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CARBON SEQUESTRATION IN NATURAL GAS RESERVOIRS: ENHANCED GAS RECOVERY AND NATURAL GAS STORAGE

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ABSTRACT

Natural gas reservoirs are obvious targets for carbon sequestration by direct carbon dioxide (CO_2) injection by virtue of their proven record of gas production and integrity against gas escape. Carbon sequestration in depleted natural gas reservoirs can be coupled with enhanced gas production by injecting CO_2 into the reservoir as it is being produced, a process called Carbon Sequestration with Enhanced Gas Recovery (CSEGR). In this process, supercritical CO_2 is injected deep in the reservoir while methane (CH_4) is produced at wells some distance away. The active injection of CO_2 causes repressurization and CH_4 displacement to allow the control and enhancement of gas recovery relative to water-drive or depletion-drive reservoir operations. Carbon dioxide undergoes a large change in density as CO_2 gas passes through the critical pressure at temperatures near the critical temperature. This feature makes CO_2 a potentially effective cushion gas for gas storage reservoirs. Thus at the end of the CSEGR process when the reservoir is filled with CO_2, additional benefit of the reservoir may be obtained through its operation as a natural gas storage reservoir. In this paper, we present discussion and simulation results from TOUGH2/EOS7C of gas mixture property prediction, gas injection, repressurization, migration, and mixing processes that occur in gas reservoirs under active CO_2 injection.

INTRODUCTION

Depleted natural gas reservoirs are promising sites for geologic carbon sequestration by direct carbon dioxide (CO_2) injection. First and foremost, natural gas reservoirs have a demonstrated integrity against gas escape shown by their long term containment of methane (CH_4) and capacity estimated at 140 GtC worldwide (IEA, 1997) and 10 to 25 GtC in the U.S. alone (Reichle et al., 1999). Second, past production activity has allowed them to be relatively well characterized and well understood. Third, there is an existing infrastructure of wells and pipelines and associated land use history amenable to an industrial process such as direct CO_2 injection. These positive aspects of natural gas reservoirs are further complemented by the fact that recovery factors for gas reservoirs average approximately 75% (Laherrere, 1997), meaning that even when they are considered depleted, many natural gas reservoirs contain significant natural gas that can be potentially recovered by enhanced gas recovery processes associated with injecting CO_2 for carbon sequestration. This additional gas recovery can be used to offset the cost of CO_2 injection. These many aspects of depleted natural gas reservoirs have led to the idea that carbon sequestration with enhanced gas recovery (CSEGR) could be a potentially important process to sequester CO_2 and enhance CH_4 production from depleting gas reservoirs.

Although the concept of CSEGR has been discussed for more than ten years (e.g., van der Burgt et al., 1992; Blok et al., 1997), CSEGR has never actually been tested in any gas reservoir. The main reason for this is that CO_2 is today still an expensive commodity and geologic carbon sequestration is not yet widely practiced. The other reason for resistance to CSEGR is the concern that injected CO_2 will rapidly mix with existing CH_4 thus degrading the natural gas resource.

Recently, we have been carrying out numerical simulations of CSEGR that suggest the process is technically feasible and that mixing between the injected CO_2 and existing CH_4 may not be extensive and that it can potentially be controlled by operational strategies (Oldenburg et al., 2001; Oldenburg and Benson, 2002; Oldenburg et al., 2003). These numerical simulation studies have been complemented by independent laboratory studies that also suggest that CSEGR is technical feasible (Mamora and Seo, 2002).

One strategy that can be used in CSEGR to avoid extensive mixing between injected CO_2 and existing CH_4 is to take advantage the much greater density of CO_2 relative to CH_4 at reservoir conditions and inject CO_2 deep in the reservoir while producing gas from high in the reservoir. In this strategy, CH_4 could be swept from the reservoir both laterally and from the bottom up. At the end of the CSEGR operation, the reservoir would contain mostly CO_2.

