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Alternating N₂ gas injection as a potential technique for enhanced gas recovery and CO₂ storage in consolidated rocks: an experimental study

Nuhu Mohammed¹ · Abubakar Jibrin Abbas¹ · Godpower C. Enyi¹ · Salihu M. Suleiman¹ · Donatus E. Edem¹ · Muhammad Kabir Abba¹

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Abstract
The promotion of enhanced gas recovery (EGR) and CO₂ storage is still shrouded in contention and is not well accepted, due to the excessive in situ CO₂ mixing with the nascent natural gas. This adulterates the recovered CH₄ and thus results in a high sweetening process cost thereby making the technique impractical. This has not only limited the field application of EGR in actual projects to a few trails but renders it uneconomical. This study aims to present, experimentally, alternating N₂ injection as a potential technique for EGR and CO₂ storage in sandstone rock cores. A laboratory core flooding experiment was carried out to simulate a detailed process of unsteady-state methane (CH₄) displacement using Bandera grey core plug. This was carried out at 40 °C, 1500 psig, and 0.4 ml/min injection rate by alternative injection of N₂ and CO₂ in succession designed to suit the application based on optimum operating conditions. The results show that both CO₂ storage capacity and CH₄ recovery improved significantly when gas alternating gas (GAG) injection was considered. The best results were observed at lower N₂ cushion volumes (1 and 2 PV). Therefore, the GAG injection method with N₂ as cushion gas can potentially increase both CO₂ storage and CH₄ recovery of the gas reservoir. This technique if employed will assert the current position and provide vital information for further researches aimed at promoting environmental sustainability and economic viability of the EGR and CO₂ sequestration processes.

Keywords Dispersion coefficient · Enhanced gas recovery · Cushion gas · Breakthrough · Concentration profile

List of symbols

| Symbol | Definition |
|--------|------------|
| yCO₂   | CO₂ mole fraction |
| yN₂    | N₂ mole fraction |
| D      | Diffusion coefficient, m²/s |
| Q      | Flow rate mil/min |
| tD     | Dimensionless time |
| xD     | Dimensionless distance |
| d      | Characteristic length scale, m |
| Kₐ     | Longitudinal dispersion, m²/s |
| L      | Core sample length, mm |
| Lₑxp   | Experimental length, m |
| µ      | Viscosity, cP |
| P      | Pressure, psig |
| T      | Temperature, K |
| u      | Interstitial velocity, m/s |
| ϕ      | Core porosity, % |
| α      | Dispersivity, m |
| τ      | Tortuosity |
| Pe     | Péclet number |
| Peₘₐ   | Medium Péclet number |
| PV     | Pore volume |
| r      | Radius of core sample, m |
| λ₉₀    | Lambda function at 90% of effluent concentration |
| λ₁₀    | Lambda function at 10% of effluent concentration |

Introduction
CO₂ emissions are generalized as a significant factor responsible for inhibiting climate change that later results to increase in environmental temperature (global warming). It was predicted that there will be a rise in world temperature and sea level from 1.9 to 3.5 °C and 18–30 cm, respectively, by the year 2100. These would be accompanied by...
many foreseen distresses (Meehl et al. 2005). The effective simultaneous enhanced gas recovery and sequestration using CO$_2$ injection are losing recognition due to high natural gas contamination, premature breakthrough, and high compression ratio, since it requires about six (6) molar volume of it to displaced one (1) molar volume of natural gas, thereby hindering its market and calorific which eventually render the process non-economically viable. In contrast, N$_2$ can be recovered mostly from the air separation unit (ASU) or as byproducts of oxygen plants. It requires less compression ratio, which is why a lower amount of it is needed to create high pressure in the CH$_4$ reservoir.

The increase in carbon credit, coupled with earlier energy demand due to population growth, has forced the exploitation of alternative sources of energy, using other fewer or zero-emission technologies (Abba et al. 2017). Natural gas is considered one of the abundant, low emission, cleanest, and affordable sources of fossil fuels (Benson et al. 2005; Al-Abri et al. 2012). Carbon dioxide (CO$_2$) underground storage for simultaneous storage and natural gas (CH$_4$) displacement is gaining attention worldwide (Ganjdanesh and Hosseini 2017; Raza et al. 2017). This underground storage can be in the form of oil and gas conventional reservoirs or deep saline aquifers (Abba et al. 2018). Conventional natural gas reservoirs have the potential to safely store anthropogenic CO$_2$, due to its proven integrity of gas storing capability (Kalra and Wu 2014). Thus, in turn, issues of CO$_2$ leakages and contamination of adjacent freshwater aquifers are minimal. This arises the need for the development of other injection techniques capable of enhancing both natural gas recovery and CO$_2$ storage (Abba et al. 2018).

