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Mechanisms of CO_2-oil-water interaction and CO_2 storage during CO_2 huff-n-puff EOR in shale reservoirs based on LB method

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ABSTRACT

CO_2 has the advantages of strong solubility, good viscosity reduction ability and low miscible pressure, which has a great potential in enhancing oil recovery (EOR). Shale has a high content of organic porous media and clay minerals, which is conducive to long-term adsorption and retention of CO_2, and is one of the best CO_2 storage environments. However, with the exploitation of hydraulic fracturing and CO_2 huff-n-puff, the water cut of the reservoir is increasing, and the mechanism of oil, water, CO_2 molecular interaction and fluid-rock molecular interaction in shale is still unclear.

In previous studies, we developed a new nano-scale porous media model based on the lattice Boltzmann method (LBM). A CO_2 huff-n-puff model including fracture and matrix (including macro poress, micro-pores (inorganic), nano-pores (organic)) were designed. The multi-relaxation Shan-Chen model was used to simulate the diffusion and adsorption behavior of CO_2 in the miscible state of shale. In this study, we additionally consider the water cut of different development stages of reservoir development to set the oil-water occurrence state in the matrix. Meanwhile, we comprehensively considered the effects of heterogeneous wettability caused by organic porous media and inorganic porous media in shale and the varying interfacial fluid viscosity of interfacial caused by adsorption on CO_2 huff-n-puff. Through the simulation of oil-water phase separation in the matrix, we set up four oil-water occurrence states (water cut is 10 %,30 %, 50 % and 70 % respectively). Water molecules mainly aggregate in inorganic matter. By injecting CO_2 into the fracture, the oil and gas density distribution and oil-water-gas three-phase distribution are analyzed. The results show that the adsorption quantity of CO_2 is reduced due to the stronger adsorption capacity of water molecules on the surface of inorganic rock particles, and the quantity of CO_2 entering the matrix in the fracture is reduced. The contact area between CO_2 and crude oil decreases with the increase of water content, and the miscibility of oil and gas slows down. We also found that the increase of water content will promote oil recovery in inorganic matter. But it will also reduce the contact area of oil and CO_2. When the water content is 0.5, the two effects are the weakest, and the oil recovery is the smallest. This paper provides theoretical guidance for the field application of CO_2 huff-n-puff mining unconventional oil and gas resources and the field application of CO_2 storage technology in different development stages.

Keywords: CO_2 huff-n-puff, Water cut, LBM, EOR, CCUS.

NONMENCLATURE

Abbreviations	
MRT	Multiple relaxation time model
EOR	Enhanced oil recovery
CSR	CO_2 storage rate
Symbols	
G	Interaction parameters between fluids
G_{ads}	Fluid-solid interaction parameters
F_{int}	Force between fluids
F_{ads}	Fluid-solid force
ψ	Potential function
e	Discrete velocity
D_c	CO_2 diffusion coefficient

1. INTRODUCTION

There are abundant shale oil and gas resources in the world. Due to the low oil flow rate in shale and the complex heterogeneity of shale formations [1], the

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production of shale oil is low. Volume fracturing is usually used on a large scale in the exploitation of early shale reservoirs. However, due to the rapid depletion of oil in fractures and the slow replenishment of oil in the matrix [2], the high initial recovery rate decreases rapidly [3], and shale reservoirs are difficult to achieve sustainable production. Because shale reservoirs contain a large amount of potential oil recovery, a slight increase in recovery can achieve huge oil production. At present, the progress and breakthrough of shale reservoir development technology is very urgent.

Global warming caused by anthropogenic emissions of greenhouse gases such as carbon dioxide (CO_2) has become a major challenge for industrialized countries. Now, carbon capture, utilization and storage (CCUS) has become a key technology that countries around the world attach great importance to and research and development, especially in the application of CO_2 enhanced oil recovery. This solution brings economic benefits to the production of shale oil reservoirs and can store a large amount of CO_2. CO_2 has a wide range of sources, and it is considered to be one of the best choices to improve the recovery of shale reservoirs because of its strong solubility, good viscosity reduction effect and low miscible pressure. Due to adsorption and other traditional capture mechanisms in shale areas, CO_2 capture is considerable [4]. Therefore, the method of injecting CO_2 into oil and gas reservoirs to improve recovery and store CO_2 can achieve a win-win situation. Due to the low permeability and poor injection capacity of shale reservoirs, the traditional methods of enhanced oil recovery based on water flooding or gas flooding are difficult to apply. As an important technology to enhance oil recovery, miscible gas flooding technology is widely accepted [5]. Miscible gas injection has been shown to be a practical technique for improving ultimate oil recovery in liquid-rich shale areas. Mohagheghian et al [6] have shown that shale surface is easier to adsorb CO_2 than other gases, which is one of the important mechanisms of CO_2 storage. Yu et al. [7] pointed out that tight oil reservoirs with low permeability are more conducive to CO_2 huff-n-puff process. Therefore, CO_2 Huff-n-Puff has become a promising technology for enhanced oil recovery and CO_2 storage.