Through the study of CO_2 properties at a range of reservoir conditions, a related use of the post-CSEGR reservoir became apparent. Namely, the post-

CSEGR reservoir could be used as a gas storage reservoir with CO_2 playing the role of a cushion gas. It turns out that the same density changes in CO_2 that occur around the critical pressure and that can be used to inhibit mixing during the process of CSEGR can be exploited in a gas storage reservoir operated around the critical pressure (Oldenburg, 2003). This potential post-CSEGR option provides further potential benefit to applying the CSEGR process in depleting natural gas reservoirs.

This paper presents a review of the CSEGR process and the potential post-CSEGR use of the reservoir as a natural gas storage reservoir. Briefly, the scenario we envision involves CO_2 injection into a depleted gas reservoir for CSEGR. Following the nearly complete removal of CH_4 from the reservoir, say over 5–10 years, the CO_2-filled reservoir can be used for gas storage.

To test these concepts, numerical experiments are carried out using a new TOUGH2 module called TOUGH2/EOS7C. The key to simulating gas reservoir processes is accurate gas mixture property prediction. We present a review of the properties of CO_2 and CH_4 gas mixtures. This is followed by conceptual descriptions of the processes of CSEGR and gas storage with CO_2 as a cushion gas. Next we present a summary of the methods we use to simulate these processes, along with a summary of the verification of our property predictions. In the last parts of the paper, we present simulation results that illustrate the processes. Finally, we discuss limitations and outstanding issues that need to be addressed in research to make progress in these areas.

GAS PROPERTIES

Phase Diagram

The phase diagram for CO_2 is shown in Figure 1, along with a typical P, T path assuming hydrostatic pressure and 25 °C km^{-1} gradients with depth in the earth. As shown, CO_2 will be supercritical (P_{crit} = 73.8 bar, T_{crit} = 31.0 °C (Vargaftik, 1975)) in gas reservoirs that are typically at depths greater than 1 km and hotter than 35 °C. In depleted reservoirs, pressures can be below hydrostatic and subcritical conditions may exist. Therefore, the simulation capability must treat both supercritical and gaseous states of CO_2.

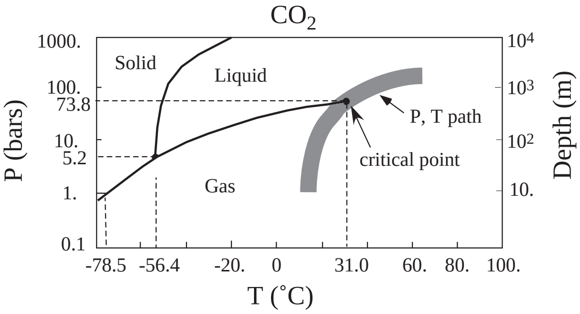

Figure 1. Phase diagram of CO_2 showing typical P, T path assuming hydrostatic pressure and 25 °C km^{-1} geothermal gradient.

Density and Compressibility Factor

At temperatures near the critical temperature, increasing pressure causes CO_2 to transition from gaseous conditions to supercritical conditions with an associated large change in density. Pure CH_4 exhibits no such drastic change in this same range of presssure and temperature and shows nearly ideal gas behavior. The degree of non-ideality shown by CO_2 can be expressed by the compressibility factor, Z, where

$$Z = \frac{PV}{nRT} \tag{1}$$

where Z = 1.0 for ideal gases, P is pressure (Pa), V is volume (m^3), n is moles, R is the universal gas constant (J mol^{-1} K^{-1}), and T is temperature (K). Plotted in Figure 2 are density and compressibility factor (Z) of CO_2-CH_4 mixtures as a function of pressure at T = 40 °C as calculated by the NIST14 database (NIST, 1992; Magee et al., 1994). As shown, the density of supercritical CO_2 can be very high, and is much larger than the density of CH_4 at all relevant gas reservoir conditions.

Viscosity

The viscosity of CO_2 also increases as pressure rises through the critical pressure. Shown in Figure 3 are density and viscosity of CO_2-CH_4 mixtures at several different pressures from subcritical to supercritical for CO_2 (NIST14, 1992; Magee et al., 1994). Even though the density of supercritical CO_2 can approach that of liquid water, the viscosities of CO_2-CH_4 mixtures across the range of compositions are always gas-like. Thus supercritical CO_2 is a highly mobile (low viscosity) fluid with very high density. Note further that CO_2 viscosity is always larger that CH_4 viscosity.