Tertiary enhanced gas recovery (EGR) and storage by CO$_2$ injection are gaining recognition within the research environment due to its greenhouse gases (GHGs) reduction potential. Therefore, both nitrogen (N$_2$) and CO$_2$ can be used to increase nascent HCs yield from oil and gas reservoirs. However, CO$_2$ drawbacks are mainly excessive mixing and high compression ratio, thus hindering the overall process uneconomically viable. In contrast, N$_2$ can be recovered virtually from the atmospheric air, through air separation units. It requires less compression ratio than CO$_2$, which is why a lower amount of it was needed to create high pressure in the CH$_4$ reservoir.

The promotion of EGR is still at its infant stage due, to the excessive mixing between the injected (displacing fluid) CO$_2$ and the nascent displaced fluid (natural gas) during the flooding process (Oldenburg and Benson 2002; Shtepani 2006; Turta et al. 2007; Sim et al. 2008, 2009; Al-abri et al. 2009; Sidiq et al. 2011; Hughes et al. 2012; Honari et al. 2013, 2015, 2016; Khan et al. 2013; Zhang et al. 2014; Patel et al. 2016). This adulterates the recovered natural gas and thus reduces its heating and market value, which results in the high cost of the sweetening process to maintain its market value (Oldenburg and Benson 2002; Sim et al. 2009).

This has not only limited the EGR project to a few pilot trials (Pooladi-Darvish et al. 2008) but also made the process apparently uneconomical due to unprecedented mixing with the displaced gas, which make the phenomenon to be poorly understood (Patel et al. 2016). Thus, finding a suitable technique for reducing such in situ mixing could be valuable at first by injecting a certain amount of nitrogen gas as cushion gas before the invention of CO$_2$, which is the concept behind gas alternating gas injection.

Several authors (Xidong et al. 2019; Hughes et al. 2012; Janssen et al. 2018; Abba et al. 2018) have carried out an extensive study on how to delay CO$_2$ breakthrough time during EGR process. Among them, Abba et al. (2018) and Xidong et al. (2019) were able to achieve reasonable improvement. Abba et al., (2018) use varying connate water concentration and was able to delay CO$_2$ breakthrough by 20 min at a concentration of 10 wt.% sodium chloride (NaCl). On the other hand, Gu et al. (2019) use different mole ratios of CO$_2$/N$_2$ mixture gases in coalbed core samples. They reveal that injection of N$_2$-rich mixtures contributes to preventing the nascent early breakthrough of injected CO$_2$ and safely stored large volumes of CO$_2$ into the shale sediment over the long term. Abba et al. (2019) achieved a high percentage of total CO$_2$ injected stored at 10 wt.% salinity using solubility trapping mechanism, but with least CH$_4$ recovery resulted from the density of connate water sealing off the narrow pore spaces within the pore matrix.

To our knowledge, no established efficient method capable of improving simultaneous natural gas recovery and CO$_2$ storage has been highlighted. This necessitated the need for an in-depth study to develop novel approaches and ways to minimizing this complex phenomenon of gas mixing during gas–gas displacements since the two gases (CO$_2$ and CH$_4$) are miscible in all outcomes (Abba et al. 2018). This study aims to highlight, experimentally, the potential of using N$_2$ as cushion gas in a novel gas alternating gas (GAG) technique to reduce or minimize excessive mixing during EGR by CO$_2$ injection, thereby improving CH$_4$ recovery while subsequently storing substantial volumes of CO$_2$ in conventional natural gas reservoirs.

**Dispersion theory and equation**

The term Péclet number, Pe, is a dimensionless measure of the level of dispersion by a solute which is defined as the ratio of advective to dispersive processes (Rose 1973) as reported by (Ho and Webb 2006). The degree or level of dispersion is inversely proportional to the magnitude of the Péclet number. At low Péclet numbers, the degree of dispersion is large. It is expressed as in Eq. (1).
\[ P_e = \frac{uL}{K_i} \]  \hfill (1)

Péclet number (ratio of convection to dispersion), \( L \) is the core sample length.

