Characterization and Origin Analysis of Heavy Oil in the Western Sag of the Liaohe Basin

Chenglong Ma,* Changhao Hu, Xingzhou Liu, Yugang Li, Jie Cui, Ying Wu, Jianhong Huang, and Shuming Li

ABSTRACT: This article systematically examines the physical characteristics, group composition characteristics, and geochemical characteristics of heavy oil in the Western Sag of the Liaohe Basin. The examination is based on the separation and quantitative analysis of crude oil and rock samples, as well as the analysis of test results from gas chromatography with saturated hydrocarbons and aromatic hydrocarbons. It also analyzes the generation mechanism and main controlling factors of heavy oil. The results show that heavy oil has low wax content (1.8–9.2%), a low freezing point (−19–38 °C), low sulfur content (0.28–0.5%), high colloid and asphaltene content, high density (0.926–1.008 g/cm³), and high viscosity (328–231910 mPa·s). The physical properties of the heavy oil in the same formation decrease from the depression’s edge toward its center and within the same area, and the physical properties in different formations also have an inverse relationship with burial depth. Biodegradation is the main reason for the formation of heavy oil. Based on the biodegradation degree, there are four types of heavy oil: undegraded, weakly degraded, moderately degraded, and severely degraded. The main controlling factors of biodegradation are temperature and the water environment. This study provides a method for studying the genetic mechanism of heavy oil, an approach for discovering similar genetic oil and gas, and a basis for the transformation of heavy oil field development.

1. INTRODUCTION

Heavy oil is an important oil reserve accounting for a significant amount of the oil and gas resources worldwide. Heavy oil is widely distributed throughout China’s numerous oil and gas basins. More than 70 heavy oil fields have been found from the Songliao Basin and the Bohai Bay Basin in the east to the Jungar Basin and the Tarim Basin in the west, and from the Erlian Basin in the north to the Pearl River Mouth Basin and the Sichuan Basin in the south.1,2 The current proven reserves are 2.06 billion tons, making heavy oil one of the most vital reservoir types in oil and gas field development. Moreover,3 the Liaohe Oilfield has the largest heavy oil production base in China.

The Liaohe Depression is located in the northeast of the Bohai Bay Basin. It is a NE-trending rift basin formed under the dual action of the Tanlu fault and mantle upper hole during the Mesozoic–Cenozoic. The three tectonic activity stages that contributed to the formation of the currently existing tectonic pattern are arch extension, rift, and depression.4 According to the characteristics of the structural units, it can be divided into six secondary structural units of “three protrusions and three depressions”, specifically three protrusions in the west, center, and east, and three depressions in the west, east, and Damintun (Figure 1). The Western Sag is the most important secondary negative structural unit in the Liaohe Depression, with an area of more than 2000 km². It is a long and narrow dustpan depression with a northeast strike and a southeast dip.5,6 It is recognized for its peaks in the north where it narrows, and valleys in the south where it widens. There is a fault in the east and an overlap in the west, and it is steep in the east and gentle in the west.5,6 The following developed strata occur from bottom to top: the Pre-Cenozoic basement, the Paleogene Fangshenpao formation, the Shahejie formation, the Dongying formation, the Neogene Guantao formation, the Minghuazhen formation, and the Quaternary Plain formation.7 The Western Sag is the most famous slope type in China that accumulates both oil and gas. In total, there are 773 oil and gas reservoirs. Both the reserves and production of oil and gas in the Western Sag play an important role.8 Heavy oil accounts for a significant proportion (more than 50%) of the proven oil geological reserves in the Liaohe Basin that are distributed in the Western Sag.9 Horizontally, the heavy oil is mainly distributed in the relatively high structure of the slopes on the
east and west sides of the depression. Vertically, heavy oil is distributed in many formations, such as the Paleozoic buried hill, the Mesozoic formation, the Paleogene Shahejie formation, and the Neogene Guantao formation. The burial depth of the reservoir is generally less than 2500 m, and the main oil and gas-bearing strata are the Shahejie formation and the Guantao formation. The establishment and evolution of heavy oil reservoirs require a lengthy geological process. Scholars from around the world have long debated the characteristics, genetic mechanisms, and main controlling factors of heavy oil in this area and have suggested that heavy oil in this area includes oil of both primary and secondary origin. However, the oil of secondary origin is predominant, which indicates that the thickening mechanism of the heavy oil mainly includes water washing, biodegradation, and oxidation.

