Optimized schemes of enhanced shale gas recovery by CO_2-N_2 mixtures associated with CO_2 sequestration

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ARTICLE INFO

ABSTRACT

In response to the growing demand for CO_2 mitigation and unconventional natural gas, the oil and gas industry is researching for the viability to enhance shale gas recovery (ESGR) and sequester CO_2 in depleted reservoirs. Previous research has shown that a CO_2-N_2 mixture can be employed as a desired injection fluid to concurrently address both industrial issues. Due to the infancy of ESGR operations, there is no thorough knowledge of field-level optimized strategies using CO_2-N_2 for the ESGR associated with CO_2 sequestration. In this study, the commercial reservoir simulator *GEM* and CMOST by CMG were adopted to conduct $\sim$ 120 simulation scenarios based on the Barnett shale formation in order to comprehend the impacts of three dominating operational parameters and propose the corresponding solutions. This study reveals four major findings. Firstly, a larger N_2 concentration corresponds to a better recovery factor of shale gas but a shorter breakthrough time that causes the worse CO_2 storage performance. Secondly, an injection rate is initially proportional to the shale gas recovery and CO_2 storage performance but beyond a threshold rate, it can inversely affect both outcomes. Thirdly, the influence of a soaking period is limited in a short term and negligible in a long term for both ESGR and CO_2 sequestration. Lastly, by selecting an appropriate combination of an injection rate and a CO_2-N_2 ratio, the income of a project can significantly be improved by 22 % under the current carbon tax policy. This study provides shale gas operators with a guideline for implementing large-scale ESGR projects employing CO_2-N_2 mixtures.

1. Introduction

According to a recent report from International Energy Agency (IEA), the demand for natural gas is increasing by 430 billion cubic meters (bcm) between 2021 and 2030 while the conventional gas production will decline by around 740 bcm by 2050, and the natural gas demand will reach 5,100 bcm which is 30 % much higher than today [1]. The rising natural gas demand and the depletion of conventional gas reserves promoted the petroleum industry to shift its direction to unconventional resources. Shale gas is one of the most reliable unconventional natural gas resources thanks to the commercialized revolution of multi-stage hydraulic fracturing and horizontal drilling development [2].

However, the current matured recovery techniques can only recover about 30 % original gas in place (OGIP) owing to the complex trapping and adsorption mechanisms of methane [3–9]. On the other hand, a rising demand of energy triggered the increasing greenhouse gas emissions which accelerated the climate change. According to IEA, the annual energy-related and industrial CO_2 emissions are predicted to achieve 36 Gt by 2030, which would lead to 2.7℃ global temperature rising by 2100 with 50 % probability [10]. As Intergovernmental Panel on Climate Change (IPCC) proved, permanently sequestrating CO_2 in the subsurface is a promising solution to mitigate the CO_2 emissions in the current energy transition and linear carbon economy [11,12]. In the oil and gas industry, an application of CO_2 injection since the 1980 s can not

