Study on Microscopic Distribution Characteristics of the Remaining Oil in Polymer-Flooded Reservoirs

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ABSTRACT: Polymer flooding is an effective enhanced oil recovery (EOR) technology used in Daqing Oilfield. Microscopic distribution of remaining oil in polymer-flooded reservoirs is more complicated in comparison with waterflooded reservoirs. In this paper, UV excitation, frozen section-laser confocal technology, and three-dimensional reconstruction technology were employed to investigate the distribution law and occurrence state of the microscopic remaining oil in polymer-flooded Daqing Oilfield. With these methods, the occurrence states of the microscopic remaining oil distribution in different washing degrees and displacement locations were analyzed, and the remaining oil distribution before and after polymer flooding was compared quantitatively. The changes and microscopic distribution characteristics of crude oil components in the process of polymer flooding were clarified, and the relationship between clay minerals and the microscopic remaining oil distribution was discussed. Based on the statistical analysis of experimental results, the remaining oil of the free state decreases gradually, while the remaining oil of the bound state increases as the washing degree increases. In addition, the remaining oil in the distributary line is more enriched than the mainstream line after polymer flooding. Compared with waterflooding, the remaining oil of the free state becomes more, while the remaining oil of the bound state becomes less after polymer flooding. The frozen section-laser confocal experimental results also indicate that the proportion and distribution characteristics of the remaining oil components have been changed, and heavy components increase while light components decrease in the polymer-flooded stage. This research performs the quantized characterization and detailed analysis of remaining oil systematically and lays the foundation for remaining oil prediction and potential tapping in polymer-flooded reservoirs.

1. INTRODUCTION

Polymer flooding is one of the most important enhanced oil recovery (EOR) technology, which can change the properties of the fluid and rock’s wettability, reduce the interfacial tension, and increase the water viscosity and mobility ratios compared with waterflooding.1−3 Druetta and Picchioni performed a two-dimensional model multiphase simulation in macroscale and microscale recovery processes to show that polymers can increase recovery up to 7.6 to 15.4% when compared to the traditional waterflooding techniques.4−5 Daqing Oilfield has entered the period of extra-high water cut period, and the remaining oil is dispersed with developmental difficulties such as low flooding efficiency and ineffective cycles after polymer flooding, and there is still a huge amount of remaining reserves after polymer flooding, so it is stringent to study the microscopic distribution characteristics of the remaining oil in polymer-flooded reservoirs.6−8 The distribution characteristics and occurrence states of remaining oil after polymer flooding are the basis of the potential tapping of remaining oil.

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At present, micromechanics analysis, physical simulation experiment, fluorescence analysis, medical CT, NMR, and confocal laser scanning microscopy methods have been carried out to study the microscopic remaining oil distribution after waterflooding and polymer flooding.\(^6\)\(^{-}\)\(^9\) Cheng has studied the remaining oil distribution in the horizontal and vertical directions with core membrane technology, showing that the behavior of remaining oil by polymer flooding is similar to that by water flooding. Shah has predicted the porosity and permeability of carbonate rocks from pore-to-core scale using medical CT, confocal laser scanning microscopy, and micro-CT, proving that micro-CT imaging cannot resolve the submicron resolution as compared to confocal imaging.\(^1\)\(^5\) Yue et al. used the microscopic visualization of physical models to study the distributions of the remaining oil ratio of the models during the different flooding stages quantitatively by gray image-processing technology.\(^1\)\(^6\) Zhou proposed an NMR interpretation method and a residual oil fluorescence analysis method for the mechanism of chemical flooding to enhance oil recovery.\(^1\)\(^7\) Cheng et al. used micro-CT and core displacement experiments to present the occurrence state of remaining oil under different flow rates.\(^1\)\(^8\)

However, the quantitative distribution law and influence factors of the remaining oil in different washing degrees and displacement locations in polymer-flooded reservoirs are not clear, neither the changes and distribution characteristics of crude oil components, after polymer flooding. Meanwhile, the effect of clay minerals on microscopic remaining oil distribution is less involved.

