then determine the productivity of shale oil. Shale oil reservoirs in the United States are mainly developed in marine strata with wide distribution and good homogeneity. Shale oil is characterized by high maturity, low wax content, and low viscosity. It is light oil and easy to flow (Miller et al., 2008). However, shale oil in China is mostly continental facies, with strong reservoir heterogeneity, low maturity, high density, and high viscosity, which makes it difficult to flow (Zhang et al., 2014a; Nie et al., 2016). Therefore, it is particularly important for the exploration of continental shale oil to study the influence of reservoir characteristics on shale oil flow and to reveal the responsible mechanism.

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In recent years, different scholars have conducted much research on the characteristics and movability evaluation of continental shale oil reservoirs. In reservoir research, based on the division of shale facies, Wang et al. (2016) elaborated the important role of lamellar shale in shale oil enrichment. Ning et al. (2017a) used micro-nano pore characterization technology to reveal the pore structure characteristics of shale and the controlling factors of pore size at different scales in the Es3l in the Zhanhua Sag. Based on high-pressure mercury injection data, Lu et al. (2018) classified the microscopic pore throats of the shale oil reservoir as microporous throat, small-pore throat, medium-pore throat, and large-pore throat, and then evaluated them accordingly. From the aspect of formation energy, Zhang et al. (2014b) combined the physical and mechanical properties of shale and oil-bearing properties to retrieve a movable oil rate change profile in the Dongying depression. Li et al. (2016) measured the adsorption capacity of different minerals onto shale oil and found that clay has the strongest adsorption capacity, which is not conducive to shale oil flow. Wang et al. (2018) used simulation experiments to find that the asphaltene of shale oil began to degrade for VRo > 1.35% in the Chang 7 group of Ordos, which improved the fluidity of shale oil. The abovementioned studies expound the shale oil reservoir and movability characteristics from different angles, which is of great significance for guiding shale oil exploration. However, it is not possible to directly combine reservoir characteristics with movability or to discuss the influence of shale reservoir characteristics on shale oil movability from dynamic mechanisms.

For this reason, this study uses N2 adsorption, high pressure mercury injection, nuclear magnetic resonance (including centrifugation), high pressure displacement imaging, and other experimental methods to investigate the shale in the Es3l Zhanhua depression. This study preliminarily reveals the influence of pore structure and fabric characteristics of shale on shale oil movability and the responsible mechanism. Furthermore, it provides a reference for the selection of favorable continental shale oil strata and the optimization of desert.

2 Geological Settings

Samples were collected from well Luo69, an important systematic coring well in the Zhanhua Depression in the lower section of Sha 3 and upper section of Sha 4. The Zhanhua Sag, which is located northeast of the Jiyang Depression in the Bohai Bay Basin (Fig. 1), is a Meso-Cenozoic rift basin that developed from the paleotopographical background of Paleozoic basement rocks. The Zhanhua Sag covers an area of 2800 km² and is bounded by the Keding uplift in the east, the Chenjiazhuan uplift in the south, the Yihezhuang uplift in the west, and the Chengdong and Chengbei uplifts in the north. The Zhanhua Sag is composed of several subsags, such as the Gubei-Zhuangxi subsag, Gunan-Fulin subsag, and Bonan-Sikou subsag. In addition, uplifts such as the Gudao uplift are present (Lin et al., 2003; Liu et al., 2012; Song et al., 2014). Generally, the basin has fault-like characteristics in the north side, while the south side has characteristics of an overlapped, east-west double fault, strong division, and concave-convex alternation (Wang et al., 2005; Wang et al., 2016). The sag contains Paleogene-Neogene sediments. Furthermore, it contains portions of the Kongdian Formation, Shaejie Formation, Dongying Formation, Guantao Formation, and Minghuazhen Formation from the lower to the upper regions.

Fig. 1. The location and the second structural distribution of Zhanhua Sag.
At the Es3l deposit, accompanied by the aggravation of crustal tension, the Zhanhua Sag experienced intense activity along the main fault, and this accelerated deposition and the deepening of the water; the depositional environment was a semi-deep lake. Meanwhile, the climate became warm and humid with thriving plants and aquatic organisms, and weakly alkaline to alkaline water was present. The environment was reductive to strongly reductive. Therefore, broad dark mudstone and shale with rich organic matter (OM) were produced and preserved at the time of the Es3l deposition in the Zhanhua Sag. This material has been widely accepted as a high-quality source rock (Li et al., 2017a). Research has revealed that clay, quartz, and calcite are the dominant minerals, while clastic limestone and lime mudstone are the main rock types in Es3l in the Zhanhua Sag (Li et al., 2015; Su et al., 2019). Horizontally bedded structures were identified through the changes in the mineral composition, layer thickness, and color, and the material was divided into that with a massive structure (no bedded structure), a layered structure (thickness of a single layer more than 1 mm), and a lamellar structure (thickness of a single layer less than 1 mm) (Liu et al., 2012; Li et al., 2015). Consequently, it represents an important shale oil exploration interval in China (Ma et al., 2012; Zhang et al., 2014b).

