Research Article

Wettability and Its Controlling Factors of Mixed Shale Oil Reservoirs: A Case Study of Permian Lucaogou Formation in Jimusar Sag

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Received 11 May 2021; Accepted 29 June 2021; Published 23 October 2021

Academic Editor: Xixin Wang

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Due to diverse mineral composition and complex lithology, mixed shale oil reservoir wettability features and the associated controlling factors remain unclear, limiting the evaluation of the oil-bearing property and sweet spot distribution. This paper examined mixed shale oil samples from the Jimusar Sag in the Lucaogou Formation. Casting thin sections, X-ray diffraction, geochemical characteristics, argon ion polishing scanning electron microscope, and high pressure mercury injection were used to analyze lithologic characteristics, mineral compositions, and pore-throat structures within the mixed shale oil reservoir. Using spontaneous imbibition and nuclear magnetic resonance, the wettability characteristics were analyzed. Impacts of organic matter abundance, mineral composition, pore-throat structure, and source-reservoir combination on wettability were discussed. The Lucaogou mixed shale oil reservoir primarily contains intergranular dissolved pores, intragranular dissolved pores, and intercrystalline pores. Three types of mercury intrusion cures were observed, including a weak platform shape (type I), gentle straight line shape (type II), and upward convex shape (type III), corresponding to intergranular, dissolved, and dissolved-intercrystalline dominant pore-throat systems, respectively. Mixed shale oil reservoirs show dual wet properties, containing both oil-wet and water-wet pores. Oil-wet pores (large pores with \( T_2 > 1 \) ms) are more common and have better connectivity than water-wet pores (small pores with \( T_2 < 1 \) ms). Dolomite-bearing siltstone and mudstone are primarily strongly oil-wet, while siltstone is primarily mixed wet. Type II and type III pore-throat systems are more oil-wet than type I for the same source-reservoir combination. Mixed shale oil reservoir wettability is primarily controlled by three factors, including organic matter abundance, source-reservoir combination, and dolomite content. The influence of the pore-throat structure is weak. High organic matter abundance, an integrated source-reservoir or adjacent source-reservoir, and appropriate dolomite content are necessary conditions for forming a strong oil-wet shale oil reservoir in the Lucaogou Formation. Stronger oil-wet is beneficial for shale oil charging and enrichment.

1. Introduction

Shale oil reserves are an abundant, widely distributed, and important type of unconventional oil and gas deposit [1]. In the narrow sense, shale oil refers to oil stored in organic-rich shale formations with nanoscale pore size [2]. In a broad sense, shale oil refers to liquid hydrocarbons found in organic-rich shales and fine-grained interlayers, such as siltstones and carbonate rocks [3, 4]. The mixed shale oil reservoirs fall into the broad sense of shale oil, which are the mixing product of multiple sources such as mechanical clastic sediment, chemical sediment, and volcanism [5]. Mixed shale oil reservoirs are commonly deposited in the deltaic front and lacustrine facies [3, 6, 7].
characterized by fine-grained sediment, complex lithology, frequent thin interbedding, and strong heterogeneity [8]. Mixed shale oil reservoirs are important for the commercial development of shale oil in China, especially the Lucaogou Formation in the Junggar basin [1]. The Lucaogou Formation mixed shale oil reservoir is significantly different from other narrowly defined shale oil reservoirs, such as the Songliao Basin and Jianghan Basin. The Cretaceous Qingshankou Formation in the Songliao basin is primarily composed of thick mudstone, which is composed of feldspar, quartz, and clay minerals with minor amounts of carbonate minerals [9]. The Eocene Qianjiang Formation in the Jianghan Basin is primarily composed of argillaceous dolomite, dolomitic mudstone, and calcareous mudstone, with low silt and feldspar content [10]. However, the Lucaogou Formation contains a set of fine grained mixed rocks including dark mudstones, siltstones, carbonates, and their associated transitional lithologies [11, 12]; the primary minerals are feldspar, quartz, and dolomite; and the clay mineral content is low [13]. The lithology of the two types of shale oil reservoirs is significantly different, and the mixed reservoirs contain more silt. Different lithologies have different diagenesis, pore type, pore size distribution, and pore-throat connectivity [11]. In recent years, the Lucaogou Formation mixed shale oil has been successfully explored and developed, and China’s first national demonstration area for continental shale oil has been established.

Wettability describes the tendency of a fluid phase to preferentially wet a rock [14, 15]. The wettability of conventional reservoirs can be divided into oil-wet, water-wet, and intermediate-wet [15]. Water (wetting phase) prefers a water-wet surface, while oil (nonwetting phase) prefers an oil-wet surface. Neither water nor oil prefers an intermediate-wet surface [16]. Wettability significantly affects hydrocarbon charging and accumulation [17–19] as well as the occurrence state and production efficiency of oil and gas [20]. For unconventional reservoirs, wettability can also affect the damage degree of the shale oil reservoir caused by fracturing fluid intrusion and retention, thus affecting fracturing plan optimization [21–23]. Therefore, studying the wettability of shale oil reservoirs is necessary.