In 1963, Perkins and Johnston denoted different termed to Péclet number called medium Péclet number (\( P_{em} \)). Its value generally determined and describes the dominant displacement fluid region as the dispersion process progresses shown in Eq. (2):

\[ P_{em} = \frac{ud}{D} \]  \hfill (2)

where \( P_{em} \) is medium Péclet number, \( u \) is the mean interstitial velocity (m/s), \( D \) is the diffusion coefficient (m²/s), and \( d \) is the porous medium characteristic length scale, termed as the medium-grain diameter of the sand pack, but it is poorly defined in consolidated medium (Hughes et al. 2012).

Ideally when \( P_{em} < 0.1 \) diffusion becomes dominates, while advective mixing dominates the dispersion process at higher medium Péclet number i.e. at \( P_{em} > 10 \).

Delgado, in 2001, uses the Lambda function, by plotting a graph of Lambda at different experimental times against the percent of displacing fluid in an arithmetic probability paper. The dispersion coefficient was then evaluated using Eq. (3).

\[ K_L = u \times L \left( \frac{\lambda_{90} - \lambda_{10}}{3.625} \right)^2 \]  \hfill (3)

where \( K_L \) = longitudinal dispersion coefficient (m²/s), \( u = \frac{Q}{\pi r^2 \phi} \) is the average interstitial velocity (m/s), and \( \phi \) is the porosity, \( L \) = length of porous media (m), \( \lambda_{90} \) and \( \lambda_{10} \) = are values of Lambda function at 10 and 90% effluent concentration. In this research, the lambda function techniques were used, as the fundamental equation was derived by considering inert gas (\( N_2 \)) as one of the displacing fluids.

**Diffusion theory and equation**

The diffusion coefficient (\( D \)) signifies the extent or magnitude at which a substance or fluid disperses through a unit area (m²) per unit time (s) at a given or defined unit concentration gradient. The proposed empirical model which relates the molecular diffusion, temperature, and pressure for empirical diffusion coefficient determination as revealed by (Hughes et al. 2012; Liu et al. 2015) was developed by Takahashi and Iwasaki in 1970. Also, this empirical equation has been tested by various researchers in determining the real and accurate diffusivity using Eq. (4) at conditions applicable to EGR by \( CO_2 \) injection. The diffusion coefficient of \( CO_2 \) in \( CH_4 \) was dignified at 298–348 K and pressures of 5–15 MPa in a porous bronze plug (Takahashi and Iwasaki 1970). The results were well within the range of conditions applicable to EGR (Abba et al. 2017).

\[ D_{CO_2,CH_4} = \frac{(-4.3844 \times 10^{-11}P + 8.5440 \times 10^{-11})T^{1.75}}{P} \]  \hfill (4)

where \( D_{CO_2,CH_4} \) is the molecular diffusion coefficient of \( CO_2 \) in pure \( CH_4 \) calculated in m² s⁻¹ with \( P \) in MPa and \( T \) in K. The absolute average deviation (AAD) of this correlation from the experimental data was 1.5% over the range of 298–348 K and 5–15 MPa (Abba et al. 2017, 2018). In this study, a different model was used to cater for the inclusion of nitrogen (\( N_2 \)) gas during the natural gas displacement and \( CO_2 \) sequestration. This model equation is presented in Eq. 5. A correlation formula obtained by Fuller et al. (1966) by means of computer-aided correlation of 340 experimental points is expressed as:

\[ D_{N_2,CH_4} = \frac{1.0110 \times 10^{-4}T^{1.75}\sqrt{\left(\frac{1}{\mu_{N_2}} + \frac{1}{\mu_{CH_4}}\right)}}{P[(\sum V_{N_2})^{1/3} + (\sum V_{CH_4})^{1/3}]} \]  \hfill (5)

where \( \sum V_{N_2} \) and \( \sum V_{CH_4} \) are the values derived from the summation of atomic diffusion volumes of \( N_2 \) and \( CH_4 \) molecules, respectively. These values and other simple molecules are presented in Table 1.