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only increase the hydrocarbon recovery factor (RF) but also mitigate the CO_2 emissions of the industry by storing CO_2 to depleted reservoirs [13–25]. In recent years, studies illustrated that it is feasible to deploy CO_2 to enhanced shale gas recovery (ESGR) due to pressure gradient changes and competitive adsorption mechanisms [26–33].
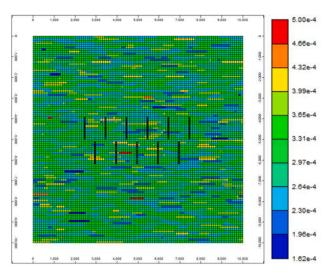
To our knowledge, only few known CO_2-ESGR projects worldwide are still in a primary recovery stage [34]. Unlike a primary recovery process which is driven mainly by pressure depletion after hydraulic fracturing, CO_2-N_2-ESGR can both increase a pressure gradient to overcome a low-permeability constraint and free up methane in an absorbed phase so that the RF can be significantly improved [35–37]. It has also been demonstrated that closure of the fracture also plays an important role in fluid recovery [38,39]. Additionally, the competitive gas adsorption follows Langmuir isotherms assuring that absorbed methane can be recovered [2]. The preferentially adsorption capacities of N_2, CH_4 and CO_2 on shale are in a ratio of 2:3:15 [37]. Injecting a mixture of high affinity gas (i.e., CO_2) and low affinity gas (i.e., N_2) can increase the RF of CH_4 because the absorption of CH_4 can be replaced by CO_2 due to the competitive adsorption mechanism and the partial pressure of CH_4 declined to promote CH_4 to a desorption state owing to N_2 injection. Theoretically, both CO_2-ESGR and N_2-ESGR can have a significant impact on ESGR, but injecting CO_2 leads to low mobility at the reservoir conditions while injecting N_2 always results in extremely short breakthrough time [34]. Thus, the injection of a CO_2-N_2 mixture can potentially extract more shale gas while avoiding individual draw-backs [34]. Furthermore, an application of CO_2-N_2 requires less separation energy from a flue gas which reduces the cost of a gas source. The research status of utilizing a CO_2-N_2 mixture for the ESGR purpose is still in an infancy stage. Prior to this work, experimental and numerical studies have proven that an application of a CO_2-N_2 mixture can achieve the goals of ESGR and CO_2 sequestration simultaneously by determining an optimal injectivity ratio [34]. Li and Elsworth proved that a mixture of CO_2-N_2 can increase the shale gas RF by ~ 20 % by controlling a component ratio [34]. On the other hand, CO_2 can be sequestrated in shale gas reservoirs mainly dominated by structural trapping and adsorption mechanisms [11,40]. Once shale gas is produced, a volume can be created to store CO_2, and the competitive adsorption of CO_2/CH_4 allows a greater CO_2 storage capacity in an absorbed phase [26,41–43]. However, the operating schemes were not well-established for CO_2-N_2-ESGR projects as they will directly impact the reservoir performance as well as the feasibility of CO_2 storage in shale gas reservoirs.

In this research, we developed a series of scenario studies based on the numerical simulator CMG-*GEM* to study the reservoir performance of CO_2-N_2-ESGR from both methane recovery and CO_2 sequestration perspectives. Optimal operating schemes and optimal gas ratios of CO_2-N_2 were selected based on different injection schemes including simultaneous flooding and cyclic flooding. The rest of this paper is structured as follows: Section 2 describes the basic reservoir properties of the numerical model and the selection of performance indicators. Section 3 discusses the results for all scenarios (~120 simulation case) as well as the impacts of operational conditions. Section 4 summarizes the key findings of this work.

2. Methodology

2.1. Reservoir model

In this section, a dual permeability 10,000 ft (L) × 10,000 ft (W) × 100 ft (H) reservoir simulation model was developed with a set of parameters according to the Barnett shale formation [44]. This heterogeneous reservoir with considering the natural fracture effect was developed based on the commercial compositional simulator *GEM* by CMG to solve a multi-component system [45,46]. The initial gas composition was assumed containing CH_4 only, and CO_2 and N_2 were defined as the injected gas components. The initial reservoir model was created and validated in the published work by Yu et al. [44], and then
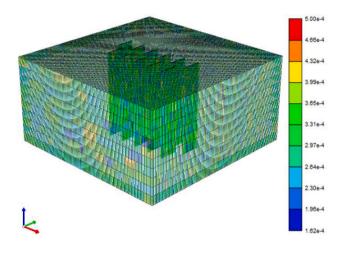

(a) 2D top view of the reservoir

(b) 3D view

Fig. 1. Heterogeneous reservoir model with colors standing for horizontal matrix permeability.

the reservoir heterogeneity and the injection parameters were modified for this study. A relationship between permeability and porosity for both matrix and natural fractures was calculated based on the Kozeny-Carman equation with assumed average permeability and porosity from the literature [7,44,47]:

$$k = \alpha \cdot \frac{\phi^3}{(1-\phi)^2} \tag{1}$$

where k is permeability [mD]; φ is porosity [unitless]; α is a regression coefficient. With the known average permeability and porosity, α can be calculated by equation (1). The porosity of each block was generated by Monte Carlo simulation with a normal distribution and average value. Therefore, the permeability of each block can be calculated by equation (1). The permeability and porosity of both matrix and fractures were calculated and input to CMG-*GEM* as described in Fig. 1 (a) and (b). The relative permeability for matrix and fractures can be found in Fig. 2 [36]. Two multistage horizontal hydraulic fractured wells were created as Fig. 1(a) shows. The space between these two wells is 1,000 ft, and the stage spacing and half-length were set at 1,000 and 500 ft, respectively. The detailed reservoir properties can be found in Table 1. Primary recovery lasted for 30 years with both wells serving as the producers with constant bottomhole pressure (BHP) assumed at 500 psi. The ESGR
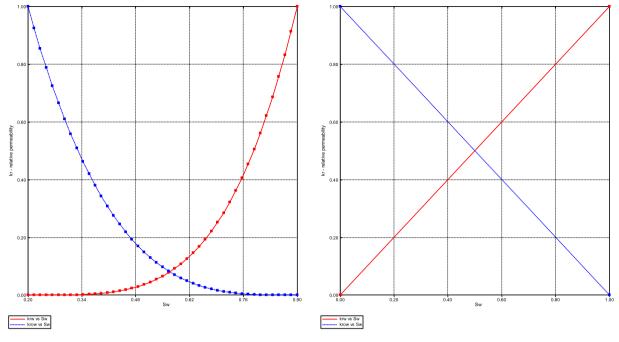
Fig. 2. Relative permeability curves for (a) matrix (left side) and (b) fractures (right side).

