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Utilization of CO₂ as Cushion Gas for Depleted Gas Reservoir Transformed Gas Storage Reservoir

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Received: 20 November 2019; Accepted: 21 January 2020; Published: 25 January 2020

Abstract: Underground gas storage reservoirs (UGSRs) are used to keep the natural gas supply smooth. Native natural gas is commonly used as cushion gas to maintain the reservoir pressure and cannot be extracted in the depleted gas reservoir transformed UGSR, which leads to wasting huge amounts of this natural energy resource. CO₂ is an alternative gas to avoid this particular issue. However, the mixing of CO₂ and CH₄ in the UGSR challenges the application of CO₂ as cushion gas. In this work, the Donghae gas reservoir is used to investigate the suitability of using CO₂ as cushion gas in depleted gas reservoir transformed UGSR. The impact of the geological and engineering parameters, including the CO₂ fraction for cushion gas, reservoir temperature, reservoir permeability, residual water and production rate, on the reservoir pressure, gas mixing behavior, and CO₂ production are analyzed detailly based on the 15 years cyclic gas injection and production. The results showed that the maximum accepted CO₂ concentration for cushion gas is 9% under the condition of production and injection for 120 d and 180 d in a production cycle at a rate of 4.05 kg/s and 2.7 kg/s, respectively. The typical curve of the mixing zone thickness can be divided into four stages, which include the increasing stage, the smooth stage, the suddenly increasing stage, and the periodic change stage. In the periodic change stage, the mixed zone increases with the increasing of CO₂ fraction, temperature, production rate, and the decreasing of permeability and water saturation. The CO₂ fraction in cushion gas, reservoir permeability, and production rate have a significant effect on the breakthrough of CO₂ in the production well, while the effect of water saturation and temperature is limited.

Keywords: underground gas storage reservoir; cushion gas; CO₂; CO₂ storage; CO₂ utilization

1. Introduction

Natural gas storage is principally used for meeting the widely fluctuating demand of gas, especially the high peak demands in winter [1–3]. Meanwhile, the development of natural gas storage is further promoted by a profitable business model, i.e., storing at a lower price and selling at a higher price based on demand loads. [4]. There are several vital strategies for natural gas storage such as gas tanks, salt caverns, and gas pipeline [5]. Owing to its advantages of a large storage capacity and

superior economy, underground gas storage reservoirs (UGSRs) has been widely used in North America and Europe, and has also gained increasing attention in China [6–11]. Underground gas storage is mostly a seasonal operation that usually is charged in summer and discharged in winter. In the operation process, cushion gas is vital to maintain suitable reservoir pressure as well as to keep a stable production operation.

In the case of depleted gas reservoir transformed gas storage reservoir, the native natural gas is commonly used as the cushioning gas [12]. This method may lead to a great deal of waste because approximately 40%–70% of stored gas works as cushion gas to provide pressure support and cannot be extracted [13]. In addition, the security of a UGSR during the CH_4 injection could be threatened dramatically by the increased reservoir pressure due to the relatively low compressibility of the cushion gas. The utilization of supercritical fluids as cushion gas is a potential strategy to tackle these issues. For supercritical fluids there is a Widom region, where some physicochemical properties such as density and compressibility exhibit anomalous behavior [14–16]. The Widom region is related to the critical pressure and critical temperature, which is 7.38 MPa and 31.04 °C, respectively for CO_2 . This critical condition exactly can be achieved in the storage applications, demonstrating the potential of utilizing CO_2 as cushion gas in the natural gas storage reservoir. In comparison with the native gas cushion, 30% more CH_4 can be stored with CO_2 as cushion gas [12]. It may also bring an additional benefit to enhance gas recovery in the post- CO_2 storage and enhanced gas recovery (CSEGR) process for a depleted gas reservoir. However, the mixing of CO_2 and CH_4 with the injection and production of working gas as shown in Figure 1 can be an obstacle for the utilization of CO_2 as cushion gas [12], which requires serious attention.

Figure 1. The schematic diagram of natural gas storage reservoir with CO_2 cushion gas.