For conventional reservoirs, three primary methods are used to measure wettability, including the contact angle measurement, the US Bureau of Mines (USBM) test, and the Amott test [24–26]. However, mixed shale has extremely low permeability as well as complex mineral compositions and rock types [13], making accurately measuring wettability difficult. Both the Amott and USBM wettability tests require displacement, and the extremely low permeability requires very large displacement force, which significantly affects the applicability of these two traditional experiments [27, 28]. Organic matter in the mature stage is oil-wet, while some rock minerals are commonly water-wet [29–32]. Hence, mixed wet is common in mixed shale, i.e., containing both oil-wet and water-wet interfaces. The contact angle method is easily disturbed by surface roughness, surface contamination, and compositional heterogeneity; therefore, it should not be used to measure the wettability of mixed shale oil reservoirs [20].

Spontaneous imbibition is a process by which the wetting phase fluid spontaneously displaces the nonwetting phase fluid do to capillary force [21, 22]. The process of imbibition is primarily related to the pore structure of the rock, the physical properties of the fluid, and the interaction between them [29–32]. Spontaneous imbibition can be used to understand the pore structure as well as predict oil and gas production behavior [33, 34]. Imbibition, a relatively simple experiment, can reveal the process of the wetting phase displacing the nonwetting phase spontaneously, reflecting the overall wettability of the rock [21, 22]. At present, many scholars have studied the wettability of shale using spontaneous imbibition experiments, revealing the influencing factors of shale oil reservoir wettability [16, 21, 27, 32, 35–47].

At present, some scholars have carried out combined experiments using nuclear magnetic resonance (NMR) and spontaneous imbibition to measure shale wettability [35, 37, 45]. This combined experiment can reveal the spatial distribution of fluid during imbibition and determine the microscopic distribution of different wet phase fluid in order to better understand the wettability mechanism [48]. NMR reflects the volume of fluid by detecting hydrogen nuclei within rock pores and indirectly reflecting the pore distribution of reservoirs. This method is convenient for measurement and will not damage the sample [49]. Previous wettability studies using the combination method primarily focused on pure shale reservoirs or tight sandstone reservoirs. However, mixed shale oil reservoirs are characterized by complex mineral composition and diverse lithology. This new combination method may shed new light on the wettability of mixed shale oil reservoirs.

This paper presents a multiphase study, including casting thin sections, argon ion polishing scanning electron microscope (AIP-SEM) analysis, organic geochemical analysis, X-ray diffraction, and high pressure mercury injection (HPMI), to reveal the lithology, mineral composition, oil-bearing properties, and pore structure of a mixed shale oil reservoir. Then, the wettability of the Lucaogou mixed oil shale is studied using both spontaneous imbibition and NMR. Finally, the influencing factors of wettability are discussed.

2. Materials and Methods

2.1. Geological Features. The Junggar Basin is one of the largest oil-gas bearing basins in Northwest China (Figure 1(a)), and the Jimsar sag, located in the eastern Junggar Basin (Figure 1(b)), is a typical dustpan structure [50] with an irregular polygon shape. The Jimsar sag is bordered by the Guxi uplift to the east, the Xidi fault and the Laozhuangwan fault to the west, the Santai fault to the south, and the Jimsar fault and Shaji uplift to the north. The Permian Lucaogou Formation in the Jimsar sag was deposited primarily in a salinized lacustrine environment, and the sediments came from a combination of terrigenous material and chemical deposition [13]. The mixed rocks in the Lucaogou Formation can primarily be classified as siltstones, mudstones, and dolomites, among which the mudstones are dominant.
The siltstones include siltstone or fine sandstone, argillaceous siltstone, calcareous siltstone, and dolomitic siltstone. The mudstones include silty mudstone, dolomitic mudstone, and shale. The dolomites include argillaceous dolomite (micrite), silty dolomite, and dolarenite [12]. The lithology contains frequent changes in the vertical direction, some of which reach the centimeter scale (Figure 1(c)). Because the provenance comes from the south, the silt content in the southern Lucaogou Formation is relatively high. Towards the north, the amount of mud-size grains or dolomite increases, and the dolomite or mudstone becomes thicker [12]. The Lucaogou Formation thickness is greater than 200 m on average, the maximum thickness can reach 350 m, and the formation generally thickens from west to east [51]. The burial depth of the Lucaogou Formation in the study area is 2500 to 4000 m, and the maturity of organic matter $R_m$ value ranges from 0.70% to 1.30% (average 0.78%) [8]. The Lucaogou Formation is divided vertically into section 1 ($P_2L_1$) and section 2 ($P_2L_2$), corresponding to the lower and upper sweet spot sections, respectively (Figure 1(c)) [1]. Silty sandstone, argillaceous siltstone, and dolarenite are the primary lithology in the upper sweet spot section. Dolomitic siltstone is dominant in the lower sweet spot section. Several horizontal wells have been drilled in these two sweet spot sections [8].

2.2. Experimental Theory

2.2.1. Spontaneous Imbibition. To describe the spontaneous imbibition process of water moving vertically upward in a water-air system, Handy [52] proposed the following equation:

$$Q_w^2 = \left( \frac{2P_c k_w \mathcal{Q} A^2 S_w}{\mu_w} \right) t,$$

(1)

where $Q_w$ is the water imbibition volume of the sample, $P_c$ is the capillary pressure, $k_w$ is the effective permeability of the sample to water, $\mathcal{Q}$ is porosity, $A$ is the core cross-sectional area, $S_w$ is the fractional water content in the pore, $\mu_w$ is the viscosity of water, and $t$ is the imbibition time. Equation (1) can be simply rewritten as

$$H = at^{0.5},$$

(2)

where $H$ is the cumulative imbibition height ($H$ equals $Q_w/A$) and $a$ is a constant.