3 Samples and experimental methods

3.1 Samples characteristics

Pore structure characteristics determine reservoir quality, and pore structure in shale is controlled directly by the mineral and OM composition. Meanwhile, the combinations of key components are also important. To investigate the influence of pore structure and fabric characteristics of shale on shale oil movability and the responsible mechanism

11 samples with different mineral compositions were selected for detailed analyses. Among them, there were three samples with a massive structure, three samples with a layered structure, three samples with a lamellar structure, and two samples with a lamellar-layered structure. These 11 samples were numbered L–1–L–11 from the deeper to upper regions, and the mineral composition and general geochemical characteristics of these samples are presented in Table 1.

Samples were high in calcite, clay minerals, and quartz. The average calcite content was 56% (range of 37% to 74%), the average clay mineral content was 17% (range of 9% to 28%), and the average quartz content was 17% (range of 13% to 23%). Furthermore, there were low levels of pyrite and dolomite in the samples, and feldspar was relatively rare. Samples had a high abundance of OM, with an average total organic carbon (TOC) content of 3.20% (range of 1.40% to 5.63%), and the main type of OM belonged to type I and III (Su et al., 2017), for which the vitrinite reflectance (Ro) average was 0.80% (range of 0.74% to 0.87%). This demonstrates that the OM has significant hydrocarbon-generating potential. Hence, this site is believed to be a good potential target for shale oil in China.

<table>
<thead>
<tr>
<th>Sample ID</th>
<th>Depth (m)</th>
<th>Bedding development</th>
<th>Clay (wt. %)</th>
<th>Quartz (wt. %)</th>
<th>Feldspar (wt. %)</th>
<th>Pyrite (wt. %)</th>
<th>Calcite (wt. %)</th>
<th>Dolomite (wt. %)</th>
<th>TOC (wt. %)</th>
<th>Ro (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>L–1</td>
<td>2947.19</td>
<td>Massive</td>
<td>23</td>
<td>16</td>
<td>2</td>
<td>2</td>
<td>51</td>
<td>5</td>
<td>2.44</td>
<td>0.83</td>
</tr>
<tr>
<td>L–2</td>
<td>2962.07</td>
<td>Massive</td>
<td>25</td>
<td>18</td>
<td>1</td>
<td>2</td>
<td>50</td>
<td>3</td>
<td>2.58</td>
<td>0.74</td>
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<tr>
<td>L–3</td>
<td>3018.55</td>
<td>Layered</td>
<td>17</td>
<td>19</td>
<td>1</td>
<td>4</td>
<td>53</td>
<td>5</td>
<td>4.12</td>
<td>0.82</td>
</tr>
<tr>
<td>L–4</td>
<td>3025.15</td>
<td>Layered</td>
<td>23</td>
<td>19</td>
<td>2</td>
<td>5</td>
<td>46</td>
<td>5</td>
<td>3.70</td>
<td>0.79</td>
</tr>
<tr>
<td>L–5</td>
<td>3030.15</td>
<td>Massive</td>
<td>28</td>
<td>23</td>
<td>2</td>
<td>5</td>
<td>37</td>
<td>5</td>
<td>3.45</td>
<td>0.77</td>
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<tr>
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<td>3045.10</td>
<td>Layered</td>
<td>11</td>
<td>15</td>
<td>1</td>
<td>3</td>
<td>66</td>
<td>4</td>
<td>2.83</td>
<td>0.77</td>
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<td>17</td>
<td>2</td>
<td>4</td>
<td>57</td>
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<td>3.05</td>
<td>0.81</td>
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<td>3055.10</td>
<td>Lamellar</td>
<td>15</td>
<td>17</td>
<td>1</td>
<td>4</td>
<td>55</td>
<td>8</td>
<td>5.63</td>
<td>0.79</td>
</tr>
<tr>
<td>L–9</td>
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<td>Lamellar</td>
<td>10</td>
<td>17</td>
<td>1</td>
<td>3</td>
<td>64</td>
<td>5</td>
<td>4.55</td>
<td>0.79</td>
</tr>
<tr>
<td>L–10</td>
<td>3109.90</td>
<td>Lamellar –layered</td>
<td>10</td>
<td>15</td>
<td>1</td>
<td>2</td>
<td>68</td>
<td>4</td>
<td>1.40</td>
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<td>L–11</td>
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<td>Lamellar –layered</td>
<td>9</td>
<td>13</td>
<td>0</td>
<td>1</td>
<td>74</td>
<td>3</td>
<td>1.42</td>
<td>0.87</td>
</tr>
</tbody>
</table>

3.2 Experimental methods

The experimental methods adopted can be categorized into two types (Chen et al., 2017). The first type classifies methods that obtain the pore structure parameters of shale, including high pressure mercury injection and low temperature nitrogen adsorption. The other type consists of methods that obtain the distribution characteristics of movable oil in shale, which include nuclear magnetic resonance (NMR) and its related centrifugation and displacement techniques.

