Study on the Ways to Improve the CO₂–H₂O Displacement Efficiency in Heterogeneous Porous Media by Lattice Boltzmann Simulation

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ABSTRACT: To improve the efficiency of CO₂ geological sequestration, it is of great significance to in-depth study the physical mechanism of the immiscible CO₂–water displacement process, where the influential factors can be divided into fluid–fluid and fluid–solid interactions and porous media characteristics. Based on the previous studies of the interfacial tension (capillary number) and viscosity ratio factors, we conduct a thorough study about the effects of fluid–solid interaction (i.e., wettability) and porous media characteristics (i.e., porosity and non-uniformity of granule size) on the two-phase displacement process by constructing porous media with various structural parameters and using a multiphase lattice Boltzmann method. The displacement efficiency of CO₂ is evaluated by the breakthrough time characterizing the displacement speed and the quasi-steady state saturation representing the displacement amount. It is shown that the breakthrough time of CO₂ becomes longer, but the quasi-steady state saturation increases markedly with increasing porosity, granule size, and non-uniformity, showing the improvement of the displacement efficiency. Furthermore, the breakthrough time of CO₂ shortens and the saturation increases significantly with increasing porosity, granule size, and non-uniformity, showing the improvement of the displacement efficiency. Therefore, enhancing the wettability of CO₂ with the surface and selecting reservoirs with greater porosity, larger granule size, and non-uniformity can all contribute to the efficiency improvement of CO₂ geological sequestration.

1. INTRODUCTION

CCS (carbon capture and sequestration) is currently recognized as an effective technology that directly reduces CO₂ emissions worldwide.1−3 Injection of CO₂ into deep saline aquifers is known to be the most feasible option for carbon storage, which has the maximum accessibility and highest storage capacity.4−7 Under the conditions of typical underground saline aquifers, CO₂ usually remains in the supercritical state or liquid state.8 When CO₂ is injected into deep saline aquifers, it displaces the formation fluid from the pore space in a complex pattern dominated by capillary and viscous forces, as well as geological heterogeneities. It is of great significance to in-depth study the physical mechanism of immiscible multiphase flow to evaluate the storage capacity of saline aquifers and improve the efficiency of CO₂ geological sequestration.

For the immiscible two-phase displacement process in porous media, the influencing factors can be divided into fluid–fluid and fluid–solid interactions and porous media characteristics.

The influencing factors on the fluid–fluid interaction mainly include the viscosity ratio, interfacial tension (capillary number), and density ratio between the two-phase fluids. For the displacement of water by CO₂, the viscosity ratio and capillary number are the main influencing factors.9 The capillary number characterizes the relationship between viscous force and interfacial tension, defined as Ca = (ηu)/γ, where η, u, and γ are the dynamic viscosity, inlet velocity, and interfacial tension of the fluid, respectively. The viscosity ratio between two fluids is defined as M = μ₂/μ₁, where μ₂ and μ₁ are the dynamic viscosities of the invading fluid and the displaced one, respectively. With the increase in Ca or decrease in M, the capillary pressure is reduced during the flow, which helps stabilize the displacement front surface, allows the invading fluid to invade more pores, and thus improves the displacement efficiency.10−14

In the process of displacement, immiscible fluids will adhere to the solid surface of the porous media and produce competition, manifesting by the wettability of the coupling between the fluid–fluid interface and the solid surface, i.e., the
2. NUMERICAL METHOD

2.1. Multiphase Lattice Boltzmann Method. The SCMC-LBM (Shan-Chen Multicomponent-LBM) model has been widely used in multiphase flow in porous media. In the present study, the two-phase flow of immiscible fluids is modeled using the SCMC-LBM model. A two-dimensional D2Q9 is considered. The particle distribution functions satisfying the lattice Boltzmann equation are introduced for each fluid phase. The fluids are subjected to the combined action of external forces during the flow, including the body force $G$, the interaction forces $F_{\text{int}}$ between fluid and solid, and $F_{\text{ext}}$ between fluid and fluid.