The main objective of this article is to determine the physical characteristics and features of the group composition of heavy oil in the study area through the geochemical analysis of a large number of heavy oil samples. This article also analyzes the genetic mechanism and controlling factors of heavy oil. Additionally, it proves that biodegradation can lead to different degrees of changes in the group composition of crude oil, which is the main reason for the formation of heavy oil. This information will help in the identification of heavy oil and gas with similar genesis.

2. EXPERIMENTAL SECTION

The heavy oil samples were collected from different areas and different depths in the Western Sag, and the sampling depth was less than 2500 m. The relevant experiments were completed at the Liaohe Oilfield Exploration and Development Research Institute laboratory. First, group components of the heavy oil samples were separated by deasphalting, and then, the hydrocarbon and non-hydrocarbon components were analyzed by gas chromatography (GC) and gas chromatography–mass spectrometry (GC/MS). The instrument used for GC/MS analysis was an Agilent 6890GC-5975iMS (Palo Alto, CA), and the chromatographic column was an HP-5MS (60 m × 0.25 mm × 0.25 μm). The sample inlet temperature was 300 °C, the detector temperature was 310 °C, He was used as the carrier gas (99.999%), the carrier gas flow rate was 1.0 mL/min, and the non-split injection method was used. When heating up, the initial temperature was 80 °C, which was maintained for 0.5 min and then raised to 150 °C at a rate of 20 °C/min, and after that, raised to 310 °C at a rate of 3 °C/min and retained for 30 min. The ion source of the mass spectrometer was electron bombardment (EI), the ionization voltage was 70 eV, and the scanning frequency was 1.3 s. Full scan (m/z50–650) mode was adopted for chromatography–mass spectrometry, and a selective ion scanning mode was implemented for some characteristic ions (selected ion monitor: including m/z177, m/z191, and m/z217).
3. RESULTS AND DISCUSSION

3.1. Physical Properties of Heavy Oil. Density, viscosity, wax content, and freezing point are important physical properties of crude oil as well as important indicators of crude oil thickening. The analyzed crude oil samples were collected from different formations and different locations in the study area, and thus the physical properties of the heavy oil were defined based on statistical analysis. The density of heavy oil at 20 °C is generally distributed between 0.926 and 1.008 g/cm³. The viscosity of heavy oil at 50 °C is generally distributed between 328 and 231,910 mPa·s. The freezing point is generally distributed between −19 and 38 °C, the wax content is generally distributed between 1.8% and 9.2%, and the sulfur content is generally distributed between 0.28 and 0.5%. The physicochemical properties of the heavy oil in the Western Sag are characterized by “three lows and three highs”; that is, low wax content, low freezing point, low sulfur content, high colloid and asphaltene content, high density, and high viscosity.

There are obvious variations in the physical properties of heavy oil in the Western Sag (Figure 2). Horizontally (Table 1), the Shuguang and Huanxiling areas, located far from the center of the depression, account for a large quantity of extra-

![Figure 2. Relationship between physicochemical properties and depth of crude oil.](https://pubs.acs.org/doi/figure/10.1021/acsomega.2c02668.28987)

![Table 1. Statistics of Plane Variation of Heavy Oil Properties in Different Blocks in the Western Sag](https://pubs.acs.org/doi/figure/10.1021/acsomega.2c02668.28987)

| Distance from the center of the depression | Block | 20°C Density (g/cm³) | 50°C Viscosity (mPa·s) | Solidifying point (°C) | Wax content (%) | Gum and asphaltene content (%) |
|------------------------------------------|-------|---------------------|------------------------|------------------------|----------------|-----------------------------|
| far                                      | Shuhuan | J612 0.9995 10224.00 20.00 | 2.58 40.50            |            |
| far                                      | H17 0.9666 1058.33 -6.57 | 3.03 30.81            |            |
| near                                     | J16 0.9315 365.65 -19.00 | 3.12 20.82            |            |

Note: *Crude oil viscosity under reservoir conditions. Those without * are the viscosity of degassed crude oil at oil layer temperature.
heavy oil and super-heavy oil, and the density is generally distributed between 0.980 and 1.000 g/cm³. The Lengjia and Gaosheng areas are located near the center of the depression and mainly contain ordinary heavy oil and extra-heavy oil, and the density is generally distributed between 0.934 and 0.966 g/cm³. Vertically (Table 2), where the depth is less than 1000 m, there is mainly extra-heavy oil and super-heavy oil. When the depth is between 1000 and 1500 m, there is mainly ordinary heavy oil and extra-heavy oil. When the depth is more than 1500 m, there is mainly thin oil and ordinary heavy oil. Generally speaking, the density, viscosity, and freezing point of crude oil in the same layer decrease gradually from the edge of the depression to the center. The density, viscosity, and freezing point of heavy oil in different layers within the same region also decline with an increase in burial depth. The wax content and colloid plus asphaltene content increase with an increase in burial depth (Figure 2).