3.2.1 High pressure mercury injection capillary pressure technique

The mercury injection capillary pressure (MICP) technique is the most common and effective method used in reservoir evaluation. The relationship between the pressure, which forces the mercury into pores, and the radius of pores can be described by the Washburn equation (Washburn, 1921). Thus, we can precisely obtain the pore...
size distribution (PSD) of samples from the injection volume under rising pressure (Cui et al., 2012). In this study, this method was implemented using an AutoPoreIV9500 capillary pressure curve determinative instrument, produced by MICROMERITICSTM. The testing range was 3 nm–1000 μm, and the precision was less than ±0.0001 mL. Samples that were tested by NMR were evaluated by the AutoPoreIV9500, in accordance with Chinese national standard GBT21650.1-2008, after being dried at 60 °C for 48 h.

3.2.2 Nitrogen adsorption

Low temperature, low pressure nitrogen adsorption is a classical method of obtaining the pore structure information of a sample, according to the theoretical model based on the adsorption of nitrogen on the surface of material pores under different partial pressures. The instrument used in this study is the Quadrasorb-SI type instrument manufactured by Quanta Company of the United States. Take 1–2 g sample and smash it to 60–80 mesh. A vacuum was applied at 110 °C for 14 h, and then a nitrogen isothermal adsorption-desorption experiment was performed under a liquid nitrogen atmosphere (77.4 K). A multi-point BET model was used to calculate the specific surface area of the shale.

3.2.3 Nuclear Magnetic Resonance

The low-field NMR T2 spectrum measured by rock-saturated single-phase fluids can reflect the inside pore structure of the rock (Brai et al., 2007; Mehana and El–Monier, 2016). Nuclear magnetic resonance combined with high-speed centrifugation is currently the main method of studying the movability of tight reservoir fluids (Zhou et al., 2016; Li et al., 2017b). The displacement experiment is also an effective means of reproducing the flow of fluid in the reservoir. In this study, the main tasks of saturated water NMR, NMR + centrifugal, NMR + displacement are completed based on nuclear magnetic resonance at Suzhou Newmial Nuclear Magnetic Testing Co., Ltd.

(1) NMR after water saturation: To obtain the surface T2 spectrum to characterize the pore structure, samples L-1–L-11 were subjected to NMR experiments of saturated water. The main steps are: first wash the oil and dry the sample, then test the rock sample volume by the drainage method. The sample was then saturated in simulated formation water with a salinity of 10 g/L (pressure 30 MPa, time 24 h) (Qi et al., 2010), and the surface water was wiped off with absorbent paper immediately after the sample was taken out. The T2 spectrum of the sample was tested in a MicroMR 12-025V nuclear magnetic resonance analyzer. The main experimental parameters are as follows: the echo interval is 0.1 ms, the waiting time is 3000 ms, the echo number is 3000, and the sampling collection time is 128.

(2) NMR + centrifugation: Because the centrifugation experiment requires the sample to be a standard one-inch small cylinder, some of the samples were broken during the sample preparation process and failed to meet the requirements of the centrifuge. Therefore, only the L-1, L-2, L-4, L-8, L-9, L-10, and L-11 samples were subjected to the saturated oil + centrifugation experiment to obtain the characteristics of the movability of shale oil. The mineral content of these seven samples are different, and the lithofacies range from massive to lamellar, which meets the analysis requirements. Therefore, the analysis of the influencing factors of shale oil movability is based on these seven samples. The main experimental steps are: first, the cylindrical sample after washing was saturated with kerosene, and then, the first nuclear magnetic test was performed; next, the sample was taken out and centrifuged at 7500 rev/min for 8 h, and then, the secondary nuclear magnetic test was performed.

It should be noted that when the speed exceeds 8000 rev/min, the shale sample is easily broken; thus, the speed was set to 7500 rev/min. The nuclear magnetic signal is essentially unchanged whether the centrifugation time is set to 8 h or 7 h, indicating that there is no movable fluid. Thus, the centrifugation time was set to 8 h.

(3) NMR + displacement: To observe the fluid migration characteristics in shale, a layered argillaceous lime sample with a depth of 3040 m was selected for the displacement experiment. The displacement fluid used was heavy water. Before starting the experiment, the sample was dried and placed in a vacuum pressure saturation device for 4 h. The kerosene was saturated with a pressure of 20 MPa for 24 h. The sample was taken out and placed in a displacement device. The displacement pressure was set to 1 MPa, 3 MPa, 5 MPa, 7 MPa, and 10 MPa, and the ring pressure was set to be 5 MPa higher than the displacement pressure. The T2 spectrum was collected each 15 min, and the corresponding spectrum was obtained. When the spectrum did not change substantially between consecutive retrievals, the next pressure point was selected.