The equation was further simplified after inserting the values of atomic diffusion volumes and the molecular weight of nitrogen and methane. The same was applied for carbon dioxide and methane displacement mechanism. These simplified equations are presented in Eqs. (6) and (7)

\[ D_{N_2,CH_4} = \frac{10.2 \times 10^{-11}T^{1.75}}{P} \]  \hfill (6)

\[ D_{CO_2,CH_4} = \frac{8.2 \times 10^{-11}T^{1.75}}{P} \]  \hfill (7)

where \( T \) and \( P \) are temperatures and pressure in Kelvin (K) and megapascal (MPa), respectively. For example, at the same temperature and pressure, Eq. (7) was validated using the experimental work of Abba et al. 2018. The molecular diffusion coefficient (\( D_{CO_2,CH_4} \)) was found to be \( 22.52 \times 10^{-8} \) m²/s. This value is 0.18% in absolute average deviation (AAD) when compared with Abba et al. (2018) findings.

**Materials description**

In this research, an experimental study approach using the core flooding system was investigated. This entities saturating the core plug with \( CH_4 \) and the injection of \( CO_2 \) at different \( N_2 \) gas cushion volumes. The core plug use was Bandera grey sandstone as presented in Table 2.
The core plug of dimension 1.0 inch diameter and 3.0 inch length was used as present in Table 2. This sandstone is considered a classic sedimentary rock primarily comprised of quartz, silica, and sand-sized minerals. For consistency, the porosity and permeability of the sandstone core samples were determined and compared with the ones provided by the supplier (Kocurek Industries INC, Hard Rock Division, 8535 State Highway 36 S Caldwell, TX 77836, Texas USA).

The research-grade CO₂, N₂, and CH₄ with a purity greater than 99.99% were sourced from BOC UK.

### Experimental method

A sequence of experimental tests was carried out to investigate the potential of N₂ as cushion gas for enhanced CH₄ recovery and CO₂ storage during core flooding experiment with Bandera grey core plug as the porous medium. Prior to the experiment, the core plug petrophysical properties were evaluated to endure in synergy with the ones from the vendor. The flow behaviour of supercritical N₂, CO₂, and N₂–CO₂–CH₄ interplay was studied using FLUIDAT® thermodynamic software. This was vital, in understanding the flow behaviour of supercritical CO₂ as it plumes transverses the pore spaces within the porous medium while displacing the nascent CH₄, especially with the N₂ as cushion gas. Followed to that, a laboratory core flooding experiment was conducted on the Bandera grey core plug to determine the dispersion coefficient, CH₄ recovery, and percent of the CO₂ injected stored of the system at different CO₂ injections and N₂ cushion volumes. Further to that, several runs were carried out at four different N₂ cushion volumes at varying CO₂ injection rates. The effluent stream rates from the core plug were recorded by the two (low and high) gas flow meters. After this, the produced gas compositions are analysed using the Agilent technologies 7890 A model gas chromatography (GC) system at an interval of 5 min elusion time through the sampling valve. The combined data are used to evaluate: one, dispersion coefficient; two, CH₄ recovery; and three; percent of total CO₂ injected stored at the test reservoir conditions of 1500 psig pressure and 40 °C temperature.

### Apparatus and procedure

#### Apparatus

The experimental set-up consists of mainly two individual units; a core lab UFS-200 core flooding system with inbuilt Smart Flood software and packed column design Agilent 7890A gas chromatography (GC) machine model. The core flooding system, designed for 2-phase liquid/gas steady or unsteady state condition displacements, was reconfigured to accommodate additional N₂ as used in the gas alternating gas injection for this study. The integrated online concentration measurement of core flooding effluents was achieved using the GC machine. These values were used in plotting the injection fluids concentration profile as the experiment progress with time. A schematic of the equipment set-up is presented in Fig. 1.

The UFS-200 core flooding system is rated to 5000 and 3750 psig overburden and pore pressure, respectively. The
injection system of the equipment is made up of a pair of dual ISCO two-barrel metering pump system (A/B and C/D) for constant flow for pulseless transition and to maintain an accurate flow rate range of 0 to 200 ml/min with a maximum pressure rating of 3750 psig. The pumps are attached to a pair of two stainless-steel floating piston accumulators which are also rated for 5000 psig working pressure and temperature of 177 °C. They are designed for injection of the fluids of interest and can withstand up to 7500 psig test pressure. The hydraulic pump with a maximum of 10,000 psig pressure output was used to set the overburden confining pressure. The Smart Flood 1.0 software forms an essential unit of the system which interfaces the UFS system and the computer data acquisition-control (DAC) system hardware and generates on-screen automatic logging of test data for all measured values like pressures, temperatures, volumes, etc., to a computer data file. A Rosemount Static DP transmitter with an accuracy of 0.0055% was responsible for measuring the differential pressures across the entire Hassler-type core holder, which was used to house the core sample. The core sample is clutch inside the core holder by a Viton rubber sleeve. A core holder heat jacket to simulate the required temperature was also employed with an accuracy of 0.1%. Dome type back pressure regulator integrated into the flooding system ensured the confinement of the desired pressures within the core holder. Such desired pressure was set using the N₂ cylinder bottle. The effluents from the back-pressure regulator pass through the mass flow controllers that measure the volume of the actual effluents produced before been analysed by the GC system in place.