The cumulative imbibition height and imbibition time form a straight line on a log-log plot, and the slope value for classical homogeneous materials is 0.5 [53, 54]. However, a slope of 0.5 in equation (2) is not suitable for natural rocks due to the complex pore connectivity. Cai et al. [31] studied the relationship between tortuosity and imbibition time (i.e., imbibition slope). According to the percolation theory, when the imbibition slope is less than 0.5, the imbibition slope decreases, and pore connectivity of tight rock worsens [55].

2.2.2. NMR Experimental Theory. NMR can directly measure hydrogen within fluids contained in reservoirs based on the strong signal and highest gyromagnetic ratio of hydrogen.
nuclei in the fluid [56]. NMR transverse relaxation in rocks consists of three parts: surface relaxation, bulk relaxation, and diffusion relaxation [57], which can be expressed as

\[ \frac{1}{T_2} = \frac{1}{T_{2s}} + \frac{1}{T_{2b}} + \frac{1}{T_{2d}}, \]

where \( T_2 \) is the proton NMR transverse relaxation, \( T_{2s} \) is the surface relaxation, \( T_{2b} \) is the bulk relaxation, and \( T_{2d} \) is the diffusion relaxation.

Diffusion relaxation and bulk relaxation can be ignored in shale oil reservoirs containing dominant nanoscale pores. The surface relaxation is proportional to the pore volume and inversely proportional to porosity proportional to the specific surface area, which can be expressed as

\[ \frac{1}{T_2} = \rho \frac{s}{v}, \]

where \( s \) is the specific pore surface area (\( \mu m^2/g \)), \( v \) is the pore volume (\( \mu m^3/g \)), and \( \rho \) is the surface relaxivity (\( \mu m/ms \)).

2.3. Experimental Method and Procedure

2.3.1. Routine Experiments. 13 mixed shale oil reservoir samples from the Lucaogou Formation in the Jimusar Sag covering the major lithologies were selected for analysis (Table 1). Cores were drilled parallel to the formation, and all experiments were carried out on the same core, in order to eliminate the influence of lithologic heterogeneity. First, the cores were cleaned with dichlorotoluene for 10 days, and then, two regular small columned samples approximately 2.5 cm and 1.5 cm long were cut and used for physical property measurement as well as subsequent spontaneous imbibition and HPMI experiments, respectively. The other fragments were used for casting thin sections and AIP-SEM analysis. The remaining samples were used for geochemical experiments.

The porosity and permeability of samples were obtained using a porosity and permeability measuring instrument (PoroPDP-200) with confining pressure of 3 MPa. The HPMI experiment was performed using an AutoPore IV 9500. The mercury intrusion and extrusion curves were measured and the pore-throat size distribution of the samples was derived from the intrusion curves. The maximum injection pressure was 200 MPa, corresponding to a minimum pore-throat radius of 3.7 nm. Through casting thin section observations, the particle size, material composition, and pore types of the rock were characterized and the lithology was determined. A Sigma 450 emission scanning electron microscope was used to analyze samples polished using an argon ion beam, in order to observe the pore structure in detail. The mineral composition of the samples was determined using an X-ray diffractometer (Zet 207 Bruker D8). The total organic carbon (TOC) content of the samples was measured using a total organic carbon analyzer (i.e., ELAB-TOC). The free hydrocarbon \( S_1 \) and pyrolysed hydrocarbon \( S_0 \) were obtained using an oil and gas display evaluation instrument (YQ-VIIIA).

2.3.2. Combined Spontaneous Imbibition and NMR Experiment. Combined spontaneous imbibition and NMR measurements were carried out on the selected samples to characterize the wettability of mixed shale. The specific steps were as follows: (1) All samples were dried at 110°C for 12 h in vacuum to remove moisture and ensure that the pore structure was not damaged [58]. Next, the mass and NMR signal of samples were measured. (2) Under indoor conditions, the dried samples were completely immersed in brine solution for 72 h, trying to balance the brine imbibition in the samples. The mass and NMR signals were recorded through time. (3) After brine imbibition, samples were centrifuged for 12 h at 8000 r/min to remove all movable fluid. The mass and NMR signal were again measured. (4) After centrifugation, samples were imbibed oil for 48 h, so that the oil imbibition volume of the samples could balance as much as possible, and the mass and NMR signal were recorded at different times.

NMR measurements were carried out using a MicroMR12-025V NMR analyzer form the Niumag Company. The waiting time \( T_{1w} \) and echo spacing \( T_{2e} \) were set to 6 s and 0.1 ms, respectively, and the number of echoes was 12000. During the experiment, a 20000 PPM sodium bicarbonate solution was used for the brine to simulate formation water, and oil was a proportional mixture of dodecane and formation crude oil (viscosity, 15 mPa·s) to simulate formation oil.