In this work, UV fluorescence excitation and frozen section-laser confocal experiments were applied to analyze the microscopic distribution characteristics of the remaining oil in polymer-flooded reservoirs. The microcosmic remaining oil can be divided into three categories containing 10 states according to the occurrence state of oil and water in pores, as shown in Section 2. The quantitative occurrence state of the microscopic remaining oil in different washing degrees and displacement locations after polymer flooding was investigated systematically, and the remaining oil distribution before and after polymer flooding was compared. The changes and microscopic distribution characteristics of crude oil components in the process of polymer flooding were analyzed, and the influence of clay minerals on the microscopic remaining oil distribution was investigated in Section 3. The main goal of this research is to clarify the microscopic distribution characteristics of the remaining oil after polymer flooding and indicate the direction and object of the potential tapping scheme of the remaining oil in polymer-flooded reservoirs.

2. EXPERIMENT AND METHODS

2.1. Materials and Instruments. The 92 cores adopted in this experiment were sandstone of the North Saertu block of Daqing Oilfield. These samples were saturated with crude oil at the beginning of the experiment. After polymer flooding by hydrolyzed polyacrylamide for 24 h, the front and back ends of the samples were intercepted for slicing analysis.

The low-temperature thermostat was DFY-1800 from Jinghua Instrument Co., Ltd., which was important for frozen section technology. The laser confocal microscope LSM 900 from Germany contained all optical microscope observation modes, as well as a high-precision confocal surface 3D imaging mode.

2.2. Experimental Methods. 2.2.1. Frozen Section Technology. The slicing process is a very important step for the laser confocal method. The thickness of the normal temperature slice method is 1 mm, whereas that of the frozen section can reach 0.03–0.05 mm. Frozen section technology can keep the original state of the fluid in samples and avoid the destruction of the initial state of oil and water distribution caused by the conventional sample section method; so, the microscopic state can be seen more accurately.

The steps for frozen slicing are listed in the following. First, the samples were frozen in liquid nitrogen and then sliced at a lower temperature to ensure that the original shape of the fluid in the pores of the sample was undamaged during this processing procedure. Second, these samples were air-dried at a lower temperature below 5 °C and cemented with glue. The last step was grinding the rock samples into thin slices to make the observation of the internal structures of the samples more convenient.\(^1\)\(^1\)

2.2.2. UV Excitation and Confocal Laser Scanning Microscopy Technology. Ordinary fluorescence makes use of blue light as the light source. There are obvious drawbacks of this method, such as unclear oil–water interface and grain edge and indistinguishable crude oil components. However, UV fluorescence excitation can distinguish between the oil and water boundaries and extract the characteristics of the pores, avoiding the problems of unclear fluorescence characteristics of oil and water.\(^9\) With the help of UV excitation, confocal laser microscopy, which is a new nondestructive multilayer morphological detection method, can not only observe the deep internal structure of the sample but also perform high-speed scanning and three-dimensional image reconstruction to reflect the 3D space.\(^8\)\(^,\)\(^1\)\(^9\) Figure 1 presents the schematic of confocal laser scanning microscopy (CLSM).

CLSM uses laser as a point light source to avoid the interference of scattered light and can detect plane to three-dimensional rock samples through multiband and multilayer tomography and receives laser excitation signals at different wavelengths.\(^1\)\(^9\) The confocal principle of CLSM is diagram-
matically presented in Figure 1. The light emitted by the laser passes through the illumination pinhole to form a point light source and then forms parallel light through the lens and dichromatic mirror. The laser can emit the specified wavelength of light that converges on the focal plane within the sample, and the wavelength of reflected light is longer than that of the incident light.\footnote{5,15} The reflected light can pass through the dichromatic mirror, refocus through the lens, and reach the detection pinhole, which is located in a plane that is conjugate to the focal plane of the objective lens.\footnote{5,15} Finally, it will be detected by the subsequent photomultiplier detector and imaged on the display. Laser confocal multiband detection technology and three-dimensional image reconstruction method are applied to detect and analyze the microscopic distribution of components of crude oil and clay minerals.