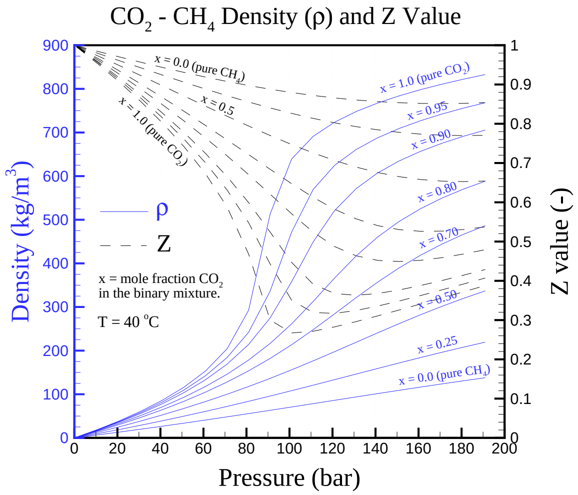

Figure 2. Density and Z factor for CO_2-CH_4 mixtures showing strongly non-ideal behavior of CO_2-rich gas.

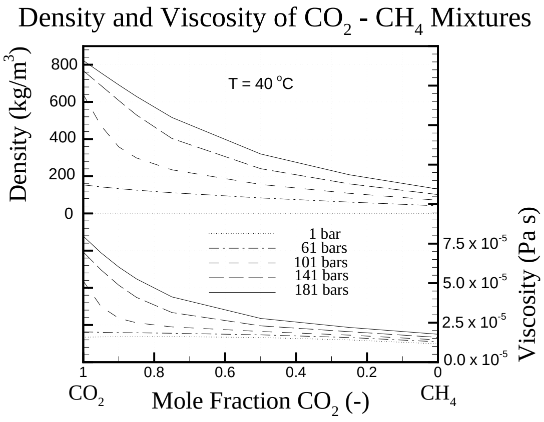

Figure 3. Density and viscosity of CO_2-CH_4 mixtures at several pressures at 40 °C.

ENHANCED GAS RECOVERY

Figure 4 shows a schematic of one scenario for CSEGR. In this scenario, CH_4 produced from a depleting gas reservoir is used to generate electricity. The flue gases from power generation and industrial activities are scrubbed and CO_2 is captured, compressed, and transported back to the CH_4 reservoir where it is reinjected at some distance from production wells. In this process, the injection of CO_2 raises the reservoir pressure and displaces CH_4 toward the production wells, thereby enhancing CH_4 production. Because of the pressure maintenance effected by the CO_2 injection, the CH_4 production can be controlled and water entry avoided in the case of water-drive reservoirs.

One critical issue in this process is the degree to which the injected CO_2 will mix with the CH_4, degrading the quality of the produced gas. The physical properties of CO_2 and CH_4 at reservoir conditions appear to discourage mixing. Specifically, the high density of CO_2 can be exploited by injecting the CO_2 in the lower parts of the reservoir while producing from higher parts. The lower mobility of CO_2 (higher viscosity) relative to CH_4 produces a stable displacement process. Furthermore, the nearly gas-like viscosity of supercritical CO_2 means that it can be injected easily. Together, the properties of CO_2 and CH_4 including density, viscosity, and viscosity ratio promote downward migration, injectibility, and stable displacements, respectively. These effects work in favor of CSEGR and will be demonstrated in numerical simulations below.

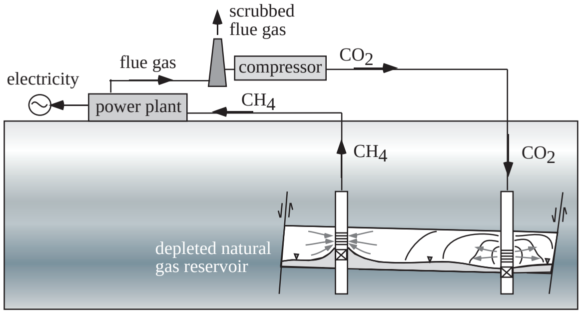

Figure 4. Schematic of CSEGR.