Some research efforts have been paid to study the mixing behavior of CO_2 and CH_4 in UGSR. Oldenburg [12] conducted a numerical simulation on a two-dimensional reservoir model, to demonstrate the suitability of CO_2 as cushion gas in natural gas storage. Based on this reservoir model, Ma et al. [17] studied the impact of geological parameters on the gas mixing behavior in UGSR with CO_2 -based cushion gas by using a hydromechanically coupled model. Their results showed that the mixing region decreases with increasing reservoir thickness and dip angle. However, the

reservoir model has been initially saturated with CO₂, which neglects the replacing process of the native CH₄ by the CO₂ during its injection into the depleted gas reservoir. In addition, it did not characterize the seasonal injection and production, as the mixing behavior has been studied only within 180 days. Moreover, the presence of residual water in the reservoir, which could affect the mixing behavior, has not been considered in their work. Niu and Tan [18] investigated the impacts of reservoir porosity and initial operation pressure on the mixing behavior of the cushion and working gases based on the three-dimensional gas-water two phase theory. Their results showed that the reservoir with high porosity and a large initial pressure is favored for limiting the formation and migration of the mixed zone. Oldenburg and Pan [19] conducted a one-dimensional radial simulation to investigate the pressurization and gas–gas mixing behavior. It was found that the pressure rise during working gas injection reduces if CO₂ is used as cushion gas. They also discovered that the impact of the CO₂ cushion on the reservoir pressure during the production period is much more significant than the one during the injection period, due to a higher production rate compared to the injection rate. The behavior of CO₂ and N₂ as cushion gases were compared by Kim et al. [20], the results showed that CO₂ is more suitable as cushion gas in depleted gas reservoirs in terms of the productivity index. However, in these works, a single well for both cushion gas injection and working gas production is very likely to accentuate the mixing of gases in a UGSR, which is not favorable in the engineering field.

For the purpose of investigating the suitability of using CO₂ as the cushion gas, the Donghae depleted gas reservoir located in Ulleung basin in Korea is used. It is important to note that the residual water is also considered in this work. After the injection of CO₂, the cyclic CH₄ production and injection was conducted over a period of 15 years, which is closer to a real engineering operation and makes it more beneficial for understanding the mixing behavior of CO₂ and CH₄ in a relatively long-term period. Additionally, the effect that the CO₂ fraction, reservoir temperature, reservoir permeability, residual water, and production rate have on the mixing behavior and gas production were analyzed in detail.

2. Simulation System

2.1. Governing Equations

The multicomponent and multiphase flow TOUGH2MP/TMVOC simulator, which could characterize the behavior of CO₂ and CH₄ in unsaturated zones, was used in this study [21]. The basic mass and energy balance equations for multi-component and multi-phase fluid flow in porous medium can be written in following form [22]:

$$\frac{d}{dt} \int_{V_n} M^k dV_n = \int_{\Gamma_n} \mathbf{F}^k \cdot \mathbf{n} d\Gamma_n + \int_{V_n} q^k dV_n, \quad (1)$$

where V_n is the control volume for an arbitrary subdomain in the flow system; Γ_n represents the closed surface of V_n ; $\mathbf{n}$ is a normal vector pointing inward into V_n ; M^k denotes the mass or energy per volume; k is the mass or energy components; $\mathbf{F}^k$ denotes the mass or heat flux; and q^k represents the sinks and sources [22].

The mass accumulation term for water, CO₂ and CH₄ in the systems can be expressed as:

$$M^k = \phi \sum_{\beta} S_{\beta} \rho_{\beta} X_{\beta}^k, \quad (2)$$

where ϕ is the porosity; S_{β} and ρ_{β} are the saturation and density of phase β , respectively; and X_{β}^k is the mass fraction of component k in phase β .

The advective mass flux $\mathbf{F}^k$ is a sum of phases which is given by:

$$\mathbf{F}^k = \sum_{\beta} \mathbf{x}_{\beta}^k \mathbf{F}_{\beta}. \quad (3)$$

The individual phase $\mathbf{F}_\beta$ is calculated by the multiphase version of Darcy's law:

$$\mathbf{F}_\beta = \rho_\beta \mathbf{u}_\beta = -K \frac{K_{r\beta} \rho_\beta}{\mu_\beta} (\nabla P_\beta - \rho_\beta \mathbf{g}), \quad (4)$$

where $\mathbf{u}_\beta$ is the Darcy velocity in phase β ; K is the absolute permeability; $K_{r\beta}$ denotes the relative permeability to phase β ; μ_β is viscosity; $\mathbf{g}$ denotes the vector of gravitational acceleration; and P_β represents the fluid pressure in phase β , which can be written as:

$$P_\beta = P + P_{c\beta}, \quad (5)$$

where P is the pressure of reference phase and $P_{c\beta}$ is the capillary pressure.