The burial depth of shale formations leads to high pressure and high temperature conditions in their reservoirs. Therefore, it is challenging and economical to study oil and gas transport behavior through experimental methods under reservoir conditions [8]. For this reason, scholars mostly use simulation methods to study. Lashgari et al. [4] studied the oil recovery mechanism of CO_2 injection in shale reservoirs by numerical simulation model. The results show that oil-gas miscibility, CO_2 adsorption and gas diffusion are important physical mechanisms for shale oil recovery by CO_2 injection. The mechanism of CO_2 and crude oil miscible enhanced oil recovery mainly includes : reducing crude oil viscosity, reducing interfacial tension, crude oil expansion mechanism, alleviating gas channeling, etc. Zhang et al. [8] characterized the miscibility and displacement of CO_2 and crude oil in the nanoslits of three minerals (quartz, calcite and kaolinite) by molecular dynamics simulation (MD). The study determined that the oil migration mode was affected by the interaction between CO_2 and solid surface. In multi-component systems, oil molecules are more likely to conduct two-way mass transfer between oil and gas. The results of CO_2-EOR mainly depend on the mixture of crude oil and CO_2. Barisik et al. [9] and Karniadakis et al. [10] found that adsorbed CO_2 molecules change the interaction between oil and pore walls, and they may also change oil transport in nanopores of unconventional reservoirs.

As a popular mesoscopic dynamics method, the Lattice Boltzmann method (LBM) has been used as an alternative to numerical simulation in the past few years to simulate physical phenomena in fluid flow by combining microscopic and macroscopic physical mechanisms. It can not only improve the computational efficiency, but also deal with the complex structural boundaries when simulating fluid flow in porous media. In particular, it has high efficiency in dealing with complex pores, and has great potential for the study of MCMP flow modeling [11，12]. Most importantly, it can capture some fluid-fluid and fluid surface molecular interactions [13,14], which is in line with our research on the mechanism of intermolecular interactions during CO_2 huff-n-puff. At present, most studies are about the migration behavior of shale gas in nanoscale, while the specific migration behavior and mechanism of shale oil in nanoscale porous media are rarely studied. Zhang et al. [15] used the generalized lattice Boltzmann method (GLBM) to simulate the migration behavior of oil in nanopores, and verified the model by MD simulation data. Wang et al. [16] established a nano-scale multiphase multi-component LBM (MCMP-LBM) model to simulate the diffusion, adsorption and miscible flow behavior of CO_2-oil in nanoporous media, and successfully verified by MD method.

According to previous studies, shale matrix contains inorganic and organic matter. Its heterogeneous wettability leads to different adsorption of oil and gas by shale. This will have a greater impact on crude oil production. When there is a certain amount of water in the matrix, the influence of different adsorption of organic matter and inorganic matter on fluid on crude oil exploitation will be further enhanced. The traditional view is that a large amount of fracturing fluid retention in the formation will significantly reduce the relative permeability of oil and gas, resulting in reservoir damage and affecting oil well productivity. However, for shale reservoirs, according to field statistics, shale reservoirs with higher water content have higher crude oil recovery [17].

Therefore, in order to better study the effect of different adsorption of organic matter and inorganic matter on CO_2 huff and puff, based on the previous research, we used the LB method to establish a CO_2 huff and puff model that can characterize the different adsorption of organic matter and inorganic matter in shale on oil, water and gas. The model includes multiple relaxation model and Shan-Chen model. It can be used to simulate the diffusion and miscible in the process of CO_2 huff and puff. Most importantly, it can well reflect the properties of organic matter and inorganic matter in shale.In this study, we simulated CO_2 huff and puff mining at different development stages of the reservoir according to different water cuts. This paper provides theoretical guidance for the field application of CO_2 huff-n-puff technology in unconventional oil and gas exploitation and CO_2 storage technology.