Table 1
Input parameters for the reservoir model.

Reservoir Property [44,48]	Value	Unit
Depth – D	6481	ft
Thickness – H	100	ft
Average porosity matrix – φ_m	0.041	–
Average porosity fracture – φ_f	0.007	–
Average permeability matrix – k_m	0.0003	mD
Average permeability fracture – k_f	1.4	mD
Anisotropy	0.1	–
Natural fracture spacing i,j,k	0.5	ft
Initial Reservoir Temperature – T_i	150	F
Initial Reservoir Pressure – P_i	4500	psi
Well Property		
Stage spacing	1000	ft
Half length	500	ft
Well spacing	1000	ft
Gas Adsorption [37]		
N_2 Lamgmuir Pressure Constant – $P_{L,N2}$	1087	psia
CH_4 Lamgmuir Pressure Constant – $P_{L,CH4}$	1596	psia
CO_2 Lamgmuir Pressure Constant – $P_{L,CO2}$	1254	psia
N_2 Lamgmuir Volume Constant – $V_{L,N2}$	11.8	scf/ton
CH_4 Lamgmuir Volume Constant – $V_{L,CH4}$	39.2	scf/ton
CO_2 Lamgmuir Volume Constant – $V_{L,CO2}$	183.6	scf/ton

started from year 31 by shifting one producer to the injector (bottom one in Fig. 1(a)) with a constant injection rate at 0.4 MMSCF/day and injection pressure assumed not to exceed the initial reservoir pressure (i.e., 4,500 psi). The OGIP contained free gas and absorbed gas with the total volume of 9.6×10^4 MMSCF. The maximum cumulative recovery factor of the primary production with both wells operating as the producers was estimated at 28.83 % over 50 years.

2.2. Gas adsorption and permeability evolution

Natural gas is usually trapped in three different thermodynamic states in shale: free compressed gas in intergranular porosity and fractures; absorbed gas on organic matters and clay materials; dissolved gas in kerogen [2]. Owing to different formations of shales, absorbed gas can account for 20 % to 85 % of total gas storage in shale reservoirs [49]. On the other hand, CO_2 is preferentially absorbed over methane and

nitrogen on both inorganic and organic micropores [26,31,41]. Therefore, taking advantage of the competitive adsorption can unlock a higher natural gas recovery factor. The classical Langmuir isotherm is the most commonly model to evaluate a monolayer gas adsorption volume at pressure P for shale gas reservoirs as shown in Equation (2) [2,34,44,50,51]:

$$V(P) = \frac{V_L P}{P + P_L} \tag{2}$$

where $V(P)$ is the gas adsorption volume at pressure P; V_L is the Langmuir volume constant corresponding to maximum adsorption capacity; P_L is the Langmuir pressure defining the curvature of an isotherm. Given the limitations of the classical Langmuir model, the BET model (Equation (3)) was developed to deal with multilayer adsorption [52]:
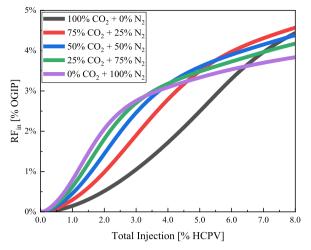
$$\frac{1}{V\left(\frac{p}{p_0} - 1\right)} = \frac{1}{V_m c} + \frac{c - 1}{V_m c}\left(\frac{P}{P_0}\right) \tag{3}$$