The diffusive flux of water, CO₂ and CH₄ in phase β are given in:

$$\mathbf{f}_\beta^k = -\phi \tau_0 \tau_\beta \rho_\beta d_\beta^k \nabla x_\beta^k, \quad (6)$$

where τ_0 is a porous medium dependent factor; τ_β is a coefficient determined by phase saturation; d_β^k denotes the diffusion coefficient of component k in phase β ; and x_β^k represents the mole fraction of component k in phase β .

A single effective multiphase diffusion coefficient can be defined as:

$$\sum_{\beta}^k = \phi \tau_0 \tau_\beta \rho_\beta d_\beta^k. \quad (7)$$

For two-phase conditions in the UGSR, total diffusive flux is then expressed by:

$$\mathbf{f}^k = -\sum_g^k \nabla x_g^k - \sum_w^k \nabla x_w^k. \quad (8)$$

The pressure and temperature dependent diffusion coefficients for CO₂ and CH₄ is given by:

$$d_\beta^k(P, T) = d_\beta^k(P_0, T_0) \frac{P_0}{P} \left(\frac{T + 273.15}{273.15} \right)^\theta, \quad (9)$$

where P_0 and T_0 denote the standard conditions, which equal 0.1 MPa and 0 °C, respectively; the temperature dependence parameter θ is 1.8. The error percentage of the diffusion coefficients for CO₂ and CH₄ calculated from Equation (9) is between 2.31% and 12.78% compared with the experimental results of Honari et al. [23] for the Estailades carbonate, Donnybrook sandstone, and Ketton carbonate, demonstrating the suitability of this empirical model. This model was implemented into the multicomponent and multiphase flow simulator TOUGH2MP that was used in this work.

2.2. Geological Model

A simple brick model with the dimension of 914.4 m × 914.4 m × 30.48 m was generated based on the geological information of the Donghae gas reservoir from the Ulleung basin in Korea, in which a slightly anticline structure is neglected [20]. As shown in Figure 2a, a typical five-spot pattern was used in this study, wherein one well is located at the lower formation for CO₂ injection, and the other four wells are located at upper formation for the CH₄ injection and production. In this symmetrical model, only a quarter model of the reservoir is selected for the simulation (Figure 2b). The applied geological properties of the reservoir are summarized in Table 1.

Figure 2. (a) Five-spot pattern depicting the CO₂ injection well and the production wells; (b) a quarter model of the reservoir in the Ulleung basin (modified from [20]).

Table 1. Reservoir parameters [20].

Parameters	Value
Porosity	0.2
Horizontal permeability (mD)	50
Vertical permeability (mD)	10
Gas saturation	80%
Initial pressure (MPa)	5.17

2.3. Operation Scenarios

Considering the low pressure of the depleted reservoir, two years of gas injection, i.e., CO₂ injection in the first year and CH₄ injection in the second year, was implemented before its operation as a gas storage reservoir, to recover the average reservoir pressure from 5.17 MPa to the original reservoir pressure of 24 MPa [24]. As shown in Figure 3, the gas storage reservoir works in one-year cycles and each cycle consists of four stages. In the first stage, CH₄ was produced at a rate of 4.05 kg/s from 1 November to 28 February the next year (120 days), followed by well shutting and facility checking from 1 March to 4 April (35 days). In the third stage, CH₄ was injected at a rate of 2.7 kg/s from 5 April to 1 October (180 days). Then the well was shut again from 2 October to 31 October (30 days).

Figure 3. Production and injection rate in a one-year cycle.

3. Results and Discussion

3.1. Properties and Behavior of the Gases in a UGSR

3.1.1. Physical Properties of the Mixed Gases

The mixing of CO₂ and CH₄ in a UGSR is affected by many factors, such as density differences, mobility ratios, molecular diffusion, and mechanical dispersion [2]. The density difference between CO₂ and CH₄ plays the most important role in the separation of the gases. Figure 4a,b shows the density and viscosity of CO₂–CH₄ mixtures at 40 °C, respectively, which were calculated by the WebGasEOS v.2.01 developed by the Lawrence Berkeley National Laboratory [25]. In comparison with CH₄, the higher density of CO₂ could lead to downward sinking of CO₂. As shown in Figure 4a, the density of the mixed gas was strongly correlated to the gas composition and pressure. Especially, with addition of small amounts the CH₄ into CO₂, the density decreased sharply. Such decreasing behavior slowed down with increasing CH₄ fraction. It should be mentioned that the sudden increase of density occurred near the critical pressure of CO₂ because of the CO₂ entering into the supercritical state.