2.2.3. Three-Dimensional Image Reconstruction Technology. In a confocal system, the sample is imaged by a point light source. In order to obtain the complete structure information of samples, the incident spot is scanned point by point or line by line along the sample on the focal plane (XY axis) perpendicular to the optical axis of the microscope, and soon afterward, the image information scanned at each point is collected, stored, processed, and converted by a computer to synthesize a two-dimensional image of a certain size, which is the basic principle of scanning. The obtained two-dimensional image is actually a planar “slice” image of the sample in a small thickness. If multiple XY plane images with different z-axis positions are scanned at a certain distance along the optical axis (Z-axis) of the microscope, the three-dimensional structure images of samples in this scanning area can be obtained by computer processing “stacking”, namely three-dimensional reconstruction. With the laser confocal 3D reconstruction technology, the distribution of oil, water, and rock in the reservoir can be observed and studied in three dimensions.

With UV excitation, frozen section technology, and a self-developed image-processing software, LSCM can distinguish the occurrence states of the microscopic remaining oil clearly and quantify the content of the remaining oil of different occurrence states in different situations and locations. Meanwhile, different components in crude oil can be distinguished, and the interaction mechanism of organic and inorganic minerals can be studied, which is of great significance to analyzing the quantitative distribution and variation law of different components of remaining oil and the relationship of clay minerals and remaining oil after polymer flooding, providing reliable technical support for further development plans.

2.2.4. Morphology of Microscopic Remaining Oil in Polymer-Flooded Reservoirs. There are various types of microscopic remaining oil in polymer-flooded reservoirs. Different displacement mechanisms result in different distribution patterns and locations of the microscopic remaining oil. According to the occurrence state of oil and water in pores, which is affected by the distance between the remaining oil and rock surface, the microcosmic remaining oil can be divided into different categories. The ordinary fluorescence method can only describe the plane distribution characteristics of the rock sample surface; therefore, remaining oil after water flooding can be classified into four types, including cluster, throat, corner-shaped, and oil film, with this method.\footnote{8} Contrastively, the LSCM method can perform high-speed scanning and three-dimensional image reconstruction to reflect the 3D space of the samples; therefore, the microcosmic remaining oil can be divided into three categories containing 10 states by this method.\footnote{11} Hence, the LSCM method allows for the perception of finer details to analyze the law of microscopic remaining oil distribution characteristics than ordinary fluorescence.

The first one is the bound state of remaining oil adsorbed on the mineral surface, which can be subdivided into a thin film on the surface of the pore, particle adsorption, and slit shape. The second one is the semibound state of remaining oil, which is the outer layer of the bound state or far from the mineral surface, mainly in the forms of cornered shape, throat, and sediments in the center of the pore. The last one is the free state remaining oil, far away from the surface of the mineral, distributed as cluster, intergranular, intragranular adsorption, and light mist.\footnote{11}

2.2.4.1. Occurrence State and Mechanism of Bound Remaining Oil. As presented in Figure 2a, the remaining oil in the form of thin film on the surface of the pore is mainly distributed on the surface of the oil-wet pore channels of the rock particles. This type of remaining oil is difficult to be displaced by the shear stress of water because the rock is oil-wet and will be greatly reduced after polymer flooding because of the stripping effect of the polymer solution. The thickness of the oil film which is widely distributed varies with the physical properties of the rock particles.