4 Experimental results

4.1 Pore structure characteristics

Pore size distribution, specific surface area, and pore connectivity are the three key parameters for evaluating the pore structure characteristics. CO2 adsorption, N2 adsorption, and MICP are often used to elucidate the pore size distribution (PSD), as in Loucks et al. (2009) and Jiang et al. (2016b). However, different results have often been obtained from the same sample during the application of CO2 adsorption, N2 adsorption, and MICP because these techniques differ from each other on a theoretical basis and are different in terms of their inversion algorithms and experimental conditions. However, these differences have not been thoroughly discussed and solved (Lu et al., 2016; Ning et al., 2017a). Therefore, the NMR data under the constraints of high pressure mercury data are used to characterize the pore size distribution of shale, and the specific surface area of

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the shale was characterized by N\textsubscript{2} adsorption experimental data.

According to IUPAC (1972), pores can be classified as micropores (< 2 nm), mesopores (2–50 nm), and macropores (> 50 nm). The categorization was initially applied to chemical products and has been widely used for shale and mudstone reservoirs worldwide (Jiang et al., 2016a; Tang et al., 2017). In the actual pore size distribution curve, different peaks are bound to correspond to internal control factors and have different effects on fluid flow. If the pore size distribution is analyzed at the boundaries of 2 nm and 50 nm, the accuracy of the analysis may be affected. Therefore, when performing statistical analyses on actual samples, the statistical interval is divided according to the actual characteristics of the pore size distribution.

### 4.1.1 Pore size distribution

Li et al. (2015) suggested that only the matched part of MICP and NMR data could be used to determine the coefficients for conversion, and this method achieved credible results. Therefore, in this study, we transformed the T\textsubscript{2} spectral data into the PSD by referring to Li et al. (2015).

The PSD determined by the NMR analysis can also be classified into three parts by taking the peak-valley marks as boundaries (Fig. 2), and the results are consistent with the capillary pressure curves from the MICP analysis. Thus, we used 50 nm and 2 \textmu m as the boundaries to separate the PSD into three parts as follows: < 50 nm, 50 nm–2 \textmu m, and > 2 \textmu m. In general (Table 2), the volume of the < 50 nm category was in the range of 0.00551–0.02182 mL·g\textsuperscript{–1}, with an average of 0.01200 mL·g\textsuperscript{–1}; the volume of the 50 nm–2 \textmu m category was in the range of 0.00085–0.00869 mL·g\textsuperscript{–1}, with an average of 0.00324 mL·g\textsuperscript{–1}; and the volume of the > 50 nm category was in the range of 0.00102–0.00752 mL·g\textsuperscript{–1}, with an average of 0.00378 mL·g\textsuperscript{–1}. The contributions of the three parts to the total volume corresponded to 61.4%, 19.6%, 19.0%, respectively, indicating that the pores with a radius of < 50 nm dominated the space within shale, while the contributions of the 50 nm–2 \textmu m and > 2 \textmu m pores were both important to the shale reservoir.

![Fig. 2. Distribution of NMR–determined pore size of the shale samples.](image)

#### 4.1.2 Specific surface area

Based on the nitrogen adsorption data, the BET model was used to calculate the specific surface area distribution of seven samples, as shown in Table 2. The specific surface area of the seven samples ranged from 3.17 to 8.67 m\textsuperscript{2}·g\textsuperscript{–1}, with an average of 5.74 m\textsuperscript{2}·g\textsuperscript{–1}. The specific surface area is related to the bedded structure development of the sample. Massive samples have the highest specific surface area (average 7.09 m\textsuperscript{2}·g\textsuperscript{–1}), while the lamellar samples have the lowest specific surface area (average 4.53 m\textsuperscript{2}·g\textsuperscript{–1}), and the layered samples are in between the two, at 6.16 m\textsuperscript{2}·g\textsuperscript{–1}. The pore specific surface area percentage of < 50 nm ranged from 97.05% to 99.64%, with an average of 98.15%. The average specific surface area of 50 nm–2 \textmu m pores is 1.83%, while the specific surface area of > 2 \textmu m pores is very small, accounting for only 0.02%.

<table>
<thead>
<tr>
<th>Sample ID</th>
<th>Depth (m)</th>
<th>Bedding development</th>
<th>Specific surface area (m\textsuperscript{2}·g\textsuperscript{–1})</th>
<th>Specific surface area percentage (%)</th>
<th>Pore volume (&lt;50nm) (mL·g\textsuperscript{–1})</th>
<th>Pore volume percentage (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>L–1</td>
<td>2947.19</td>
<td>Massive</td>
<td>8.674</td>
<td>99.429, 0.567, 0.003</td>
<td>15.01</td>
<td>76.79, 16.41, 6.80</td>
</tr>
<tr>
<td>L–2</td>
<td>2962.07</td>
<td>Massive</td>
<td>5.497</td>
<td>99.638, 0.353, 0.008</td>
<td>16.62</td>
<td>38.17, 52.28, 9.56</td>
</tr>
<tr>
<td>L–4</td>
<td>3025.15</td>
<td>Layered</td>
<td>6.161</td>
<td>97.051, 2.938, 0.011</td>
<td>14.62</td>
<td>37.66, 45.88, 16.45</td>
</tr>
<tr>
<td>L–8</td>
<td>3055.10</td>
<td>Lamellar</td>
<td>4.315</td>
<td>97.632, 2.309, 0.059</td>
<td>20.16</td>
<td>55.50, 7.23, 37.27</td>
</tr>
</tbody>
</table>

