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Research paper

What role would the pores related to brittle minerals play in the process of oil migration and oil & water two-phase imbibition?

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ABSTRACT

This study aimed to investigate the geometry distinction of the pores formed by brittle minerals in shale oil reservoir and dig out its significance in hydrocarbon migration and exploitation.

14050 pores related to brittle minerals in typical shale oil reservoir samples are selected as the research objects, high-resolution field emission scanning electron microscopy (FE-SEM), imbibition test, Nuclear Magnetic Resonance(NMR), high-resolution image technique, construction of new parameters, statistics and analysis of large numbers and other means are used to discover this issue.

Results firstly show that the development of pores in the two directions (Angle between two directions is 90 degrees or approximately 90 degrees) are complementary and promote each other. Reversely, the data points begin to be scattered, which means that the development of the pore in the above two directions begins to "lose stability", rather than promote each other all the time. This could prove that the rectangle (width, height), Legendre ellipse (major axis and major axis) and maximum and minimum Feret diameter of pores can all be used as the effective criteria for the development of pore shape related to brittle minerals. Sufficient evidences could prove that the pores related to brittle minerals contribute to oil and gas migration and oil & water imbibition.

The conclusion of this study will provide an important theoretical basis for clarifying the oil occurrence mechanism of shale oil reservoir and looking for the geological and engineering dessert from the microscopic viewpoint.

1. Introduction

With a large number of crude oil began to be extracted from tight sandstone, shale, oil shale and other unconventional reservoirs, unconventional resources have become the new main battlefield for energy exploration and development (Law and Curtis, 2002; Holditch, 2013; Scanlon et al., 2014; Liu et al., 2017a,b; Pan et al., 2019; Xiao et al., 2020).

As a typical representative of unconventional energy reservoir, shale oil reservoir is characterized by quasi continuous distribution, small pore-throat scale, and difficulty in seepage. As to the shale oil in China, the source and reservoir rock are integrated and frequently interbedded, with complex lithology and no obvious boundary between source and reservoir rock. It is essentially different from "Bakken marine formation" in North America and "Ordos continental formation" in central China. In terms of sedimentary characteristics, tight oil in North America is located in the large, wide and gentle craton dominated by marine facies, with a large area of source and reservoir distribution. Reversely, structural sedimentary background of China is complex, dominated by continental facies, with relatively limited source and reservoir distribution. Compared with North America, China's shale oil resources are poor in scale, liquidity and economy. Therefore, how to characterize the complex reservoir space of shale oil reservoir and its impact on oil and gas migration and production carefully and accurately has become an urgent scientific problem (Stevens et al., 2013; Zou et al., 2015; Du et al., 2018a,b, 2019a; Du, 2019).

The brittle minerals in sedimentary rocks mainly include quartz, feldspar, calcite and zeolite, mainly because they are mostly siliceous and calcareous (Perez and Marfurt, 2013). The higher the content of brittle minerals in tight reservoirs, the greater the potential of hydraulic fracture formation (Perez Altamar and Marfurt, 2014). Pore characterization has always been a hot topic in the study of unconventional reservoir geology in recent years. Previous researchers used imaging technology and

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fluid testing technology to describe the pore development and the fractal characteristics of unconventional reservoirs in detail, and obtained a series of new conclusions. However, most of the existing ideas of reservoir characterization were focused on the characterization of pore size (Clarkson et al., 2012, 2013; Ghanizadeh et al., 2015). For example, we are good at using the equivalent circle radius to represent the actual pore size, which to some extent ignores the importance of pore geometry (Alfi et al., 2019; Hassan et al., 2019), which is not appropriate. Because for some types of pores with the same equivalent circle radius, the mineral types that make up the pore boundary are not necessarily the same, and the degree of compaction and diagenetic transformation are not necessarily the same, resulting in different geometry. So the seepage characteristics of these types of pores must be different, which cannot be generalized. Therefore, it is necessary to pay attention to the particularity of pores in the types of boundary minerals that make up them. This is an important theoretical premise for us to pay special attention to the pores related to brittle minerals.

In addition, during the growth and diagenetic transformation of brittle minerals, pores with different origins will be formed, including primary pores and secondary pores related to brittle minerals (Gao et al., 2019). The primary pore is mainly formed by the boundary "building" of brittle minerals and their combinations. Therefore, the size, structure, shape and configuration of pore-throat are closely related to the external crystal shape and the diagenesis process of brittle minerals, which is generally the product of the external crystal combination. The secondary pore is mainly formed by the boundary "building" of crystals' internal defects caused by brittle fracture, dissolution and metasomatism of brittle minerals. Therefore, the size, structure, shape and configuration of pore-throat are closely related to the internal lattice characteristics of brittle minerals, diagenesis process and other complex factors (Wanniarachchi et al., 2019; Sun et al., 2019). Therefore, due to the high elastic modulus and low Poisson's ratio of brittle minerals, oil and gas will inevitably cause the deformation of mineral boundary formed by such minerals in the flow process, which would have a more important impact on the oil and gas flow. Therefore, the pores related to brittle minerals should be treated as a special type of pores, which also need special attention.

A lot of effective studies have been done in the pores related to brittle minerals. Khidir and Catuneanu (2010) also believed that the potential of a sandstone to serve as the sandstone reservoir for producible hydrocarbons is strongly related to the quartz overgrowths. Hakimi et al. (2012) and Al Areeq et al. (2016) both concluded that the widespread occurrence of early calcite cement could suggests that the sandstones lost significant primary porosity at an early stage of its diagenetic history. To some extent, the formation of calcite in diagenetic process has a negative effect on the primary pores of the reservoir. Mørk (2013) found that diagenesis and the distribution of quartz cement is influenced by lithofacies and detrital compositions which would cause the low permeability of the reservoir rock. Lai et al. (2015) and Nolansnyder and Parnell (2019) both indicated that feldspar had higher surface area fractions than volumetric fractions in sandstone which means that the hydrocarbon would flow by these type of minerals more frequently. Saïag et al. (2016) found that feldspar alteration released silica and aluminum into the reservoir promoting the development of dickite and illite-rich I/S mixed layers, which tended to destroy porosity and permeability, as calcite cements and quartz overgrowths. Xiong et al. (2016) proved that calcite cementation acts as a main controlling factor on the reservoir quality in the Flemish Pass reservoir sandstones. Over 75% of initial porosity was lost due to the early calcite cementation. Liu et al. (2017a,b) found that mostly feldspar and pyrite affect the total pore volume of samples from Middle Bakken Formation whereas clay dominates the total pore volume of samples from Upper/Lower Bakken Formation. Anovitz and Cole (2019) believed that the onset of feldspar dissolution increased both porosity and surface area, and these increased again near the surface with the initiation of chlorite and illite dissolution.