**Procedure**

The core sample was dried overnight in an oven at 105 °C for moisture removal and other volatile compounds. The dried sample was wrapped with cling film and in foil paper before inserted into a heat shrink. This is vital to avoid viscous fingering and the penetration of the gases through the sleeve into the ring-shaped core holder. It was then loaded into the core holder and staple with clamps from both ends. Hydraulic oil was then pumped into the ring-shaped core holder to provide the desired overburden pressure, which was kept 500 psig above the pore pressures to in order to avoid fracturing of the core sleeve. The heat jacket was then installed on the core holder and the temperature step-up was observed. The backpressure was engaged and CH₄ was slowly injected into the core sample from the CH₄ cylinder to saturate the core plug until the GC constantly read methane > 99%. N₂ is then injected as cushion gas for about 20 min (1 PV) before the invention of CO₂ at 0.4 ml/min injection rate. Further runs were carried out at increasing N₂ cushion volumes. The experiment elapsed when the methane concentration was insignificant from the GC reading or the CO₂ concentration was > 99%. At each injection time of the GC, the time was noted and the effluent composition is then used to evaluate the dispersion coefficient, CH₄ recovery,
and percent of total CO₂ injected stored was recorded. The investigation was carried out at 1500 psig pressure and 40 °C temperature. This condition was chosen based on a normal gas pressure reservoir with a gradient of 0.451 psi/ft, an average reservoir depth of 1 km, and a geothermal temperature of 35–40 °C/km. The core sample was dried overnight in an oven at 105 °C for moisture removal and other volatile compounds. The dried sample was wrapped with cling film and in foil paper before inserted into a heat shrink.