2. PROBLEM DEFINITION AND MATHEMATICAL MODEL

2.1 Problem statement

In this study, according to the real shale structure, a shale model containing cracks and matrix is set as shown in **Fig.1**. The matrix part is divided into two parts : organic matter and inorganic matter. According to the wettability difference between organic matter and inorganic matter, we set nanopores as oil-wet, macro pores and micro pores as water-wet. At the same time, we set up micro pores and nano pores to adsorb CO_2, while macro pores repel CO_2. The three pore diameter ratios are 5:2:1. The upper part of the fracture and the left, right and lower parts of the matrix are set as rocks (shown in the red part), and the left and right sides of the fracture are open. We also consider the combined effects of oil and gas diffusion, miscibility, competitive adsorption and other factors in the process of CO_2 huff and puff. The model lattice unit data are shown in **Fig.1**, where 1 lu represents 1 nm.

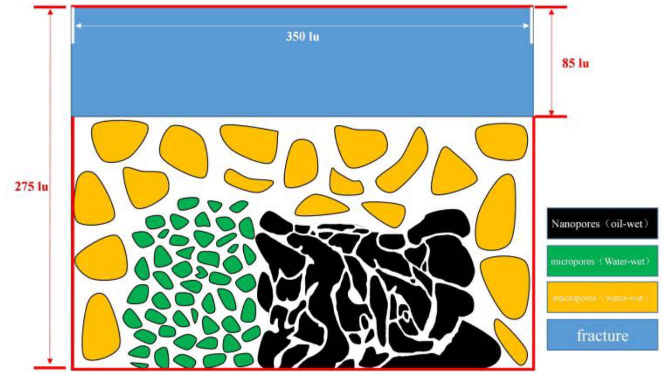

Fig. 1. Shale structure diagram (including fracture, matrix (organic matter, inorganic matter)).

2.2 Lattice Boltzmann model

In previous studies, we established a nano-scale LBM model of oil-water-gas three-phase, considering the inter-component force, cohesion, and fluid-solid force. In this study, we improved the previous model by using the multiple relaxation model (MRT) and Shan-Chen model to establish a CO_2 huff and puff model including oil-water-gas three-phase. The characterization methods of diffusion coefficient and adsorption in the new model are as follows.

2.2.1 Diffusion coefficient

Based on the Shan-Chen model [18,19], three kinds of forces are mainly considered in the multiphase multi-component system : fluid-fluid interaction force, fluid-solid wall interaction force and bulk phase force. The force between fluids is calculated as follows :

$$F_{int,\sigma}(x,t) = -\rho_\sigma(x,t) G \sum_\alpha w_\alpha \rho_{\bar{\sigma}}\left(x + e_\alpha \delta_t\right) e_\alpha \qquad (1)$$

where G is the interaction strength between different components. In the LBM simulation, the diffusion and miscibility between CO_2 and oil can be achieved by changing the value of *G*. According to the study of Wang et al., the diffusion coefficient D_c of CO_2 is negatively correlated with the oil-CO_2 interaction parameter G_{og}, while the miscible degree (or interfacial tension) is positively correlated with the oil-CO_2 interaction parameter G_{og}. Therefore, the diffusion coefficient of oil and gas can be controlled by the force between fluids.

2.2.2 adsorption

The fluid-wall interaction parameters can be used to control the wall wettability and the attraction or repulsion of the wall to the fluid molecules [20] :

In the formula, G_{ads} controls the fluid-surface interaction strength, that is, the fluid-solid interaction parameter. The positive value is repulsion and the negative value is attraction. It can be used to simulate the adsorption parameters in the model.

2.3 Boundary conditions

In this study, a half-way rebound boundary condition [21] was used for the solid boundary, and periodic boundary conditions [22] were used on the left and right sides of the fracture.

2.4 Initial oil-water distribution

The engineering background of this study is CO_2 huff and puff at different stages of reservoir development. Aiming at the water content of different development stages of the reservoir and the heterogeneous wettability of organic matter and inorganic matter, we first carried out oil-water phase separation simulation for the matrix part. In this study, we set the density of crude oil to 0.81, the density of water to 1.04, and the density of CO_2 in fractures to 1.2 (dimensionless density). Oil and water were randomly distributed in the matrix, and the proportion of total oil and water content was different (water content was 0.1,0.3,0.5,0.7). Due to the influence of heterogeneous wettability, when the model reaches stability, the initial oil-water distribution with different water content can be obtained. As shown in **Fig.2**, the oil-water density distribution at a water content of 0.3. The water in organic matter and inorganic matter has different wetting angles. The water content in organic matter is much less than that in inorganic matter.

