

Article

# Experimental Study on Feasibility of Enhanced Gas Recovery through CO<sub>2</sub> Flooding in Tight Sandstone Gas Reservoirs

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**Abstract:** The development of natural gas in tight sandstone gas reservoirs via CH<sub>4</sub>-CO<sub>2</sub> replacement is promising for its advantages in enhanced gas recovery (EGR) and CO<sub>2</sub> geologic sequestration. However, the degree of recovery and the influencing factors of CO<sub>2</sub> flooding for enhanced gas recovery as well as the CO<sub>2</sub> geological rate are not yet clear. In this study, the tight sandstone gas reservoir characteristics and the fluid properties of the Sulige Gasfield were chosen as the research platform. Tight sandstone gas long-core displacement experiments were performed to investigate (1) the extent to which CO<sub>2</sub> injection enhanced gas recovery (CO<sub>2</sub>-EGR) and (2) the ability to achieve CO<sub>2</sub> geological storage. Through modification of the injection rate, the water content of the core, and the formation dip angle, comparative studies were also carried out. The experimental results demonstrated that the gas recovery from CO<sub>2</sub> flooding increased by 18.36% when compared to the depletion development method. At a lower injection rate, the diffusion of CO<sub>2</sub> was dominant and the main seepage resistance was the viscous force, which resulted in an earlier CO<sub>2</sub> breakthrough. The dissolution of CO<sub>2</sub> in water postponed the breakthrough of CO<sub>2</sub> while it was also favorable for improving the gas recovery and CO<sub>2</sub> geological storage. However, the effects of these two factors were insignificant. A greater influence was observed from the presence of a dip angle in tight sandstone gas reservoirs. The effect of CO<sub>2</sub> gravity separation and its higher viscosity were more conducive to stable displacement. Therefore, an additional gas recovery of 5% to 8% was obtained. Furthermore, the CO<sub>2</sub> geological storage exceeded 60%. As a consequence, CO<sub>2</sub>-EGR was found to be feasible for a tight sandstone gas reservoir while also achieving the purpose of effective CO<sub>2</sub> geological storage especially for a reservoir with a dip angle.

**Keywords:** CO<sub>2</sub> flooding; supercritical CO<sub>2</sub>; CO<sub>2</sub> geological storage; tight sandstone gas reservoirs; enhanced gas recovery

## 1. Introduction

“Gas flooding” typically using CO<sub>2</sub>, N<sub>2</sub>, or air has become one of the leading enhanced oil recovery (EOR) technologies for residual oil development in conventional reservoirs [1–4]. Unfortunately, “gas flooding” for natural gas reservoirs is currently only in the research and development stage. Tight gas reservoirs are one of the most important areas of unconventional natural gas exploration and development in the world and they have rich resource reserves [5]. However, a tight gas reservoir is characterized by poor reservoir properties, strong heterogeneity, and complicated pore-throat structures. The main traditional method for gas recovery is depletion development, but the recovery is

only approximately 35% [6,7]. In order to increase recovery from tight gas reservoirs, a new method of enhanced gas recovery (EGR) is urgently required. The phase state of CO<sub>2</sub> is easily transformed into the supercritical state [8–10] when the temperature exceeds the critical temperature (31.26 °C) and the pressure exceeds the critical pressure (7.29 MPa). Due to tight/shale gas reservoirs generally having great depths, it is easy for CO<sub>2</sub> to reach the supercritical state if it is injected into these reservoirs. Theoretically, supercritical CO<sub>2</sub> effectively displaces the natural gas and improves gas recovery in tight/shale gas reservoirs due to its higher density, higher viscosity, and lower diffusion rate.

There have been a substantial number of detailed investigations on gas adsorption characteristics in recent years, which aim to understand the mechanism of CH<sub>4</sub> displacement by CO<sub>2</sub> in coal reservoirs. Littke [11] studied the adsorption and desorption abilities of CO<sub>2</sub> and CH<sub>4</sub> under various temperature and pressure conditions. The adsorption capacity of CO<sub>2</sub> was higher when compared to CH<sub>4</sub>. Liang [12] experimentally investigated the displacement mechanism underlying the driving out of coal-bed methane by gaseous CO<sub>2</sub> and discovered that the permeability of CO<sub>2</sub> was beyond two orders of magnitude higher when compared to CH<sub>4</sub>. This result was explained by the differences in the physical properties of the two gases, which are combined with the competitive adsorption effect. Zeng [13] theoretically established an internally consistent adsorption-strain-permeability model to describe the adsorption capacity of coal reservoirs to CH<sub>4</sub> and CO<sub>2</sub> and the displacement process of CH<sub>4</sub> by CO<sub>2</sub>. The results indicated that the adsorption capacity of CO<sub>2</sub> was two to five times that of CH<sub>4</sub>. In general, CO<sub>2</sub> is chosen for injection into tight sandstone gas reservoirs to achieve EGR based on the following aspects. First, CH<sub>4</sub> in tight gas reservoirs primarily exists in an adsorbed state and CO<sub>2</sub> has a stronger adsorption capacity than CH<sub>4</sub> under the same conditions [14,15]. Second, since the mixing speed of CO<sub>2</sub> and CH<sub>4</sub> is slower when compared to pressure recovery, the injection of CO<sub>2</sub> can increase the formation pressure and displacement pressure gradient. Consequently, the flow velocity increases, which effectively gathers and drives the flow of CH<sub>4</sub> in the reservoirs [16]. Moreover, the maintenance of pressure can also provide pressure support to prevent the formation of subsidence and water invasion [17]. In addition, injecting CO<sub>2</sub> into tight sandstone gas reservoirs not only can achieve the purpose of EGR but also realize CO<sub>2</sub> geological storage, which is of great significance for mitigating the global greenhouse effect [18,19]. For the previously mentioned reasons and the fact that CO<sub>2</sub> flooding can increase coal seam recovery, CO<sub>2</sub> flooding is feasible for EGR from tight sandstone gas reservoirs, theoretically. According to reports, only three pilot projects have existed globally [20,21]. CO<sub>2</sub> storage pilot experiments were carried out in three gas fields including Beihai K12-B in Holland [22], Budafa in Hungary [23], and Algeria [10], but all were mainly concerned with achieving CO<sub>2</sub> geological storage. CO<sub>2</sub>-EGR is widely utilized in medium-permeability and high-permeability gas reservoirs both domestically and overseas while experimental studies on CO<sub>2</sub>-EGR in tight sandstone gas reservoirs have rarely been reported.