Results and discussion

Flow behaviour of supercritical N₂, CO₂, and N₂–CO₂–CH₄ during simulated EGR

The actual flow behaviour of supercritical CO₂ as it plumes traverses the pore spaces within the core sample to displace the in situ methane is quite complex, especially with the inclusion of inert nitrogen gas. Investigating these complexities of displacing fluids (CO₂ and N₂) with regard to the nascent CH₄ is vital in understanding the trends and expected outcomes of the displacement process knowing that these gases in their supercritical state have unique behaviour by exhibiting the density of a liquid but retain the viscosity of a gas (Abba et al. 2018). CO₂ is 2–6 times denser than methane at all relevant reservoir conditions. Besides, CO₂ had a lower mobility ratio compared to methane; thus, it was considered as a high viscosity component (Al-Hasami et al. 2005). Due to the favourability of these properties, CO₂ would be migrated downwards, and this relatively would stabilize the displacement process between the injected CO₂ and methane initially in place (Oldenburg and Benson 2002). The supercritical conditions of CO₂, N₂, and CH₄ are (31.05 °C and 73.80 bar), (~146.9 °C and 33.90 bar), and (~82.55 °C and 46 bar), respectively. The experimental conditions employed in this study are well above that of the supercritical temperature and pressure of each single species. The fluids exhibit excellent behaviour due to the response on their supercritical temperature and pressure of each single species. A simulation of their respective properties at elevated operational conditions was carried out using FLUIDATR software to check the effects of temperature and pressure on the individual gas densities and viscosities at the stated conditions as presented in Fig. 2, 3, and 4. There are pronounced differences in their properties, with CO₂ being much higher and more extreme than those of N₂ and CH₄. The density was said to increase as the gas pressure increases. This was more significant with CO₂ especially at 500–1400 psig range, after which become constant as shown in Fig. 2. This makes CO₂ approximately six (6) times denser than N₂ or CH₄, which signifies the possibility of storing more of it at a supercritical state. However, N₂ and CH₄ exhibited similar behavioural trends as the pore pressure raises, justifying why the recovered CH₄ onsite mostly contain higher traces of N₂ than CO₂ during exploration (Xidong et al. 2019). The kinetic energy of a gas is proportional to its temperature due to the increased rate of collision with the container wall. The reverse was the case to that of liquid fluids due to high inter-molecular forces keeping them close to each other. In general, gas viscosity increases with pressure raise. However, at higher pressures (1300–2000) psig CO₂ maintained high viscosity with a density like that of liquid as observed in Figs. 2 and 3. Also, at
a lower temperature (18–50 °C) CO₂ experienced viscosity reduction similar to liquid as evidence of an about 66% reduction in viscosity at 50 °C, shown in Fig. 4. Thus, in turn, based on the proceeding statement, CO₂ demonstrated strange properties behaviour compared to those of N₂ and CH₄ at conditions of temperature and pressure (40 °C and 1500 psig) applicable to EGR. This justifies the selection of the experimental conditions as also, reported by Abba et al. 2018. Thus, it makes the application of supercritical CO₂ for the EGR process to be well accepted globally. Such unique esteem property will grossly affect the flow behaviour of the gases as will be presented in the successive sections.
The viscosity is said to be increasing at the beginning of the displacement process until the mobility of the displacing phase is less than that of the displaced phase; this makes the mobility ratio to be less than one (unity). Such a state does maximize gas recovery and sweeps efficiency, due to negligible premature mixing, by providing a sharp viscosity gradient at the displacement front without experiencing viscous fingering effect. A condition in which a combination of two fluids escape some part of the reservoir as it progresses along, forming irregular, or fingered profile. Fingering is relatively a routine problem in reservoirs with gas-injection wells (Al-Abri et al. 2012). Fingering effects result mostly in an unproductive sweeping action, which bypasses many recoverable gas volumes, with a premature breakthrough of displacing fluids. In Fig. 5, both the N₂ injection (CH₄–N₂) and CO₂ injection (CH₄–CO₂) display a lower mobility ratio at their respective critical conditions. The mobility ratio of CH₄–N₂ displacement was low compared to CH₄–CO₂ at pressure (100–800 psig) below CO₂ critical points (33.9 °C and 1070 psig). This means the effect of viscous fingering was minimal as the N₂ plume transverse through the core spaces as against that of CO₂. The effect of pressure on the CH₄–CO₂ mobility ratio was insignificance above its supercritical state and remains almost constant thereafter. Overall, the supercritical state of gases plays an important role in investigating their flow behaviour with maximum recovery efficiency, creating an even flood front with minimum risk to viscous fingering effect.

On the other hand, the mobility of CH₄–CO₂ displacement was ascending as the temperature increase due to an increase in density with temperature and pressure down the reservoir. However, a reverse scenario was observed for that of the CH₄–N₂ process as the temperature increases to 100 °C as observed in Fig. 6. This is so because as the temperature increases CH₄–N₂ system experienced a high rise in diffusion coefficient compared to those of CH₄–CO₂ and CO₂–N₂ as shown later in Fig. 7. Thus, in turn, CH₄–CO₂ system mobility is more sensitive to change in temperature and pressure compared to that of CH₄–N₂ during EGR and storage.

The developed Eqs. (6), (7) derived from the fundamental (first principle) correlation [Eq. (5)] as reported by Fuller et al. (1966) were used to simulate the effect of pressure and temperature for N₂–CH₄, CO₂–CH₄, and CO₂–N₂ interaction behaviours. This simulation was carried out at constant temperatures of 30, 40, and 50 °C, respectively, at a varying pressure of 100–2000 psig for the stated interactions. In Fig. 7, the molecular diffusion coefficient increases with a roughly constant value of 1.5 m²/s at constant temperatures of 30, 40, and 50 °C over the pressure ranges. These values were more pronounced for CH₄–N₂ interaction than that of CH₄–CO₂ due to the high density and molecular weight of carbon dioxide compared to that of nitrogen at reservoir condition. Meanwhile, above 1500 psig of pressure, the diffusion coefficient trend was the same for all the temperatures and the decline rate was less notable.