GAS STORAGE WITH CO_2 AS CUSHION GAS

Once the reservoir is filled largely with CO_2, it will serve as an effective carbon sequestration site. Such a reservoir may have additional potential uses, specifically as a natural gas storage reservoir (Oldenburg, 2003). Natural gas storage is used to smooth the natural gas supply (Katz and Tek, 1981). The approach allows storage in reservoirs near metropolitan areas of natural gas (CH_4) delivered by pipeline from far-off production fields during periods of low demand, and rapid CH_4 production from the storage reservoir during periods of high demand.

Critical to the use of gas storage reservoirs is the idea of a cushion gas, i.e., a gas whose compression allows additional storage of working gas, and whose expansion helps drive production of the working gas. A schematic showing a gas storage reservoir is shown in Figure 5. Often, the cushion gas is remnant CH_4 in a depleted gas reservoir, although inert cushion gases such as nitrogen have also been used (Laille et al., 1988). Just about any gas will work as a cushion gas.

Table 1. Properties of CO_2-CH_4 gas mixtures and aqueous solubility at 40 and 100 bars, 40 °C.

P = 40 bars, T = 40 °C	gas phase				aqueous phase	
Prediction Source	x_g^{CH4}	x_g^{CO2}	ρ (kg m^{-3})	μ (Pa s)	x_l^{CH4}	x_l^{CO2}
TOUGH2/EOS7C	0.	1.	85.45	1.70×10^{-5}	0.	1.62×10^{-2}
Reference Values	0.	1.	83.79 [1]	1.73×10^{-5} [1]	0.	1.37×10^{-2} [2]
TOUGH2/EOS7C	0.5	0.5	51.97	1.44×10^{-5}	3.73×10^{-4}	8.07×10^{-3}
Reference Values	0.5	0.5	51.33 [1]	1.67×10^{-5} [1]	3.66×10^{-4} [3,4,5,6]	6.74×10^{-3} [3,4,5,6]
TOUGH2/EOS7C	1.	0.	26.42	1.21×10^{-5}	7.43×10^{-4}	0.
Reference Values	1.	0.	26.10 [1]	1.23×10^{-5} [1]	7.22×10^{-4} [3,4,5,6]	0.
P = 100 bars, T = 40 °C	gas phase				aqueous phase	
TOUGH2/EOS7C	0.	1.	566.00	4.35×10^{-5}	0.	4.03×10^{-2}
Reference Values	0.	1.	631.90 [1]	5.04×10^{-5} [1]	0.	2.19×10^{-2} [2]
TOUGH2/EOS7C	0.5	0.5	155.16	1.81×10^{-5}	9.43×10^{-4}	2.00×10^{-2}
Reference Values	0.5	0.5	153.97 [1]	1.94×10^{-5} [1]	7.95×10^{-4} [3,4,5,6]	1.21×10^{-2} [3,4,5,6]
TOUGH2/EOS7C	1.	0.	71.57	1.41×10^{-5}	1.86×10^{-3}	0.
Reference Values	1.	0.	70.03 [1]	1.41×10^{-5} [1]	1.54×10^{-3} [3,4,5,6]	0.

[1]NIST, 1992. [2]Wiebe and Gaddy, 1940. [3]Spycher and Reed, 1988. [4]Johnson et al., 1992. [5]Shock et al., 1989. [6]Wagman et al., 1982.

However, CO_2 that is compressed through its critical pressure near the critical temperature is a very effective cushion gas because of its large compessibility. This can be seen in Figure 2 by the large change in density or corresponding small value of Z. Thus the use of a CO_2 cushion gas will allow more gas to be stored for a given pressure rise than is possible with a native gas or nitrogen cushion (Oldenburg, 2003). This process will be demonstrated by numerical simulation below.

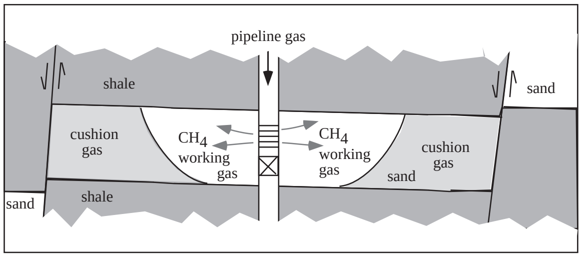

Figure 5. Schematic of gas storage showing cushion gas that is never produced.