It should be pointed out that in the quantitative study of pores, we mostly pay attention to how much storage volume it can bring to hydrocarbon or how much flow space volume it could provide for hydrocarbon. This is necessary and, frankly, insufficient. The formation of all kinds of pores in underground reservoirs is due to the active or passive construction of the boundaries of various minerals or organic matters. Of course, this kind of boundary is likely to be transformed later, which often does not reflect the initial standard crystal shape of minerals. After a series of geological processes such as weathering, erosion, transportation, deposition, compaction, dissolution and diagenesis, the crystal shape of the boundary will change from the approximate straight line to the curve. When these minerals accumulate or dissolve, they form various pores. Therefore, the zigzag feature of the boundary crystal is an indispensable material basis for us to study a series of geological processes experienced by the reservoir. This is the significance of pore geometry research.

2. Background

Jimsar shale oil is located in Jimsar depression in the east of Junggar Basin in Western China, 450 km away from Karamay City, Xinjiang Uygur Autonomous Region, 150 km away from Urumqi City (Su et al., 2018).

The main mining objective in the study area is Permian Lucaogou Formation, with stratum thickness of 25 to 300 m, average 200 m, buried depth of 800 m to 4800 m, average 3570 m. According to the law of crude oil enrichment, it has overall oil-bearing and two-stage concentration. There are two desserts developed in the main production layer, with stable horizontal distribution. The thickness of the upper desserts is 8 to 26 m, and the thickness of the lower desserts is 12 to 40 m. The structure is a monocline with high in the East and low in the west, the dip angle of the main part of the stratum is 3–5°, and the fault is not developed (Yang et al., 2017; Su et al., 2018). The observation of shale oil reservoir samples in the study area shows that the oil saturation is high (usually higher than 70%), and the oil content is good, which provides a very important material basis for crude oil development. The lithology is complex, with rapid longitudinal change and thin interbedding (Fig. 1).

Previous research shows that shale oil reservoir in the study area has poor permeability, developed secondary micro to nano pore-throat system and good connectivity The average porosity is 11% under overburden pressure, and the average overburden permeability is $0.01 \times 10^{-3}\ \mu m^2$, the proportion of samples less than $0.1 \times 10^{-3}\ \mu m^2$ is more than 90%. The main type of reservoir space is dissolved pore, and the microfractures are underdeveloped. Nano pore-throat accounts for more than 65% of the reservoir space, mercury saturation can reach more than 90%, pore-throat connectivity is good. Fine siltstone is better than arenaceous dolomite. The mineral composition of reservoir rock is various, most of which are calcite, quartz, feldspar and other brittle minerals. The natural fracture is not developed and the two horizontal stress difference is large, so it is necessary to subdivide the formation to carry out the hydraulic fracturing. Good brittleness, and weak water sensitivity of reservoir make it suitable for large-scale fracturing development (Wei et al., 2017; Ding et al., 2017; Ma et al., 2018).

According to the statistics of permeability values of all samples (Fig. 2), the permeability distribution of shale oil reservoir in the

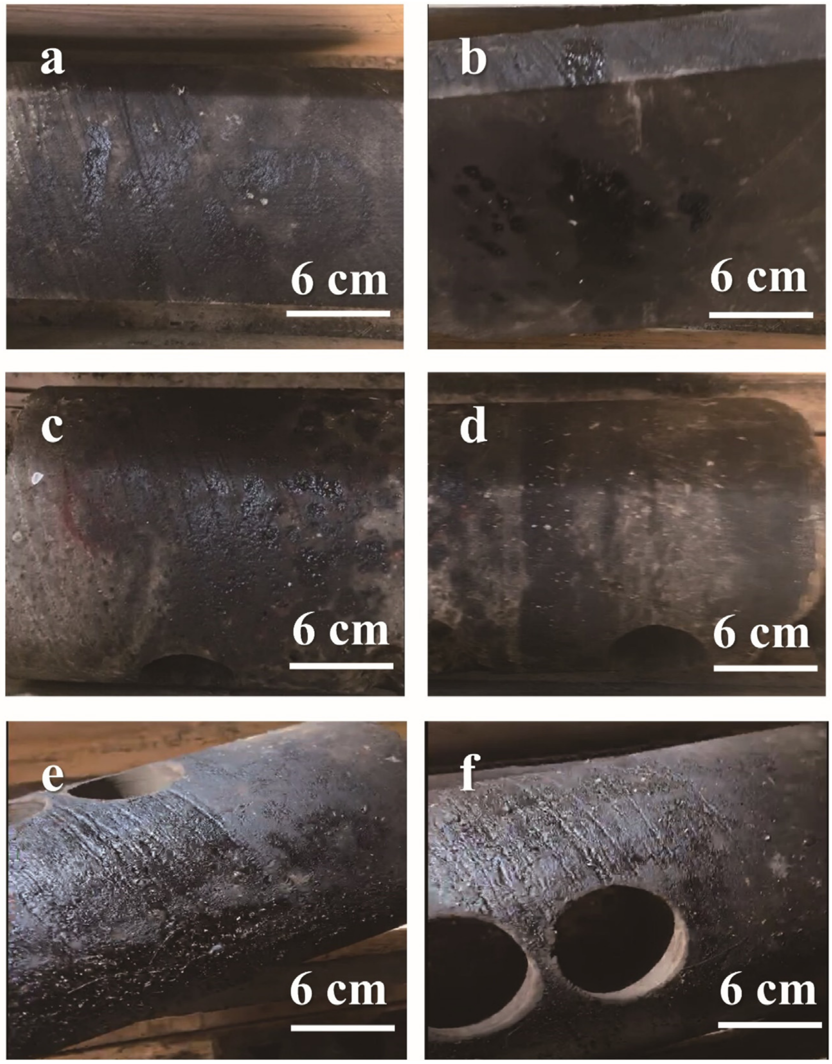

Fig. 1. Observation of fresh shale oil reservoir samples in Jimsar depression, Junggar Basin.

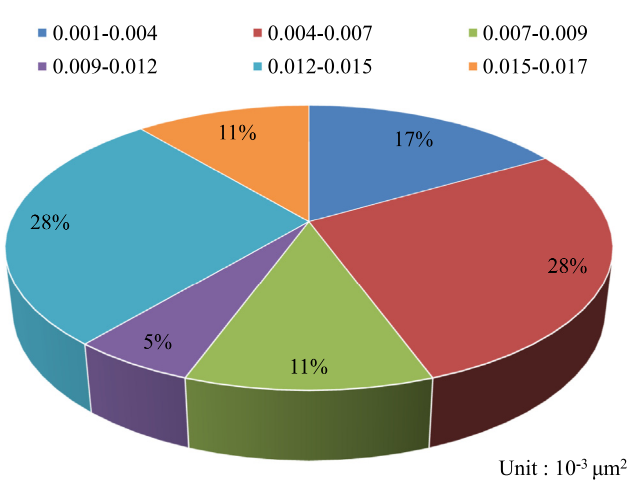

Fig. 2. Pie chart of permeability frequency distribution of shale oil samples.

study area is between 0.001∼0.017 × 10^{-3} μm^2, in which the highest distribution frequency is 0.012∼0.015 × 10^{-3} μm^2 and 0.004∼0.007 × 10^{-3} μm^2 which can reach 28%; the second is 0.001∼0.004 × 10^{-3} μm^2, the frequency can reach 17%; followed by 0.015∼0.017 × 10^{-3} μm^2 and 0.007∼0.009 × 10^{-3} μm^2, the frequency can reach 11%, the last is 0.009∼0.012 × 10^{-3} μm^2. It fully proves that the permeability of shale oil reservoir is relatively low. The contribution of different pores to oil–water flow should be quantified.