3.2. Group Composition Characteristics of Heavy Oil.
The physical properties of heavy oil are a reflection of its chemical composition. The group composition of heavy oil results from the effects of integrated actions, which include the composition of organic facies, thermal evolution and reservoir physicochemical processes during oil and gas migration, and the accumulation and preservation of source rocks in sedimentary basins. Therefore, it is an important basis for understanding the origin, type, reservoir forming conditions, and distribution law of heavy oil. Compared with normal crude oil, heavy oil has higher colloid and asphaltene content, but the content of saturated hydrocarbons is significantly reduced. The group composition of heavy oil is controlled by reservoir depth (Figure 3). As the depth increases, the saturated hydrocarbon content gradually increases, the colloid and asphaltene content decreases drastically, and the aromatic hydrocarbon content also decreases.

The density, viscosity, and freezing point are all affected by the chemical composition of the crude oil. Based on the statistical analysis of the relationship between the density, viscosity, and freezing point of the crude oil in the Liaohe Basin and its wax content and colloid and asphaltene content (Figures 4–6), the density gradually increases with an increase in the colloid and asphaltene content, and the freezing point gradually increases as the wax content increases. The initial boiling point of crude oil also shows a clear correlation with the chemical composition of crude oil and increases as the colloid and asphaltene content rises. The correlation between the wax content and the colloid and asphaltene content is also clearly noticeable, where an increase in colloid and asphaltene content leads to a reduction in wax content, exhibiting a clear negative correlation.
These observations show that the density and viscosity of heavy oil are mainly associated with the content of polar heteroatomic compounds such as colloids and asphaltene. In the process of secondary origins, the light components such as alkanes are gradually depleted, while the heavy components such as polar NSO heteroatomic compounds with high molecular weight are gradually concentrated, and the strong polarity enhances the mutual attraction between molecules, clay minerals, and water molecules. Finally, heavy oil with a high density and high viscosity is formed. The freezing point is mainly correlated with the wax content because wax is a high-density and high-viscosity material. The freezing point is also affected by the molecular weight, which is gradually concentrated, and the strong clay minerals, and water molecules. Finally, heavy oil with a high density and high viscosity is formed.

Biodegradation also influences the physicochemical properties of heavy oil, especially the colloid and asphaltene content. Thus, biodegradation is one of the important formation mechanisms of heavy oil. Many scholars believe that oxidation and water washing are also heavy oil formation mechanisms. However, oxidation is completed with the help of microorganisms, and the main role of water washing is to provide microorganisms with activity carriers and nutrients needed for survival. Therefore, the root of these formation mechanisms is biodegradation. Most of the heavy oil in the Western Sag has endured biodegradation to varying degrees. The concentration of saturated hydrocarbons decreases due to biodegradation. This results in an increase in the colloid and asphaltene content, as well as an increase in its density and viscosity.

Different crude oil components have different anti-biodegradation abilities. Consequently, the chemical composition of crude oil varies with different degrees of degradation. The degradation of alkane components by microorganisms can range from C1 (methane) to C35 (including steroidal terpenes). As shown in Table 3, once the degradation is enhanced, the oil components change accordingly. With the intensification of biodegradation, the changes in various crude oil biomarkers reflect the anti-biodegradation ability of different compounds: n-alkane < single branched alkane, alkyl cyclohexane, and alkyl cycloalkane < isomonoalkane < C14–C16 bicycloalkane < normal sterane < rearranged sterane < pentacyclic triterpenes < tricyclic terpene. This represents the biodegradation series of weak degradation → medium degradation → severe degradation.