3. Results

3.1. Petrology and Geochemical Characteristics. The lithology of the mixed shale oil reservoir in the Lucaogou Formation is complex and diverse. The selected samples in this study include dolomite-bearing siltstone, siltstone, mudstone, and dolomite (Table 1). The porosity of the sample ranges from 7.29% to 19.68%, with an average of 13%. Permeability varies from 0.02 mD to 1.582 mD, with a mean value of 0.278 mD. A weak positive correlation exists between permeability and porosity, and the permeability of siltstone with the same porosity is higher than the overall (Figure 2(a)), which reflects the strong heterogeneity of the pore structure. Quartz, feldspar, and dolomite are the primary mineral types in the Lucaogou samples. Quartz content ranges from 11.9 wt % to 35.1 wt %, with an average of 23.5 wt %. Feldspar content ranges from 15.3 wt % to 71.6 wt %, with an average of 48.9 wt %. The feldspar content of siltstone is the highest, with an average of 63.7 wt %. Dolomite is generally developed throughout the mixed shale oil reservoir, ranging from 0 to 72.8 wt %, with an average of 18.7 wt %. The cements are primarily calcite and clay minerals. Calcite content ranges from 0 to 20.1 wt %, with an average of 6 wt %. Clay mineral content is low, with an average of 2.5 wt %. Mineral composition has a certain control over porosity. Quartz and feldspar content positively correlate with porosity. Also, calcite and clay mineral content negatively correlate with porosity. When dolomite content is less than 20 wt %, it positively correlates with porosity, and when the dolomite content is greater than 20 wt %, it negatively correlates with porosity (Figure 2(b)).
| Sample ID | Depth (m) | Lithology | Por. (%) | Per. (mD) | TOC (wt %) | $S_1$ (mg/g) | $S_1 + S_2$ (mg/g) | Quartz (wt %) | Feldspar (wt %) | Dolomite (wt %) | Calcite (wt %) | Clays (wt %) | Section |
|-----------|-----------|-----------|----------|-----------|------------|-------------|------------------|--------------|---------------|----------------|----------------|-------------|---------|
| 10        | 3161.20   | Silty mudstone | 12.3     | 0.02      | 2.72       | 8.57        | 22.79            | 35.1         | 48.8          | 3.3            | 5.9            | 6.8         |         |
| 12        | 3177.03   | Cal-b fine sandstone | 7.3     | 0.032     | 1.06       | 5.59        | 13.15            | 13.9         | 62.0          | 4.1            | 20.1           | 0.0         | Upper sweet spot |
| 41        | 3482.2    | Siltstone | 14.3     | 1.009     | 1.87       | 5.85        | 14.01            | 26.6         | 61.8          | 7.8            | 2.1            | 0.0         |         |
| 43        | 3484.00   | Silty dolomite | 7.7     | 0.038     | 1.23       | 5.40        | 14.46            | 11.9         | 15.3          | 72.8           | 0.0            | 0.0         |         |
| 52        | 3494.75   | Fine siltstone | 8.6     | 1.42      | 6.34       | 17.70       | 28.0             | 59.1         | 0.0            | 12.9           | 0.0            | 0.0         | Lower sweet spot |
| 8         | 3487.33   | Dol-b arg siltstone | 12.6    | 0.036     | 2.53       | 7.09        | 27.08            | 18.8         | 45.0          | 12.4           | 14.8           | 9.0         |         |
| 26        | 3697.70   | Dol-b arg siltstone | 19.7    | 0.374     | 3.92       | 20.34       | 60.39            | 27.5         | 51.7          | 14.8           | 3.8            | 2.3         |         |
| 28        | 3699.70   | Dol-b arg siltstone | 16.7    | 0.077     | 3.53       | 12.47       | 44.22            | 26.7         | 56.1          | 15.0           | 0.0            | 2.3         |         |
| 31        | 3706.22   | Dol-b fine siltstone | 14.9    | 0.039     | 3.29       | 13.05       | 39.67            | 26.2         | 48.5          | 20.6           | 0.0            | 4.7         |         |
| 35        | 3687.50   | Dol-b siltstone | 11.2     | 0.055     | 3.1        | 7.34        | 32.47            | 31.5         | 48.1          | 20.4           | 0.0            | 0.0         |         |
| 37        | 3695.7    | Arg siltstone | 16.8     | 1.582     | 3.23       | 14.33       | 43.42            | 20.5         | 71.6          | 7.9            | 0.0            | 0.0         |         |
| 47        | 3647.37   | Dolomitic mudstone | 11.8    | 0.021     | 2.69       | 10.10       | 32.30            | 14.5         | 26.5          | 36.1           | 17.9           | 3.5         |         |
| 51        | 3641.32   | Dol-b silty mudstone | 15.1    | 0.050     | 3.11       | 11.49       | 40.11            | 24.7         | 41.5          | 28.1           | 0.0            | 4.4         |         |
| AVG       | 13        | 0.278     | 2.59      | 9.84      | 30.91      | 23.5         | 48.9             | 18.7         | 6             | 2.5            |               |             |         |

Notes: Cal-b: calcite-bearing; Dol-b: dolomite-bearing; arg: argillaceous; Por.: porosity; Per.: permeability; AVG: average.
The TOC of unwashed oil samples ranges from 1.06 wt% to 3.92 wt%, with an average of 2.59 wt%. Dolomite-bearing siltstone and mudstone have a relatively high TOC. $S_1$ of the sample ranges from 5.4 mg/g to 20.34 mg/g, with an average of 9.84 mg/g. $S_1 + S_2$ ranges from 13.15 mg/g to 60.39 mg/g, with an average of 30.91 mg/g. Porosity positively correlates with TOC, $S_1$, and $S_1 + S_2$, suggesting that the better the physical properties of the sample, the better the oil-bearing property.