High Pressure Mercury Injection (HPMI) test could obtain four main parameters: maximum radius of pore-throat, median radius of pore-throat, average radius of pore-throat and displacement pressure. Statistical analysis of the maximum, minimum, average and homogeneity coefficient of the above four types of parameters of all samples (Table 1) shows that the values of the four types of parameters are respectively 30∼2070 nm (average 510 nm), 10∼190 nm (average 63 nm), 10∼520 nm (139 nm on average), 0.36∼21.96 mpa (8 MPa on average), and the homogeneity coefficient distribution of the four parameters is between 0.25∼0.36. As a whole, the effective pore-throat size of shale oil reservoir is very low, which will have different effects on oil–water displacement and imbibition.

3. Method and application

3.1. Pore characteristics of brittle minerals in shale oil reservoir

Field emission scanning electron microscopy (FE-SEM) observation on a large number of samples shows that quartz, calcite

Table 1
Statistics of pore related parameters in High Pressure Mercury Injection (HPMI) test.

Statistics	Properties			
	Maximum radius/nm	Median radius/nm	Average radius/nm	Displacement pressure/MPa
Maximum	2070	190	520	21.96
Minimum	30	10	10	0.36
Average	510	63	139	8
Homogeneity index	0.25	0.33	0.27	0.36

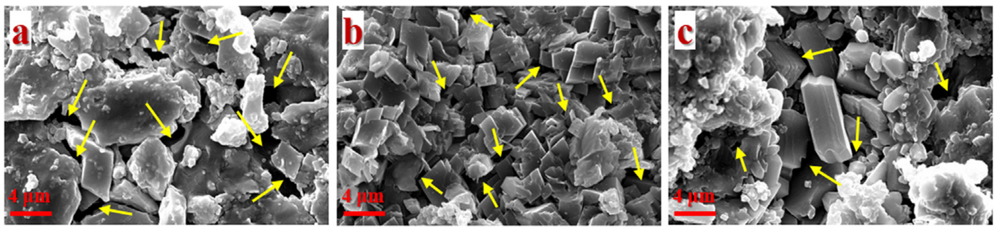

Fig. 3. Microscopic FE-SEM characterization of pores related to brittle minerals in shale oil reservoir of Jimsar sag, Junggar Basin. (The electron microscope photo data were provided by Research Institute of Exploration & Development and Experimental Detection Research Institute of Xinjiang Oilfield. a-calcite grain and intergranular dissolution pore in matrix; b-calcite and granular quartz crystal in matrix; c-plate zeolite and granular quartz distributed in the matrix).

and other brittle minerals, as important mineral components, are mostly autogenetic and widely exist in shale oil reservoirs in Junggar basin. Due to diagenesis process, in-situ stress and other factors, dissolution and fracture are relatively common. From Fig. 3, we can see that there are three types of pores related to brittle minerals: granular accumulation pore of quartz-calcite, accumulation pore of quartz–quartz and intergranular dissolution pore of calcite. At the same time, we can see the obvious oil immersion from Fig. 1(c, d). This provides us with the extremely important geological evidence for the presence of crude oil in such pores. Similarly, crude oil will pass through such pores in both migration and seepage process. This is also the necessity of the research on pores related to brittle mineral for oil and gas exploration and development.

14 050 pores related to brittle minerals in shale oil reservoir were selected as research objects, and their equivalent pore radius were all measured and counted. The results show that (Fig. 4), the radius of pores related to brittle minerals is mainly distributed between 6 to 341 nm, and the peak radius is about 62 nm, accounting for nearly 70%, which is higher than the lower limit radius of crude oil flow (50 nm). It could prove that the pores related to brittle minerals have the space size required for the flow of crude oil.

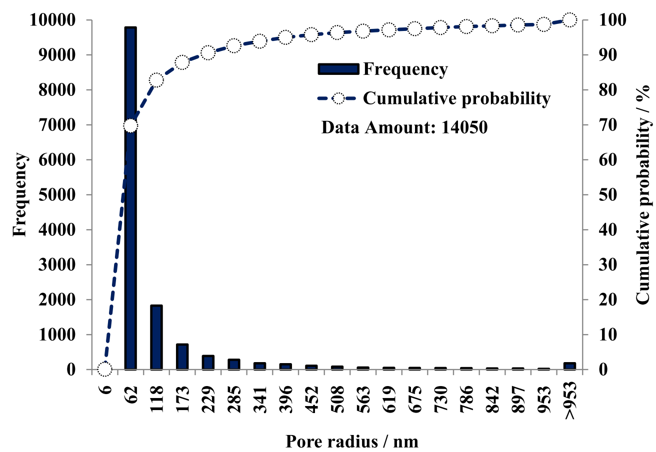

Fig. 4. Distribution histogram of equivalent radius of pores related to brittle minerals in shale oil reservoir of Jimsar depression, Junggar Basin (Data Amount: 14 050).

3.2. Pore geometry characterization and development mechanism

In order to fully characterize the geometric characteristics of the pores related to brittle minerals, we have established four types of geometric parameters, which is the width and height of the external rectangle the major and minor axes of Legendre ellipse, the maximum and minimum Feret diameters, and the shape factor and solidity, respectively. For these four kinds of geometric parameters, we try to give the relative qualitative physical meaning, respectively. For pores with the same equivalent radius, if these four kinds of geometric parameters are different, then these parameters can better show the unique properties of pores, which is equivalent to adding several new evaluation indexes for the accurate description of pores, and more conducive to our in-depth understanding and research of the relative geological process.

3.2.1. Minimum width and height of external rectangular of pores

The minimum circumscribed rectangle refers to the maximum range of several two-dimensional shapes (such as points, lines, polygons) expressed in two-dimensional coordinates, that is, the rectangle with the maximum abscissa, the minimum abscissa, the maximum ordinate and the minimum ordinate in each vertex of the given two-dimensional shape to determine the lower boundary. It is a very important concept in GIS (Geographic Information System) and computer graphics (Kwak and Habib, 2014). From the viewpoint of energy and geology, under the condition of uniform stress, sedimentary compaction and diagenetic transformation, the width and height of the rectangle outside each pore should also show uniform changes. However, this is not the case, which is determined by the complexity of geological activities.