The particulate-adsorbed remaining oil mostly occurs in locations with high contents of mud impurities or clay minerals. This kind of remaining oil is adsorbed on the surface of the particle, pore wall, and interststitial material, the strong adsorption capacity of which makes this kind of remaining oil unable to be flushed out easily, as shown in Figure 2b. The slit-shaped remaining oil in Figure 2c mostly exists in the layers with low maturity of detrital components and the mineral-dissolved pores of feldspar, mica, and other flaky minerals as well as the cracks in the flaky minerals of kaolinite and chlorite particles.

2.2.4.2. Occurrence State and Mechanism of Semibound Remaining Oil. Figure 3a shows that corner-shaped remaining...
oil is mainly distributed in the unconnected parts of pores and occurs in the nooks and crannies of the complex pore space. One side of the corner-shaped remaining oil is attached to the contact angle of particles, and the other side is the free state in an open space. This type of remaining oil is pulled and stripped by polymer molecules and displaced partly in the stage of polymer flooding.

Throat-like remaining oil (Figure 3b) mainly occurs in a bound state by capillary action and remains in the small throat connected with the pores and curved throats formed in mosaic formation where clastic particles are in close contact. This type of remaining oil is widely and continuously distributed in small pores and medium- or low-permeability parts that are not swept by water. It is also widely distributed in weak washed and unwashed cores with poor physical properties but only narrowly distributed in throats in medium or strong washing cores.

As displayed in Figure 3c, the remaining oil in the form of throats and sediments in the center of the pore is made up of colloid and asphaltene with macromolecule and high viscosity and precipitates in the center of the pore. This type of remaining oil mainly exists in tiny pores and poorly connected pores, surrounded by compact particles; therefore only a small part of light components can be displaced by the polymer.

### 2.2.4.3. Occurrence State and Mechanism of Free Remaining Oil

As shown in Figure 4a, cluster-like remaining oil occurs in the pore space in the form of clusters, clumps, oil droplets, and even in a motion state. This type of remaining oil usually remains in the small throat pore clusters surrounded by large pore channels after water flooding and can form a stable oil silk channel after polymer flooding. A large amount of remaining oil is left in the pores in the polymer flooding stage.

The intragranular remaining oil (Figure 4b) is mainly distributed in intragranular pores in the layer where the maturity of clastic components is low and intragranular dissolved pores are developed. Due to diagenesis, it is formed in the dissolution pores of flake minerals such as feldspar and mica.

The remaining oil of intergranular adsorption (Figure 4c) is mainly distributed in places with a high content of clay minerals or mud base. Although this type of remaining oil is in open pores, it is still attached to the clay mineral surface and may be displaced by injected water, which may cause crystal fragmentation and deformation, and blocking the throat may even reduce the reservoir permeability and affect the recovery of remaining oil. It can be carried out with the flow of the reservoir fluid.

As displayed in Figure 4d, the remaining oil in the form of light mist is almost stripped away by displacement after sufficient and repeated scouring by the displacement fluid in severely water-flooded layers, where brown and yellow-brown remaining oils are adsorbed on the surface of some debris particles, and a small amount of dissolved hydrocarbons is distributed in a light mist.

### 3. RESULTS AND DISCUSSION

#### 3.1. Distribution Characteristics of Microscopic Remaining Oil Distribution

#### 3.1.1. Occurrence State of Microscopic Remaining Oil Distribution in Different Washing Degrees

Washing degree refers to the degree of core displacement by fluid injection in the process of flooding. Usually, it can be divided into three grades, including strong, medium, and weak washing degrees. The qualification method of washing degree is mainly based on the observation of cores and water cut and oil displacement efficiency. In this paper, if displacement efficiency is less than 35%, it can be judged as weak washing; if displacement efficiency is between 35 and 55%, it can be judged as medium washing; and if oil displacement efficiency is greater than 55%, it can be judged as strong washing.

On the whole, the microscopic remaining oil after polymer flooding presents a distribution pattern of overall dispersion and local enrichment, and different types of remaining oil show obvious regular distribution characteristics. Based on the statistical analysis of experimental results in Table 1, it is observed that with the increase of washing degree, the movable free state remaining oils such as intergranular adsorption and cluster shape gradually decrease, while the immovable bound...
state remaining oil such as pore surface film shape gradually increases. The specific manifestations are listed as follows.