where P/P_0 is the relative pressure; V_m is the monolayer adsorption capacity; c is a characteristic energy constant. The extended Langmuir model was used by CMG-*GEM* to consider the multicomponent adsorption under a heterogenous surface condition as shown in Equation (4) [53,54]:
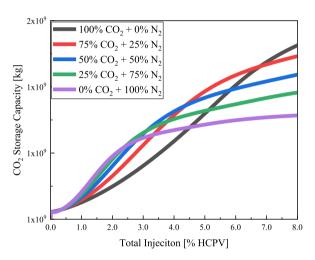
$$\omega_i = \frac{\omega_{i,max} B_i y_{i,aqu}}{1 + \sum_j B_j y_{j,aqu}} \tag{4}$$

where B_i is a parameter for a Langmuir isotherm relation; ω_i indicates the moles of an adsorbed component per unit mass of rock; $\omega_{i,max}$ is the maximum moles of the adsorbed component i per unit mass of rock; $y_{i,aqu}$ is the molar fraction of the adsorbed component i in the aqueous phase, where the aqueous phase is the gas phase for a shale gas reservoir as there is no other phases included in this study.

The permeability evolution described an effect of changing matrix and fracture permeability over time with gas injected to a shale gas reservoir. When injecting CO_2, the matrix permeability increases, and the fracture permeability decreases over time owing to the competitive adsorption CO_2 over CH_4 on shale triggering the matrix swell and fracture shrinkage. However, the injection of N_2 will lead to a decreasing matrix permeability and increasing fracture permeability due to its

(a) Total Injection vs. RF_{in} with respect to different CO_2-N_2 ratio
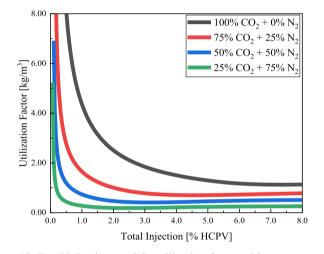
(b) Total injection vs. CO_2 retention and CO_2 stored with respect to different CO_2-N_2 ratio

(c) Total injection vs. CO_2 storage capacity with respect to different CO_2-N_2 ratio

(d) Total injection vs. CO_2 utilization factor with respect to different CO_2-N_2 ratio

Fig. 3. Impact of CO_2-N_2 ratio.

effect of desorbing CH_4 by lowering its partial pressure and causing the matrix shrinkage and fracture widening [34].

2.3. Computational indicators

In this section, the calculational methods were demonstrated from the perspectives of shale gas recovery and CO_2 sequestration. To evaluate the performance of shale gas recovery, the RF [%] was adopted by taking a ratio of cumulative gas production over the OGIP as shown in Equation (5):

$$RF_{in} = \frac{P_{cum} - E[P_{cum}]}{OGIP}$$ (5)

where P_{cum} is the cumulative shale gas produced and $E[P_{cum}]$ is the expected cumulative shale gas production without ESGR. The total gases injection including CO_2 and N_2 at the ESGR stage is presented as the percentage of a hydrocarbon pore volume (HCPV) at the surface condition defined by Equation (6):

$$HCPV_{inj} = \frac{\sum V_{\alpha,sc}}{V_{HCPV}}$$ (6)

where $V_{\alpha,sc}$ is the volume of the injected α gases at the surface condition, where α can include CO_2 and N_2; V_{HCPV} is the total HCPV; and $HCPV_{inj}$ is the unitless injected gas in the HCPV scale.

Besides the shale gas recovery, CO_2 sequestration is another major consideration in the context of greenhouse gases emission reduction. As described in the previous sections, injecting CO_2 into shale gas reservoirs can not only improve the gas recovery, but also permanently sequestrate CO_2 to mitigate the CO_2 emissions. In addition, successful CO_2 storage can earn a carbon credit to improve the revenues of a project. According to IEA, the carbon price will rise CAD $15 per tonne annually to CAD $170 per tonne by 2030 [55]. In this work, the CO_2 storage capacity and CO_2 retention factor were considered to evaluate the performance of CO_2 sequestration in a reservoir as illustrated in Equations (7) and (8), respectively. The CO_2 storage capacity can be calculated based on the modified USDOE method with considering contributions from both free trapping and adsorption mechanisms; an impact of hysteresis was neglected in this study [40,56–59]. The total volume that can be occupied by CO_2 is the volume of free trapped CO_2 plus the additional absorbed CO_2 in a shale formation owing to the competitive adsorption effect and deducting the volume that is occupied by nitrogen. The retention factor of CO_2 in percentage can be calculated by taking a ratio of retained CO_2 over total injected CO_2 as illustrated in Equation (8).