In this work, the reservoir characteristics and fluid properties of the Sulige Gasfield were chosen as the research platform while displacement experiments using combined natural long-cores were performed to investigate the variables affecting CO<sub>2</sub>-EGR. The extent of CO<sub>2</sub>-EGR and CO<sub>2</sub> geological storage was measured by using various injection rates, dry and aqueous cores, and formation dip angles. In this scenario, the feasibility of CO<sub>2</sub>-EGR in tight sandstone gas reservoirs using the displacement mechanism is examined and the geological storage of CO<sub>2</sub> is also discussed and explained.

## 2. Experimental Section

### 2.1. Cores

During experimentation, natural tight cores were obtained from the Sulige Gasfield of Ordos Basin. In order to simulate the actual geological conditions of tight gas sandstone reservoirs, long-core displacement experiments were carried out. Since a natural tight long-core with a length of 1 m was obviously unrealistic for the experiments, a series of natural small-sized cores were combined to

form a long-core, which was used to perform the displacement experiments of natural gas with CO<sub>2</sub>. The long-core consisted of 32 natural tight short-cores that were  $3.0 \times 10^{-4}$  m in length and  $2.5 \times 10^{-4}$  m in diameter. The total long-core was approximately 1 m in length. Even though this combination of natural short-cores might result in the end effect, the placement of a piece of filter paper between every two short-cores could effectively ameliorate it [24]. Using weighted averages, the average permeability and porosity of the long-core were  $0.325 \times 10^{-3} \mu\text{m}^2$  and 9.36%, respectively. In addition, a piece of tight sandstone core was used to perform the capillary pressure tests. The diameter of the core was  $2.54 \times 10^{-2}$  m and the length was  $5 \times 10^{-2}$  m. The core permeability and porosity were  $0.307 \times 10^{-3} \mu\text{m}^2$  and 9.41%, respectively.

## 2.2. Fluid

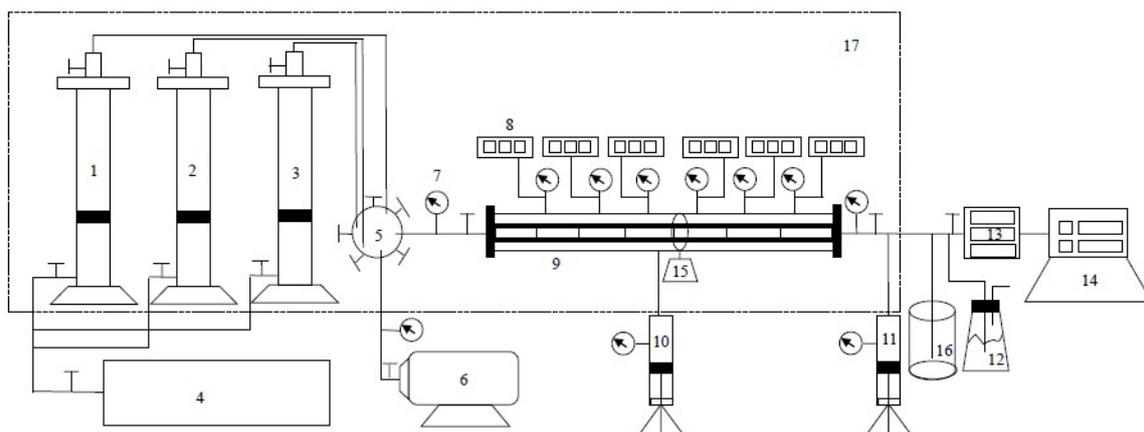
The gas for the experiments was CH<sub>4</sub> of high purity with the purity exceeding 99.95%. The CO<sub>2</sub> purity for the experiments exceeded 99.9%. The simulated formation water for the experiments was prepared according to the ion content and salinity of the actual formation water in the Sulige Gasfield, which is presented in Table 1.

**Table 1.** Mineral composition of the water sample in experiments.

K <sup>+</sup>	Na <sup>+</sup>	Ca <sup>2+</sup>	Mg <sup>2+</sup>	Sr <sup>2+</sup>	Cl <sup>-</sup>	SO <sub>4</sub> <sup>2-</sup>	CO <sub>3</sub> <sup>2-</sup>	HCO <sub>3</sub> <sup>-</sup>	OH <sup>-</sup>	Total Salinity
(mg/L)										
1541	15,221	12,344	2539	650	59,774	744	none	257	none	102,000

## 2.3. Experimental Setup

The experimental setup for the long-core displacement experiments was produced in France and it was mainly composed of an injection system, a displacement system, and a production system. A total of six pressure measurement points were set up from the injection end to the outlet end and the pressure variation throughout the entire process was observed by a monitoring system. The experimental installation is presented in Figure 1.



**Figure 1.** Schematic diagram of the experimental setup (1. Intermediate container (high purity CH<sub>4</sub>), 2. Intermediate container (simulated water), 3. Intermediate container (high purity CO<sub>2</sub>), 4. Pump, 5. Sluice with five gates, 6. Vacuum pump, 7. Pressure gage, 8. Electronic pressure gauge, 9. The long-core, 10. Confining pressure pump, 11. Back pressure pump, 12. Conical flask, 13. Electronic pressure gauge, 14. Chromatographic analyzer, 15. Angle regulator of core, 16. Beaker, 17. Thermostat box).

In addition, a high-pressure semipermeable plate instrument was utilized for capillary pressure testing.

## 2.4. Experimental Method

The experiments were conducted under the conditions of 110 °C and 30 MPa, which match the conditions in the actual tight gas reservoir of the Sulige Gasfield.

### 2.4.1. Measurement of Capillary Pressure

The effect of the injection rate on the results of CO<sub>2</sub> flooding CH<sub>4</sub> can be interpreted by comparing the viscous pressure drop generated by the flow and the capillary pressure. Therefore, we conducted an experiment using the semipermeable plate method to measure the capillary force [25] and then we measured the correlation of the capillary force with water saturation. First, the core was saturated with water. Then the displacement pressure difference between the two ends was established by using vacuum extraction. This pressure difference is balanced with a certain capillary force and different displacement pressure differences correspond to different capillary forces. Under a displacement pressure difference, the non-wet phase fluid (gas) displaces the wet phase fluid (the formation water) in the core. Therefore, the saturation of the wet phase (water) decreases as the displacement pressure difference increases. According to the displacement pressure balanced with the capillary pressure and the corresponding wet phase (water) saturation in the core during the displacement process, the correlation of the capillary force with the wet phase (water) saturation can be obtained. According to the research of Zou et al. [26,27], for a tight sandstone gas core, a hydrophilic semipermeable plate with a threshold pressure of 3 MPa should be selected. Thus, after the wet phase fluid saturates the semipermeable plate, the wet phase fluid can only pass through the semipermeable plate as a result of the capillary pressure until the displacement pressure is not less than the threshold pressure of the semipermeable plate. It is worth noting that the equilibration time should exceed 72 h for each pressure point.