SIMULATION METHODS

For the simulation of CSEGR and gas storage with CO_2 as a cushion gas, we are using a new module called TOUGH2/EOS7C. This module was developed by extending EOS7R (Oldenburg and Pruess, 1995; Pruess et al., 1999) by changing one of the volatile components to CO_2, and by changing the air to CH_4. Thus, EOS7C considers five mass components (water, brine, CO_2, gas tracer, CH_4) and heat. Solubility subroutines for CO_2 and CH_4 were taken from EWASG (Battistelli et al., 1997; Cramer, 1982). For the key gas mixture properties, new real gas mixture subroutines were developed to calculate density and enthalpy departure in the system H_2O-CO_2-CH_4 using the Peng-Robinson equation of state (e.g., Poling et al., 2001). Accurate gas mixture viscosities are calculated using the method of Chung et al. (1988). A summary of gas mixture properties is presented in Table 1 for verification of the gas properties module. As shown, TOUGH2/EOS7C predicts density and viscosity very well, but overpredicts gas solubility. Current efforts are underway to improve the solubility model. Future effort is needed to test and verify the non-isothermal capabilities of TOUGH2/EOS7C.

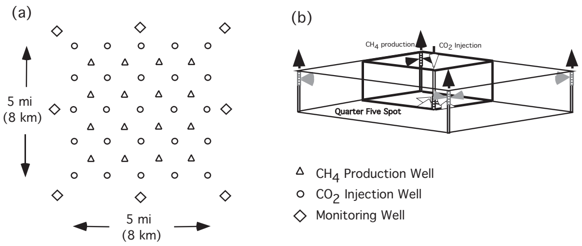

Figure 6. Schematic of model gas reservoir well pattern and quarter five-spot simulation domain.

CSEGR

We have used TOUGH2/EOS7C to carry out a number of CSEGR simulation studies over the last several years (e.g., Oldenburg et al., 2001; Oldenburg and Benson, 2002; Oldenburg et al., 2003). Here we present a representative result that demonstrates the concept of injecting CO_2 deep in the reservoir while producing from higher in the reservoir.

A schematic of the model well pattern and five-spot reservoir geometry are shown in Figure. 6, while properties of the model reservoir are presented in Table 2. The reservoir properties and injection and production rates are based loosely on the Rio Vista gas field in California (Cummings, 1999; Johnson, 1990). Someday carbon dioxide could be sent by pipeline from electricity generation and industrial sources in the San Francisco Bay Area to the Rio Vista gas field.

Table 2. Properties of the quarter five-spot domain.

Property	Value
Quarter five spot size	6.4×10^5 m^2
(21×21 gridblocks in X-, Y-dir.)	(160 acres)
Reservoir thickness	50 m (160 ft)
(10 gridblocks in Z-dir.)	
Porosity	0.30
Permeability (isotropic)	1×10^{-12} m^2
Residual liquid saturation	0.20
Relative permeability	
Liquid	Immobile.
Gas	Equal to gas saturation.
Diffusivity in gas and liquid	1.0×10^{-5} m^2 s^{-1}, 1.0×10^{-10} m^2 s^{-1}
Temperature (isothermal)	75 °C (167 °F)
Pressure at start of CSEGR	50 bars (725 psi)
CO_2 inj. rate (per full well)	3 kg s^{-1} (260 t/day)
CH_4 prod. rate (per full well)	0.56 kg s^{-1} (48 t/day)
Pressure (after 15 years)	60 bars (870 psi)

Simulation results are shown in Figure 7 for the 3-D quarter five-spot domain in which CO_2 is injected in the lower 10 m of a 50 m thick reservoir while CH_4 is produced from the top 10 m. In this configuration, the high density of CO_2 makes it remain in the lower parts of the reservoir, and CH_4 displacement is to some degree from the bottom up. As shown in Figure 7, the CO_2 breakthrough occurs after approximately 15 years, before which time high CH_4 production rates could be maintained by means of CO_2 injection and sustaining high pressure in the reservoir. Although not modeled specifically in this study, pressure maintenance would help prevent water from entering the reservoir and discourage land subsidence that is observed at some gas fields.