Both Equations (7) and (8) are calculated at the standard conditions:

$$M_{CO_2} = \rho_{CO_2,sc} \cdot \left(RF_{cum} \times OGIP + V_{CO_2,absorbed} - V_{CH_4,absorbed} - V_{N_2,retained} \right)$$

(7)

$$R_{CO_2} = \frac{M_{CO_2,inj} - M_{CO_2,prod}}{M_{CO_2,inj}}$$

(8)

The utilization factor normalized the recovery efficiency of natural gas production by applying CO_2. It defines an amount of CO_2 utilized to recover a unit volume of natural gas. Since CO_2 is an expensive source for field applications decades ago, minimizing the CO_2 usage was always expected to maximize project revenues [60,61]. However, with a large-scale implementation of an emission trading market, the industrial direction will shift to store CO_2 as much as possible so that a carbon credit can be received to maximize the project revenues. Hence, the utilization factor can be given in Equation (9):

$$UF_{CO_2} = \frac{M_{CO_2,stored}}{RF_{in} \times OGIP}$$

(9)

where UF_{CO2} is the utilization factor [M/L^3]; RF_{in} is the incremental recovery factor. The UF_{CO2} demonstrated that an amount of CO_2 was stored in mass when recovering an additional volume of natural gas from a reservoir. We used the mass unit of CO_2 and the volumetric unit of natural gas as the unit market prices for CO_2 and natural gas are in ton and m^3, respectively. The utilization factor can be adopted to describe the reservoir performance from both resource recovery and CO_2 sequestration perspectives.
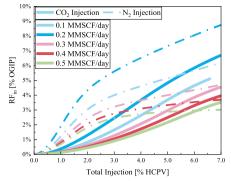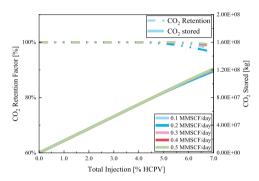
3. Result and discussion

3.1. Continuous flooding

In this section, an optimal CO_2-N_2 ratio and injection rate were investigated based on a series of reservoir simulations. The constant injection rate operation was assumed at the ESGR stage with the injector BHP not exceeding its initial reservoir pressure. With the total injection of a gas mixture at 8.0 HCPV, the RF_{in} of shale gas, CO_2 storage capacity, CO_2 retention factor and utilization factor were compared for all scenarios.

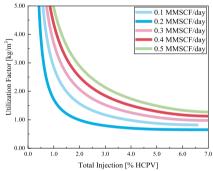### 3.1.1. Impact of a CO_2-N_2 ratio

As illustrated in Fig. 3, Fig. 3(a) demonstrated the incremental recovery factor after the gas mixture is injected into the reservoir. As a result, the pure application of nitrogen has a better performance at the early stage owing to its impact on increasing the matrix permeability so that CH_4 molecules can flow to fractures. However, due to the smallest adsorption effect of N_2 compared to CH_4 and CO_2, the recovered amount of absorbed CH_4 is limited, and the breakthrough time of N_2 injection is shorter compared to the CO_2 injection. The injection of a CO_2-N_2 mixture can postpone the breakthrough time compared with pure N_2 injection and a larger adsorption effect of CO_2 can recover the absorbed CH_4 with the help of N_2 lowering the partial pressure of CH_4. We further observed that the injected CO_2 was absorbed to the shale formation at an early stage of the injection so that the matrix permeability was not increased, and hence the incremental gas recovery was maintained at a lower factor. Therefore, the pure CO_2 injection can perform better at a late stage because of the increasing fracture permeability and much more absorbed CH_4 replaced by CO_2 owing to the competitive adsorption mechanism. Fig. 3(b) showed the evaluation of CO_2 sequestration, and the results illustrated that all injected CO_2 can be stored as long as the breakthrough time is not achieved. A higher concentration of CO_2 can trigger a slower breakthrough time. The CO_2 storage capacity curves (Fig. 3(c)) were in the same shape as RF_{in} due to a correlation between the storage capacity and cumulative recovery factor. Thus, the N_2 application will result in less CO_2 storage capacity and a shorter

(a) Total injection vs. RF_{in} with respect to different injection rate of CO_2 and N_2

(b) Total injection vs. CO_2 retention & CO_2 stored with different CO_2 injection rate

(c) Total injection vs. CO_2 utilization factor with respect to different CO_2 injection rate

Fig. 4. The impacts of an injection rate.