Figure 4. (a) Density of CO₂–CH₄ mixtures at 40 °C and (b) viscosity of CO₂–CH₄ mixtures at 40 °C.

The mobility differences in CO₂–CH₄ displacement was primarily caused by different dynamic viscosities. Figure 4b shows the dynamic viscosities of CO₂–CH₄ mixtures, whose tendency is similar as that of the density in Figure 4a. The difference of viscosities between CO₂ and CH₄ is also favorable for limiting the gas mixing [26], but it may lead to an unstable contact interface [17].

The dispersion coefficient of CO₂ in CH₄ normally ranges from 0.01 to 0.3 cm²/min [27], which is a relatively slow velocity compared with the advective and convective transport. It should be pointed that the diffusion effect is proportional to concentration gradient, thus its effect will decrease along with the mixing process of gases. The mechanical dispersion is controlled by the movement of formation fluids [2].

3.1.2. Spatial Distribution of CO₂ and CH₄

Figure 5 shows the spatial distribution of CO₂ at the end of the CH₄ production stage, well shutting stage, CH₄ injection stage, and the second well shutting stage, i.e., the time of 120 d, 155 d, 335 d, and 365 d in the 1st, 5th, 10th, and 15th year, respectively for the gas storage reservoir with 10% CO₂ as the cushion gas. It can be seen that the CO₂ was concentrated at the bottom of the reservoir due to the large density compared with CH₄. The concentration of CO₂ in the reservoir decreased gradually with time, resulting from the production of CO₂ and the mixing of CO₂ with CH₄. Specifically, the maximum concentration of CO₂ in the UGSR decreased from 100% to 92.3%, 68.7%, 53.5%, and 43.6% at the end of the 1st, 5th, 10th, and 15th year, respectively.

Figure 5. (a) CO₂ saturation in the gas phase in the 3D model and (b) CO₂ saturation in the gas phase in the diagonal section, which consists of the CO₂ injection well and the CH₄ injection and production well.

The distribution of CO_2 changes periodically over time corresponding to the operation scenarios (Figure 3). In the first stage (CH_4 production), CO_2 moved towards the production well due to the pressure gradient. Meanwhile, the gases in the reservoir were significantly mixed until the end of gas production. In the second stage (well shutting), the behavior of the mixed gases was controlled dominantly by the density difference and diffusion effect. However, the effect on mixing was still minor due to the relatively low diffusion and short well shut-in time. In the third stage (CH_4 injection), CO_2 migrated towards the CO_2 injection well under the displacement effect of CH_4 injection. The mixed zone of gases became smaller in this stage. The last stage, i.e., the second well shutting stage, also had a negligible effect on the gases mixing.

3.2. Effect of the CO_2 Fraction

To figure out the suitability of CO_2 as cushion gas, the UGSR with a CO_2 concentration varying from 0% to 20% for the cushion was investigated. The reservoir average pressure against time is shown in Figure 6. It can be seen that all of the reservoir average pressure changed periodically over time, decreasing in the production stage, remaining constant during well shutting stage, and increasing again in the injection stage. In the first cycle, the minimum value of reservoir pressure decreased with the increasing CO_2 concentration due to the higher compressibility of CO_2 compared with CH_4 . With the operation of the UGSR, the maximum reservoir average pressure increased gradually as the produced CO_2 was replaced by the injected CH_4 , which had a lower compressibility, and the increase in CO_2 concentration led to pressure increment.

Figure 6. Average reservoir pressure for the underground gas storage reservoirs (UGSRs) with different concentrations of cushion gas CO_2 .