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4.2 Movable oil characteristics
Comparing the T<sub>2</sub> spectra before and after centrifugation at a speed of 7500 rev/min, the numerical reduction is the movable oil, and the saturation of immovable oil can be calculated accordingly (Table 3). The movable oil saturation of shale oil in the lower third member of the Shahejie formation (Es3l) of the Zhanhua depression is generally low, averaging only 21.50%. The lowest is layered sample L-4 at only 8.03%, the highest is lamellar sample L-8 with a movable fluid saturation of 44.86%, and the movable fluid saturation of the Lamellar-layered sample is between the two, averaging 25.92%.

Table 3 Movable and immovable oil saturation of 7 samples from Es3l, Zhanhua Sag

<table>
<thead>
<tr>
<th>Sample ID</th>
<th>Depth (m)</th>
<th>Bedding development</th>
<th>Movable oil saturation (%)</th>
<th>Immovable oil saturation (%)</th>
</tr>
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<tbody>
<tr>
<td>L-1</td>
<td>2947.19</td>
<td>Massive</td>
<td>15.06</td>
<td>84.94</td>
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<td>Massive</td>
<td>18.00</td>
<td>82.00</td>
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<td>Layered</td>
<td>8.03</td>
<td>91.97</td>
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<td>3055.10</td>
<td>Lamellar</td>
<td>44.86</td>
<td>55.14</td>
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<td>L-9</td>
<td>3060.10</td>
<td>Lamellar</td>
<td>12.74</td>
<td>87.26</td>
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<tr>
<td>L-10</td>
<td>3109.90</td>
<td>Lamellar-layered</td>
<td>34.17</td>
<td>65.83</td>
</tr>
<tr>
<td>L-11</td>
<td>3119.90</td>
<td>Lamellar-layered</td>
<td>17.66</td>
<td>82.34</td>
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<tr>
<td>Average</td>
<td></td>
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<td>21.50</td>
<td>78.50</td>
</tr>
</tbody>
</table>

5 Discussion

Based on the above experimental data, the occurrence characteristics and the migration and flow path of shale oil are first analyzed. Then, the influence of pore structure characteristics on shale oil flow is discussed. Finally, the influence of mineral fabric characteristics of the reservoir on shale oil flow is addressed.

5.1 The occurrence and flow characteristics of shale oil
5.1.1 The dependence of oil movability on porosity
Micro-nanometer pores develop in shales, whose sizes range from several micro-nanometers to microns, even up to millimeters. Oil and gas molecules are subject to different viscosities and intermolecular forces in pores of different sizes (Zou et al., 2012), and the degree of the ease of flow varies.

For the T<sub>2</sub> spectrum characteristics before and after centrifugation, the overall forms of different samples are similar, but the data are different. For this case, sample L-11 is illustrated (Fig. 3). Based on spectrum characteristics, the spectra are divided into three regions: short relaxation, medium relaxation, and long relaxation—with the boundaries of T<sub>2</sub> = 2 ms or 50 ms. As the time of centrifugation increases, the T<sub>2</sub> spectrum amplitude gradually decreases, indicating that the fluid is gradually “thrown out” under the action of centrifugation force. After a centrifugation of approximately two hours, the amplitude change was the most significant, and the peak in the long relaxation region almost completely disappeared, indicating that the fluid for the large-sized pores is prioritized to flow out. After approximately five hours of centrifugation, the amplitude in the medium relaxation region remarkably goes down, indicating that the fluid for the medium-sized pores begins to flow out significantly. When the time of centrifugation reaches eight hours, the pattern of the spectrum is not much different from that of five hours, indicating that the movable fluid almost completely flows out, and what remains is immovable fluid. Comparing the spectra of saturated oil and eight-hour centrifugation, it is found that the part of the short relaxation portion changes little, only declining approximately 2.3%, the medium relaxation declines approximately 59.0%, and the long relaxation declines approximately 92.5%, corresponding to the proportions of movable oil in small pores, medium pores, and large pores.
Fig. 3. $T_2$ curve after centrifuge of L-11 m sample in Luo69 well, Zhanhua Sag.

As you can see from Fig. 4, the movable oil saturation is positively correlated with pore volume ratios greater than 2 μm, and the immovable oil saturation is positively correlated with pore volume ratios less than 2 μm (Fig. 5). These rules are found in different lithofacies samples, indicating that the percentage of the large pores governs the movable oil content of the shale oil, and the percentage of the small pores has vital influence on the immovable oil content of the shale oil.

Fig. 4. Relationship between movable oil saturation and pore volume percentage (> 2 μm).