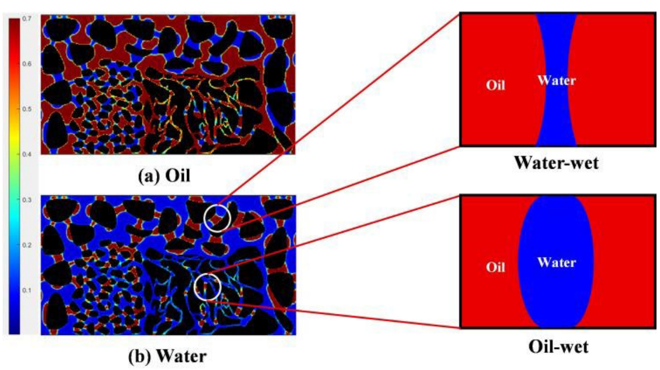

Fig. 2. Dimensionless density distribution in matrix (Heterogeneous wettability).

$$\mathbf{F}_{\mathrm{ads},\sigma}(\mathbf{x},t) = -G_{\sigma s}\psi(\mathbf{x},t)\sum_{\alpha} e^{-|\mathbf{x}-\mathbf{x}_{ads}|/\lambda}\mathbf{e}_{\alpha} \qquad (1)$$

3. RESULTS AND DISCUSSION

3.1 Mechanism of CO_2 huff and puff under water condition

According to our previous studies, the interaction force between CO_2 molecules and crude oil molecules under miscible conditions can affect the diffusion coefficient D_c of CO_2. In this paper, the diffusion rate of CO_2 is controlled by CO_2-crude oil interaction parameter (G_{og}).

With the decrease of G_{og}, the smaller the force between oil and gas molecules, the greater the diffusion rate. The size of the fracture will affect the contact area of oil and gas, and then change the rate of oil and gas miscible, and ultimately affect the effect of CO_2 huff and puff. Based on the above research results, we simulated CO_2 huff and puff at different development stages. As shown in **Fig.3**, when the water content is 0.3, the oil and gas density and three-phase distribution figue change with time.

When T=1, CO_2 contacts with crude oil at the bottom of the fracture. Because CO_2 is difficult to dissolve in water, CO_2 diffuses into crude oil mainly through the middle channel of the pore. When T=500, CO_2 is miscibled with crude oil and enters the matrix in large quantities. Due to crude oil expansion, wettability and pressure difference. The water in the macro pores is driven by a large amount of CO_2, moving in the form of a liquid bridge to the water-wet micron pores. When T=1000, the main migration channels of CO_2 and water are formed. In the whole process of huff and puff, the main mechanisms are:oil-gas miscibility, diffusion, adsorption, oil-water phase separation, crude oil expansion, etc. As shown in **Fig.4**, it is the main migration channel of CO_2 and the migration path of water when the water content is 0.3.

When T=500, there is still a lot of water in the macro pores. Due to the downward migration of water under pressure and the influence of heterogeneous wettability, when T=30000, the main water-bearing area becomes a micro pore area. The nanopores contain only a very small amount of water. Due to the repulsion of macro pores to CO_2, CO_2 mainly migrates through the middle of the pore channel with large pores and no obstruction. Due to the water wettability of inorganic matter, the water in the fracture migrates in the form of adsorbed water film on the rock wall, and accumulates in the form of liquid bridge between rock particles along the green arrow direction to the micro pore area. Oil in micron pores gradually migrates to nanopores and macro pores due to the large accumulation of water phase. The crude oil in the macro pores diffuses into the fractures.

the matrix with larger fractures at the same time. In this study, CO_2 and water are not miscible. The effect of water is similar to that of smaller fractures, both of which hinder the contact area between CO_2 and crude oil. **Fig.6** shows the density distribution of CO_2 when T=30000. The boundary of CO_2 density of 0.6 was selected for comparison. When the water content was 0.1, the diffusion degree of CO_2 was significantly greater than that when the water content was 0.3. When the water content is 0.5, the diffusion of CO_2 into the matrix is very small, and most of it is blocked by water. Therefore, only the limit of CO_2 density of 0.7 can be found in the figure.