### 2.4.2. Measurement of CH<sub>4</sub> Recovery and CO<sub>2</sub> Storage Efficiency through CO<sub>2</sub> Flooding

Generally, higher water saturation always exists in tight sandstone gas reservoirs in China. As a consequence, irreducible water saturation under conditions of confining pressure and a displacement pressure drop should be established in the experiment first. The simulated formation water was injected into the long-core and CH<sub>4</sub> was used to displace the water until the simulated formation water did not flow out of the long-core outlet.

The experiments were performed according to the following procedures.

- (1) The short tight sandstone cores were added to the long-core holder in order, which was placed into the thermostat box. The displacement flow path was connected, according to the experimental device diagram presented in Figure 1.
- (2) The long-core was vacuum-pumped. The thermostat box was set to 110 °C and the long-core confining pressure was set to 30 MPa.
- (3) The irreducible water saturation status of the combined tight long-core was obtained under conditions of confining pressure and a displacement pressure drop. The simulated formation water was injected into the closed long-core holding system from intermediate container 2. Consequently, high-purity CH<sub>4</sub> gas was injected into the system from intermediate container 1. In this process, the simulated formation water was displaced by CH<sub>4</sub> until it did not flow out of the long-core outlet, which indicated that this procedure was over.
- (4) Pure CH<sub>4</sub> was continuously injected into the closed long-core holding system with a constant pressure of 8 MPa. The tight sandstone long-core was fully saturated with CH<sub>4</sub> and the outlet valve was closed during the entire saturation process. The CH<sub>4</sub> saturation process was considered to be completed when the inlet pressure was stable for more than 12 h.
- (5) The intermediate container 3, which was filled with high-purity CO<sub>2</sub>, was connected to the displacement device system. The pressures of the long-core inlet and outlet were 12 MPa and 8 MPa, respectively. This meant that the displacement differential pressure was 4 MPa. Each time

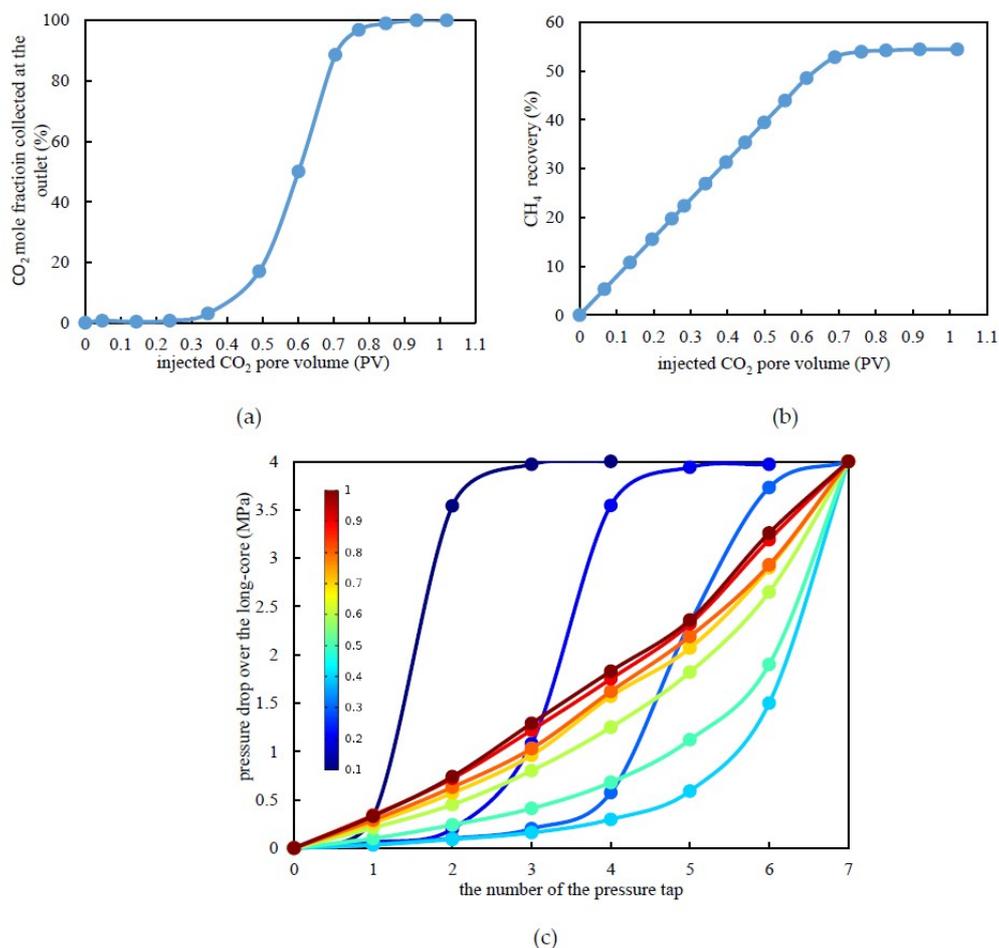
that a 0.1 pore volume (PV) of CO<sub>2</sub> was injected into the long-core, the pressure at each pressure point was recorded. Furthermore, the gas production at the outlet was also recorded and the gas contents of CH<sub>4</sub> and CO<sub>2</sub> were analyzed with the chromatography analyzer. The characteristics of CO<sub>2</sub> migration and breakthrough of the front edge were monitored in real time. Additionally, the displacement efficiency of CO<sub>2</sub> flooding CH<sub>4</sub> was calculated. When the gas content of CO<sub>2</sub> at the outlet exceeded 98%, the experiments were over.

- (6) Subsequent to each group of experiments, the tight sandstone long-core was vacuum-pumped and steps (2)–(5) were repeated.

### 3. Results and Discussion

#### 3.1. CH<sub>4</sub> Recovery through CO<sub>2</sub> Flooding and CO<sub>2</sub> Storage Efficiency under Constant Pressure Displacement

Under a confining pressure of 30 MPa, the irreducible water saturation of the combined tight long-core was 41.05% in this experiment. According to the previously mentioned experimental procedures, CO<sub>2</sub> was injected to displace CH<sub>4</sub> at a constant pressure of 4 MPa until the gas content of CO<sub>2</sub> at the outlet exceeded 98%. The CO<sub>2</sub> mole fraction collected at the outlet and the CO<sub>2</sub> recovery calculated are shown in Figure 2a,b. Under the experimental temperature and pressure conditions, CO<sub>2</sub> was in the supercritical state. The results demonstrated that the displacement front of supercritical CO<sub>2</sub> would be miscible with CH<sub>4</sub>, which requires an extended duration of this process.