**Fig. 5** CH₄–N₂ and CH₄–CO₂ mobility ratios as a function of pressure at 40 °C
Temperature plays a significant role in determining the extent of diffusion when two fluids are in contact with each other especially of the same phase. The higher the temperature, the more the kinetic energy due to high collision velocity, which invariably results in the high molecular diffusion coefficient. Such evidence can be seen in Eqs. (6), (7) with the temperature component (in Kelvin) being the rise to the order of 1.75. Viscosities and atomic diffusion also contribute to measuring the overall diffusion coefficient of binary mixtures. Further to that, atoms or molecules with higher viscosities and diffusion volumes do result in lower molecular diffusion coefficient value compared to those with lower viscosities and diffusion volumes, respectively. Figure 8 presents a plot of diffusion coefficient against temperature. The CH$_4$–N$_2$ binary mixture shows a high periodic increase as the temperature rise to 120 °C (393.15 K). However, a lower rise was experienced for the case of the CH$_4$–CO$_2$ mixture. This was due to high density, viscosity, and diffusion volume of CO$_2$ at the supercritical condition as against that of N$_2$. Also, a similar trend was observed for that of the CO$_2$–N$_2$ binary mixture.
Core flooding experiment

The recovery efficiency and percent of total CO₂ injected stored were analysed by a laboratory displacement system of an experiment to evaluate the concentration profiles of the interacting gases. This involved the injection of N₂ and CO₂ into the Bandera grey core plug at a condition reasonable for the EGR process.

The concept of N₂ as a cushion gas during enhanced gas recovery

The principle behind the whole concept of the core flooding experiment for CH₄ displacement in a porous medium is well understood using the concept of dispersion theory and its governing equations. For example, establishing the dominant mechanism of displacement as the displacing gases (N₂ and CO₂) transverses the core samples are prerequisites to avoid excessive mixing during the EGR process. If the medium Péclet number in Eq. (2) is less than 0.1, diffusion dominates and the flow is driven by the concentration gradient, and the transport is influenced by the mobility ratio, as evident in most of the experimental EGR process. However, when the Péclet number is above 10 advective mixing dominates due to the turbulence and eddy current effects and the flow is driven by velocity gradient as seen in most displacement at higher injection velocity. In this experiment, the Péclet number was 0.02, meaning the dominant displacement is by diffusion. The rate of gas miscibility during the natural gas displacement process mostly depends on the injection rate of the displacing species. The higher the rate of mixing will be recorded, and invariably more contaminated CH₄ will be recorded. This was why most researches on CO₂ injections were tailored toward storage rather than CH₄ recovery.

The injected amount of N₂ prior to the CO₂ injection acts as a barrier by creating a thin film layer between CO₂ and CH₄, making it difficult for the carbon dioxide to penetrate and disperse into the methane due to the blanketing nature of nitrogen. This affirmed the use of N₂ for reservoir pressure maintenance for decades. The introduction of N₂ displaces a larger amount of the CH₄ until it reached its breakthrough; this allows most of the CO₂ later injected to be trapped within the rock space without mixing with the nascent CH₄. More so, at the time the CO₂ reaches its breakthrough a substantial volume of CH₄ has been recovered already since the CO₂ will find it difficult to disperse itself into the methane due to the presence of nitrogen gas which acted like a barricaded wall between the CO₂ and CH₄. A decline in the dispersion coefficient was observed as the cushion gas volume increases; thus, less gas miscibility was noticed with higher CO₂ storage compared to the conventional CO₂ flooding. This signifies the feasibility and potential N₂ as a cushioning medium on CH₄ swept recovery efficiency and CO₂ storage for both social and economic benefits.

The variation of effluents against the total injected gases in pore volumes

The breakthrough indicates the first contact point at which the injected gas species (CO₂ and N₂) trespass the length scale of the core sample during the laboratory experimental runs. The later the breakthrough the more the sweep efficiency and invariably the volumes of CO₂ stored. On the other hand, the shorter the breakthrough, the larger the dispersion coefficient; an indication for excessive mixing and product contamination. This results in natural gas production with low calorific value and high purification cost, rendering the process uneconomical. The breakthrough
points were determined from the plots of the concentration profiles against total gas injection as presented in Fig. 9. The effluents mole fraction was measured online using the integrated GC machine as seen from the experimental setup. Five sets of breakthrough points were measured at 1, 2, 3, and 5 PV of injected gas species. It was observed that there was a delay in the CO₂ breakthrough as the cushion gas volume increases from 1 to 5 PV. This was so because injecting N₂ into the gas-bearing formation can cause a large volume of nascent methane displacement from the reservoir by lowering the partial pressure of CH₄ due to its high conductivity and invariably increase its recovery (Jessen et al. 2008). This accounted for approximately 150% delayed in breakthrough compared to conventional CO₂ injection as evidence in Fig. 9.