### 3.2. Pore Types

Based on the casting thin sections and AIP-SEM images, six types of pores were distinguished, including residual intergranular pores, intergranular dissolved pores, intragranular dissolved pores, intracrystalline pores, clay-related pores, and organic matter pores. Among them, intergranular dissolved pores, intragranular dissolved pores, and intracrystalline pores are the most developed. Dissolved pores are the most important pore types in Lucaogou shale oil reservoirs. The primary cause of dissolved pore formation is the dissolution of unstable minerals (such as feldspar, carbonate minerals, and rock debris) due to acid fluid [59]. Intergranular dissolved pores dissolve from the grain edge toward the grain center (Figure 3(b)). They form in the shape of an embayment and are commonly greater than 25 $\mu$m.
μm in size. Intragranular dissolved pores are mostly elliptical and densely distributed throughout the granules or minerals (Figures 3(c) and 3(d)), and individual pores are primarily less than 1 μm in size. Filamentous illite cements can be seen in the vicinity of feldspar dissolved pores (Figure 3(d)) [60]. A large number of intercrystalline pores are developed between crystals such as dolomite and clay minerals (Figure 3(e)). Intercrystalline pores are more densely distributed, and the apertures are commonly less than 100 nm.

Residual intergranular pores are the residual of primary intergranular pores [61]. Mineral crystals such as secondary quartz and dolomite are common, but chlorite is rarely visible (Figures 3(a), 3(c), and 3(e)). Intergranular pores are dispersed, but the pore size is greater than 1 μm. Their existence indicates that the rock has strong ability to resist compaction and weak cementation [62]. Clay minerals develop multiscale pores (Figures 3(d) and 3(f)), including interlamellar cracks between clay particles (>100 nm) to intergranular pores between clay minerals (10 nm to 100 nm) [60]. However, due to low clay mineral content (Table 1), the contribution of clay-related pores is relatively weak. The TOC of the Lucaogou Formation is high, but maturity is low [63]. Organic pores, which are primarily organic matter margin shrinkage cracks, are sporadically developed due to the low organic matter maturity in the Lucaogou Formation (Figure 3(e)).

3.3. Pore-Throat Structure and Size Distribution. The size distribution and connectivity of the pore-throat can be analyzed using HPMI. The shale oil samples can be divided into three types based on the mercury injection curves (Figure 4). Type I is characterized by low displacement pressure (less than 5 MPa) and a "weak platform-shaped" intrusion curve (Figure 4(a)). In other words, mercury injection begins at low pressure, and with increasing pressure, mercury injection volume shows a concave shape, which is fast at first and then slows. The pore-throat size distribution of type I samples (e.g., sample 41) shows multiple peaks, in which...
the right peak is significantly higher than the other peaks and is around 100 nm to 2 μm (Figure 5). The casting thin section and AIP-SEM images show numerous intergranular pores and intergranular dissolved pores (Figure 3(a)), showing a combination of “larger pores connected with narrow throats” [60]. The mercury withdrawal efficiency of type I samples is commonly greater than 30%, indicating that increased connectivity correlates to a smaller pore-throat ratio. For type II samples, displacement pressure is around 10 MPa, and the mercury injection curve shows a straight line with a gentle slope (Figure 4(b)). For a small interval of the mercury injection pressure, the mercury injection saturation increases rapidly, indicating that the pore-throat type is relatively single. AIP-SEM images show that type II samples primarily contain dissolved pores (Figure 3(b)), and the combined pore-throats have a short, ducted shape. The pore-throat distribution of type II samples (e.g., sample 47) is primarily unimodal, and the peak is around 100 nm (Figure 5). Mercury withdrawal efficiency ranges from 20% to 40%, indicating a relatively large pore-throat ratio. (3) Type III samples are characterized by higher displacement pressure (greater than 10 MPa) and an upward convex-shaped injection curve (Figure 4(c)). Type III samples contain numerous intercrystalline pores and a small amount of intragranular dissolved pores. Also, the pore-throat ratio is relatively large, leading to lower withdrawal efficiency (less than 20%). The pore-throat distribution of type III samples (e.g., sample 43) is primarily unimodal, and the peak ranges from 10 nm to 100 nm (Figure 5).

With increasing physical properties, the mercury injection types gradually change from type III to type I, indicating that the pore-throat becomes larger and the proportion of intergranular pores and intergranular dissolved pores increases.

3.4. Spontaneous Imbibition and NMR Experimental Results. Figure 6 shows the imbibition curves for brine imbibition and oil imbibition of the 13 samples. The horizon ordinate is the cumulative imbibition height, which is the ratio of imbibition volume to rock surface area, representing fluid imbibition volume per unit of rock surface [55]. Both brine and oil cumulative imbibition curves initially increase rapidly and with the increase rate slowing near the end (Figure 6). The imbibition height change for oil is more pronounced compared to brine; it gradually slows down after approximately 380 min, and that of brine slows down after approximately 760 min.