**Figure 2.** Experimental results under constant differential pressure displacement. (a) CO<sub>2</sub> mole fraction at the outlet versus the injected CO<sub>2</sub> pore volume. (b) CH<sub>4</sub> recovery by CO<sub>2</sub> flooding. (c) Pressure drop over a tight long-core versus the pore volume during CO<sub>2</sub> flooding (Note: the colored lines represent the various injected CO<sub>2</sub> pore volumes).

According to Figure 2a, as the pore volume (PV) of the injected CO<sub>2</sub> increased to 0.4 PV, a current of CO<sub>2</sub> was detected at the outlet, which indicated that displacement front breakthrough had occurred. Following this, the CO<sub>2</sub> mole fraction collected at the outlet increased almost linearly with the pore volume of the injected CO<sub>2</sub> until it reached 0.7 PV. When it reached approximately 0.8 PV, almost no CH<sub>4</sub> was detected at the outlet, which indicates that the CH<sub>4</sub> recovery had reached its maximum. It can be clearly observed from Figure 2c that the pressure drop was mainly diminished at the displacement leading edge. Before breakthrough of the leading edge, the higher the pore volume of the injected CO<sub>2</sub>, the further the pressure propagation, and, for the same pressure test point, the pressure drop decreased. As the injected CO<sub>2</sub> reached 0.4 PV, breakthrough occurred at the displacement leading edge. Subsequently, with the increase in the pore volume of injected CO<sub>2</sub>, the distribution of the pressure drop was more uniform over the whole long-core. With the formation of continuous flow channels, the pressure drop tended to be stable. When the injected CO<sub>2</sub> pore volume was 1.0 PV, the pressure drop curve was close to linear.

As seen in Figure 2b, the CH<sub>4</sub> recovery from CO<sub>2</sub> flooding was approximately 53.36%, which is an 18.36% increase when compared to the depletion development method for tight sandstone gas reservoirs. Therefore, it can be concluded that it is feasible to achieve EGR in tight sandstone gas reservoirs through CO<sub>2</sub> flooding.

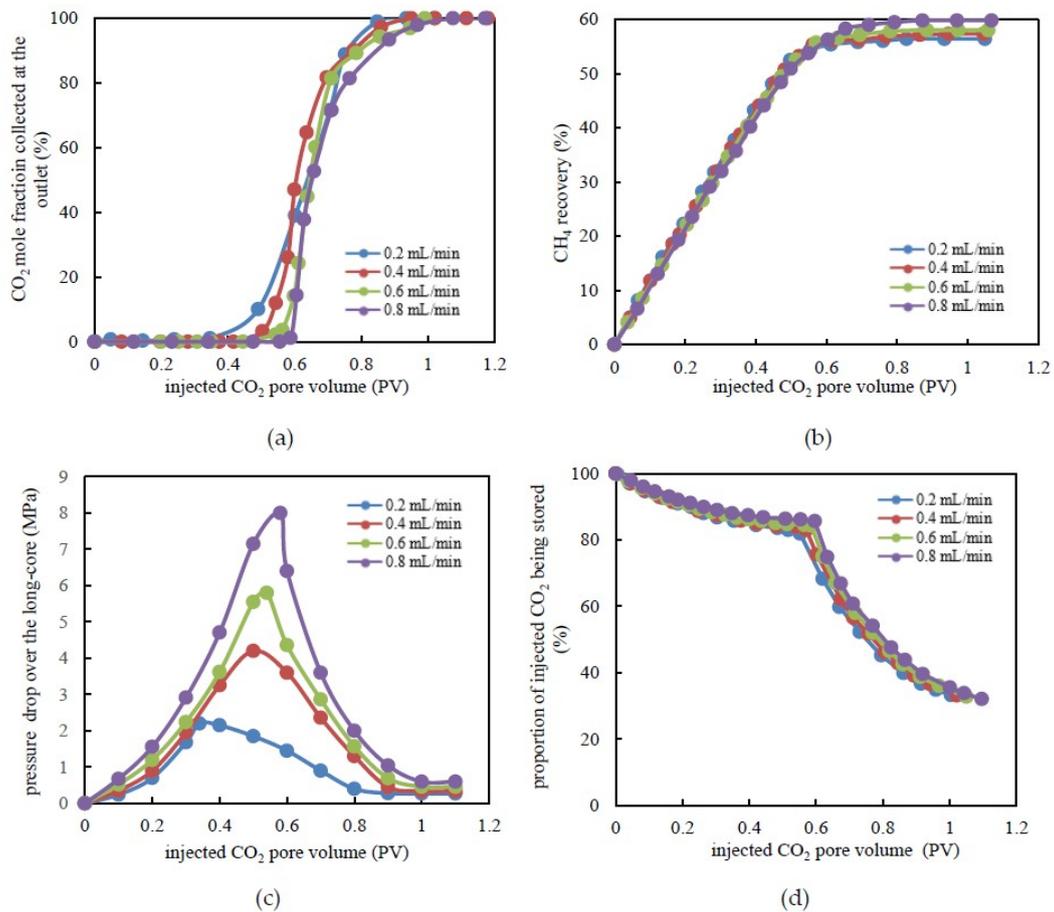
### 3.2. CH<sub>4</sub> Recovery through CO<sub>2</sub> Flooding and CO<sub>2</sub> Storage Efficiency under Various Injection Rates

In order to determine the effect of the injection rate on EGR and CO<sub>2</sub> storage efficiency, four groups of experiments were conducted using injection rates of 0.2, 0.4, 0.6, and 0.8 mL/min, respectively. The experimental results are presented in Figure 3.