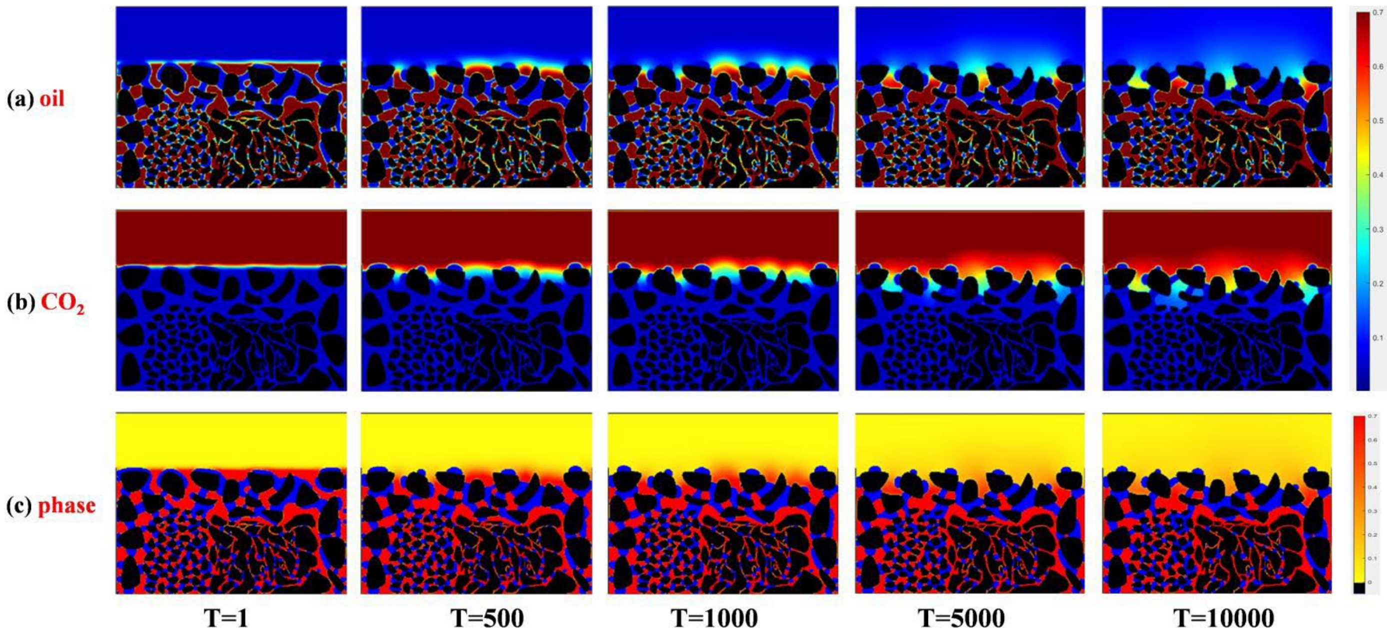

Fig. 3. Oil-water density and three-phase distribution change with time.

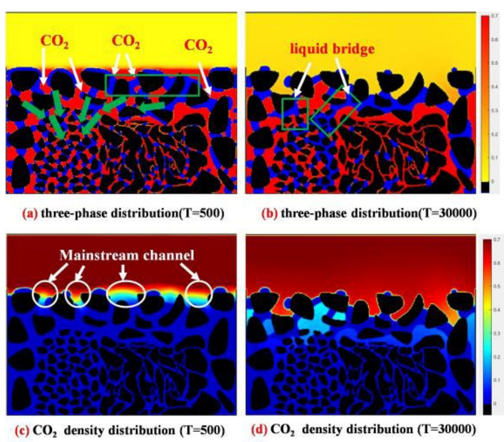

Fig. 4. The main migration pathways of CO_2 and water.

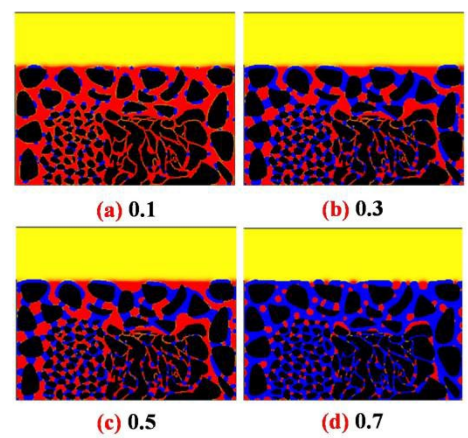

Fig. 5. Initial oil-water-gas three-phase distribution under different water content conditions.