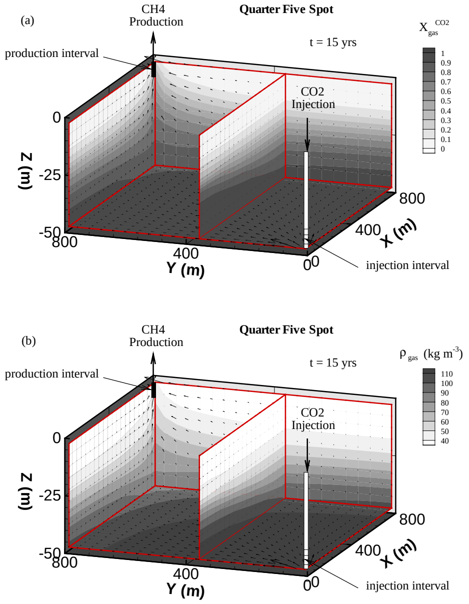

Figure 7. Simulation results of mass fraction CO_2 and gas-phase density.

Table 3. Properties of the model gas storage reservoir.

Property	Value
Area (X-, Y-direction)	4 km $\times$ 1 km (1.5 mi^2)
Thickness (Z-direction)	22 m
Gridblocks in X-, Y-dir.	1×200
Gridblocks in Z-dir.	11
Porosity	0.30
Permeability (isotropic)	1.0×10^{-12} m^2
Relative permeability	Van Genuchten (1980)
$m, S_{lr}, S_{ls}, S_{gr}$	0.2, 0.27, 1, 0.01
Liquid is immobile	$S_l \approx 0.26 < S_{lr}$
Diffusivity gas, liquid	1.0 x 10^{-6}, 1.0 x 10^{-10}
Temperature	40 °C (isothermal)
Initial pressure	60 bars
CH_4 injection rate	73.5 kg s^{-1}

SIMULATIONS OF CO_2 AS CUSHION GAS

Simulations of an idealized two-dimensional gas storage reservoir with CO_2 as a cushion gas have also been carried out (Oldenburg, 2003). The idea here is that after CSEGR is completed, the reservoir could be used for gas storage. We present in Table 3 properties of the model gas storage reservoir idealized to represent a reservoir that is an elongated dome structure with a line of wells along the axis. Note that the reservoir starts at 60 bars and will exceed the CO_2 critical pressure (73.8 bars) during the storage cycle.. Shown in Figure 8 are contours of CO_2 mass fraction in the gas at three times during the storage cycle. The working gas is injected in the upper left-hand corner of the model domain. Figure 9 shows the corresponding gas density for the same simulation. Note that the CO_2 cushion gas density increases from just below 200 kg m^{-3} to just below 600 kg m^{-3}. As shown, the CO_2 cushion drastically compresses, allowing more volume for CH_4 for a given pressure increase.

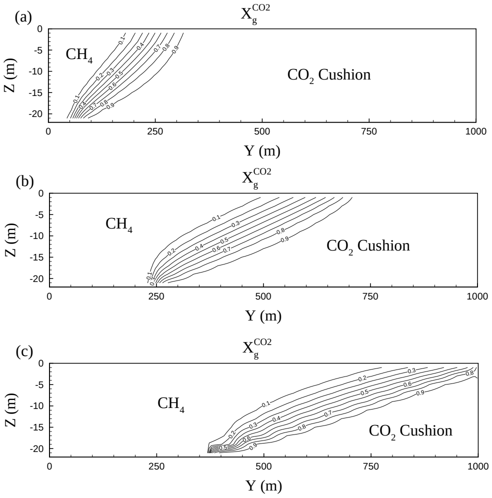

Figure 8. Two-dimensional simulation results of CO_2 mass fraction where CO_2 is the cushion gas: (a) 30 days; (b) 90 days, and (c) 180 days.