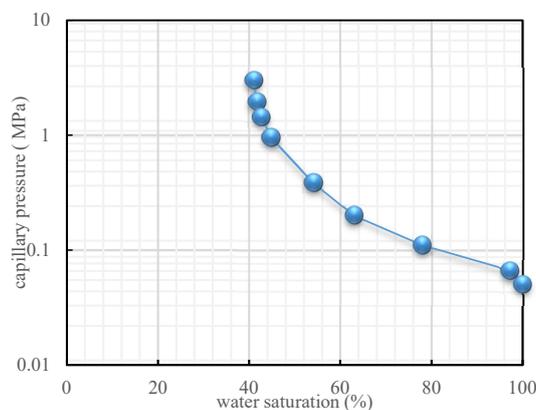
The results demonstrated that, with the injection rate conditions of 0.2, 0.4, 0.6, and 0.8 mL/min, displacement front breakthrough occurred when the injected CO<sub>2</sub> pore volume was 0.34, 0.42, 0.47, and 0.60 PV, respectively (Figure 3a). The lower the displacement velocity is, the earlier the occurrence of displacement front breakthrough is. This result occurred because the diffusion of CO<sub>2</sub> dominated under a lower injection rate. Moreover, the viscous pressure drop, which was generated by the flow and the capillary pressure between the gas and the water, constituted the percolation resistance. At a lower injection rate, because the water in the tiny capillaries could barely be driven and the long-core was already in a state of near-irreducible water saturation, the capillary pressure had little effect on the gas flow. CO<sub>2</sub> mainly flowed in the channels formed by larger-sized pores and throats. The main seepage resistance was the viscous force between the gas and the water attached to the pore surface while the viscous force was relatively lower at a lower injection rate. This was also a reason for gas breakthrough occurring sooner. Under a higher injection rate, in order to overcome higher seepage resistance, the injection pressure was apparently higher, which caused displacement of the additional water in the tiny capillaries. Consequently, the water saturation further decreased and the capillary pressure increased (Figure 4). When the water saturation decreased from 41.5% to 40.75%, the capillary force rapidly increased from 2.09 to 3.52 MPa. That means that the water saturation only decreased by less than 1.8% but caused an increase in a capillary force up to 168%. In this case, the capillary pressure and the viscous force increased simultaneously, but the seepage resistance was too high, which requires more time for the breakthrough to occur.

Figure 3b shows the changing degrees of CH<sub>4</sub> recovery with varying injection rates. There were large differences in the degree of CH<sub>4</sub> recovery for different time frames of CO<sub>2</sub> breakthrough. At CO<sub>2</sub> leading edge breakthrough, the CH<sub>4</sub> recovery was 43.24%, 46.10%, 50.46%, and 56.25%, which corresponds to the CO<sub>2</sub> injection rates of 0.2, 0.4, 0.6, and 0.8 mL/min. The difference between the maximum and the minimum degree of recovery was 13.01%. However, at the end of displacement, CH<sub>4</sub> recovery corresponding to the four injection rates above was 56.39%, 57.32%, 58.00%, and 59.80%. The CH<sub>4</sub> recovery difference was only 3.4% over the entire displacement range. This phenomenon demonstrated that the miscible behavior between supercritical CO<sub>2</sub> and CH<sub>4</sub> mainly occurred at the leading edge of the displacement and it required more time to occur under lower displacement velocity

conditions. Therefore, the miscible band range was also relatively higher. In addition, a continuous flow channel gradually formed following breakthrough of the leading edge and, subsequently, no additional water outflow occurred at the outlet. This meant that the capillary pressure was essentially constant and the viscous force was slightly decreased. In general, slight changes existed in the swept volume of CO<sub>2</sub> flooding at various injection rates. Consequently, the injection rates had slight differences during EGR by CO<sub>2</sub> flooding.



**Figure 3.** Experimental results under various injection rates. (a) CO<sub>2</sub> mole fraction at the outlet versus injected CO<sub>2</sub> pore volume. (b) CH<sub>4</sub> recovery by CO<sub>2</sub> flooding. (c) Pressure drop over tight long-core versus the pore volume during CO<sub>2</sub> flooding. (d) The proportion of injected CO<sub>2</sub> being stored versus an injected CO<sub>2</sub> pore volume.



**Figure 4.** Curve of capillary pressure versus water saturation.

During CO<sub>2</sub> flooding, the increase in the pressure drop accelerated with the injected CO<sub>2</sub> pore volume prior to the displacement front breakthrough (Figure 3c). The pressure drop over the long-core attained its maximum value prior to the moment of the breakthrough. At the different breakthrough points, the pressure drop over the long-core was 2.12, 4.07, 5.89, and 8.16 MPa. Subsequently, the pressure drop decreased gradually and the speed decreased more quickly for higher injection rates of displacement under the same injected CO<sub>2</sub> pore volume conditions. At a lower injection rate, breakthrough of the leading edge mainly depended on the diffusion of CO<sub>2</sub> molecules. Consequently, the pressure drop became slower after breakthrough. As a flowing single gas gradually becomes a mixture of gases, the pressure drop was mainly concentrated in the latter half of the long-core. The closer to the outlet the pressure drop concentration is, the higher the resistance to seepage is.

The proportion of CO<sub>2</sub> being stored is defined as the ratio of the amount of CO<sub>2</sub> storage to the total amount of CO<sub>2</sub> injected. In the initial stage, most of the injected CO<sub>2</sub> remained in the long-core and the proportion of CO<sub>2</sub> being stored was higher (Figure 3d). From the injection of CO<sub>2</sub> to breakthrough of the leading edge, the proportion of the stored CO<sub>2</sub> decreased from 100% to 84.85%, 85.07%, 86.76%, and 87.64%, which corresponds to CO<sub>2</sub> injection rates of 0.2, 0.4, 0.6, and 0.8 mL/min, respectively. Following CO<sub>2</sub> breakthrough, the proportion of stored CO<sub>2</sub> decreased rapidly and it was almost unaffected by the injection rate. The final proportion of CO<sub>2</sub> being stored was 32.50%. This occurred due to the fact that both CO<sub>2</sub> and CH<sub>4</sub> had sufficient time to achieve sufficient miscibility with a lower injection rate. Therefore, additional CO<sub>2</sub> was stored subsequently to the leading edge breakthrough. To a certain extent, a higher CO<sub>2</sub> injection rate could improve gas recovery. However, it would also lead to larger pressure loss in a short time, which is not conducive to sustainable, stable production. As a consequence, the actual production system for developing tight sandstone gas reservoirs should take into account the joint determination of EGR and the economic payback period.