To quantify the spatial extent of the mixing region, the mixed thickness, defined as the distances between the CO_2 fraction level of 10% and 90% along the diagonal line crossing both the CO_2 injection well and the CH_4 production well (see in Figure 7), were compared in Figure 8 under various initial concentrations of CO_2 the cushion gas. The mixed thickness was used to characterize the interface between the working and cushion gases [19]. Clearly, the thickness of the mixed zone increased with the increasing of the CO_2 initiated concentration. The curve of the mixed zone's thickness for the UGSR with initial CO_2 concentration ranging from 8% to 10% was similar, in which the representative curve for the initial 9% of CO_2 could be divided into four stages over the lifetime of project (see in Figure 9c). Besides, the distance between the CO_2 injection well and 10% CO_2 ($r_{0.1}$), a 90% CO_2 ($r_{0.9}$) concentration point along the same diagonal line is plotted in Figure 9a,b against time, respectively. The variation of $r_{0.1}$ is harmonized with the periodical operation scenarios (Figure 3). It shows that $r_{0.1}$ was mainly driven by the gas displacement effect. However, this value was limited in a narrow range

from 189.9 to 193.7 m. Therefore, it could be concluded that the abrupt change of mixed zone thickness ($r_{0.1} - r_{0.9}$) was mostly caused by the variation of $r_{0.9}$.

Figure 7. Schematic diagram of the mixed zone in the diagonal section of the reservoir.

Figure 8. Thickness of the mixed zone for the UGSR with different concentration of CO₂ as cushion gas.

The stage I corresponds to the first injection in the first cycle. In which, $r_{0.1}$ increased gradually (Figure 9a), while $r_{0.9}$ fell very fast from 123.5 to 64.4 m (Figure 9b), thus the thickness of the mixed zone increased dramatically due to the mixing of CO₂ and CH₄ driven by the pressure gradient and the diffusion effect. In stage II, $r_{0.9}$ decreased only by 4.5 m and $r_{0.1}$ slightly decreased by about 2 m, so the thickness changes in this stage was only 2.5 m in 245 days, which corresponds to the period from the first well shutting to the second well shutting stage in the first operation cycle. Therefore, the thickness in the second stage behaved smoothly. Although the change of $r_{0.9}$ was not significant, the concentration of CO₂ near the CO₂ injection well decreased gradually. With the continuous mixing of CO₂ and CH₄, the concentration of CO₂ in the UGSR decreased to a value lower than 90%, thereby $r_{0.9}$ decreased to 0 and the distance of the mixed zone substantially increased again in a short time, which was assigned to stage III. In stage IV, the distance of the mixed zone equals $r_{0.1}$ and changes cyclically with the injection and production of CH₄.

Unlike the above curve (Figure 9b), there are two stable sections in the $r_{0.9}$ curve for the UGSR with 20% CO₂ (Figure 10), which corresponds to the two stable sections of the curve of the mixed zone (Figure 8). Figure 11 shows the CO₂ concentration in the diagonal of the reservoir for the UGSR with 20% CO₂. It can be seen that the mixed zone thickness maintained at 131.1 m from 0.35 to 0.84 a, corresponding to the first stable section in Figure 8. During this period, the concentration of CO₂ in the region away from the CO₂ injection well decreased to the threshold value of 90% gradually, then led to an increasing of the mixed zone. It also shows that the mixed zone thickness changed only from 189.7 to 195.9 m during the period of 0.95 to 2.22 a, corresponding to the second stable section in Figure 8.

During this period, the concentration of CO₂ in the region close to CO₂ injection well decreased to lower than 90% gradually, then led to the increasing of the mixed zone again.

Figure 9. (a) The distance between the CO₂ injection well and the point with 10% CO₂ in the diagonal section of the reservoir during production; (b) the distance between the CO₂ injection well and the point with 90% CO₂ in the diagonal line of the reservoir; and (c) the thickness of the mixed zone during production.

Figure 10. The relationship of $r_{0.9}$ with time for the UGSR with 20% CO₂ as cushion gas.

Figure 11. The CO₂ concentration in the diagonal of the reservoir for the UGSR with 20% CO₂.

In every production cycle, the CO₂ concentration in the produced gas mixture (CO₂–CH₄) over 15 years is shown Figure 12. Obviously, the CO₂ concentration in the produced gas mixture increases in every production cycle and reached up to the maximum at the end of each production. The maximum CO₂ concentration increased sharply, when CO₂ migrated to the production well in the first few years. Then the maximum concentration of CO₂ decreased gradually. This is a result of the decreasing amount of CO₂ in the UGSR. The maximum amount of the produced CO₂ concentration in the whole operation process was found at an earlier time with the increasing of the CO₂ cushion gas. It occurred at the 6th and 10th year for the UGSR with 20% and 9% CO₂ cushion gas, respectively. According to the national natural gas standards of China, the concentration of CO₂ in the second class natural gas for civil fuel is not allowed higher than 3% [28]. Therefore, it can be found in Figure 12, that the optimal CO₂ concentration for the cushion gas in this operation scenario was 9%, which would be used as the base case in following studies.