breakthrough period of injected gas. The cumulative CO_2 production is positively correlated with the N_2 percentage. However, it can improve the methane recovery factor at an early stage for shale reservoirs. To analyze from both natural gas recovery and CO_2 sequestrating perspectives, we adopted the utilization factor of CO_2 as shown in Fig. 3(d). The utilization factor declines rapidly starting from the injection of gas and approximates a constant value after the injected gas breakthroughs. The final utilization factor of CO_2 is in a range from 0.26 to 1.16 kg/m^3 increasing with the CO_2 purity indicating that 1 m^3 of CH_4 recovered can store up to 1.16 kg CO_2. Overall, the injection of 75 % CO_2 plus 25 % N_2 less than 7.0 HCPV can reach the highest gas recovery and CO_2 storage goals with the CO_2 utilization factor at 0.8 kg/m^3 according to our observations.

3.1.2. Impact of an injection rate

Besides the CO_2-N_2 ratio, the variation of an injection rate can also significantly influence the natural gas recovery as well as the CO_2 storage according to the results in Fig. 4. The injection rates in a range from 0.1 to 0.5 MMSCF/day were studied accordingly to address the impact of changing an injection rate. Increasing an injection rate can improve the shale gas recovery before the injected gas breakthroughs
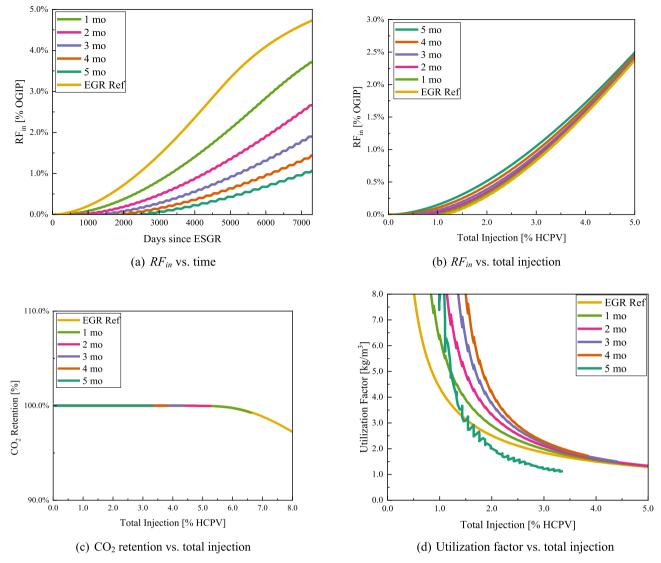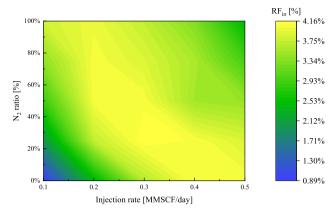
(a) RF_{in} vs. time

(b) RF_{in} vs. total injection

(c) CO_2 retention vs. total injection

(d) Utilization factor vs. total injection

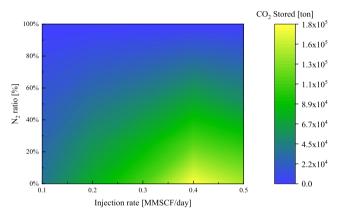**Fig. 5.** Performance comparison of cyclic flooding scenarios.

within the same time frame. However, when controlling the total mass of injected gas there was a threshold injection rate at which a higher injection rate led to a lower recovery factor. For example, the optimal injection rate in this study was 0.2 MMSCF/day for both pure CO_2 and N_2 as the ESGR fluids showed as the solid and dash-dot lines in Fig. 4(a), respectively, with the same amount of total injection fluids. The reason is that a higher injection rate requires a higher BHP of the injector and hence reduces breakthrough time owing to the higher reservoir pressure and permeability. Thus, the production rate of CH_4 declines rapidly after the injected gas breakthroughs as more CO_2 was being produced, and result in a lower sweep efficiency of CH_4 afterward. Hence, a longer breakthrough time can secure a higher recovery factor. On the other hand, if the injection rate was extremely low, the injected gas remained immobile so that the free CH_4 molecules cannot flow to the producer. Additionally, the fracture permeability cannot be effectively improved with a great increasing rate of gases beyond the threshold injection rate. Therefore, the natural gas cannot be extracted. The results of CO_2 retention and the amount of CO_2 stored (Fig. 4(b)) were similar as the previous results stated. As long as the breakthrough point is not reached, the injected CO_2 can be successfully sequestrated. The CO_2 utilization factors were in a range from 0.65 to 1.43 m^3/kg. A higher injection rate can store more CO_2 per cubic meter of CH_4 recovered, but it also indicated less efficiency of CO_2 utilization. To sum up, the 0.2 MMSCF can