3.2 Effect of different water content on CO_2 huff and puff mechanism

In order to simulate CO_2 huff and puff in different development stages of the reservoir, we set different initial water content, and the oil-water-gas three-phase distribution is shown in **Fig.5**.

According to our previous study on the effect of fracture size on CO_2 huff and puff, fracture size affects the contact area between crude oil in the matrix and CO_2 in the fracture, which in turn affects the rate of oil and gas miscible and diffusion, resulting in more CO_2 entering

3.3 Effect of different water content on CO_2 huff and puff

In order to more clearly study the effect of water content during CO_2 huff and puff, we calculated the

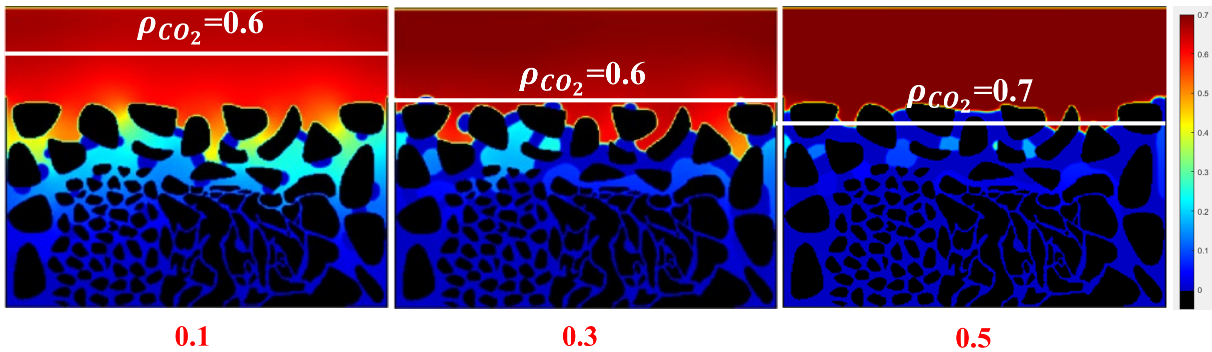

Fig. 6. Comparison of CO_2 density distribution with different moisture content.

crude oil recovery and CO_2 storage rate in each pore area and the total, and compared with the recovery and storage. **Fig.7** shows the oil-water-gas distribution map and crude oil density distribution map at T=30000 under different water cut conditions.

When the water content is 0.1, the matrix is basically a mixture of crude oil and CO_2. When the water content is 0.3, micro pores and nano pores are the main oil-bearing areas. When the water content is 0.5 and 0.7, nanopores are the main oil-bearing areas. **Fig.8** shows the curves of crude oil recovery, recovery, CO_2 storage rate and CO_2 storage with time in different regions.