2.2. Initial and Boundary Conditions. The computational domain is $500 \times 500$ lu$^2$ (lu: lattice unit), where CO$_2$ is injected from the left side and outflows through the right side. At the initial time, the pore space of the calculation domain $(0-1 \text{ lu})$ is filled with CO$_2$ and the rest with water, both with zero initial velocities. The inlet velocity and outlet pressure boundary conditions are given by Zou–He relations, where the nonzero inlet velocity is $u_x$ in the flow direction. The nonslip bounce-back boundaries are applied to the top, bottom, and internal skeleton structure of porous media.

2.3. Samples of Porous Media. For the two-phase flow, the bounce-back boundary condition used by the LBM method requires a certain number of lattices between solids, and too few lattices will lead to numerical instability, resulting in scattering or low accuracy. Therefore, the minimum channel width in the physical model of porous media should be greater than four lattices. As a result, the applicability of the LBM for the two-phase flow depends on the minimum channel width. Based on the above-mentioned requirement and the reconstruction method of the porous media in this paper, the solid granules are not in contact with each other so that the pore spaces are completely connected, and the porosity is limited to greater than 0.6 in the simulation. The flow mechanism with lower porosity could be inferred according to the numerical simulations.

In summary, the factors of the fluid–fluid interaction have been more comprehensively studied, such as the capillary number. However, the research on the factors affecting the fluid–solid interaction is not comprehensive enough, which needs to be further studied, including the wettability of CO$_2$ to the solid surface. Furthermore, the study of the influencing factors of the porous media characteristics in the heterogeneous case is less, i.e., non-uniform granule size.

The researchers use a variety of methods to study the two-phase displacement process, including experiments and numerical simulations. Due to the limitation of experimental conditions, numerical simulation is much more effective in predicting multiphase flow in porous media and supplementing experimental conditions. Compared with other numerical methods, the LBM (lattice Boltzmann method) at mesoscopic scales, which can handle various complex geometric boundaries and accurately capture the evolution of phase interfaces, has great advantages in solving complex fluid flow problems. Therefore, the LBM is applied to investigate the influence of fluid–solid interaction and porous media characteristics on the immiscible two-phase displacement process and efficiency, specifically including the wettability, porosity, granule size, and non-uniformity of the heterogeneous porous media.
The 500 × 500 lu² computational domain corresponds to the physical size of 5 × 5 mm², with each lattice being 10 μm. The permeability $k$, an inherent property of porous media independent of the properties of the fluid, is calculated by Darcy’s law as $k = \frac{u_d \mu \nu}{\Delta p}$, where $u_d$ is the average velocity in the flow field, $\nu$ is the kinematic viscosity of the fluid, and $\Delta p$ is the pressure difference at the inlet and outlet.

The 15 samples of porous media are presented in Table 1, where samples 1–5 contribute to the study of porosity factor for the same average granule size, samples 11–15 contribute to the study of uniform granule size factor for the same porosity, samples 3, 7, 8, 9, and 10 contribute to the study of average granule size factor for the same porosity, and samples 3, 6, and 11 contribute to the study of non-uniformity factor for the same porosity and average granule size.

Figure 2 visually shows the porous media structures of samples 1, 3, 6, and 11. By comparing the four samples, although the average granule sizes are the same, the porosity or granule size is different, manifesting various skeleton structures.

### 2.4. Model Validation.

The basic code is improved by adding the fluid–fluid force and fluid–solid force and correcting the equilibrium velocity of each fluid component based on the two forces through the C++ language in the Linux environment. The SCMC-LBM model is validated using two standard benchmarks tests: the Young–Laplace test and the static contact angle test.

#### 2.4.1. The Young–Laplace Test.

For the SCMC-LBM model, the interfacial tension $\gamma$ between the two fluid components needs to be indirectly calculated using the droplet