3.3.1. Undegraded Heavy Oil. Most of the undegraded heavy oil samples are ordinary heavy oil, with a relatively low density, generally less than 0.94 g/cm³. A complete series of normal alkanes and isoalkanes are developed and the distribution characteristics of steranes, terpanes, and sesqui-terpanes are normal. The steranes are mainly C27–C30 regular steranes and 4-methylsteranes, except for the rearrangement of 13α (β) (H), 17α (β) (H), and C21 and C23 steranes (Figure 7A). This kind of heavy oil is the product of short-distance migration and accumulation of crude oil with high colloid and asphaltene content.

3.3.2. Weakly Degraded Heavy Oil. The density of weakly degraded heavy oil samples is relatively low compared with non-degraded heavy oil, generally between 0.93 and 0.95 g/cm³. Despite the fact that it is still rich in n-alkanes with a weak parity advantage, the distribution of tricyclic terpanes and pentacyclic triterpenes does not change much. Nevertheless, the relative abundance of low carbon n-alkanes is

| Biodegradation Types | Chemical Composition and Variation Characteristics |
|----------------------|--------------------------------------------------|
| Undegradation        | Rich alkanes                                     |
| Weak Degradation     | Light hydrocarbons are removed                   |
| Medium Biodegradation| Ninety percent N-alkanes are removed; cyclohexane and alkyl naphthalene are removed; naphthalene is reduced; isoprenoid alkanes and methylpentane are removed. |
| Severe Biodegradation| C21–C25 cycloalkanes are removed; fifty percent 20R-ααα sterane is removed; demethylhopane is dominant. |
Figure 7. Biodegradation series of heavy oil in the Western Sag of the Liaohe Basin. (A) Undegradation, T1-26-32 well, S4 Horizon, 2066.8 m. (B) Weak degradation, L46-56-S well, S1+2 Horizon, 1347.0 m. (C) Medium degradation, S1-52-23 well, S4 Horizon, 1578.6 m. (D) Severe degradation, J45-17-12 well, S4 Horizon, 906.5 m.
significantly reduced and the abundance of isopentadiene alkanes increases. In particular, pristane (Pr) and phytane (Ph) become the major peak of saturated hydrocarbons by gas chromatography (reconstructed ion chromatograph), where Pr/nC17 and Ph/nC18 increase and the distribution of steroid and terpene biomarkers is normal (Figure 7B) with no rearrangement of steranes and little pregnancy steranes and homopregnanes. This indicates that the degradation of steranes may start from regular steranes, reducing the overall content and enabling the detection of low-carbon steranes. This kind of heavy oil is formed by weak biological degradation with high colloid and asphaltene content.

3.3.3. Moderately Degraded Heavy Oil. The density of moderately degraded heavy oil is slightly higher than that of weakly degraded heavy oil, which is typically above 0.94 g/cm³. Most of the n-alkanes and isopentadiene alkanes are degraded. Normal steranes, especially those with biological configuration, are preferentially degraded and the relative abundance of rearranged steranes increases significantly. The 25-norhopane appeared, which is formed by removing the methyl group at the C-10 position of hopane (Figure 7C).

3.3.4. Severely degraded heavy oil. The density is normally over 0.95 g/cm³, the viscosity is high, the content of colloid and asphaltene is high (usually > 45%), and the wax content is low. N-alkanes and isopentadiene alkanes are all degraded, and steranes and terpanes are severely degraded, while the abundance of normal steranes is significantly low or almost all degraded, and the content of rearranged steranes increases. The 25-norhopane disappeared (Figure 7D).

Steranes and rearranged steranes disappear with an increase in degradation. In addition, all pregnancy steranes, including those in ascending order, have been degraded. Also, as the strength of sesquiterpene/pentacyclic triterpene weakens, it becomes more difficult to distinguish the two apart.

The characteristics of C27−C29 steranes and the samples shown in Figure 7A–D are basically the same. Although the content of C27, C28, and C29 may be slightly different, they are mainly C29 steranes, indicating that the source of organic matter is basically the same. The organic matter maturity is not high, and there are no parameters indicating the organic matter maturity achieving balance. The samples are low maturity oil. However, due to the relatively severe biodegradation of sample D, the sterane content was significantly reduced, resulting in a drift of the baseline. At the same time, a large number of rearranged steranes were formed, and the relative content of regular steranes was relatively low.