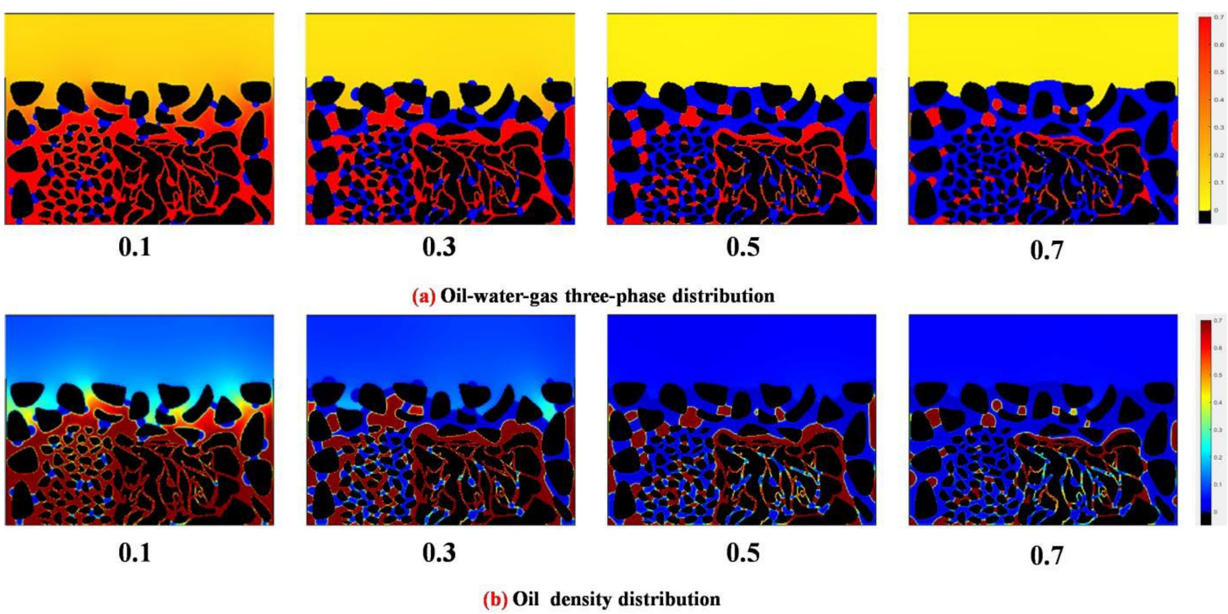

Fig. 7. Oil-water gas distribution and crude oil density distribution when T=30000.

When the water content is 0.1, similar to our previous study, the crude oil in the macro pores area is fully miscible with CO_2 and diffuses into the fracture. Under the action of pressure, the crude oil at the lower part of the macro pores will enter the micro-pores and nano-pores, resulting in the increase of crude oil content. When the water content is 0.3, due to the increase of water content and the micro-pores are wet, the water squeezed downward by CO_2 in the macro pores accumulates in the micro-pores, while the crude oil is repelled into the nano-pores or mixed with CO_2, and diffuses into the macro poress, and then is produced into the fractures. When the water content is 0.5, the phenomenon in micron pores and nano pores is similar to that when the water content is 0.3, but it is more affected by water. In order to more intuitively see the mining effect of each region, we drew a histogram of the mining effect of each region as shown in **Fig.9**.

It can be seen that water has two main effects on CO_2 huff and puff in this model. Firstly, the presence of water affects the contact area and migration channel of oil and gas. With the increase of water content, the effect of CO_2 on crude oil exploitation is weakened. The other is that the aggregation of water in the inorganic substance will reduce the crude oil content in the inorganic substance. With the increase of water content, the crude oil extraction effect in the inorganic substance increases. When the water content is 0.5, the comprehensive effect of the two effects is the worst, so the total oil recovery is the lowest when the water content is 0.5. When the water content is 0.7, the second effect is the strongest, and the mining effect in macro pores and micro pores (inorganic matter) is better than that in water content of 0.3 and 0.5. This is consistent with the data of some on-site shale oil exploitation. When the water content is high, the recovery rate of crude oil increases instead. We speculate that when shale contains more micron pores in this study, its ultimate recovery will be higher.