Fig. 5. Relationship between immovable oil saturation and pore volume percentage (< 2 μm).

As is shown above, based on the changes in centrifugal NMR spectra and statistical analysis, it can be determined that the movable fluid is mainly distributed in the larger pores, corresponding to the long relaxation and the medium relaxation behavior, while the immovable fluid is more distributed in the smaller pores, corresponding to the short relaxation results.

5.1.2 Shale oil flow path

The hydrogen in the heavy water used in the displacement experiment is deuterium ($^2$H), consisting of one proton and one neutron, and does not generate a signal in nuclear magnetic resonance experiments. Therefore, when heavy water is used for the high-pressure displacement of saturated oil samples, the nuclear magnetic signal of the sample gradually weakens, as heavy water enters the sample to drive the kerosene. The reduced
signal corresponds to the storage space where the water is concentrated and also corresponds to the storage space of the movable oil. 

Figure 6 shows the $T_2$ spectrum at different displacement pressures and times. 0 MPa refers to the saturated oil state. The $T_2$ spectrum represents the ability to store oil and is divided into three parts: short relaxation, medium relaxation, and long relaxation parts, at boundaries of 2 ms and 50 ms. After 45 min of displacement at 1 MPa, the short relaxation peak decreased significantly, and the medium-long relaxation peak had a tendency to migrate to the right, indicating that heavy water displaced some kerosene from smaller pores to larger pores. After 45 min of displacement at 3 MPa, the short relaxation curve decreased again, the middle relaxation peak decreased, and the long relaxation peak increased. This indicated that the kerosene in the medium-scale pores decreased, while the kerosene in the large-scale pores increased, and the peak of the medium-long relaxation migrated to the left, caused by the kerosene in the small pores being continuously displaced to the larger pores. When the displacement was 45 min at 5 MPa, the short relaxation part also decreased, but the amplitude was not as large as that for 1 MPa-45 min or 3 MPa-45 min, indicating that displacement became increasingly difficult. The medium-long relaxation peak migrated to the right and increased its peak value from 3 MPa-45 min, indicating that the kerosene in the small pores gradually flows into the large pores. At 7 MPa-30 min, the peak value of short relaxation was weak, and the peak of medium-long relaxation was significantly reduced, indicating that heavy water had difficulty in displacing kerosene in small pores. After heavy water replaced the kerosene in larger pores, the medium-length relaxation exhibited significantly reduced features. At 10 MPa-15 min, the mid-relaxation peak further decreased and migrated from 20 ms to 50 ms, indicating that the kerosene in the pores continuously moves to the outside through the large pores. It is worth noting that at 7 MPa-30 min and 10 MPa-15 min, the signal gradually increased at 50 ms, probably because heavy water opened up new migration channels under high pressure.

![Fig. 6. $T_2$ curve after displacement of 3040 m sample in Luo69 well, Zhanhua Sag.](image)

Figure 7 shows the imaging of the sample oily abundance at different displacement pressures and times. As the pressure increases, the red area becomes increasingly smaller, indicating that the kerosene in the core decreases with increasing time during the displacement experiment. Heavy water displacement experiments reproduce the fluid migration process under high pressure. The fluid in the small pores first seeps into the larger pores and finally migrates to the outside. It can be seen that the existence of large pores can be used not only as an important reservoir space but also as a migration channel for fluids.
5.2 Effect of pore structure on shale oil movability

Reservoir characteristics comprise two types: the characteristics of the reservoir organization, including the content of mineral components constituting the reservoir and its distribution arrangement, and the pore structure characteristics, directly controlled by the structural features. The influence of shale reservoir characteristics on the shale oil movability also comes from the two aspects. The pore structure is the most fundamental factor affecting fluid flow in the reservoir, and the organization characteristics affect the shale oil movability by controlling the pore structure of the reservoir. To clarify the phenomena, the effects on shale oil movability were demonstrated from the inside to the outside of the reservoir.

5.2.1 Effect of specific surface area on shale oil movability

Figure 8 shows that as the specific surface area increases, the immovable oil saturation increases. This also means a reduction in the saturation of the movable oil, which is clearly detrimental to the flow of shale oil. Although shale oil is the same as shale gas, it can be classified as an adsorption state and a free state, depending on the aggregation state. However, shale gas in the adsorption state will desorb and release when the temperature and pressure change, eventually becoming an important part of industrial output (Ji et al., 2014; Jiang et al., 2016b). The abundant specific surface area can increase the shale reservoir storage capacity for shale gas (Tang et al., 2019). For shale oil, this is not the case. Adsorbed shale oil largely exists on the pore surface through physical or chemical adsorption, and it is typically not flowable. The flowable part is generally the free shale oil. The pore specific surface provides a place for the adsorption shale oil, so the specific surface area is unfavorable for shale oil flow.
5.2.2 Effect of pore connectivity on shale oil movability