not only recover the most amount of CH_4 but also effectively store CO_2 with a utilization factor of 0.65 kg/m^3 compared with other scenarios for pure CO_2.
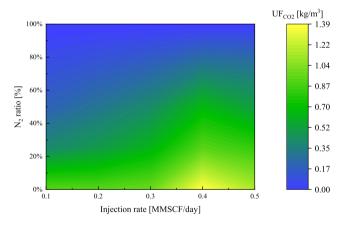
3.2. Cyclic flooding

In this section, we studied five cyclic injection scenarios to address the impact of soaking time on the reservoir performance. As known, a Huff and Puff (HnP) process can be applied to shale reservoirs and the soaking time can significantly impact a recovery factor [62]. To analyze the impact of a soaking period on the shale gas recovery and CO_2 sequestration, we developed the five cyclic injection scenarios with a soaking period from 1 to 5 months as shown in Fig. 5. For each scenario, the producer and injector are alternatively shut in for a specific time so that the soaking impact of a CO_2-N_2 mixture can be addressed. The optimal CO_2-N_2 mixture with a ratio of 75 % CO_2 plus 25 % N_2 was deployed, as proven in Section 3.1. According to Fig. 5(a), a longer soaking period requires longer time to inject the same amount of gas to the reservoir and trigger a lower RF_{in} within the same timeframe. However, Fig. 5(b) shows that when the total injections were same, a longer period can improve the shale gas recovery as more absorbed CH_4 can be extracted especially in an early stage of operation. With the total injected fluid increasing, the RF_{in} of each scenario was approximate to

(a) The optimization solution for RF_{in}

(b) The optimization solution for CO_2 sequestration

(c) The optimization solution for CO_2 utilization factor

Fig. 6. Optimization scheme from engineering perspective.

each other as the impact of a soaking time decays. Fig. 5(c) shows that a soaking period has no impact on the breakthrough time as all scenarios were sharing the same curve of CO_2 retention. Similarly, the utilization factor of CO_2 was different at an early stage of the simulation owing to the distinct recovery factors and approached the same value with injecting an increasing amount of the gas mixture. Understanding the impact of a soaking period can provide a sense to formulate an economic evaluation of operation as to whether it is worthwhile to recover an extra amount of CH_4 by an impact of the soaking mechanism in a longer timeframe with less total injected CO_2-N_2 mixture. In brief, the soaking period of injected gas had limited impact on both ESGR and CO_2 sequestration according to our simulation results.
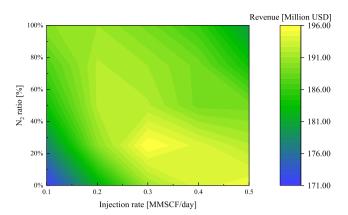

Fig. 7. Optimization scheme of project revenues.

3.3. Optimized schemes

In this section, an optimized operation scheme was determined based on simulations using the CMOST module. An injection rate, CO_2-N_2 ratio, and soaking period were considered initially as the optimizers. However, as discussed in the previous section, a soaking period has no impact after 4.0 HCPV of injected gases. The cyclic injection schemes were eliminated in this section and the continuous ESGR flooding was assumed for over 40 years so that at least 4.0 HCPV of a gas mixture can be injected to the reservoir. By selecting different combinations of an injection rate and a CO_2-N_2 ratio, the optimal ESGR and CO_2 sequestration performance can be determined according to Fig. 6 (a)-(c) with ~ 100 simulation cases performed by CMOST. In Figs. 6 and 7, a N_2 ratio defines the percentage of N_2 of the injected gas. For example, the 100 % N_2 ratio indicates that the injected gas is comprised of N_2 only, whereas the 0 % N_2 ratio indicates that the injected gas is comprised of CO_2 only. As shown in Fig. 6(a), the highest RF_{in} can reach 4.16 % by injecting 30 % N_2 and 70 % CO_2 at an injection rate of 0.3 MMSCF/day. A higher injection rate with a higher N_2 ratio result in an inefficient recovery factor, as the absorbed natural gas cannot be displaced to the producer. Furthermore, a low injection rate and high purity of CO_2 cannot effectively achieve the greatest ESGR recovery goal, as the matrix permeability was not increasing owing to a larger adsorption capacity of CO_2. Fig. 6(b) illustrated the performance of CO_2 sequestration by estimating the total amount of CO_2 stored to the reservoir. As discussed in the previous sections, the injected CO_2 can be successfully stored in the reservoir before the gas breakthrough. A larger injection rate can result in a shorter breakthrough time. Once the injected gas was produced, the decreasing CO_2 retention factor suggested that the storage efficiency was decreasing. An optimal combination to store CO_2 associated with the ESGR process was at an injection rate of 0.4 MMSCF/day and 100 % CO_2 concentration of the injected gas. The result proved that the maximum of 180 thousand tonnes CO_2 can be stored in the reservoir according to the simulations. The CO_2 utilization factor can be calculated consequently as shown in Fig. 6(c). Owing to the greatest CO_2 storage, the pure CO_2 injection at a rate of 0.4 MMSCF/day also leads to the highest utilization factor, which is 1.39 kg/m^3 indicating that one cubic meter of natural gas recovery can contribute to 1.39 kg of CO_2 storage during the ESGR process. However, a higher utilization factor also suggests the less efficiency of CO_2. Thus, a project optimization scheme should not only consider the reservoir performance but also economic revenues.