3.4. Main Controlling Factors of Heavy Oil Biodegradation.

(1) Temperature: The formation temperature governs the survival and reproduction of microorganisms. When the temperature exceeds 60−80 °C, the growth of microorganisms is inhibited. The main controlling factor of formation temperature is depth. The relationship between formation temperature and burial depth is \( t = 10 \times 3.3 \times H/100 \). With an increase in depth, the formation temperature also increases. It can also be verified from Figure 2 that with an increase in depth, the colloid and asphaltene content is reduced, and the density and viscosity also decrease. Furthermore, the figure illustrates how temperature/depth controls the degree of biodegradation.

(2) Water environment: Some research results show that the Western Sag has the characteristics of vertical flow. When near the surface, the surface water is continuously injected, mixing with formation water. Other findings suggest that fault and fracture are the direct passage between surface water and groundwater, which results in high oxygen content in the water environment and promotes the growth and reproduction of a large number of aerobic microorganisms. Specifically, the shallow layer is mainly an environment dominated by aerobic microbial degradation. When the burial depth increases, the abundance of aerobic microorganisms gradually decreases and there is a conversion to anaerobic microorganisms, and crude oil biodegradation is dominated by anaerobic microorganisms. It is inferred that for the same number of aerobic microorganisms and anaerobic microorganisms, there is more biodegradation with aerobic microorganisms than with anaerobic microorganisms. Conversely, in the process of cyclical contact between formation water and surface water, the number of shallow aerobic microorganisms is much greater than that of anaerobic microorganisms under deep burial conditions. Therefore, it is observed that the shallower the burial depth, the more serious the degradation of heavy oil.

In addition, formation water can provide nutrients and electron receptors through diffusion to organisms (bacteria) near the oil−water interface, and this ability is controlled by the relative size between formation water volume and oil column volume. Hence, the contact degree between crude oil and formation water and the relative volume of formation water directly control the range and degree of biodegradation. That is, the greater the oil−water contact area, the larger the plane range of biodegradation; the higher the volume of oilfield water, the more nutrients and electron receptors are provided, and the greater the vertical biodegradation range of the oil column. During the upward migration of oil and gas from source rocks, with an increase in migration distance, the contact time between oil and gas and formation water increases accordingly, which leads to the gradual deepening of crude oil microbial degradation. Therefore, the shallower the buried oil layer, the farther the oil and gas migrate, the longer the contact time with the formation water, and the deeper the degradation.

4. Conclusions

The physicochemical properties of the heavy oil in the Western Sag of the Liaohe Basin are characterized by “three lows and three highs”; that is, low wax content, low freezing point, low sulfur content, high colloid and asphaltene content, high density, and high viscosity. The density increases with an increase in polar heteroatom content, such as colloid and asphaltene content. Biodegradation is the most important mechanism of heavy oil formation. The result of biodegradation leads to a reduction in saturated hydrocarbon content and growth in colloid and asphaltene content. Based on the physical properties of heavy oil and the change law of biomarkers, heavy oil is divided into four types: undegraded, weakly degraded, moderately degraded, and severely degraded. At the same time, the main controlling factors affecting biodegradation include the formation temperature and the water environment. The formation temperature corresponds to the formation depth. The shallower the depth, the lower the
temperature, the more suitable for microbial reproduction and survival, and the higher the degree of biodegradation. The water environment mainly refers to the volume of formation water, oil—water contact area, oil—water contact time, and oxygen content, which are positively correlated with the degree of biodegradation.

■ AUTHOR INFORMATION

Corresponding Author
Chenglong Ma – China University of Geosciences (Beijing), Beijing 100083, China; Exploration & Development Research Institute of Liaohe Oilfield Company, Panjin 124010, China; Email: mhwhgo125@126.com

Authors
Changhao Hu – Exploration & Development Research Institute of Liaohe Oilfield Company, Panjin 124010, China
Xingzhou Liu – Exploration & Development Research Institute of Liaohe Oilfield Company, Panjin 124010, China
Yugang Li – Exploration & Development Research Institute of Liaohe Oilfield Company, Panjin 124010, China
Jie Cui – Exploration & Development Research Institute of Liaohe Oilfield Company, Panjin 124010, China
Ying Wu – China Oilfield Services Ltd., CNOOC, Beijing 101149, China
Jianhong Huang – Exploration & Development Research Institute of Qinghai Oilfield Company, Dunhuang 736200, China
Shuming Li – Exploration & Development Research Institute of Liaohe Oilfield Company, Panjin 124010, China

Complete contact information is available at: https://pubs.acs.org/10.1021/acsomega.2c02668

Notes
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