### Table 1. Characteristics of Porous Media

| sample | porosity $\varepsilon$ | granule size $D$/($\mu$m) | dimensionless granule size range $D^* \times 10^3$ | average granule size $D$/($\mu$m) | dimensionless average granule size $D^*$ | permeability $k$/($\mu$m$^2$) |
|--------|------------------------|--------------------------|-----------------------------------------------|-----------------------------------|-----------------------------------------|-------------------------------|
| 1      | 0.60                   | 30, 40, 50, 60, 70       | 6, 8, 10, 12, 14                              | 49.98                            | 0.01                                    | 59.16                         |
| 2      | 0.62                   |                          |                                               | 49.99                            |                                         | 70.82                         |
| 3      | 0.65                   |                          |                                               | 49.99                            |                                         | 99.45                         |
| 4      | 0.65                   |                          |                                               | 49.98                            |                                         | 154.04                        |
| 5      | 0.70                   |                          |                                               | 49.99                            |                                         | 180.14                        |
| 6      | 0.65                   | 10, 20, 30, 40, 50, 60, 70, 80, 90 | 2, 4, 6, 8, 10, 12, 14, 16, 18 | 49.99                            |                                         | 141.23                        |
| 7      | 0.65                   | 40–80                    | 8, 10, 12, 14, 16                              | 59.99                            | 0.012                                   | 142.03                        |
| 8      | 0.65                   | 50–90                    | 10, 12, 14, 16, 18                             | 69.98                            | 0.014                                   | 196.04                        |
| 9      | 0.65                   | 60–100                   | 10, 12, 14, 16, 18, 20                         | 79.98                            | 0.016                                   | 254.77                        |
| 10     | 0.65                   | 70–110                   | 14, 16, 18, 20, 22                             | 89.97                            | 0.018                                   | 335.79                        |
| 11     | 0.65                   | 50                       | 10                                             | 50                                | 0.01                                    | 93.28                         |
| 12     | 0.65                   | 60                       | 12                                             | 60                                | 0.012                                   | 126.47                        |
| 13     | 0.65                   | 70                       | 14                                             | 70                                | 0.014                                   | 174.10                        |
| 14     | 0.65                   | 80                       | 16                                             | 80                                | 0.016                                   | 222.59                        |
| 15     | 0.65                   | 90                       | 18                                             | 90                                | 0.018                                   | 304.61                        |
The Young–Laplace test is to verify that the model developed can accurately determine the interfacial tension at various fluid–fluid interaction strength coefficients $G_c$.

The 200 × 200 lu² droplet model is established, where the periodic boundaries are applied in the x- and y-directions of the simulation domain. The $G_c$ values are 0.8, 0.9, 1.0, and 1.1, respectively. Five different droplet radii are initialized as $R_i = [20, 25, 30, 40, 50]$. The density ratio of water to CO2 is set to 1, and the viscosity ratios are set to 8. The relationship between the pressure difference across the interface and the final size of the droplet is determined, as shown in Figure 3.

![Figure 3](image.jpg)

**Figure 3. Relationship between $\Delta P$ and $1/R$.**

The simulation results satisfy Laplace’s law. The slope of the fitting line is the interfacial tension increasing with increasing fluid–fluid interaction strength coefficient $G_c$. As observed in Figure 3, the interfacial tension at different $G_c$ values corresponds to 0.104, 0.128, 0.151, and 0.175.

### 2.4.2. The Static Contact Angle Test

The static contact angle test is to confirm that the established model can accurately determine the contact angle $\theta$ between two fluid components and the solid surface under different fluid–solid interaction strength coefficients $G_{ads}$. A 300 × 200 lu² computational domain is selected in which supercritical CO2 and water are placed above a solid surface and different wettability values are considered. The main and associated dissolved densities of both fluids are set to $\rho_{main} = 2.0$ and $\rho_{dissolve} = 0.06$, respectively, and the $G_c$ is 0.9. The half-step bounce-back boundary conditions (no-slip zero velocity) are implemented at the top and bottom walls and the periodic boundary conditions for both left and right boundaries.33

Under the effect of interfacial tension and the interaction between the solid surface and the fluid components, the contact angle can be measured when the steady state of the fluids is finally reached.

Simulation results of three typical static contact angles are shown in Figure 4, where blue indicates supercritical CO2 as fluid component 1 ($\sigma = 1$), and red indicates water as fluid component 2 ($\sigma = 2$). The contact angles formed at the bottom boundary for fluid components are labeled as $\theta_1$ and $\theta_2$, respectively. The two-phase fluid exhibits opposite wettability to the surface so that $\theta_1 + \theta_2 = 180^\circ$. For water, the three typical simulations are as follows: (a) high, (b) neutral, and (c) low wetting, as demonstrated in Figure 4.