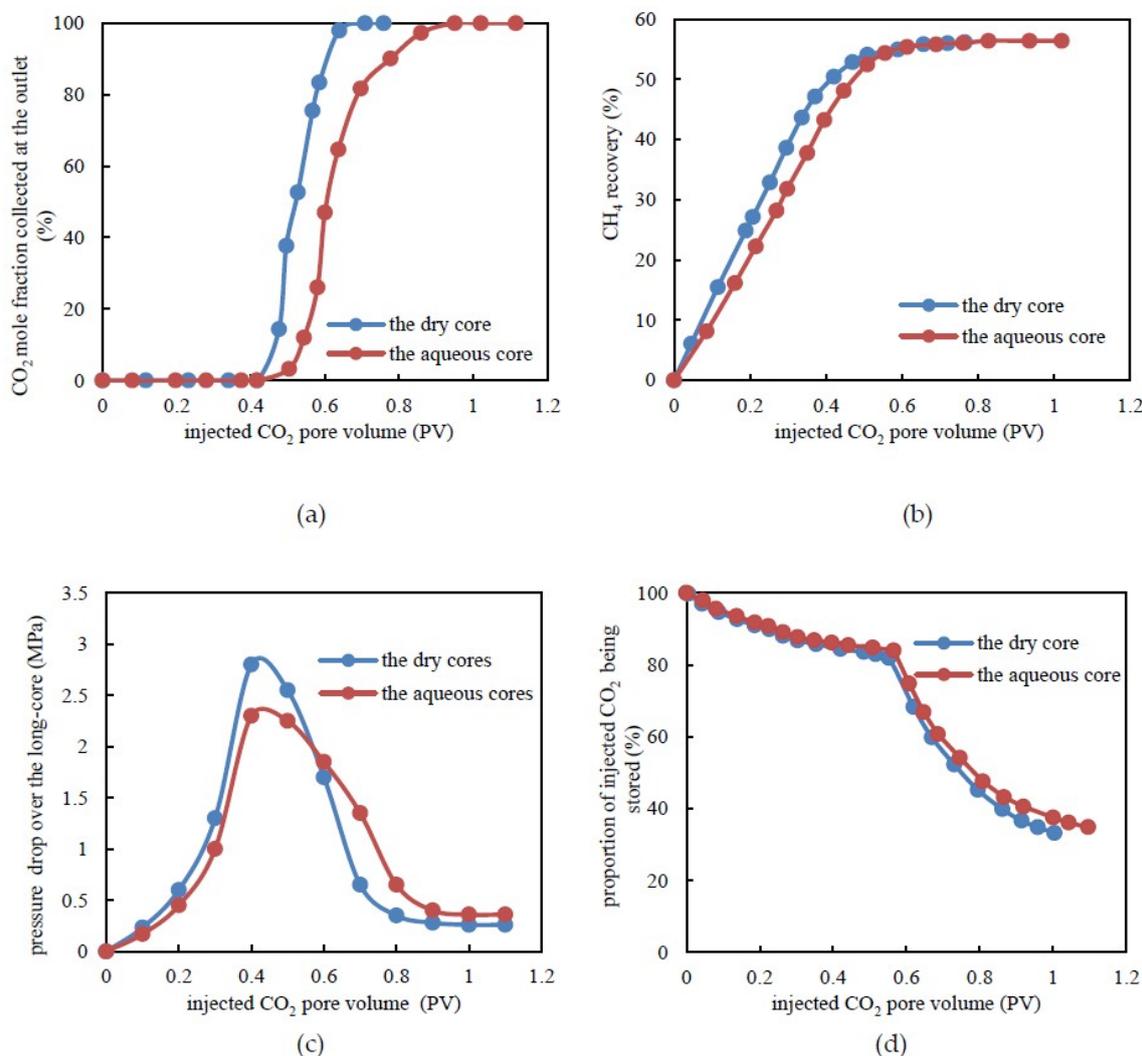
### 3.3. CH<sub>4</sub> Recovery through CO<sub>2</sub> Flooding and CO<sub>2</sub> Storage Efficiency under Dry and Aqueous Cores

In general, higher water saturation is common in the tight sandstone gas reservoirs of China [5]. In one group of experiments, the tight long-core was saturated with high-purity and dry CH<sub>4</sub>. In the other group, an aqueous long-core with an irreducible water saturation of 41.5% existed under the confining pressure of 30 MPa. The injection rate was 0.2 mL/min.

The results demonstrated that the displacement front broke through when the injected CO<sub>2</sub> pore volume was 0.4 PV for the dry long-core while it was up to 0.5 PV for the aqueous core (Figure 5a). For the former, when the injected CO<sub>2</sub> was 0.76 PV, the CO<sub>2</sub> mole fraction collected at the outlet was 100% and no more CH<sub>4</sub> was displaced. For the latter, this situation did not occur until the injected CO<sub>2</sub> was 0.95 PV. Due to the presence of water, a portion of injected CO<sub>2</sub> initially dissolved and formed unstable carbonic acid in the water. In the same area, when the carbonic acid was saturated in the aqueous solution, CO<sub>2</sub> began to displace CH<sub>4</sub>. Consequently, leading edge breakthrough occurred slightly later in time for the aqueous long-core with CO<sub>2</sub> flooding. When the CO<sub>2</sub> leading edge broke through, the CH<sub>4</sub> recovery was 48.78% and 52.48%, which corresponds to the dry long-core and the aqueous core. However, at the end of displacement, the corresponding CH<sub>4</sub> recovery was 54.68% and 56.23%, respectively (Figure 5b). In previous studies [28], it was shown that, due to the strong adsorption capacity of the tight sandstone rock surface, a tiny fraction of pores and throats might be blocked by water. Consequently, CH<sub>4</sub> exists in relatively larger-sized pores and throats, so it is more easily displaced. The amount of gas in this part was quite limited. Therefore, CO<sub>2</sub>-EGR for the aqueous long-core was slightly enhanced when compared to the dry long-core.

As shown in Figure 5c, there was less variation in the range of pressure drop values for the aqueous long-core when compared to the dry long-core. Due to a portion of injected CO<sub>2</sub> being dissolved in the formation water, under the same injected CO<sub>2</sub> pore volume, the pressure drop was relatively lower when compared to the dry long-core. The higher the pressure, the higher the proportion of dissolved CO<sub>2</sub> in the formation water. At the point of CO<sub>2</sub> leading edge breakthrough, the pressure drop over the dry long-core and the aqueous long-core was 2.85 and 2.32 MPa. Following breakthrough of the

leading edge, for the aqueous long-core, the seepage resistance was lower when compared to the dry long-core. Therefore, the pressure drop decreased more slowly.



**Figure 5.** Experimental results under tight dry and aqueous cores. (a) CO<sub>2</sub> mole fraction at the outlet versus injected CO<sub>2</sub> pore volume. (b) CH<sub>4</sub> recovery by CO<sub>2</sub> flooding. (c) Pressure drop over tight long-core versus the pore volume during CO<sub>2</sub> flooding. (d) The proportion of injected CO<sub>2</sub> being stored versus an injected CO<sub>2</sub> pore volume.