After image processing of each pore, the smallest rectangle enclosing the selection which is short for "bounding rectangle" could be identified. Then the width and height of the bounding rectangle of each pore could be calculated. Results of 14 050 pores were calculated and the cross-plot of the width and height of each pore was also drawn (Fig. 5).

It can be clearly seen from Fig. 5 that for the pores related to brittle minerals in shale oil reservoir of Jimsar depression in Junggar basin, the width and height of the rectangle outside the pores are mainly distributed in 10 nm∼10 μm and 4 nm∼10 μm, respectively. When the width is less than the height (green points in Fig. 5) or the width height ratio is between 1 and 5 (red points

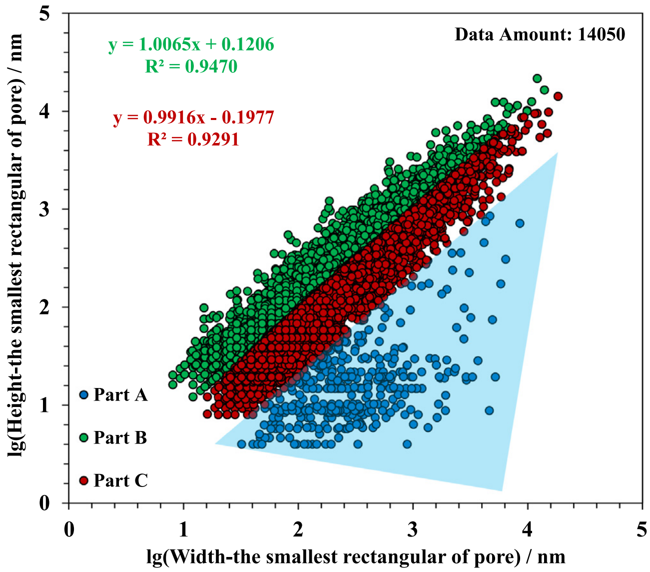

Fig. 5. Correlation diagram of the width and height of the external rectangle of the pores related to brittle minerals in the shale oil reservoir of Jimsar depression, Junggar Basin (Data amount: 14 050). . (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

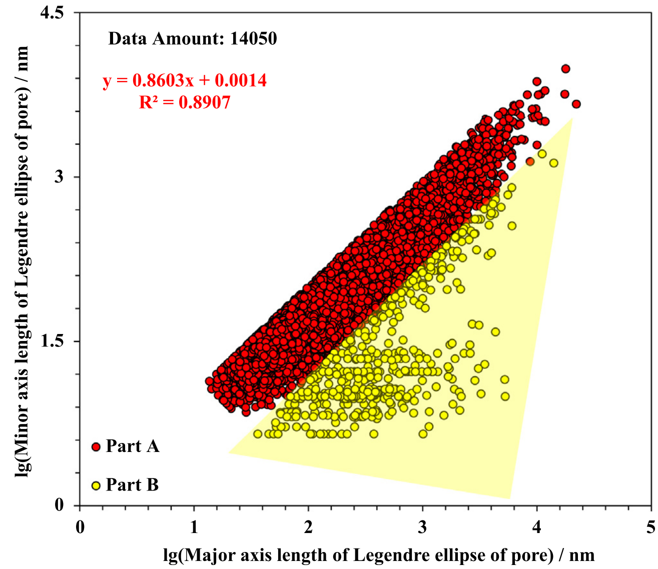

Fig. 6. Correlation diagram of Length of major axis and minor axis in Legendre ellipse of brittle mineral related pores in shale oil reservoir in Jimsar depression, Junggar Basin (Data Amount: 14 050).

in Fig. 5), the relationship between the width and height of the rectangle will be linear obviously. The square of the correlation coefficient could be up to above 0.9.

In Fig. 5, Part A represents the data points should satisfy the inequality (1) as following:

$$\lg(\text{Height}) \leq (0.94 \times \lg(\text{Width}) - 0.51) \quad (1)$$

Part B represents the data points should satisfy the inequality (2) as following:

$$\lg(\text{Height}) > \lg(\text{Width}) \quad (2)$$

Part C represents the data points should satisfy the inequality (3) as following:

$$(0.94 \times \lg(\text{Width}) - 0.51) < \lg(\text{Height}) < \lg(\text{Width}) \quad (3)$$

It is shown that in this numerical range, the development of pores in the vertical and horizontal directions are complementary and promote each other, and the heterogeneity of the development is mainly reflected in the change rate of the width and height of the external rectangle. When the data points satisfy the inequality (1) (Part A in Fig. 5), the correlation coefficient between the width and height of the rectangle of the pore begins to decrease significantly, the data points begin to be scattered, which proves that in this numerical range, the development of the pore in the vertical and horizontal directions begins to "lose stability", rather than promote each other all the time. This shows that the numerical value and ratio of the width and height of the external rectangle of the pore can be used as an effective criterion for the development of pore shape.

3.2.2. Major axis and minor axis of Legendre ellipse of pores

In this process, the best fit ellipse would be found. The ellipse will have the same area, orientation and centroid as the original selection. Major and Minor are the primary and secondary axis of the best fitting ellipse. The same fitting algorithm is used to measure the major and minor axis lengths (Mikli et al., 2001). Results of 14 050 pores were calculated and the cross-plot of major axis and minor axis of Legendre ellipse of each pore was also drawn (Fig. 6). Similarly, it can be clearly seen from Fig. 6 that for pores related to brittle minerals in shale oil reservoir of Jimsar depression in Junggar basin, length of major axis and minor axis of Legendre ellipse are mainly distributed in 20 nm ∼10 μm and 4 nm∼6 μm respectively. When length of minor axis and major axis of Legendre ellipse meet the inequality in (4) (Part A in Fig. 6):

$$\lg(\text{Minor}) \geq (1.02 \times \lg(\text{Major}) - 0.81) \quad (4)$$

The length of the minor axis and the major axis of Legendre ellipse will show a very significant linear correlation, and the square of the correlation coefficient can reach about 0.83. Similarly, this also proves that in this numerical range, the development of pores in the direction of the two axes of the ellipse is complementary and mutually reinforcing, and the heterogeneity of their development is mainly reflected in the rate of change.

On the contrary, when the length of the major axis and the minor axis of Legendre ellipse meet the inequality (5) (Part A in Fig. 6):

$$\lg(\text{Minor}) < (1.02 \times \lg(\text{Major}) - 0.81) \quad (5)$$

The relationship between the length of the minor axis and the length of the major axis of Legendre ellipse began to decrease significantly, and the data points began to be scattered, which proved that within this numerical range, the development of pores in the direction of the two axes of the ellipse began to lose stability, rather than promote each other all the time. This shows that the length of the major axis and the minor axis of Legendre ellipse can also be used as an effective criterion for pore shape development.