The relationship between the contact angle $\theta_2$ and $G_{ads}$ is shown in Figure 5 for water. It is found that the simulation results are in good agreement with the analytical solution proposed by Huang et al.33 i.e.,

$$\cos \theta = \frac{G_{ads,1} - G_{ads,2}}{G_{ads,1} + G_{ads,2}}$$



![Figure 4](image.jpg)

**Figure 4. Interaction of two fluids with a surface: (a) $G_{ads} = -0.4$, $\theta_2 = 26.8^\circ$; (b) $G_{ads} = 0$, $\theta_2 = 90^\circ$; (c) $G_{ads} = 0.4$, $\theta_2 = 153.5^\circ$ (red, water; blue, CO2).**

![Figure 5](image.jpg)

**Figure 5. Relationship between $G_{ads}$ and contact angle $\theta_2$.**

### 3. RESULTS AND DISCUSSION

During the simulation, the viscosity ratio $\gamma$ between water and CO2 is 8, the inlet velocity $u_i$ is supposed to be $10^{-3}$, $G_c$ and corresponding interfacial tension $\gamma$ are 0.9 and 0.128, respectively, and dynamic viscosity $\eta_{CO2}$ is 0.1. The study is carried out under $Ca = 7.81 \times 10^{-4}$. Neglecting the effect of gravity, the density ratio of CO2 to water taken as 1 (i.e., $\rho_{CO2} = \rho_{H2O}$) has little effect on the results.40 The contact angles $\theta_2$ of water and the surface are $26.8^\circ$, $45.1^\circ$, $62.2^\circ$, $74.2^\circ$, $90^\circ$, $104.7^\circ$, $117.8^\circ$, $133.6^\circ$, and $153.5^\circ$, respectively.
In this section, the effects of the surface wettability, porosity, and granule size of porous media on the CO₂–water displacement process are discussed. The displacement efficiency is evaluated by two parameters. One is the breakthrough time of CO₂, \( t_{b} \), defined as the time required for the invading fluid to reach the outlet. The other is the saturation of CO₂, \( S_{CO₂} \), at a quasi-steady state, defined as the percentage of CO₂ in pore volume when the saturation difference of CO₂ per 10⁵ iterations is less than 10⁻³.

### 3.1. Effect of Surface Wettability

A series of numerical simulations are conducted to study the influence of surface wettability on the CO₂–water displacement process. The displacement process takes place in the porous media of sample 3, where the porosity and average granule size are 0.65 and 0.01, respectively.

Figure 6 shows the flow process of CO₂ (red) in pores under different surface wettability values. Approximately at \( 0° < \theta_2 < 75° \), the solid surface exhibits hydrophilic properties so that water is prone to adhere to the solid surface and form a liquid film. Therefore, it is easier for CO₂ to flow along the center of the pore channel. The irregularity of the porous media leads to different capillary pressures in each pore. The CO₂ front preferentially forms multiple protrusions and invades some large pore channels in porous media and also merges and separates continuously. Gradually, the dominant channels are large pore channels in porous media and also merges and preferentially forms multiple protrusions and invades some pore bodies (the second type of residual water, II), and a very minor amount of residual water adheres to the solid surface in the form of a water film (the third type of residual water, III). Since the driving force needs to overcome the capillary pressure of the throat, the water clusters are very difficult to remobilize once trapped by CO₂ in the throats. In the hydrophilic case, for example, \( \theta_2 = 26.8° \) in Figure 7a, a certain amount of CO₂ can be observed to form a continuous flow path in the form of thin and long linked large clusters. Three types of residual water exist in the porous media simultaneously. The second type of residual water exists in the throats. The reason is that CO₂, as a nonwetting phase, needs to overcome large capillary breakthrough pressure when entering the throat due to the smaller geometric width of the throat. Therefore, the nonwetting phase tends to occupy larger pores rather than the smaller ones. Unlike the pore-fluid distribution observed in the highly hydrophilic case, as shown in Figure 7b at \( \theta_2 = 90° \), it depicts a wide continuous flow path of CO₂. In addition, both CO₂ and residual water can occupy the pore bodies or throats, demonstrating that the form of the second category of residual water has changed. In addition, the third type of residual water has also been observed in this case. Figure 7c shows the hydrophobic case, for example, \( \theta_2 = 153.5° \), where CO₂ sweeps the flow channel in a large area, the second type of residual water only occupies the pores, and the third type of residual water is not presented.