Parts of the remaining oil between the grains of weakly washed samples are displaced out, but most remaining oil is still mainly in the free state in the form of cluster and intergranular adsorption and bound state in the form of thin film on the surface of the pore. Most of the remaining oil in the semibound state of the medium-washed sample is driven out, while the free state and bound state remaining oil account for 46.5 and 42.9%, respectively. Most of the remaining oil in the semibound state remaining oil of the strongly washed samples is displaced out, while the bound state remaining oil in the form of thin film on the surface of the pore is massively left. Meanwhile, small throats of strong water flooding degrees and poorly connected narrow throats can be swept. In strongly washed rock samples, flake minerals such as feldspar and mica are dissolved into the solution pores and gaps in the grain, and part of crude oil migrate into these solution pores during the process of displacement, causing a high proportion of the remaining oil in the grain.

3.1.2. Occurrence State of Microscopic Remaining Oil Distribution at Different Locations. Both geological factors and development factors can affect the microscopic distribution of remaining oil after polymer flooding, forming various types of the microscopic remaining oil. Geological factors mainly include structure, sedimentary microfacies, reservoir physical properties, and heterogeneity, while development factors mainly include injection—production relationship, degree of perfection, well pattern, and production measures. The development factor is the main external cause of microscopic remaining oil formation, and the geological factor is the internal cause. 30 For the polymer flooding well pattern with relatively uniform well pattern deployment, there is a great difference in microscopic remaining oil at different displacement sites after polymer flooding,3 as shown in Figure 5. The mainstream line is the flow line connecting the injection well and the production well center point, and the distributary line is the flow line between two producing wells that separates the seepage from left and right. In general, the macroscopic remaining oil is mainly distributed in the distributary line, while little remaining oil is left in the mainstream line due to serious flooding after polymer flooding. Remaining oil is also distributed in areas without diversion lines.

As presented in Table 2, the content and occurrence state of the microscopic remaining oil in different displacement positions change a lot after polymer flooding. The microscopic remaining oil in the distributary line is mainly in a bound state in the form of particle adsorption and thin film on the surface of the pore and in a free state in the form of clusters and intergranular adsorption. The bound state and free state remaining oil account for 25.2 and 71.8% respectively in the distributary line. The remaining oil in the mainstream is mainly in the form of particle adsorption, intergranular adsorption, and cluster. The total amount of free state remaining oil is only 49.5% in the mainstream, which is 22.3% lower than that in the distributary line, while the bound state remaining oil in the mainstream is 20.7% higher than that in the distributary line. On the whole, the free state microscopic remaining oil in the distributary line is more than about 20% higher than that in the mainstream line, mainly in the form of clusters and intergranular adsorption states, which is the main object of potential tapping and injection—production relationship adjustment of the remaining oil after polymer flooding.

3.1.3. Comparison of Microscopic Remaining Oil Distribution before and after Polymer Flooding. Comparing the confocal laser images before and after polymer flooding in Figure 6, it can be seen that the oil displacement effect of polymer flooding is better than that of water flooding. After polymer flooding, the proportion of all kinds of microscopic remaining oil decreases, and the main remaining oil types are pore surface film, particle adsorption, and intergranular

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Table 1. Proportion of Different Occurrence States of the Microscopic Remaining Oil in Three Washing Degrees

| washing degree | weak | medium | strong |
|----------------|------|--------|--------|
| bound state    | 18.3 | 30.4   | 58.0   |
| semibound state|      |        |        |
| particle adsorption | 2.5 | 10.5  | 2.3    |
| slit shape     | 0    | 2.0    | 7.0    |
| corner-shaped  | 12.0 | 6.0    | 4.0    |
| throat         | 4.4  | 2.5    | 0.5    |
| sediments in the center of the pore | 0.1 | 2.1 | 4.3 |
| free state     | 39.0 | 28.0   | 5.0    |
| cluster        | 0.2  | 7.3    | 10.0   |
| intergranular  | 23.5 | 8.0    | 3.9    |
| intragranular adsorption | 0 | 3.2 | 5.0 |
| light mist     | 0    | 3.2    | 5.0    |