Generally speaking, for the Legendre ellipse fitted by the pores of shale oil reservoir, the length of the major axis and the minor axis of the ellipse also has the obvious positive correlation trend, which shows that when the compaction, pressure dissolution and other sedimentary diagenesis cause the pores to be approximately elliptical distribution, the pores show better elastic characteristics.

3.2.3. Maximum and minimum feret diameter of pores

The maximum and minimum of Feret's diameter indicates the longest and shortest distance between any two points along

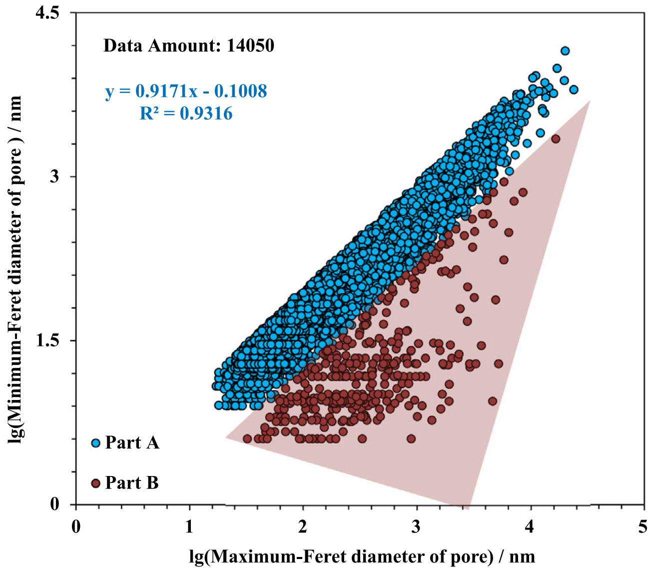

Fig. 7. Correlation diagram of maximum and minimum Feret diameters of pores related to brittle minerals in shale oil reservoir of Jimsar depression, Junggar Basin (Data Amount: 14 050).

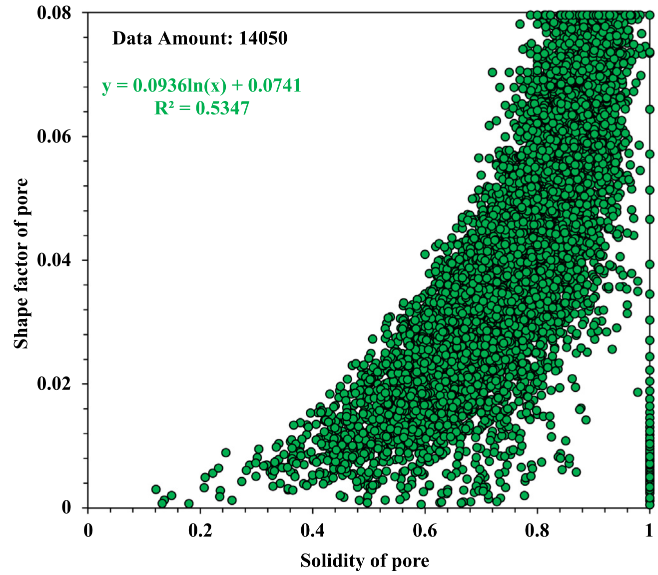

Fig. 8. Correlation diagram between shape factor and solidity of pores related to brittle mineral in shale oil reservoir of Jimsar depression, Junggar Basin (Data Amount: 14 050).

the fracture boundary (Gregorova and Pabst, 2007). Results of 14 050 pores were calculated and the cross-plot of maximum and minimum Feret diameter of each pore was also drawn (Fig. 7).

Similar to the rectangle (width, height) and Legendre ellipse (major axis and major axis) of pores, it can be clearly seen from Fig. 7 that for pores related to brittle minerals in shale oil reservoir of Jimsar depression in Junggar basin, the maximum and minimum Feret diameters are mainly distributed in 20 nm∼10 μm and 4 nm∼8 μm respectively When meeting the condition (6) with the minimum Feret diameter (Part A in Fig. 7):

$$\lg(\text{Minimum Feret}) \geq (0.92 \times \lg(\text{Maximum Feret}) - 0.47) \quad (6)$$

There will be a very significant linear correlation between the maximum diameter and the minimum diameter of the pore, and the square of the correlation coefficient can reach about 0.87. This proves that the development of pores in the direction of maximum and minimum diameter of the pores is complementary and mutually promoting, and the heterogeneity of the development can also be reflected in the numerical change rate for the third time.

On the contrary, when the maximum and minimum Feret diameters of pores meet the condition (7) (Part B in Fig. 7):

$$\lg(\text{Minimum Feret}) < (0.92 \times \lg(\text{Maximum Feret}) - 0.47) \quad (7)$$

The relationship between the maximum and minimum Feret diameters of the pores began to decrease significantly, and the data points began to be scattered, which proved that in this numerical range, the development of the pores in the two directions of the maximum and minimum Feret diameters of the pores began to lose stability, rather than promote each other all the time. This shows that the maximum and minimum diameter of the pore can also be used as an effective criterion for the development of pore shape.

Generally speaking, for the Feret diameter for pores in shale oil reservoir, the maximum and minimum Feret diameter show a significant positive correlation trend, which further proves the unity process of homogeneity and heterogeneity of pore development.

3.2.4. Shape factor and solidity of pores

The shape factor is the ratio of the actual area of pore to the square of perimeter (Miao et al., 2017), which can be used to quantify the curvature of pore boundary. The lower the shape factor, the higher the curvature of pore boundary. Pore solidity refers to the ratio of actual pore area to convex hull area (Du, 2020; Du et al., 2020), which can be used to quantify the degree of pore compression. The lower the pore solidity, the greater the degree of compression.

For the pores related to brittle minerals in shale oil reservoir of Jimsar depression in Junggar basin, the shape factor and solidity are mainly distributed in 0∼0.08 and 0.3∼1, respectively. The correlation analysis shows that (Fig. 8), with the continuous compaction process, the pore solidity decreases gradually, and at the same time, the shape factor also decreases gradually, which means that the pore tortuosity increases, which may be caused by the dissolution of pore boundary formed during compaction. Of course, it cannot be ignored that there are a few pores without effective compression (the value of pore solidity equals to 1), and their shape factors are distributed between the maximum and minimum values. This shows that compression is only one of the factors that lead to the change of shape factor in the process of sedimentary diagenesis, and there are other important factors that will change the shape factor.