Figure 7 demonstrates the variation of the CO₂ saturation in porous media over time with different wettability values. At the same contact angle, the growth process of CO₂ volume fraction can be divided into three stages: (1) rapid growth stage; (2) slow growth stage; (3) stabilization stage. In the early stage of the displacement process, the available pore space of CO₂ in the porous media is relatively large, and the saturation of CO₂ in the pores increases rapidly. When CO₂ flows out of the outlet, CO₂ flows preferentially along the dominant channel. However, CO₂ is still able to break through some small pores due to the action of viscous force, leading to a slow growth rate of its saturation. At the later stage of the displacement process,
the inflow and outflow of CO₂ are basically stable and its saturation remains steady. In addition, with the enlargement of the contact angle, the surface changes from hydrophilic to hydrophobic, and the duration of the rapid growth stage and slow growth stage increases.

As shown in Figure 9, the breakthrough time and final saturation of CO₂ with the contact angle θ₂ confirm the function relationship. The breakthrough time t* of CO₂ prolongs with the increase in contact angle. Specifically, the breakthrough time t* of CO₂ increases from 0.161 to 0.254 with the change of the contact angle from 26.8° to 153.5°. The final saturation of CO₂ increases with the rise of contact angle from 54.35 to 77.4%. This can be attributed to the fact that when the porous media are hydrophilic, water forms a liquid film on the wall, which acts as lubrication, thus making CO₂ flow faster and shortening the breakthrough time. The smaller the contact angle is, the much stronger the adhesion of water to the surface will be. This adhesion makes it more difficult for CO₂ to displace water, resulting in a lower CO₂ displacement efficiency. Conversely, the flow channels in porous media with smaller porosity are narrower, while the inflow space is smaller. To sum up, with the porosity increasing, the flow becomes more continuous. Moreover, regardless of the porosity, the form of residual water in the porous media is still dominated by the first type. Also, a slight change in porosity will cause a large difference in the volume fraction of residual water in the pores.

In addition, as shown in Figure 10, for ε = 0.60, the case of θ₂ = 26.8° (hydrophilic) maintains a "fingering mode", and while the contact angle increases to 153.5° (hydrophobic), the flow pattern changes to the "stable displacement mode". The two modes coexist in the condition of neutral wetting. However, as ε increases from 0.60 to 0.70 for a fixed contact angle, the number of dominant channels established by CO₂ in the porous media rises and the sweep area widens significantly with the porosity increasing. At the same average granule size, the porous media with larger porosity have a smaller solid surface area, providing more pore channels for two-phase fluid flow. Conversely, the flow channels in porous media with smaller porosity are narrower, while the inflow space is smaller. To sum up, with the porosity increasing, the flow becomes more continuous. Moreover, regardless of the porosity, the form of residual water in the porous media is still dominated by the first type. Also, a slight change in porosity will cause a large difference in the volume fraction of residual water in the pores.
angle, the displacement mode of CO\textsubscript{2} is not affected by the change of porosity.

Figures 11 and 12 show the porosity as a function of CO\textsubscript{2} breakthrough time and final saturation under different $\theta_2$ values. For example, at $\theta_2 = 26.8^\circ$, the porosity of porous media increases from 0.60 to 0.70, the breakthrough time $t^*$ of CO\textsubscript{2} decreases from 0.314 to 0.094, and the final saturation increases from 48.74 to 58.63%. Similarly, when the porosity varies within the same range at $\theta_2 = 153.5^\circ$, the breakthrough time of CO\textsubscript{2} is shortened from 0.362 to 0.164, and the final saturation increases from 71.78 to 85.02%. This is because as the porosity increases for a fixed average granule size, it leads to the increase in mean pore throat size. As a result, the permeability of porous media increases significantly, and the resistance to the displacement process decreases. Therefore, the breakthrough time is shortened and the final saturation is higher.