Figure 12. CO₂ concentration in the produced gas for the UGSR with different concentrations of CO₂ as cushion gas.

3.3. Effect of the Reservoir Temperature

Figures 13–15 show the average reservoir pressure, mixed zone thickness, and CO₂ concentration in produced gas over 15 years of operation under different temperatures, respectively. Logically, temperature had a positive influence on reservoir pressure change, and the cyclical changes were synchronized with the operation scenarios (Figure 3). Figure 14 shows that the thickness of the mixed zone decreased with the rising temperature in the initial stage, which is in accordance with the results from Ma et al. [17]. This result states that the high reservoir pressure caused by increasing temperature increased the dynamic viscosity of the gases. Likewise, the rapidly increased stage (stage III in Figure 9) occurred earlier with a lower temperature. After that, the ranking of the mixed zone thickness reversed eventually in stage IV. The mixing region increased with increasing temperature due to the high diffusion coefficient. Figure 15 shows that the impacts of temperature on the produced CO₂ concentration. It can be seen that the effect of temperature on the CO₂ concentration in the produced gas could be neglectable ranging from 30 to 50 °C.

Figure 13. Reservoir average pressure for different temperature.

Figure 14. Thickness of the mixed zone for different temperature.

Figure 15. CO₂ concentration in the produced gas for different temperatures.

3.4. Effect of the Reservoir Permeability

The permeability of depleted gas reservoirs may range from a few to hundreds of millidarcy. To quantify the impact of reservoir permeability on the mixing behavior of the gases, some cases with the reservoir horizontal permeability of 50, 70, 100, and 120 mD were carried out. As shown in Figure 16, the reservoir permeability had only a minor effect on the formation pressure, but the effect on the mixing behavior of CO₂ and CH₄ was much more significant. Figure 17 shows that the mixing thickness was positively correlated to permeability and this relationship was inversed after entering stage IV. In the production stage of the first year (stage I in Figure 9c), higher permeability will accelerate the migration of CO₂, further promoting the mixing behavior. With continuous operation, more CO₂ was produced along with CH₄ production in the case with higher permeability, which made the CO₂ concentration point of 10% quickly return to the CO₂ injection well. In other words, $r_{0.1}$ and the corresponding thickness of the mixed zone decreased much more quickly with higher permeability. Similar to that mentioned above, the thickness of the mixed zone changed periodically. The gases were much easier to mix in the reservoir with higher permeability. In stage IV (Figure 9c), the mixing zone was dominantly determined by $r_{0.1}$, the displacing effect of CH₄ injection was more significant in the permeable reservoir, thus the mixing zone was decreased with increasing permeability and varied in a large amplitude.

Figure 16. Reservoir average pressure for different reservoir horizontal permeability.

Figure 17. Thickness of the mixed zone for different reservoir horizontal permeabilities.

The CO_2 concentration in the produced gas for different reservoir horizontal permeability was compared in Figure 18. Clearly, CO_2 concentration in produced gas was positively correlated to the permeability. The high CO_2 production rates between 3 and 8 years for the reservoir with a permeability of 100 and 120 mD affected the distribution of CO_2 in the reservoir a lot and led to the non-uniform fluctuation of mixed region as shown in Figure 17. Therefore, the CO_2 concentration in a highly permeable reservoir might be less than that of a lowly permeable reservoir in the late period of an operation if no CO_2 was re-injected for supplementation. Likewise, the maximum CO_2 concentration was found at the earlier time in a highly permeable reservoir.

Figure 18. CO₂ concentration in the produced gas for different reservoir horizontal permeabilities.

3.5. Effect of the Residual Water

Figure 19 depicts the formation pressure for the reservoirs with different residual water saturations. The formation pressure increased with the growing residual water saturation, because incompressible water can build up higher pressure in the reservoir. Figure 20 shows the thickness of the mixed zone. In the production stage of the first year (stage I in Figure 9c), the mixing zone in the reservoir tended to increase with the expanding residual water saturation, one reason is that the occupied pores promote the injected CO₂ traveling deeper into the reservoir, i.e., higher mixing thickness under the same injection volume. Another reason is that the residual water also generates more tortuous flow-paths for the gases as shown in Figure 21, which was discovered in the result of nuclear magnetic resonance during a core flood experiment [29]. Therefore, the rapidly increasing stage (stage III in Figure 9c) occurred earlier with higher residual water saturation. For the same reason, the mixing thickness increasing happened firstly, in the case with higher residual water saturation after initiation.