Porosity connectivity is a prerequisite for shale oil flow. Considering the relationship between movable fluid saturation and total pore connectivity ratio (Fig. 9a), the 0–50 nm pore connectivity ratio (Fig. 9b) and the 50 nm–2 μm pore connectivity ratio (Fig. 9c) are positively correlated, indicating that the better the pore connectivity is, the higher the proportion of movable fluid. The value of the fitting coefficient $R^2$ reflects the correlation between different variables to some extent. The fitting coefficient $R^2$ of the movable fluid saturation and the pore connectivity ratio of 0 to 50 nm is 0.50, and the $R^2$ of the pore communication ratio of 50 nm to 2 μm is 0.65. This shows that the influence of 50 nm–2 μm connectivity on fluid saturation is more significant. It is not difficult to understand that the small pores have a stronger resistance to shale oil movability because of the small pore throat, and therefore, the shale oil movability is inevitably adversely affected. Therefore, better connectivity is beneficial to the flow of shale oil, and the effect of large pores on shale oil movability is more significant.
(c) Relationship between movable oil saturation and pore connectivity (50 nm–2 μm).

Fig. 9. Relationship between movable oil saturation and connectivity ratio of different scale pores in Es3l, Zhanhua Sag.

5.2.3 Effect of pore size on shale oil movability

In terms of pore size, the above control effect of shale oil movability has been clarified by NMR + centrifugation and NMR + displacement, and the cause of this phenomenon can be explained from at least three aspects:

(1) Small pore are lipophilic and macroporous are hydrophilic

Because of the difference in wettability among the pore surfaces, oil saturation and water saturation of the same sample will be different. A parallel sample is saturated with oil and water, and then subjected to NMR experiments. If the content of saturated oil is higher than that of saturated water, the pore is considered to undergo oil-wetting, while if the opposite is true, then the water-wetting characteristic is observed.

The abscissa of Fig. 10 is the T2 spectrum, and the ordinate is the normalized difference between the saturated water and the saturated oil. The curve below the X-axis indicates that the pore is hydrophobic, while the curve above the X-axis indicates hydrophilicity.

Six samples showed negative values in the short relaxation region and positive values in the long relaxation region. This shows that the small pores of different samples experience oil-wetting, while the large pores undergo water-wetting. Figure 10 shows that the high TOC content corresponds to a lower curve amplitude, which indicates stronger lipophilicity. This is because organic matter is the parent material of raw oil, and the pores associated with organic matter naturally exhibit strong lipophilicity. Minerals such as clay mixed with organic matter exhibit oil-wetting characteristics under the infiltration of organic hydrocarbons. Ma et al. (2017) found that the amount of retained oil in the hot simulated samples has good correspondence with the pores of organic matter, confirming, to some extent, the binding effect of pores related to organic matter on shale oil due to its lipophilicity. The interstitial pores/cracks of organic matter developed, and the intercrystalline pores of clay minerals mixed with organic matter are relatively developed in the Es3l of the Shahejie Formation in the Zhanhua sag (Ning et al., 2017a), which may be the primary type of oil-wetting pores. Oil-wetting pores have a stronger binding force to shale oil, which is not conducive to shale oil flow. Water-wetting pores have a stronger binding force to water, which is more conducive to shale oil flow. This is the reason that large pores contain movable oil, while immovable oil is contained inside small pores.

(2) Small pores are richer than the specific surface area

Fig. 10. Distinguish graph for wettability of different scale pores in Es3l, Zhanhua Sag.
The full pore characterization of the shale pore structure of the Longmaxi Formation shale in the southeastern Sichuan Basin shows that micropores and mesopores (<50 nm) provide most of the specific surface area (Jiang et al., 2016). The shale of the Es3l of the Shahejie Formation in the Zhanhua Sag also showed similar characteristics. The seven samples with pores smaller than 50 nm provided a specific surface area of 99.78% (Table 2). Surface physicochemical theory shows that the fluid on the surface of the material will undergo a physical chemical reaction on the surface of the medium to form a bounded fluid. Under conventional centrifugal displacement, it is clearly immovable (Shen et al., 2012). The abundant specific surface area provides a large number of sites for the adsorption of shale oil, while the adsorbed shale oil is generally immovable. This is the other reason that large pores contain movable oil, and immovable oil resides inside small pores.

(3) Small pores have poor connectivity
Ning et al. (2017b) combined NMR and high-pressure mercury injection to quantitatively evaluate the pore connectivity of different shale lithofacies in the Es3l of Shahejie Formation in Zhanhua Sag. The results show that in the shale of the different lithofacies, small pores have poor connectivity and macropores exhibit good connectivity (Figure 11). In addition, the slope of the spontaneous infiltration curve of the same sample in the first half is larger than that in the second half, which also indicates that the connectivity of the macropores is better than that of the small pores. It is not difficult to understand that when comparing pores with the same degree of bedding development, large-scale pores tend to extend farther, and thus, it is easier to connect with layered cracks to improve connectivity. The small-pore channel is narrow and, thus, is not easy to communicate with other pores. Therefore, the connectivity is poor, which is not conducive to shale oil flow.