Increasing the porosity of porous media can significantly reduce the breakthrough time and increase the final saturation of CO\textsubscript{2}. The results show that selecting reservoirs with higher porosity is helpful in improving the CO\textsubscript{2} displacement efficiency.

3.3. Effect of Granule Size. The porous media with uniform granule size are the simplest model to study the effect of granule size on the displacement process, but they cannot reflect the heterogeneity of the real porous media. The non-uniform porous media have a variety of granule sizes, and the effect of granule size can be studied by controlling the overall average granule size and considering the non-uniformity of granule size, which better reflects the heterogeneity of the real situation.

3.3.1. Effect of Uniform Granule Size. The samples of porous media are 11, 12, 13, 14, and 15 with the granule sizes of 0.01, 0.012, 0.014, 0.016, and 0.018, respectively, when the porosity is 0.65. For uniform granule sizes, that is, the size of solid granules in porous media is consistent, and the effect of different granule sizes on the CO\textsubscript{2}−water displacement process is studied.

Figure 13 illustrates the distribution of CO\textsubscript{2} in porous media at different granule sizes and wettability values when the displacement process reaches the quasi-steady state. When $\theta_1 = 26.8^\circ$ and $D^* = 0.01$, much CO\textsubscript{2} flows along the surface, which causes the water film on the surface to be very thin or even disappear, and CO\textsubscript{2} penetrates water along the center of the flow path only in part of the pores. In addition, in the flow process, it can be found that CO\textsubscript{2} breaks through a few small throats and is subjected to a large capillary pressure. As the average granule size increases, for example, when $D^* = 0.018$ for the fixed contact angle, it can be clearly observed from the figure that there are thick water films between the surface and CO\textsubscript{2} in several places, and CO\textsubscript{2} flows through the water layer along the center in several flow channels. The reason is that on the premise of the same wettability and porosity, the seepage network in the porous media with small granule size is
characterized by numerous exceedingly fine seepage channels; in addition, the proportion of main channels is low, and each channel is distorted.

From the results shown in Figure 13, it can be clearly observed that the residual water morphology is still dominated by the first type. The volume fraction of the first and second types of residual water can be significantly reduced by increasing the size of solid granules in porous media, but the volume fraction of the third type of residual water increases. However, the increase in the volume fraction of the third type of residual water is extraordinarily smaller compared to the decrease in the volume fraction of the first and second ones of residual water.

In the case of $D^* = 0.01$ and $\theta_2 = 26.8^\circ$, the porous media maintain a "fingerering pattern". Meanwhile, when $\theta_2 = 153.5^\circ$, the $\text{CO}_2$ displacement mode changes to the "stable displacement pattern". The coexistence of the two modes can clearly be captured in the condition of neutral wetting. As $D^*$ increases from 0.01 to 0.018, the displacement mode of $\text{CO}_2$ remains unchanged under the same wettability. The results show that the granule size has no effect on $\text{CO}_2$ displacement mode in porous media.

Figures 14 and 15 show the $\text{CO}_2$ breakthrough time and final saturation as a function of granule size under different $\theta_2$ values. At $\theta_2 = 26.8^\circ$, the granule size $D^*$ increases from 0.01 to 0.018, the breakthrough time of $\text{CO}_2$ is shortened from 0.168 to 0.05, and the saturation increases from 49.21 to 65.6%. At $\theta_2 = 153.5^\circ$, when the granule size changes within the same range, the breakthrough time of $\text{CO}_2$ is shortened from 0.283 to 0.073, and the saturation increases from 74.83 to 89.11%. When $\theta_2$ changes from 26.8° to 153.5°, the saturation values of $\text{CO}_2$ increase by 25.62% for $D^* = 0.01$, 24.18% for $D^* = 0.014$, and 23.51% for $D^* = 0.018$. The results show that under the same wettability, the breakthrough time of $\text{CO}_2$ is an approximately quadratic function, and the final saturation of $\text{CO}_2$ is linear with the granule size. The reason is that as the granule size increases, the channel becomes wider and straighter, the resistance of $\text{CO}_2$ during the flow process in porous media is reduced, and the flow velocity becomes faster, so the time to reach the outlet is shortened and the final saturation is higher.