The final oil imbibition proportion (the ratio of oil imbibition volume to total pore volume calculated by porosity and density) after 48 h varies from 19.6% to 75.1%, with a mean value of 51.7%, and the average proportion of the final brine imbibition after 72 h is 26.5% (Table 2), which is significantly less than that of oil. The oil and brine imbibition shows that the mixed shale oil samples from the Lucaogou Formation have mixed wettability. Both oil-wet pores and water-wet pores are developed. Overall, the oil-wet pores are more developed.

Figure 7 shows the comparison of $T_2$ distributions of samples after brine and oil imbibition, eliminating the interference of NMR signals of dry samples and centrifuged samples, respectively [45]. The distribution of corresponding $T_2$ spectra after brine and oil imbibition is divided into three categories: (1) The $T_2$ spectral amplitudes of brine and oil imbibition are approximately equivalent, showing a wide range, such as in samples 41 and 12 (Figure 7). The oil and brine had a similar imbibition volume, and the larger pores with $T_2$ (greater than 1 ms) were both water-wet and oil-wet. These samples were primarily siltstone with the lowest dolomite content (Table 1). (2) The $T_2$ spectral amplitude
of oil imbibition was significantly higher than that of brine imbibition, in which the $T_2$ distribution range of brine imbibition was completely covered by that of oil imbibition, such as samples 8, 10, 31, 35, and 52. The imbibition volume of oil was significantly higher than that of brine. The small pores could both imbibe oil and brine, and the larger pores primarily imbibed oil. Most of them corresponded to type II mercury injection characteristics (Figures 4(b) and 5(b)), and the dominant pore types were dissolved pores and dolomite intercrystalline pores, indicating that smaller dolomite intercrystalline pores were oil-wet. (3) The third type is similar to the second type, but the position of the $T_2$ spectra of oil imbibition was significantly to the right of the brine imbibition spectra, such as samples 26, 28, 37, 43, 47, and 51, indicating that the smaller pores were primarily water-wet, while the larger pores were primarily oil-wet. Type I and type II mercury injection curves were developed in this type of samples, indicating that intergranular dissolved pores were more developed. Rock composition and pore structures both affected the volume and distribution of brine and oil imbibition.

Figure 8 shows the change characteristics of $T_2$ spectra during brine imbibition and oil imbibition, illustrating the relative pore size changes during brine and oil imbibition.

### Table 2: Imbibition characteristics, wettability index, and type of pore-throat system of mixed shale oil samples.

| Simple ID | Slop (brine) | Slop (oil) | Pore volume (brine) (%) | Pore volume (oil) (%) | Wettability index | Pore-throat system | Section       |
|-----------|--------------|------------|-------------------------|-----------------------|------------------|--------------------|---------------|
| 41        | 0.29         | 0.21       | 63.3                    | 19.6                  | 0.38             | I                  | Upper sweet  |
| 12        | 0.27         | 0.36       | 40.8                    | 37.0                  | -0.30            | II                 | Upper sweet  |
| 52        | 0.25         | 0.43       | 56.2                    | 60.7                  | -0.73            | II                 | Upper sweet  |
| 10        | 0.32         | 0.37       | 36.5                    | 57.9                  | -0.70            | III                | Upper sweet  |
| 43        | 0.44         | 0.41       | 46.5                    | 59.3                  | -0.58            | III                | Upper sweet  |
| 26        | 0.34         | 0.32       | 4.9                     | 75.1                  | -0.87            | I                  | Lower sweet  |
| 37        | 0.39         | 0.44       | 6.8                     | 38.5                  | -0.81            | I                  | Lower sweet  |
| 51        | 0.28         | 0.37       | 14.4                    | 52.1                  | -0.84            | II                 | Lower sweet  |
| 8         | 0.35         | 0.39       | 22.5                    | 54.7                  | -0.95            | II                 | Lower sweet  |
| 28        | 0.36         | 0.41       | 12.6                    | 55.1                  | -0.96            | II                 | Lower sweet  |
| 31        | 0.37         | 0.38       | 14.2                    | 55.1                  | -0.96            | II                 | Lower sweet  |
| 35        | 0.27         | 0.41       | 9.1                     | 46.2                  | -0.90            | II                 | Lower sweet  |
| 47        | 0.34         | 0.38       | 16.1                    | 61.3                  | -0.95            | II                 | Lower sweet  |
| AVG       | 0.33         | 0.37       | 26.5                    | 51.7                  |                  |                    |               |

**Notes:** AVG: average.
Both samples 35 and 37 are analyzed as examples. Sample 35 belongs to the second type of imbibition characteristics. The oil imbibition $T_2$ distribution is significantly wider than that of brine, and some of the smaller pores can also imbibe oil. Sample 37 belongs to the third type of imbibition characteristics. After smaller pores are occupied by brine, the sample is practically unable to imbibe oil. The peak $T_2$ spectral value of brine imbibition mixed shale oil samples is commonly less than 1 ms, and that of oil imbibition is largely distributed between 1 ms and 10 ms, indicating that the oil-wet pores are more abundant than the water-wet pores. During imbibition, the $T_2$ peaks of water and oil imbibition remain in the same range, with only a slight offset to the right, indicating that imbibition of oil and water occurs in different pores, and the imbibition mode is a piston type, which confirms the mixed wettability of the mixed shale oil samples.