3.2.5. Connectivity of pores related to brittle minerals

Pore coordination number refers to the number of throat connected by each pore in rock or soil, which directly reflects the connectivity between pores and throats. When the coordination number is zero, it means that the pore is not connected. When the coordination number is greater than or equal to 1, the pores are connected. The higher the coordination number, the better the reservoir quality (Vogel, 1997).

Some scholars have studied the connectivity conditions of various two-dimensional and three-dimensional network models, closely combining the research of pore structure with petroleum reservoir rock, which also makes the research of pore structure more in-depth (Dullien, 2012). The results show that the coordination number is closely related to recovery, and the higher the coordination number is, the better the connectivity is. The effect

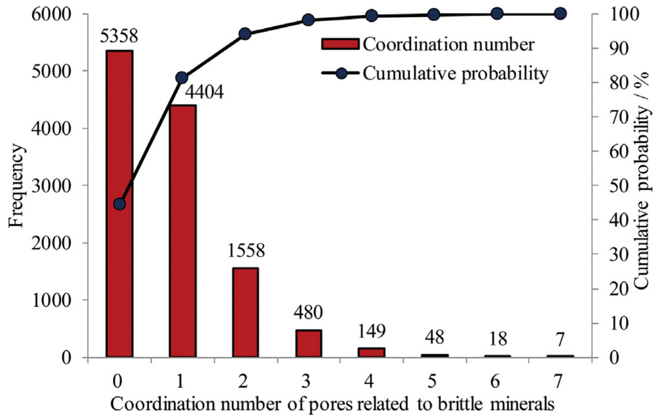

Fig. 9. Coordination number and cumulative probability distribution of pores related to brittle minerals.

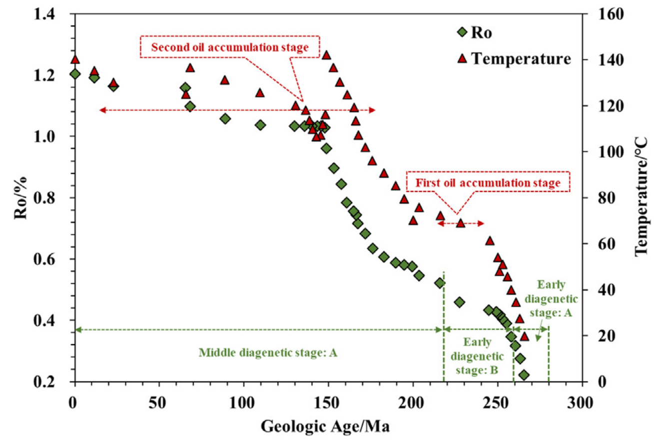

Fig. 10. Diagenetic evolution stage (a) and hydrocarbon filling and accumulation stage (b) of shale oil reservoir in Jimsar sag, Junggar Basin. Revised after (Zheng et al., 2018).

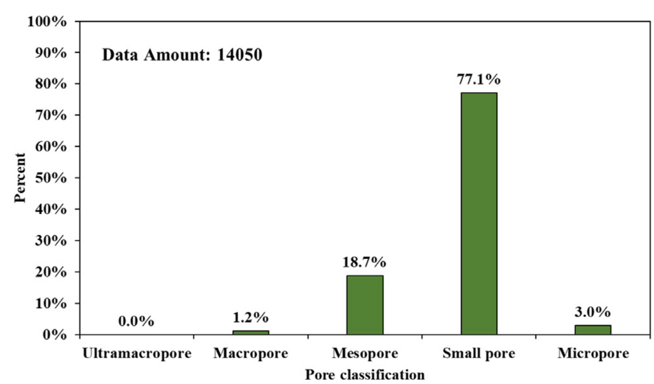

Fig. 11. The distribution of pore levels related to brittle minerals in shale oil reservoir of Jimsar depression, Junggar Basin (Data Amount: 14 050).

of coordination number on oil recovery could not be compared with the pore & throat ratio, because although the coordination number is high, the oil recovery will not be very good if more thin throats are connected (King Jr. et al., 2015; Sakhaee-Pour and Bryant, 2015). Therefore, we calculated the coordination number of 12 022 pores related to brittle minerals and drew a histogram (Fig. 9).

Fig. 9 shows that the total connectivity of pores related to brittle minerals is 55.43%, among which 36.63%, 12.96%, 3.99%, 1.24%, 0.40%, 0.15% and 0.06% of pores with coordination numbers of 1, 2, 3, 4, 5, 6 and 7 respectively. This proves that more than half of the pores related to brittle minerals are connected, and they may play an important role in the process of oil–gas migration and oil–water two-phase imbibition, which is worthy of further exploration.

4. Significance of pores related to brittle minerals

4.1. Significance in oil and gas migration

It can be seen from the above analysis that the pores related to brittle minerals in shale oil reservoirs are mainly related to the growth and metasomatism of authigenic calcite, quartz and other minerals. Therefore, it is worth discussing whether the pores related to brittle minerals contribute to hydrocarbon migration, which is one of the important stages in the formation of oil and gas reservoirs. If the formation period of pores related to brittle minerals overlaps before or during the period of oil and gas migration and accumulation, it is proved that hydrocarbons are most likely to pass through the pores related to brittle minerals in the process of oil and gas accumulation, that is to say, pores related to brittle minerals would contribute to oil and gas migration.

As can be seen in Fig. 10, Zheng et al. (2018) carried out the homogenization temperature of fluid inclusion and vitrinite reflectance (Ro) test of Junggar shale oil reservoir, and found that the generation and metasomatism of authigenic calcite mainly occurred from the Late Triassic to the present (220∼0 Ma). At the same time, the source rocks entered the hydrocarbon generation stage from the Late Triassic, and there are two major migration and accumulation processes, namely, Jurassic and Cretaceous Paleogene. These two periods are both after the Late Triassic. Therefore, we have sufficient evidences to prove that the pores related to brittle minerals contribute to oil and gas migration.

4.2. Significance in oil migration in oil & water two-phase imbibition

As we know, shale oil reservoir is very tight, it is difficult to establish an effective displacement pressure system in actual development. In other words, displacement alone cannot effectively solve the shale oil development completely (Wang et al., 2011).

In recent years, imbibition oil recovery has become a hot topic. It is a physicochemical process of effectively extracting crude oil through the replacement of oil and water by the difference of capillary absorption and wettability of quasicrystal state (Birdsell et al., 2015).

In order to furtherly study whether oil–water displacement occurs in the pores related to brittle minerals in oil & water imbibition, we need to carry out imbibition experiments. If we find that there is a change process of crude oil in the distribution range of pore radius related to brittle minerals, we can conclude that such pores could contribute to the process of oil & water imbibition and absorption. For the convenience of research, shale oil pores are divided into ultramacropore (≥10 μm), macropore (10∼1 μm), mesopore (1 μm∼100 nm), small pore (100 nm∼10 nm) and micropore (<10 nm). According to the statistics of 14 050 pores related to brittle minerals in shale oil reservoir (Fig. 11), they are mainly small pores, followed mesopores. Micropores and macropores only occupy very small parts. In addition, there are no ultramacropores.