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Table 2. Proportion of Different Occurrence States of Microscopic Remaining Oil at Different Locations

| displacement location | mainstream line | distributary line |
|-----------------------|-----------------|-------------------|
| bound state           |                 |                   |
| thin film on the surface of the pore | 9.7 | 7.0 |
| particle adsorption   | 35.1            | 16.2              |
| slit shape            | 1.1             | 2.0               |
| corner-shaped         | 2.2             | 1.1               |
| throat                | 1.9             | 1.6               |
| sediments in the center of the pore | 0.5 | 0.3 |
| free state            |                 |                   |
| cluster               | 21.5            | 37.8              |
| intergranular         | 2.1             | 0.4               |
| intragranular adsorption | 23.7 | 30.1 |
| light mist            | 2.2             | 3.5               |
adsorption, accounting for more than 30% of the total remaining oil. The remaining oil of these three types decreases more obviously after polymer flooding than after waterflooding. There is no obvious change in the overall proportion of particle and interparticle remaining oil, indicating that polymer flooding has no effective effect on these two types of remaining oils, which is the main target of potential tagging in the next step. Compared with waterflooding, in polymer flooding, the bound state remaining oil is relatively less, the relative proportion of free state is more, while the semibound state remaining oil changes a little bit, as shown in Figures 7 and 8.

3.2. Changes and Distribution Characteristics of Crude Oil Components after Polymer Flooding. The recovery of polymer-flooded reservoir is related to sweeping efficiency and displacement efficiency, which can be expressed as:

$$\eta = E_D E_V$$

where $\eta$ is the recovery, %; $E_D$ is the oil displacement efficiency, %; and $E_V$ is the sweep efficiency.

The main components of crude oil are oil, colloid, asphaltene, and carbonaceous. Due to the surface activity of the interphase, heavy components of crude oil can adsorb to the rock surface, make the rock hydrophobic, and form interfacial layers with high viscosity and shear elasticity, resulting in a large portion of the remaining oil left in the formation in the form of film and decrease of oil displacement efficiency, sweep efficiency, and oil recovery.

3.2.1. Changes of Light and Heavy Components in Different Washing Conditions after Polymer Flooding. Based on the laser confocal scanning analysis results of water-washed samples from the polymer flooding reservoirs in Daqing Oilfield, the ratio of light and heavy components with different water cuts can be accurately obtained. Hydrocarbons present fluorescence characteristics. Fluorescence wavelengths of 510–600 nm can receive light components, while fluorescence
wavelengths of 600−800 nm can receive heavy components in crude oil such as colloid and asphaltene. Therefore, different components and viscosity of hydrocarbon can display different images by laser scanning confocal technology clearly, so CLSM can be used to observe the distribution of different organic components of crude oil quantitatively.

As displayed in Figure 9, the light components decrease, while the heavy components increase when the washing degree increases after polymer flooding, and the ratio of light to heavy components decreases rapidly from 1.20 to 0.9−0.66, which is caused by the pore structure. In the weak and medium water-washing stages, pores in the free state of lightweight components and heavy components are displaced out at the same time, so the ratio of light to heavy components varies from 1.28 to 1.18, and the proportion of light and heavy components changes a little. Due to the hydrodynamic effect of polymer erosion, the light components on the pore surface are gradually separated, and most of the heavy components are absorbed on the surface and remain in formation with the increase of the water-washing degree. Therefore, this phenomenon is helpful to make targeted measures when adjusting the later development scheme.