The economic revenues without considering the time value of money (a discount rate) can be estimated by summing up the sale revenues of natural gas plus receiving a tax credit for the carbon sequestration as in Equation (10):

$$Revenue = p_{NG} \times Q_{NG} + p_{CO_2} \times Q_{CO_2} \qquad (10)$$

where p_{NG} is the unit price of natural gas assumed at \$6.85/MCF in 2022 USD according to the U.S. energy information agency (USEIA); p_{CO2} is the carbon price for geologically sequestrated CO_2 with EOR (enhanced oil recovery) at the value of \$25/ton in 2022 USD according to 45Q; Q_{NG} and Q_{CO2} are the cumulative produced CH_4 and cumulative stored CO_2, respectively, according to numerical simulations [63,64]. Fig. 7 illustrates the revenue variation with combinations of an injection rate and a CO_2-N_2 ratio. As a result, the highest total revenue requires the largest amount of natural gas production and storing CO_2 as much as possible in the current price scenario. A successful implementation of CO_2-N_2 ESGR can earn up to 7 % to 22 % more revenues for a project, and 13 % of the additional revenues is from the carbon credit. Therefore, under the current carbon price, the CO_2 sequestration can significantly improve the revenues of an ESGR project. By selecting the optimal operation strategy, the revenues of a project can be improved by 15 %.

4. Conclusions

In this work, we deployed the *GEM* and CMOST of CMG to investigate the feasibility of a CO_2-N_2 mixture for the purpose of ESGR and proposed the optimized operating strategies to overcome the dual challenges of maximizing the natural gas recovery as well as permanently sequestrating CO_2 in shale formations. A total of 120 simulation scenarios were conducted to address the impacts of a CO_2-N_2 ratio, injection rate and cyclic soaking period, and an optimal combination of a CO_2-N_2 ratio and an injection rate was concluded to maximize project revenues consequently. The competitive adsorption mechanism was considered during an ESGR phase, and the performance of a reservoir was evaluated from the natural gas recovery and CO_2 sequestration perspectives. The limitation of this study includes that the CO_2 storage mechanisms do not include a mineralization process. As the results indicated, we summarize the following contributions:

(1) A larger concentration of N_2 corresponds to a higher methane recovery factor at an early stage but caused less amount of CO_2 storage owing to a shorter breakthrough period.

(2) A higher injection rate does not always indicate that a greater amount of natural gas can be recovered associated with great performance of CO_2 sequestration.

(3) A soaking period only impacts the ESGR performance at an early stage and has no impact to both gas recovery and CO_2 sequestration in a long term.

(4) An optimized scheme can be determined by varying an injection rate and a CO_2-N_2 ratio simultaneously to achieve the max revenues with earning a carbon credit at a significant level. In our case, the optimal combination is 70 % CO_2 plus 30 % N_2 at an injection rate of 0.33 MMSCF/day with the continuous flooding.

(5) The optimized scheme shows that a project can earn up to 22 % more revenues under the current carbon tax policy. By controlling an injection rate and a gas mixture ratio, the project revenues can be improved by 15 %.

Declaration of Competing Interest

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

Data availability

Data will be made available on request.

Acknowledgements

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