We selected fresh oil samples in the study area, and used water shielded from hydrogen signal as the imbibition liquid to carry out oil–water two-phase imbibition experiment.

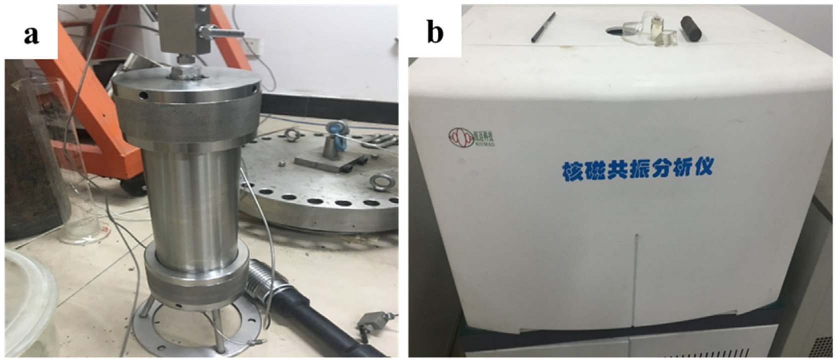

Fig. 12. Main test instruments of imbibition experiment (a-imbibition vessel; b-nuclear magnetic resonance instrument).

The details of the experimental process and the instrument installation are as follows (Fig. 12)

(1) Prepare water-based imbibition liquid and add $MnCl_2$ solid into the liquid to shield the hydrogen signal in the water and ensure that the signal amplitude change explored in the experiment only indicate the change of crude oil.

(2) The device is composed of a pneumatic pump (the gas source is inert gas such as helium), which is connected with the intermediate container with a piston to form a set pressure on the intermediate container. Put the prepared water-based imbibition liquid in the intermediate container, place each sample in the intermediate container in turn, close the intermediate container, connect the air pump, apply pressure through gas source injection and piston control, and carry out the pressure imbibition determination of shale oil reservoir samples.

(3) The samples were taken out at 18 time points from 0 h, 2 h, 5 h, 8 h, 11 h, 14 h, 39 h to 762 h of imbibition, respectively, and the T2 relaxation time spectrum curves at different times were measured.

At the same time, we choose different time of imbibition to measure the NMR T_2 spectrum of the sample, and discover the oil & water displacement process through the change characteristics of crude oil signal. The experimental results are shown in Figs. 13–15.

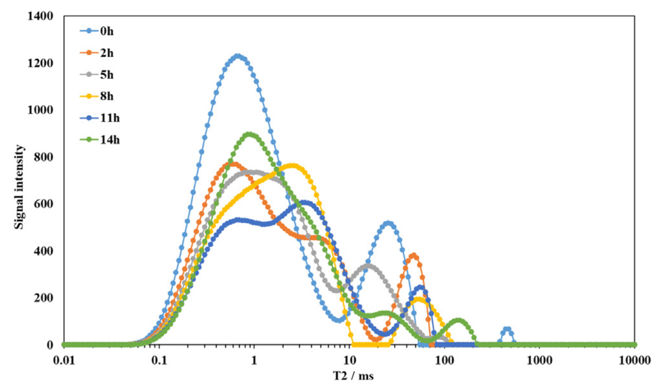

Fig. 13. Long term imbibition-NMR joint measurement results of shale oil reservoir in Jimsar sag, Junggar Basin (0∼14 h).

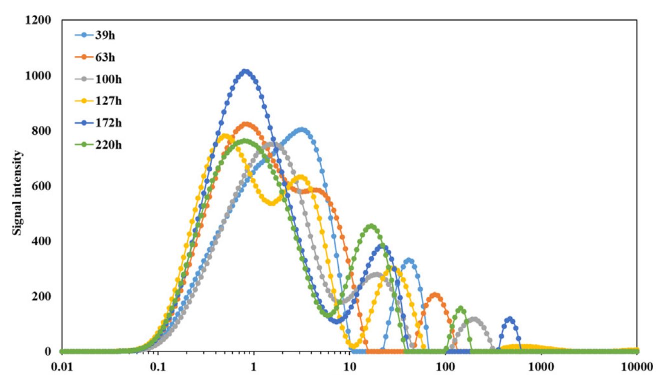

Fig. 14. Long term imbibition-NMR joint measurement results of shale oil reservoir in Jimsar sag, Junggar Basin (39∼220 h).

Figs. 13–15 show that the most types of pores related to brittle minerals (100 nm∼10 nm) are observed. It can be found that the crude oil signal curve changes unsteadily in the period of 0 h to 762 h within this range of radius, which fully shows that the pores within this range of radius have a very unstable oil & water displacement process, which has an important impact on oil & water imbibition and absorption. From the viewpoint of minerals, quartz and calcite are mainly water wet minerals, so it is conducive to the suction of water and the discharge of oil. However, it is also limited by the capillary force.

The total oil discharge efficiency of 18 time points in the process of imbibition is calculated, and the change curve is drawn as shown in Fig. 16. It can be seen very clearly that in the first two hours of imbibition, the oil drainage process is very active, the amount of imbibition and oil drainage is very large, and the oil discharge efficiency can reach about 26%. In the range of 2∼5 h, the total oil discharge efficiency is stable for a short time, and then in the range of 5 h∼11 h, the oil discharge efficiency continues to rise rapidly, reaching about 36.7% at 11 h. From 11h on, the oil discharge efficiency began to change periodically. At 266h, the oil discharge efficiency reached the highest (about 36.8%), and the final oil discharge efficiency stayed in 32.5%.

This is mainly due to the complexity of pore-throat size distribution, connectivity and wettability of shale oil reservoir and the reasons of experimental design, which lead to the previously discharged oil to be sucked into the rock again. To some extent, this is in line with the reality, because the mineral composition and wettability in the actual rocks are extremely complex, so the "oil displacement behavior" and "oil absorption behavior" would be both existed in different levels of pores in the process of imbibition. In addition, this may be related to the change of wettability and even the reversal of wettability caused by long-term fluid contact.