3.2.2. Distribution Characteristics of Light and Heavy Components of Crude Oil in Microscopic Pores. By analyzing the microscopic three-dimensional images of remaining oil distribution utilizing laser confocal scanning, the distribution law of light and heavy components of crude oil in microscopic pores can be obtained, and the relationship between the remaining oil distribution and pore structure as well as clay minerals can be investigated. Based on the distribution of remaining oil in the strongly washed rock samples after polymer flooding (Figure 10), it is observed that the components of crude oil that contact the rock surface are mainly heavy components displayed in blue color, the distribution of which is wide and continuous. This is because the interphase of the heavy components such as colloid and asphaltene are surface-active and can be adsorbed on the surface of the rock, making the rock hydrophobic and become a high-viscosity and shear elastic interface layer. The intergranular light components are in the state of small flake and scattered continuously with local enrichment relatively. Meanwhile, the quantity of light components is less than the heavy components. There is a spot of light components, where slightly heavy components are distributed on the surface of clay minerals. However, the light components are dominant, and the heavy components are supplemented in a “cluster” symbiotic pattern in the solution pores, which may be caused by the migration of crude oil during polymer flooding.

3.3. Effects of Clay Minerals on the Distribution of Microscopic Remaining Oil. Clay mineral plays an important role in reservoir development. On the one hand, it can cement the skeleton particles of the reservoir; on the other hand, influenced by the physical and chemical effects due to the dynamic hydrodynamic geological process of reservoir development, the physical and chemical properties of the clay mineral evolve dynamically and can change its crystal lattice. Moreover, clay minerals can affect the pore throat network and
seepage characteristics of the reservoir and thus can influence and control the distribution of the microscopic remaining oil.

3.3.1. Content and Distribution Characteristics of Clay Minerals. The content and dispersion occurrence of clay minerals are the main factors affecting the microscopic distribution of the remaining oil. Meanwhile, clay content has an important effect on the polymer flooding efficiency, and higher clay content leads to higher polymer adsorption and low displacement efficiency. The swelling capacity of different clays also matters to the formation permeability, the impact of which should be considered to choosing the polymer with appropriate molecular weight in polymer flooding. Therefore, the content and dispersion occurrence of clay minerals play an important role in the design of polymer flooding schemes, including the choice of oil displacement agents, optimization of injection parameters, and so forth.

As shown in Table 3, clay minerals in the principal producing formation of Daqing Oilfield are mainly kaolinite with a content of more than 67%, followed by illite and chlorite with the content between 6 and 13%, while the contents of the mixture of illite and smectite and the mixture of chlorite and smectite are less than 8%, except in unwashed formation. Kaolinite which is a flake crystal aggregate and vermicular is affected by mechanical and chemical weathering and denudation of injected water in the long term, so its aggregate crystal frame is destroyed to form fine particles. Driven by the reservoir development fluid, these particles are easily transported out of the reservoir to the surface with the produced fluid, resulting in a decrease in the relative content of kaolinite in the formation, especially in the case of high injection production intensity. However, the clay content in the polymer flooding reservoir of Daqing Oilfield does not decrease with the increase of washing degree, but the relative content of kaolinite increases. The reason for this result is that the permeability of sandstone tends to increase significantly as the kaolinite content increases. Although some kaolinite in the reservoir will dissolve with the increase of injection intensity, the relationship between the kaolinite content and permeability does not change basically.

3.3.2. Relationship between Clay Minerals and Distribution of Microscopic Remaining Oil. The occurrence of clay minerals can be divided into three basic types, namely, filling type, padding type, and bridging type, which can be seen in the Daqing oilfield. They are generally distributed on the surface of particles, micropores in grains, and microchannels between grains. Illite, chlorite, and the mixture of illite and smectite are the main thin-film clay minerals on the grain surface, resulting in the reduction of the original intergranular pores in the reservoir. Similarly, bridging clay minerals at the microchannels between grains cause pore throat clogging and reduce reservoir permeability, which is one of the most important reservoir properties affecting the distribution of microscopic remaining oil.