As the granule size increases, the breakthrough time of $\text{CO}_2$ is significantly shortened and the final saturation increases. Moreover, compared with the number of flow channels in uniform porous media, the displacement efficiency of $\text{CO}_2$ is more sensitive to the width of the flow channels. Also, in the case of the same flow channel width, as the contact angle increases, the displacement efficiency of $\text{CO}_2$ increases.

### 3.3.2. Effect of Non-uniform Granule Size

In the study of Section 3.3.1, the granule size in the same porous media is consistent. In this section, we consider the case of non-uniform granule size, which is of multiple granules with different sizes in the same porous media. But it is ensured that the porosity and average size of the granules of the porous media are 0.65 and 0.01, respectively, consistent with the parameters used in Section 3.3.1, to study the influence of the non-uniformity of porous media on the $\text{CO}_2$–water displacement process. The porous media samples are 3, 7, 8, 9, and 10, with the average size ranging from 0.01 to 0.018.

Figure 16 illustrates the distribution of $\text{CO}_2$ in porous media at different average granule sizes when the displacement process reaches the quasi-steady state in the case of non-uniform granules. Comparing Figures 13 and 16, we find that under any fixed wettability, the morphological characteristics of residual water are consistent regardless of the granule size and average granule size for the flow characteristics of the displacement process, but the flow of $\text{CO}_2$ in non-uniform porous media is more continuous than that in uniform ones. As shown in Figure 16, at $\theta_2 = 26.8^\circ$, it can be clearly observed that the main channel of $\text{CO}_2$ is basically established along the vicinity of the large granule size, while the small granule size only disturbs the flow of the main channel. At $\theta_2 = 153.5^\circ$, the distribution of $\text{CO}_2$ in non-uniform porous media is more uniform, and the volume fraction of $\text{CO}_2$ adsorbed on the solid surface around the smaller pores is significantly reduced. The reason that can be described is that the non-uniformity of granule size affects the flow channel morphology so that large pores and wider channels appear near the large-size granule, and the resistance of $\text{CO}_2$ during the flow is reduced.
As shown in Figures 17 and 18, in non-uniform porous media, the breakthrough time of CO$_2$ and the average granule size still conform to the law of quadratic function, and the final saturation presents a nearly linear relationship. It is also found that the breakthrough time of CO$_2$ in non-uniform porous media is shorter, and the final saturation is higher than those in uniform porous media for other factors fixed. The reason is that the proportion of wider flow channels in non-uniform porous media increases relative to uniform porous media, and CO$_2$ tends to flow preferentially along these flow channels; therefore, the time to reach the outlet is shortened and the final saturation is higher. In summary, the non-uniformity of porous media can improve the CO$_2$ displacement efficiency.

### 3.3.3. Effect of Non-uniformity of Granule Size

As shown in Figures 17 and 18, in non-uniform porous media, the breakthrough time of CO$_2$ and the average granule size still conform to the law of quadratic function, and the final saturation presents a nearly linear relationship. It is also found that the breakthrough time of CO$_2$ in non-uniform porous media is shorter, and the final saturation is higher than those in uniform porous media for other factors fixed. The reason is that the proportion of wider flow channels in non-uniform porous media increases relative to uniform porous media, and CO$_2$ tends to flow preferentially along these flow channels; therefore, the time to reach the outlet is shortened and the final saturation is higher. In summary, the non-uniformity of porous media can improve the CO$_2$ displacement efficiency.

### 3.3.3. Effect of Non-uniformity of Granule Size

From the results in Section 3.3.2, we find that although the average granule size of the porous media is the same, the non-uniformity of granule size seems to have a certain impact on the displacement process and efficiency. In this section, we ensure that the porosity and average granule size of the porous media are consistent, $\varepsilon = 0.65$ and $D^* = 0.01$, respectively. To quantitatively study the effect of the non-uniformity of granule size, the standard deviation is applied using $\kappa = \sqrt{\frac{1}{n} \sum_{i=1}^{n} (D^i - \overline{D}^*)^2}$, where $n$ is the total number of granules. The standard deviations of porous media samples 11, 3, and 6 are calculated as 0, 1.284, and 2.168, respectively. The influence of the standard deviation (i.e., non-uniformity) of the porous media on the displacement process and efficiency is discussed.