From the injection of CO<sub>2</sub> to the breakthrough of the leading edge, the proportion of the stored CO<sub>2</sub> decreased from 100% to 84.03% and 84.80%, which corresponds to the dry long-core and the aqueous core, respectively (Figure 5d). Following breakthrough of CO<sub>2</sub>, the proportion of CO<sub>2</sub> being stored decreased rapidly for the two situations mentioned above. At the end of displacement, the proportion of CO<sub>2</sub> being stored was 33.07% and 34.92%. In summary, due to the dissolution of CO<sub>2</sub> in the formation water, the total CO<sub>2</sub> being stored in the aqueous tight long-core was about 5% higher than that of the dry long-core during the whole displacement process. For the aqueous long-core, CO<sub>2</sub> was mainly stored by two mechanisms. One was to displace CH<sub>4</sub> and occupy its former position and the other way was to dissolve in the formation water. Accordingly, a slightly improved geological storage effect existed for CO<sub>2</sub> in tight sandstone reservoirs with formation water. This indicates that CO<sub>2</sub> flooding is more suitable for an aqueous tight sandstone gas reservoir (such as in China) since it results in a higher degree of enhanced gas recovery and improved CO<sub>2</sub> storage. In addition, with a stable injection rate, since the CO<sub>2</sub>-EGR method can more or less mitigate the hindrance of water

relative to the gas flow, controlling the water saturation could be regarded as a major factor rather than a primary factor in the development process of actual tight sandstone gas reservoirs.

#### 3.4. CH<sub>4</sub> Recovery through CO<sub>2</sub> Flooding and CO<sub>2</sub> Storage Efficiency under Various Formation Dip Angle Conditions

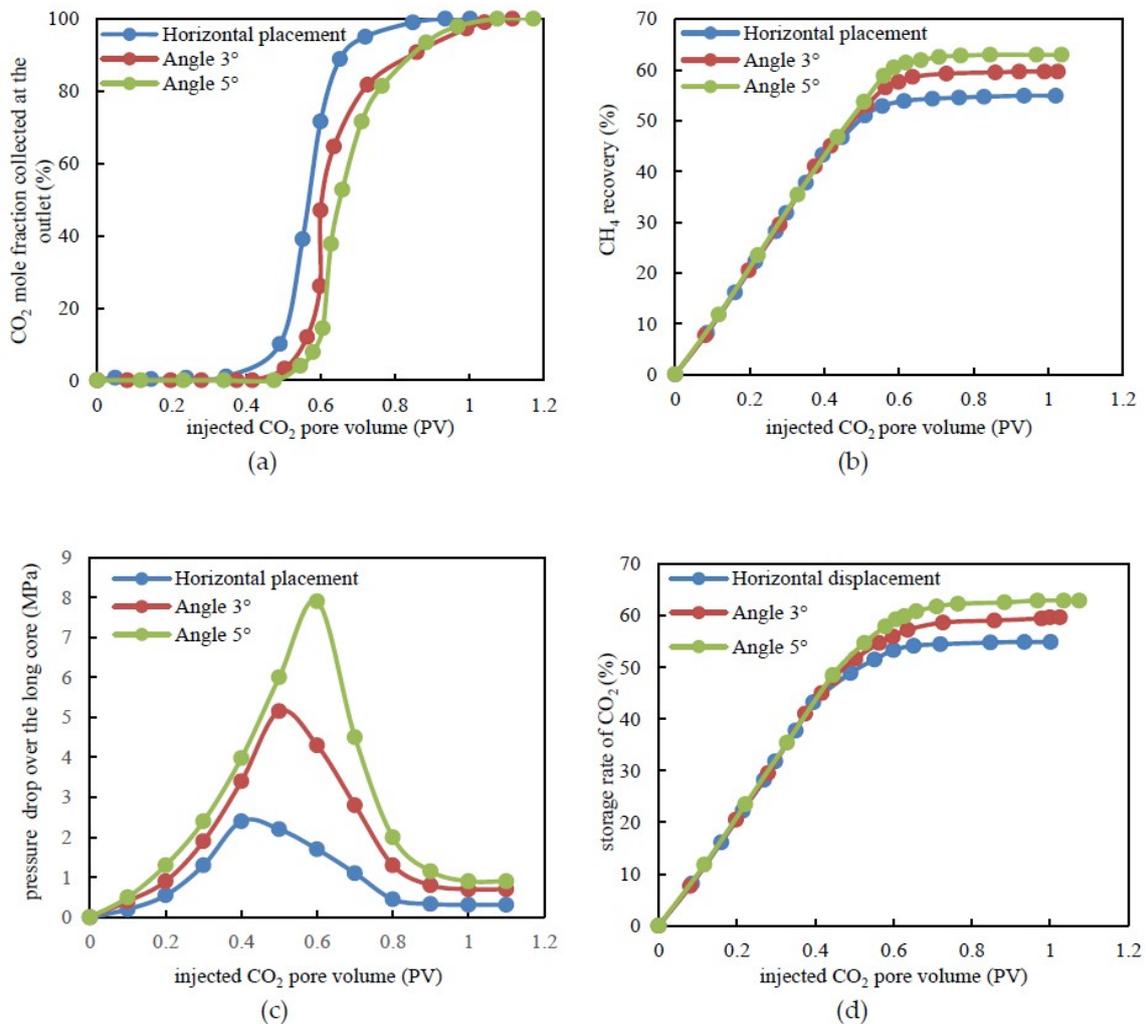
Since the density of supercritical CO<sub>2</sub> was higher when compared to CH<sub>4</sub>, the injected CO<sub>2</sub> tended to deposit at the reservoir bottom. Therefore, three groups of experiments were carried out to study the formation angle effect on EGR through CO<sub>2</sub> flooding. In the displacement experiments, the injection end of the long-core was at a lower position and the corresponding outlet was at a higher position with formation dip angles of 3° and 5°, respectively, relative to the actual formation conditions. In addition, a group of experiments was designed for comparison with the long-core placed horizontally. The injection rate was 0.2 mL/min.