4. Discussion

4.1. Imbibition Slope and Pore Connectivity. In a dual logarithmic coordinate system, the brine imbibition curves of mixed shale samples are primarily distributed in a one-stage form (Figure 9), indicating that it has primarily experienced the imbibition stage. Oil imbibition curves show a two-stage form, representing both the imbibition stage and the diffusion stage. According to previous research results, the slopes of the imbibition stage were calculated (Table 2), which can reflect the internal pore connectivity of mixed shale oil reservoirs [55]. The diffusion front of rocks with good pore connectivity is advanced along the square root direction of time, while the front of rocks with sparse pore connectivity is advanced along a quarter direction of time [64]. In other words, the closer the imbibition slope is to 0.25, the worse the pore connectivity is. The closer the
imbibition slope is to 0.5, the better the pore connectivity is. Imbibition slopes of 13 mixed shale oil samples are less than 0.5 (Table 2), indicating that microcracks or clay hydration are not developed in the samples. Brine imbibition slopes range from 0.25 to 0.44, with an average of 0.33, and oil imbibition slopes range from 0.21 to 0.44, with an average of 0.37, suggesting that the connectivity of oil-wet pores is generally better than that of water-wet pores.

In this experiment, the imbibition height and the imbibition slope of mixed shale samples generally match (Figures 7 and 9). However, some of the overall oil-wet mixed shale samples, such as sample 26, have a lower oil imbibition slope and a higher brine imbibition slope (Table 2). Sample 26 belongs to the third type of imbibition (Figure 10). $T_2$ distribution corresponding to water-wet pores ranges from 0.1 ms to 1 ms, while that for oil-wet pores is wider, ranging from 0.1 ms to 50 ms, but the primary range is from 1 ms to 50 ms. This indicates that the main oil-wet pores are much larger than the water-wet pores. Furthermore, the oil (or brine) imbibition is synchronized in all the oil-wet (or water-wet) pores (Figure 8). The capillary force of a large pore is smaller than that of a small pore according to the Washburn equation [65]. As a result, the oil imbibition slope of sample 26 is lower than that of brine. Therefore, the imbibition slope is not only affected by the pore connectivity but also related to the size of pore-throats involved in the imbibition.

4.2. The Characteristics and Influence Factors of Wettability

4.2.1. Wettability Index. The wettability index ($I_w$) of each sample is obtained by combining wettability discrimination formula (5) [28, 49]. The relationship between the imbibition mass and the $T_2$ spectral amplitude for oil and water is established, respectively; then, the amplitude, measured using NMR, can be transformed into the pore volume.

$$I_w = \frac{NMR(S_w) - NMR(S_{\text{do}})}{NMR(S_w) + NMR(S_{\text{do}})}$$

where $NMR(S_w)$ and $NMR(S_{\text{do}})$ are the ratio of pore volume of brine imbibition and oil imbibition to the bulk pore volume, respectively, as measured by the helium porosity.

The wettability index $I_w$ varies from -1 to 1. -1 and 1 mean complete oil-wet and water-wet, respectively, while 0 means intermediate-wet. A value between -0.5 and 0.5 commonly indicates mixed wet. A value between -0.5 and -1 indicates strong oil-wet, and a value between 0.5 and 1 indicates strong water-wet [35].
The mixed shale oil reservoir of the Lucaogou Formation is dominated by strong oil-wet (e.g., samples 8, 31, and 47) (Table 2). Some samples are oil-wet-dominated mixed wet (e.g., samples 12 and 43), and only sample 41 is water-wet-dominated mixed wet. $I_w$ of the upper sweet spot section ranges from -0.7 to 0.4, with an average value of -0.4. $I_w$ of the lower sweet spot section ranges from -1 to -0.8, with an average value of -0.9, indicating that shale oil reservoirs in the lower sweet spot section are more oil-wet. In terms of lithology, the $I_w$ of siltstone ranges from -0.8 to 0.4, with an average value of -0.4. $I_w$ of dolomite-bearing siltstone ranges from -1 to -0.9, with an average of -0.9. $I_w$ of mudstone ranges from -0.9 to -0.7, with an average of -0.8. $I_w$ of dolomite is -0.6, indicating that siltstone is more water-wet than other lithologies.

4.2.2. Influence Factors of Wettability. The original sedimentary rocks were largely water-wet originally, and the present strong oil-wet state is caused by the change in the wettability of the mineral surfaces and crude oil charging. Therefore, the wettability of mixed shale oil reservoirs should be closely related to crude oil charging and mineral composition [59]. In the early diagenetic stage, organic matter is immature to semimature [8], and primary inorganic pores are the primary pore type. Water-wet pathways with good connectivity were initially present, making the reservoir primarily water-wet [66]. At present, the mixed shale oil reservoir of the Lucaogou Formation is in the middle stage of diagenesis, and the organic matter is low mature to mature [8]. Compaction decreases the amount of primary inorganic pores, and the heavy components generated by the cracking of organic matter change the wettability of the rock surface. Secondly, the cracking of organic matter forms organic matter pores and produces the oil-wet pathway, allowing for oil-wet and mixed wet to occur.