Figure 19 shows the distribution of CO$_2$ in porous media under different standard deviations in the quasi-steady state at $\theta_2 = 26.8^\circ$. With the further expansion of the standard deviation, it can be clearly found that the CO$_2$ channel is still established in the vicinity of the large-size granules, the flow of CO$_2$ is more continuous, and the large-area residual water is reduced. However, the seepage network characteristics of CO$_2$ in porous media have a great difference, embodied in the width and number of main channels. The reason is that as the standard deviation increases, the distribution of solid granules becomes more and more uneven. Therefore, the number of large-size granules increases, resulting in the increasing proportion of a wider flow channel, and the number of small-size granules increases, leading to the weakened or even disappearing disturbance effect on the flow channel of CO$_2$.

We quantify the breakthrough time and final saturation of CO$_2$ to illustrate the impact of granule unevenness on displacement efficiency as shown in Figures 20 and 21. As the standard deviation increases, the breakthrough time is

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**Figure 16.** Distribution of CO$_2$ in non-uniform porous media for different wettability values and average sizes of solid granules at the quasi-steady state (red, CO$_2$; blue, water; black, skeleton structure) with a porosity of 0.65.

**Figure 17.** Relationship between CO$_2$ breakthrough time and average granule size under different contact angles.

**Figure 18.** Relationship between the final saturation of CO$_2$ and average granule size under different contact angles.

**Figure 19.** Distribution of CO$_2$ in non-uniform porous media under different standard deviations in the quasi-steady state at $\theta_2 = 26.8^\circ$ (red, CO$_2$; blue, water; black, skeleton structure), with a porosity of 0.65 and an average granule size of 0.01.
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water displacement process at the pore scale is simulated by 
porosities and granule sizes, the immiscible two-phase CO2 
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changes from 
uniformity of granule size is bene 
0 to 2.168, the breakthrough time is shortened from 0.168 to 
significantly shortened and the final saturation increases. For 
example, at θ2 = 26.8°, with increasing standard deviation from 
0 to 2.168, the breakthrough time is shortened from 0.168 to 
0.097, and the final saturation increases from 49.21 to 59.04%. 
The results show that choosing a reservoir with a greater non-
uniformity of granule size is beneficial to improve the efficiency 
of CO2 displacement.

Figure 20. Relationship between the final saturation of CO2 and 
wettability under different standard deviations.

4. CONCLUSIONS
In this paper, based on two-dimensional heterogeneous porous 
media models numerically reconstructed with different 
porosities and granule sizes, the immiscible two-phase CO2-
water displacement process at the pore scale is simulated by 
the LBM method, and the displacement efficiency is evaluated 
under different conditions. It is concluded as follows:

With the enhancement of CO2-surface wettability represent-
ing the fluid–solid interaction, the CO2 displacement mode 
changes from fingerling to stable displacement, where residual 
water tends to occupy the pores instead of the throats, and 
the saturation of CO2 at the quasi-steady state increases 
significantly. Although the breakthrough time becomes longer, 
the overall displacement efficiency shows a trend of improve-
ment.

For the porous media characteristic factors, with the increase 
in porosity, granule size, and non-uniformity of granule size in 
the heterogeneous porous media, the sweeping area of the CO2 
mainstream channel increases and the flow becomes more 
continuous, where the breakthrough time of CO2 is shortened, 
the final saturation at the quasi-steady state increases 
significantly, and the displacement efficiency shows a trend of 
improvement.

In summary, wettability is the primary mechanism to 
I improve CO2 displacement efficiency. Even if porous media 
maintain a low porosity or small average granule size, CO2 can 
still maintain a high displacement efficiency in hydrophobic 
porous media. The reservoirs with high porosity, large granule 
size, and great non-uniformity of granule size have greater 
storage capacity, which can help improve the geological storage 
efficiency of CO2.

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Notes
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