In order to furtherly clarify the importance of each level of pores for crude oil recovery, we calculated the oil recovery of four types of pores at different times, and drew the change curve. As can be seen in Fig. 17, the increase of oil discharge efficiency of small pores and micropores in the initial period (0–11 h) is relatively obvious, and then enters the unsteady state change stage. The average oil recovery rates of small pores and micropores were 47% and 51%, respectively. The oil discharge

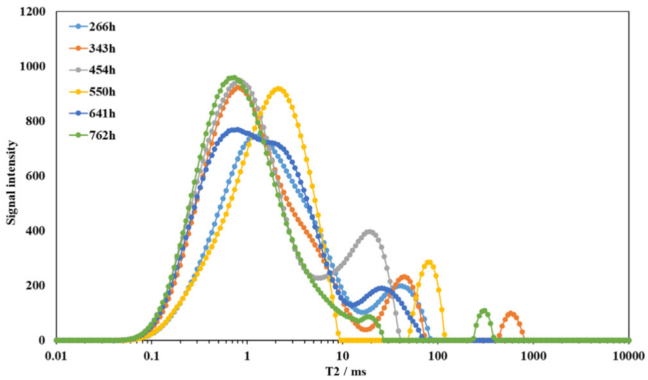

Fig. 15. Long term imbibition-NMR joint measurement results of shale oil reservoir in Jimsar sag, Junggar Basin (266∼762 h).

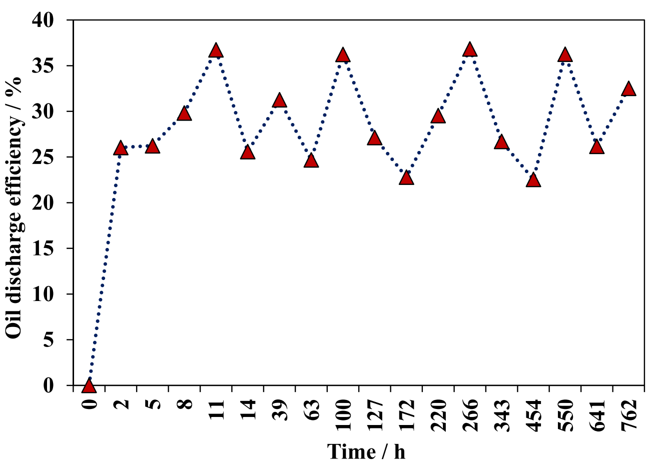

Fig. 16. Curve of total oil discharge efficiency of shale oil samples with time.

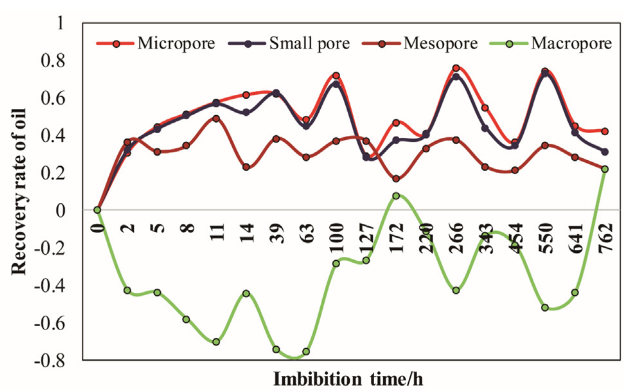

Fig. 17. Oil recovery curve of different levels of pores related to brittle minerals in shale oil reservoir of Jimsar sag, Junggar Basin.

efficiency of the mesopores has been in a fluctuating state, with an average value of 31%. Macropores receive crude oil from other pores for most of the time, so the crude oil volume increases instead of decreasing, and the oil displacement rate keeps at - 36% on average. For pores related to brittle minerals, they are mainly small pores and mesopores, so the oil displacement rate of these pores should be between 31% and 47%, which contributes higher than other level of pores in the oil discharge.

In fact, in the process of two-phase imbibition, the stability of oil & water displacement of each level of pore is different. We quantify the stability of this process by calculating the coefficient

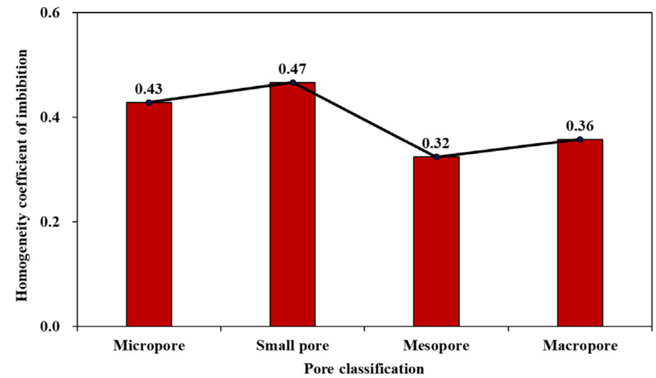

Fig. 18. Distribution histogram of the replacement activity (homogeneity coefficient of absolute replacement rate at each time) in different levels of pores related to brittle mineral in shale oil reservoir of Jimsar sag, Junggar Basin.

of homogeneity (the ratio of the average value to the maximum value) of the recovery at all times. The larger the homogeneity coefficient is, the more stable the process is.

From Fig. 18, we can see that the degree of stability of imbibition and absorption process is sorted as small pores, micropores, macropores and mesopores, respectively. As the pores related to brittle minerals are mainly small pores, followed by mesopores, the role of pores of brittle minerals in imbibition is complex. The pores in brittle mineral pores are relatively stable in the process of imbibition, while the mesopores are relatively unstable. This is a very important phenomenon.

It should be noted that, as for how to reflect the contribution of nanopores in NMR, the highest test accuracy range of the NMR instrument used in this time is about 60 nm. According to a large number of research results, the lower limit radius of shale oil flow in the study area is about 60 nm, too. Therefore, the data obtained from NMR test could meet our research objectives and requirements in advance, which can support our conclusion.

In addition, as for the issue of imbibition of large pores, it is obvious that it is difficult to saturate the dry shale, and the properties of the simulated oil are also different from the underground crude oil, so this is not appropriate. Therefore, in order to ensure the accuracy of the experimental results, we use the original oil-bearing core (but not fully saturated crude oil), and the original water content of such core is very low (almost negligible). Therefore, the crude oil in the original oil-bearing core and large pores may not saturated, so it can receive crude oil from other pores in the process of imbibition.

5. Conclusions

The rectangle (width, height), Legendre ellipse (major axis and major axis) and maximum and minimum Feret diameter of pores can all be used as the effective criteria for the development of pore shape related to brittle minerals. Sufficient evidences to prove that the pores related to brittle minerals contribute to oil and gas migration. Meanwhile, this type of pores have a very unstable oil & water displacement process, which has an important impact on oil & water imbibition and absorption.

Complexity of pore-throat size distribution, connectivity and wettability of shale oil reservoir and the reasons of experimental design, which lead to the previously discharged oil to be sucked into the rock again.

Generally speaking, for the brittle minerals in the reservoir, more attention has been paid to the influence of the formation of the brittle minerals on the physical properties (including porosity and permeability) of the reservoir. However, for the pores formed after the formation of the brittle minerals such as authigenic calcite and authigenic quartz in the diagenetic stage, there is currently a lack of clear characterization. The development mechanism of such special pores is also concerned. Research on the above issue has the great theoretical significance.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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