The results demonstrated that the displacement front breakthrough occurred when the injected CO<sub>2</sub> pore volume was 0.37, 0.51, and 0.60 PV, which corresponds to the horizontally placed long-core and dip angles of 3° and 5°, respectively (Figure 6a). The higher the dip angle, the longer the miscible band and the later CO<sub>2</sub> breakthrough occurred. Since the stage before CH<sub>4</sub> breakthrough is an important period for CO<sub>2</sub> storage, such experimental results indicated that the CO<sub>2</sub> storage rate could be increased by at least around 1.5 times at this stage for a tight sandstone gas reservoir even with a small dip angle. When the CO<sub>2</sub> leading edge broke through, the CO<sub>2</sub> recovery was 40.49%, 53.03%, and 59.06%, which corresponds to the horizontally placed long-core and dip angles of 3° and 5°, respectively (Figure 6b). In addition, at the end of displacement, the corresponding CH<sub>4</sub> recovery was 54.49%, 59.65%, and 62.95%, respectively. Due to the higher density of supercritical CO<sub>2</sub>, it was easier for CH<sub>4</sub> to move from the bottom to the higher part. Moreover, the gravitational differentiation between CO<sub>2</sub> and CH<sub>4</sub> limited the mixing of gases in the vertical direction, which was beneficial to the displacement. Furthermore, as the viscosity of supercritical CO<sub>2</sub> was higher when compared to CH<sub>4</sub>, a favorable mobility ratio could improve the displacement stability. An additional 5% to 8% recovery of CH<sub>4</sub> could be achieved when compared to the horizontally placed long-core due to the dip angles (Figure 6b).

In terms of a pressure drop over the long-core, a relatively higher seepage resistance existed in the long-core with a dip angle at the early stage, which leads to a higher pressure drop for the same injected CO<sub>2</sub> pore volume (Figure 6c). At the point of CO<sub>2</sub> leading edge breakthrough, the pressure drop over the long-core was 2.40, 5.15, and 7.9 MPa, which corresponds to the horizontally placed long-core and dip angles of 3° and 5°, respectively. Subsequent to leading edge breakthrough, the pressure difference decreased faster in the slanted long-core. For the long-cores with a dip angle, the gravity increased the flow pressure at the injection end while the CH<sub>4</sub> at the bottom was displaced to a higher position by CO<sub>2</sub>. The higher the dip angle, the higher the pressure drop over the long-core and the larger the stored volume of CO<sub>2</sub>. The geological rate of CO<sub>2</sub> refers to the ratio of the amount of CO<sub>2</sub> storage to the pore volume of the core. Consequently, CO<sub>2</sub> could occupy the additional space that previously belonged to CH<sub>4</sub>. Furthermore, the storage rate of injected CO<sub>2</sub> would be higher under the same conditions (Figure 6d). From the injection of CO<sub>2</sub> to breakthrough of the leading edge, the stored rate of CO<sub>2</sub> gradually reached 40.49%, 52.68%, and 56.29%, which corresponds to the horizontally placed long-core and dip angles of 3° and 5°, respectively. At the end of the displacement, the corresponding storage rate of CO<sub>2</sub> was eventually 54.88%, 59.65%, and 62.90%. Thus, the geological rate of CO<sub>2</sub> increased by 4.77% and 8.02% for the long-cores with dip angles of 3° and 5°. Therefore, the CH<sub>4</sub> recovery and CO<sub>2</sub> storage could be improved CO<sub>2</sub> flooding in tight gas sandstone reservoirs with a dip angle.

In addition, before CO<sub>2</sub> breakthrough, the pressure drop over the long-core with a dip angle of 5° was approximately four times that without a dip angle under the same injection rate and it stored a large amount of displacement energy. This suggested that the initial bottom-hole pressure should be appropriately higher in tight sandstone gas reservoirs with a dip angle. The purpose is to reduce the

rapid coning entry of  $\text{CH}_4$  and  $\text{CO}_2$  after the gas leading edge breakthrough. Then the bottom-hole pressure should be slowed as the development progresses in order to effectively extend the effective period of  $\text{CO}_2$ -EGR and improve the storage rate of  $\text{CO}_2$ .



**Figure 6.** Experimental results under various formation dip angle conditions. (a)  $\text{CO}_2$  mole fraction at the outlet versus injected  $\text{CO}_2$  pore-volume. (b)  $\text{CH}_4$  recovery by  $\text{CO}_2$  flooding. (c) Pressure drop over tight long-core versus the pore-volume during  $\text{CO}_2$  flooding. (d) The storage rate of  $\text{CO}_2$  versus injected  $\text{CO}_2$  pore-volume.

#### 4. Conclusions

In this work, the extent of  $\text{CO}_2$ -EGR and  $\text{CO}_2$  geological storage in a tight sandstone gas reservoir were investigated by carrying out a series of long-core displacement experiments with the purpose of analyzing the feasibility of  $\text{CO}_2$ -EGR. The effects of the injection rate, the water content of the core, and the ability to achieve  $\text{CO}_2$  geological storage are discussed in detail and their implications for  $\text{CO}_2$ -EGR are summarized. It is found that the  $\text{CH}_4$  recovery through  $\text{CO}_2$  flooding is approximately 53.36%, which is an 18.36% increase when compared to the depletion development method for tight sandstone gas reservoirs. Additionally, the  $\text{CO}_2$  geological rate is around 60%. The  $\text{CH}_4$  recovery and  $\text{CO}_2$  geological rate are related to the time of  $\text{CO}_2$  leading edge breakthrough and the pressure drop distribution along the long-core. First, a higher injection rate improves the  $\text{CH}_4$  recovery and  $\text{CO}_2$  geological rate to some extent. However, a higher  $\text{CO}_2$  injection rate leads to greater pressure loss. Therefore, a reasonable developing strategy for tight sandstone gas reservoirs should consider the

combined effect of EGR and economic payback period. Second, for the aqueous tight long-core due to the dissolution of CO<sub>2</sub> in the formation water, the CH<sub>4</sub> recovery and the CO<sub>2</sub> geological rate are increased by 1.5% and 5%, respectively. The water phase reduces the pressure loss and extends the time to the gas leading edge breakthrough. Therefore, when a CO<sub>2</sub>-EGR method is utilized to develop a tight sandstone gas reservoir, controlling water saturation could be regarded as a major factor rather than a primary factor. Third, compared with the horizontally placed long-core, the CH<sub>4</sub> recovery increases by an additional 5% to 8% for an inclined long-core with dip angles of 3° and 5°. In addition, the geological rate of CO<sub>2</sub> increases by 4.77% and 8.02%. Additionally, based on the pressure drop over the long-core before CO<sub>2</sub> breakthrough, the initial bottom-hole pressure should be appropriately higher in tight sandstone gas reservoirs with a dip angle. Then the bottom-hole pressure should be slowed as the development progresses in order to extend the effective period of CO<sub>2</sub>-EGR and improve the geological rate of CO<sub>2</sub>. This research is of great significance for guiding a successful CO<sub>2</sub>-EGR process for the actual development of tight sandstone gas reservoirs.

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