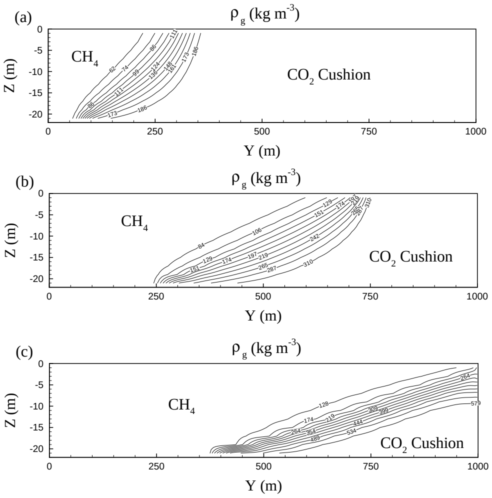

Figure 9. Two-dimensional simulation results of gas-phase density where CO_2 is the cushion gas: (a) 30 days; (b) 90 days, and (c) 180 days.

A comparison of the pressure increase during gas storage over a half a year for the case of CO_2 as a cushion gas, CH_4 as a cushion gas, and for CH_4 as cushion gas using 70% of the CH_4 injection rate, is presented in Figure 10. This figure shows that using CO_2 as a cushion gas allows approximately 30% more gas to be stored for a given pressure increase relative to using a native CH_4 cushion gas.

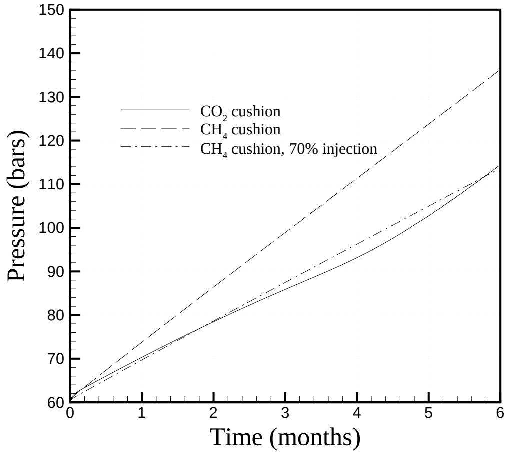

Figure 10. Pressure vs. time for one cycle of CH_4 injection showing the lower pressure rise for CO_2 cushion gas relative to a native gas cushion.

The simulations carried out to date include significant numerical dispersion, making a mixing zone that is artificially large. For a rectangular grid with full upstream weighting, the numerical dispersion is approximately one-half the grid spacing multiplied by the gas velocity. For these simulations, numerical dispersion in on the order 10^{-4} m^2 s^{-1} (5 m/2 x 700 m/6 months), approximately two orders of magnitude greater than molecular diffusion. Nevertheless, gas mixing will be a concern in actual practice since the injection and production cycle operate repeatedly over the years, unlike CSEGR which is a one-time displacement. Considerable additional research will have to be done before CO_2 can be considered a practical cushion gas. Nevertheless, the unique properties of CO_2 make it an intriguing cushion gas option. Other configurations and applications, for example, for gas storage in an open tank or solution-mined cavity with a physical barrier separating the two gases, are being investigated to avoid the mixing problem.

CONCLUSIONS

Simulation studies suggest that CSEGR and gas storage with CO_2 as a cushion gas may be technically feasible uses of depleting gas reservoirs. In both processes, the properties of CO_2 and CH_4 favor limited mixing of the gases. In particular, the large density and viscosity of CO_2 relative to CH_4 favor lack of mixing for CO_2 displacing CH_4 in CSEGR. The large effective compressibility of CO_2 for pressures around the critical pressure favors the use of CO_2 as a cushion gas. We envision a scenario where depleting gas reservoirs near CO_2 sources are first used for approximately a decade for CSEGR, and then after CO_2 breakthrough, they can be used as gas storage reservoirs. Additional simulation capability development to consider nonisothermal effects and further detailed reservoir characterization will have to be carried out to investigate these processes further. In the meantime, a field pilot experiment of CSEGR should be carried out to verify the simulation results and further test the idea.

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