Table 3. Clay Content of the Reservoir in Different Washing Degrees after Polymer Flooding

| washing degree   | permeability ($\mu m^2$) | clay content (%) | clay mineral relative content (%) |
|------------------|--------------------------|-----------------|----------------------------------|
| unwashed         | 0.035                    | 8.63            | illite 67                        |
| strong washing   | 0.887                    | 7.82            | kaolinite 13                     |
| medium washing   | 1.651                    | 6.55            | chlorite 10                      |
| weak washing     | 3.587                    | 6.05            | mixture of illite and smectite 2  |
|                  |                          |                 | mixture of chlorite and smectite 3|

After polymer flooding, the clay minerals on the particle surface are washed away by fluid, and heavy components such as colloid and asphaltene in the crude oil adhere to the particle surface, symbiotic with the thin-film clay minerals. As displayed in Figures 11 and 12, further away from the particle surface, the content of heavy components gets lower. The content of heavy components is high in the cracks that are filled with clay minerals while low in the cracks that are filled with polymers and formation water. The distribution of clay minerals is scattered at the micropores in grains and the microchannels in grains, symbiotic with light components. However, there is no crude oil in the fractures, and micropores are completely filled with clay minerals in the grains. The distribution characteristics of crude oil may be related to the surface activity of heavy components and the strong diffusion ability of light components.

4. CONCLUSIONS

This article focused on the occurrence, law, and changes of microscopic remaining oil, providing a scientific and reliable basis for the further exploitation and potential tapping of remaining oil in polymer-flooded reservoirs with the LSCM method. Based on the experiment and statistical results of image recognition by a self-developed software, the main conclusions are as follows:
(1) The distribution pattern of the microscopic remaining oil presents an overall dispersion and local enrichment in polymer-flooded reservoirs. The occurrence states of the remaining oil are mainly free state in weak water-washing degree condition, dominant bound state and complementary free state in medium water-washing degree condition, and bound state of the thin oil film on the particle surface in strong water-washing degree condition.

(2) After polymer flooding, the microscopic remaining oil at the distributary line is mainly in a bound state in the form of particle adsorption and thin film on the surface of the pore and in a free state in the form of clusters and intergranular adsorption. Meanwhile, the free state microscopic remaining oil at the distributary line is the main potential tagging objective of the remaining oil and the adjustment target of injection–production relationship after polymer flooding. Comparison results of the microscopic remaining oil distribution before and after polymer flooding indicate that polymer flooding has no effective effect on the proportions of particle and interparticle remaining oils, which are the main target of potential tagging in the next step.

(3) Changes and distribution characteristics of crude oil components after polymer flooding should be considered to take pertinent measures to maximize recovery. As the flooding degree increases, the proportion of light components decreases significantly, while the proportion of heavy components increases, especially when the reservoir enters the strong water-washing stage, and this phenomenon is more obvious. The heavy components are distributed on the surface of rock particles in a contiguous form, while the light components are distributed in the intergranular pores in the form of small flakes and in an overall scattered and locally enriched state.

(4) The content and dispersion occurrence of clay minerals are the main factors affecting the microscopic distribution of the remaining oil and play an important role in the design of polymer flooding scheme. The content of kaolinite accounts for more than two-thirds of clay minerals in the oil layer of the Daqing Oilfield. When the washing intensity increases, the clay content decreases, but the relative content of kaolinite increases after polymer flooding. Clay minerals and light components coexist in the micropores and microchannels between grains, while there is no crude oil in the fractures and micropores filled with clay minerals.

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**Notes**

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Abbreviations

EOR: enhanced oil recovery
UV: ultraviolet
CT: computed tomography
NMR: nuclear magnetic resonance
CLSM: confocal laser scanning microscopy
LSM: laser scanning microscopy
3D: three-dimensional
Co., Ltd.: company limited

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