Figure 19. Reservoir average pressure for different residual water saturations.

Like the effect of permeability, the mixing thickness was inversely correlated to the residual water saturation after entering stage IV. This was caused by the dissolution of CO₂ in residual water. As shown in Figure 22, less CO₂ existed as gas in the reservoir with higher water saturation, due to more CO₂ dissolved in the residual water. Figure 22 also shows the dissolved CO₂ changed periodicity

in a certain range during operation. These are results from the dissolution of CO_2 , which is principally pressure- and temperature-dependent. The CO_2 concentration in the produced gas for different residual water saturation is shown in Figure 23. It demonstrated that the effect of the residual water on the CO_2 concentration in the produced gas was still minor, which was due to the differences of the gaseous CO_2 in the UGSR being not very significant as shown in Figure 22.

Figure 20. Thickness of the mixed zone for different residual water saturations.

Figure 21. (a) Fluid flow in unsaturated porous rock and (b) fluid flow in water saturated porous rock [29].

Figure 22. CO_2 in gaseous and aqueous in the UGSR for different residual water saturations.

Figure 23. CO₂ concentration in the produced gas for different residual water saturations.

3.6. Effect of the Production Rate

The average reservoir pressure under different production rates were plotted against time in Figure 24. Noticeably, average reservoir pressure changed periodically and decreased with the increasing production rate. Comparing the mixing thickness shown in Figure 25, the impact on the mixing behavior of CO₂ and CH₄ was limited, especially in the early operation stages I–III. Considering the relatively significant difference of the mixed zone at the same operation rate (same pressure gradient) while at a different temperature and residual water saturation (different diffusion effect) in Figures 14 and 20, it could be inferred that the mixing of the gases in these stages is controlled by the diffusion effect rather than pressure gradient. In the subsequent operation, stage IV, the mixing zone had a slight decrease with an increasing production rate due to the impact of the pressure gradient. As shown in Figure 26, production also has a positive influence on the produced CO₂. The maximum amount of produced CO₂ was limited to under 3%, which still satisfies the required standard [28]. Therefore, the production rate at 4.2 kg/s could be used in this case, to improve the efficiency of the UGSR.

Figure 24. Reservoir average pressure for different production rates.

Figure 25. Thickness of the mixed zone for different for different production rates.

Figure 26. CO₂ concentration in the produced gas for different production rates.

4. Conclusions

The Donghae gas reservoir was used to study the suitability of utilizing CO₂ as the cushion gas for a depleted gas reservoir transformed gas storage reservoir. The maximum amount of CO₂ concentration for cushion gas was 9% under the condition of production and injection for 120 d and 180 d in a production cycle at a rate of 4.05 kg/s and 2.7 kg/s, respectively.

The typical curve of the mixing zone thickness could be divided into four stages. They were the increasing stage, the smooth stage, the suddenly increasing stage, and the periodic change stage. It should be mentioned that there existed two smooth stages and two suddenly increasing stages when the CO₂ concentration in the cushion gas increased to 20%. In the periodic change stage, the mixed zone thickness increased with the increasing of the CO₂ fraction, temperature, production rate, and the decreasing of permeability and water saturation. The correlation of the mixing zone thickness with reservoir temperature, permeability, and residual water was inverse in the former stages.

The CO₂ fraction in the cushion gas, reservoir permeability, and production rate had a significant effect on the breakthrough of CO₂ in the production well, while the effect of water saturation and temperature was relatively minor. For the purpose of utilizing more CO₂ as cushion gas in the UGSR, CO₂ should be injected for supplementation during the operation of UGSR, which will be discussed in our future work.

Author Contributions: C.C. and Z.H. proposed the main idea; C.C. and J.L. conducted the simulation and performed the analysis; C.C. and J.L. wrote the paper; H.X., F.M. and X.W. contributed to the development and discussion of the results. All authors have read and agreed to the published version of the manuscript.

Funding: This research was funded by the China Scholarship Council, grant number 201808080067, 201708080145.

Conflicts of Interest: The authors declare no conflict of interest.

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