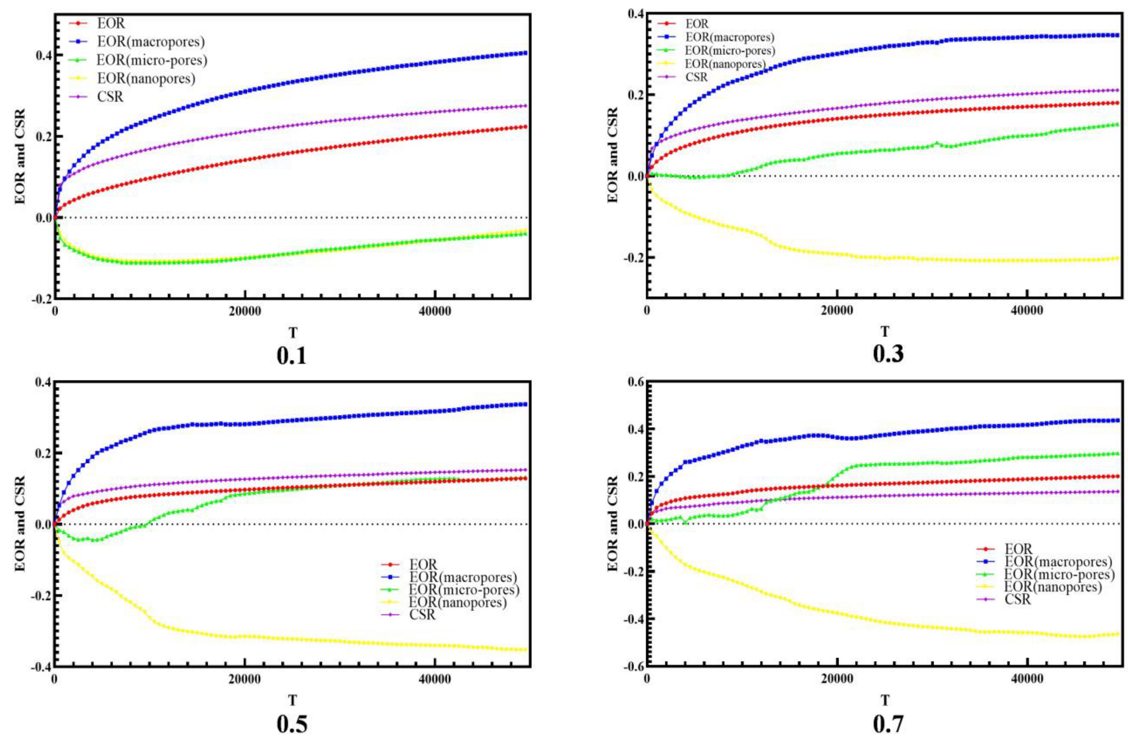

Fig. 8. Oil-water gas distribution and crude oil density distribution when T=30000.

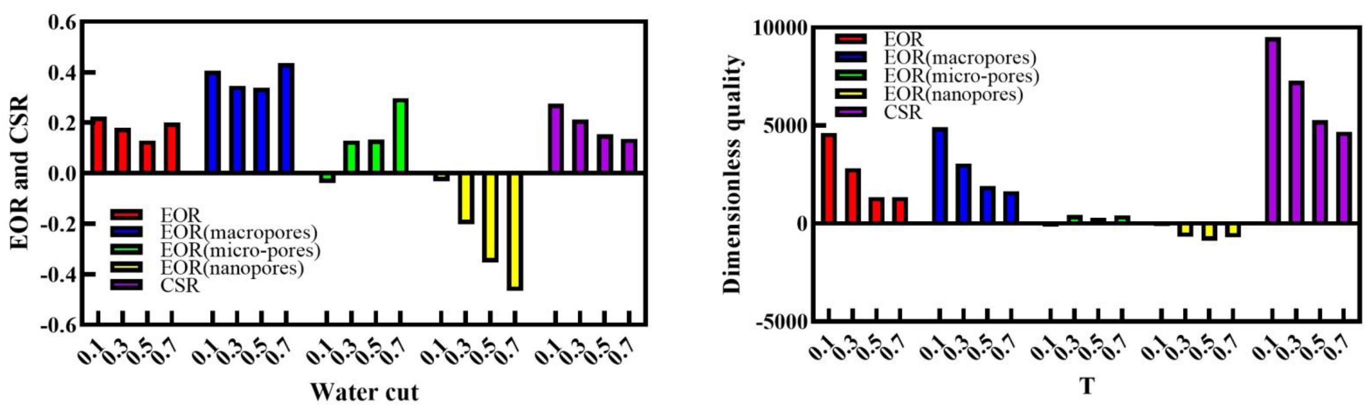

Fig. 8. Oil-water gas distribution and crude oil density distribution when T=30000.

4. CONCLUSIONS

In this work, we used the Lattice Boltzmann method to establish a CO_2 huff and puff model that can characterize the different adsorption of organic matter and inorganic matter in shale on oil, water and gas. The model includes multiple relaxation model and Shan-Chen model. We simulated CO_2 huff and puff mining at different development stages of the reservoir according to different water cuts.

（1）The heterogeneous wettability of our shale model was verified by phase separation simulation in the matrix. The water content in inorganic matter is much more than that in organic matter. Its oil-water distribution conforms to the actual conditions.

（2）Due to the repulsion of macro pores to CO_2, CO_2 mainly migrates through the middle of the pore channel with large pores and no obstruction. Due to the water wettability of inorganic matter, the water in the fracture migrates in the form of adsorbed water film on the rock wall, and accumulates in the form of liquid bridge between rock particles along the green arrow direction to the micro pore area.

（3）The results show that the adsorption quantity of CO_2 is reduced due to the stronger adsorption capacity of water molecules on the surface of inorganic rock particles, and the quantity of CO_2 entering the matrix in the fracture is reduced. The contact area between CO_2 and crude oil decreases with the increase of water content, and the miscibility of oil and gas slows down. We also found that the increase of water content will promote oil recovery in inorganic matter. But it will also reduce the contact area of oil and CO_2. When the water content is 0.5, the two effects are the weakest, and the oil recovery is the smallest.

This paper provides theoretical guidance for the field application of CO_2 huff and puff technology in unconventional oil and gas exploitation and CO_2 storage technology..

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DECLARATION OF INTEREST STATEMENT

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. All authors read and approved the final manuscript.

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