5.3 The influence of mineral fabric characteristics on shale oil movability
The mineral fabric characteristics consist of mineral composition and distribution arrangement. Compared with the microscopic pore structure, the mineral fabric characteristics are easier to observe, which is of greater direct significance for the optimization of favorable shale oil zones.

5.3.1 The influence of mineral compositions on shale oil movability
Calcite and clay minerals are the two main minerals in the study area. The saturation of movable oil is positively correlated with the calcite content, with a correlation coefficient R^2 as high as 0.88, indicating that the calcite content significantly controls the movable oil (Fig. 12). A preliminary study has unveiled that the calcite content has great influence on the fracture density (Ning et al., 2017a). However, the micro-fracture size is larger than that of the micro-nanometer pores of shales. Moreover, owing to the wetting of water, the binding force on shale oil is relatively weak, which is beneficial to the flow of shale oil. This is the internal reason for this phenomenon. As seen in Fig. 13, the clay minerals have an essential influence on the saturation of immovable oil. Intercrystalline pores provided by the minerals of the shales in the Sha 3 formation in Zhanhua Sag are nanoscale pores, which are not beneficial to the flow of shale oil. In addition, clay minerals block the throat (Guo et al., 2014) so that the connectivity of pores worsens. The result is that a sufficiently large specific surface area (Ross et al., 2008) could trap more oil and gas, resulting in an immovable adsorption state. These all are internal factors that contribute to the hindrance of clay minerals in the flow of shale oil.
5.3.2 The influence of the bedding on shale oil movability

The bedding is the most intuitive aspect of the change in the arrangement of the different minerals. In the 35 industrial shale oil run wells in the Jiyang Depression, most of the industrial oil run is obtained from non-interlayer shales that developed lamellar strata (accounting for 73%), and the horizontal permeability of the lamellar strata is 1–4 orders of magnitude higher than that of the vertical strata, as the horizontal seepage performance is superior (Wang et al., 2016). According to the statistics of 7 samples in Table 2, the average movable oil saturation of massive, layered, layered-laminar, and lamellar shale is 16.53%, 8.03%, 25.92%, and 28.80%, respectively. As a result of the limited sample quantity, the movability of lamellar samples is not highest. However, the more developed the bedding is, the better the shale oil can flow (Fig. 14). The pore volumes (> 2 μm) of massive, layered, layered-laminar, and lamellar shale samples in Zhanhua Sag increase regularly (Table 2), indicating that the more developed the bedded structure, the more developed the bedding fractures, which provide more large-scale storing space and can store more free shale oil. Furthermore, without bedding fractures, many small-scale pores are more likely to become isolated small pores. When the bedding fractures are developed, some surrounding smaller pores are more easily extended and connected to the bedding fractures, naturally increasing the connectivity of pores at different scales (Fig. 11, large red arrows). These are the internal reasons that the bedding is beneficial to the flow of shale oil.
5.4 Geological significance

The above experimental result and analysis demonstrate that the pore structure of shales is the fundamental factor influencing and controlling the flowability of shale oil. Specifically, small pores have a strong binding effect on shale oil, as result of their lipophilicity and abundant specific surface area. Meanwhile, large pores, as the main storing space of movable oil and the seepage channel of shale oil, have an important control effect on the flow production of shale oil. The mineral fabric affects shale oil movability macroscopically. Brittle calcite is more likely to form micro-fractures and has a positive effect on the flow of shale oil. Clay is bad for the flow of shale oil on account of its huge specific surface area and its filling and blocking effect. On the one hand, the development of laminates increases the storing space at large scales; on the other hand, it improves the connectivity of pores at different scales, which is conducive to the flow of shale oil. Therefore, under the condition that the oil generation capacity is satisfied, the optimal selection of favorable exploration sections and the determination of favorable areas of shale oil should focus on the development of large pores in shale oil reservoirs at the micro-level, rather than the total porosity. At the macro-level, brittle minerals and bedded structure developing segments and zones should be selected.

6 Conclusions

(1) The movable oil saturation of the Zhanhua Sag is only 21.50% on average, and the movability is poor. Movable oil is mainly distributed in the large pores, while the small pores are mainly favorable for immovable oil. The gradual migration from small pores to large pores is the main path for movable oil flowing.

(2) The root cause of shale oil movability is related to the pore structure. Large pores provide more pore space for the storage of movable oil and can also serve as a seepage channel to facilitate shale oil flow. The oil-wetting characteristics and large specific surface area facilitate the enrichment of adsorbed oil by small pores, while poor connectivity makes it unfavorable for shale oil flow.

(3) Macroscopically, mineral fabrics affect shale oil movability. Calcite increases shale brittleness, which is more conducive to the formation of cracks and has a positive effect on shale oil movability. Because clay has a strong adsorption effect and easily blocks the pore throat, the occurrence of clay does not facilitate the flow of shale oil. Bedded structure increases the storage space and also improves pore connectivity, which is beneficial to the flow of shale oil.

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