$S_1$ represents the amount of residual hydrocarbon, which can directly reflect the oil content of shale [67]. TOC is an important indicator of oil production capacity and is closely related to $S_1$ [67]. $S_1 + S_2$ represents the amount of hydrocarbon in the reservoir [68]. Therefore, the combination of TOC, $S_1$, and $S_1 + S_2$ can reflect the oil-bearing property of mixed shale. $I_w$ negatively correlates with TOC, $S_1$, and $S_1 + S_2$ (Figures 11(b)–11(d)), indicating that there is a good correlation between wettability and oil-bearing property. The better the oil-bearing property, the more oil-wet the mixed shale oil reservoir is. Heavy components of

![Figure 11: The relationship of wettability index with TOC, $S_1$, $S_1 + S_2$, and dolomite content.](http://pubs.geoscienceworld.org/gsa/lithosphere/article-pdf/doi/10.2113/2021/9190823/5437031/9190823.pdf)
crude oil are more likely to precipitate on water-wet pore surfaces, altering the wettability of the minerals [69, 70]. Therefore, the existence of crude oil charging is key to changing the wettability of the mixed shale oil reservoir. A good oil-bearing property indicates that a large amount of crude oil charging has occurred.

For example, sample 41 (water-wet-dominated mixed wet) has relatively low TOC, $S_1$, and $S_1 + S_2$ despite its relatively large porosity and pore-throat radius, indicating that the wettability of rocks has not changed obviously due to lack of hydrocarbon-generating matter or sufficient crude oil charging. In the mixed shale oil reservoirs of the Lucaogou Formation, large-scale crude oil charging cannot easily occur when the reservoir is far from the source rock [1]. Therefore, a better source-reservoir combination (e.g., integrated source-reservoir or adjacent source-reservoir) can facilitate wettability change. The shale oil reservoirs in the lower sweet spot section have better source-reservoir combinations compared to the upper sweet spot sections and therefore are more oil-wet (Table 2).

The impacts of pore-throat size on wettability are not obvious. For the upper sweet spot section samples, type I, type II, and type III pore-throat systems are all developed. Among them, the $I_w$ of type II and type III is smaller than that of type I (Table 2). For samples in the lower sweet spot, the $I_w$ of the type II pore-throat system is lower than that of type I. In general, samples with type II and type III pore-throat systems are more oil-wet than those with type I, indicating that the pore-throat size has no significant effect on wettability due to the wide distribution range of pore-throats in type I pore-throat system samples.

The essence of the wettability change is the change in zeta potential [71, 72]. Oil has a negative surface potential [72]; therefore, if the mineral surface carries a positive charge, it will attract negatively charged oil, and the surface will be oil-wet. The formation water in the Lucaogou Formation is neutral to weakly alkaline ($\text{pH} 7$ to 8), and carbonate minerals have a positive charge when the $\text{pH}$ value exceeds 8 [73]. Therefore, the surface of carbonate minerals such as dolomite is more likely to attract oil molecules under the current formation conditions. $I_w$ and dolomite content show a “V”-shaped relationship (Figure 11(a)). When dolomite content is low, $I_w$ negatively correlates with dolomite, indicating that the oil-wet property is positively proportional to dolomite content. When dolomite content is greater than 30 wt %, $I_w$ positively correlates with dolomite content, indicating that dolomite content has a two-sided effect on wettability of oil-wet rocks. On the one hand, when dolomite content is relatively low, with increasing dolomite content, the oil-wet components in the pore space increase, and the rock becomes more oil-wet. On the other hand, when dolomite content is greater than a certain critical value, the pore space is gradually filled, the pore spaces become smaller, and the oil-wet property becomes worse.

5. Conclusions

(1) The mixed shale oil reservoirs of the Lucaogou Formation primarily contain intergranular dissolved, intragranular dissolved, and intercrystalline pores. Three types of mercury injection curves are formed including a weak platform shape (type I), gentle straight line shape (type II), and upward convex shape (type III), corresponding to intergranular-dominated, dissolution-dominated, and dissolution-intercrystalline-dominated pore-throat systems, respectively.

(2) The mixed shale oil reservoir of the Lucaogou Formation shows dual wet characteristics. Overall, more oil-wet pores are present, which primarily correspond to large pores ($T_2 > 1\text{ ms}$). Water-wet pores primarily correspond to small pores ($T_2 < 1\text{ ms}$). The connectivity of oil-wet pores is generally better than that of water-wet pores.

(3) Overall, the mixed shale oil reservoir is strongly oil-wet, with a small amount of oil-wet-dominated mixed wet and minor water-wet-dominated mixed wet, which is primarily developed in siltstones. The reservoirs in the lower sweet spot section of the Lucaogou Formation are significantly more oil-wet than those in the upper sweet spot section. Type II and type III pore-throat systems are more oil-wet than type I for the same source-reservoir combination.

(4) The wettability of mixed shale oil reservoir is primarily controlled by organic matter abundance, source-reservoir combination, and dolomite content. A high organic matter abundance or favorable source-reservoir combination (integrated source-reservoir or adjacent source-reservoir) is the prerequisite for the development of oil-wet pores. An appropriate dolomite content is beneficial for increasing the oil-wet property of the reservoir.

Data Availability

All data in this study are analyzed by the authors. Associated tests were performed at the Key Laboratory of Unconventional Oil and Gas Accumulation and Development of Heilongjiang Province, Northeast Petroleum University, and Key Laboratory of Deep Oil and Gas, China University of Petroleum (East China). We guarantee the authenticity and unrepeated use of data.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

Acknowledgments

This paper was financially supported by the National Natural Science Foundation of China (No. 41972139, No. 41922015, and No. 41602141) and Fundamental Research Funds for the Central Universities (No. 18CX02069A).

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