The changes in effluent concentration with displacement were ascertained by conventional and gas alternating gas (GAG) displacement simulation (Figs. 10, 11). The N₂ breakthrough occurred when about 6 PV of it been injected into the core sample. This value was roughly 2 times that of the conventional CO₂ injection. The injected pore volume of the CO₂ was earlier detected by the gas chromatography at the same injection temperature and pressure, due to its high diffusion volume. The diffusion volumes of CO₂ and N₂ are 26.9 and 18.5 cm³, respectively (Fuller et al. 1966). In comparison with conventional N₂ displacement, the breakthrough time of N₂ increases when the cushion gas was employed. The increase was highest at 2 PV cushion gas volume. As expected, more product contamination of N₂ was recorded as the volume of N₂ injected into the system increases. The least contamination was noticed at lower cushion volume with 19% nitrogen contaminants compared to 75% contamination at 5 PV of injected cushion gas. Due to the high cost of natural gas purification, designing an experiment with high product purity is paramount for the economic viability of such a process. Thus, the level of product contamination will be considered in selecting the best and optimum cushion gas volume. Similarly, a prolonged CO₂ breakthrough time was recorded at the highest cushion gas volume (5 PV), this was 5.8 PV more than conventional CO₂ flooding. This also resulted in a higher volume of total injected CO₂ stored due to lower penetration and dispersion coefficient as later present in Table 4. The combined effect for all the runs is presented in Fig. 12. Thus, in turn, the breakthrough of CO₂ can be delayed by increasing N₂ cushion gas volume. Overall, a minimum of 3.2 PV delayed longer than the traditional CO₂ injection was recorded across all the cushion gas volume tested. It evidence that the presence of impurity (N₂) causes large changes in supercritical CO₂ behaviour as reported by several authors (Xidong et al. 2019; Hughes et al. 2012; Janssen et al. 2018; Abba et al. 2018). To reduce the high cost of gas separation, a longer breakthrough with minimum miscibility is preparable for experimental gas injection (Xiangchen et al. 2018). It is worth noting that higher displacement efficiency is obtained at lower cushion volume. The optimization of breakthrough time and displacement efficiency should be expected for the success of the GAG injection process.

Fig. 9 CO₂ breakthrough time of all the experimental runs
Dispersion coefficient and dispersivity

Dispersion can be defined as an irreversible mixing that exists during miscible displacements reported by Adepoju et al. (2013). This phenomenon occurs mainly due to molecular diffusion and mechanical dispersion as the experiment progress, while the displacing fluids CO₂ and N₂ tend to mix with the displaced CH₄ as shown in the concentration profile plots. The displacement efficiency in miscible flooding is grossly affected by in situ mixing taking place inside the core spaces of the core plug. Also, a transition or mixing zone called ‘displacement front’ develops when the concentration of methane decreases from > 99% to < 1% as aforementioned in Sect. 4. The dispersion observed reported in Table 5 was used to quantify the nascent mixing as the CO₂ plumes transverses through the porous media. These values were evaluated using empirical evaluation [Eq. (3)] and laboratory experiments by analysing the concentration of CO₂ relative to that of CH₄ in the produced effluent stream with the aid of the GC machine. Several repeated experimental displacement tests were carried out to check for uncertainty and repeatability of the research method and set-up. However, such results were demonstrated in our previous publication. The medium Péclet number (Peₘ) was determined using Eq. (2), in that the characteristic length scale of mixing, d (µm), was obtained from the work of Abba et al. (2019). This value was found to be 57.15 µm for Bandera grey, such value was used in Eq. (2) to determine the dominant phenomenon of displacement mechanism. The Peₘ value was calculated to be 0.02, meaning diffusion is the dominant mechanism since its value is < 0.1 as stated in Sect. 2.1. Also, the dispersivity (α), an empirical property of a porous medium, is responsible for characteristic dispersion of the medium by comparing the components of pore velocity to that of dispersion coefficient. This value was 0.0007 m as reported in our previous work. Thus, in turn, both the medium Péclet number and dispersivity were well within the range obtained by (Abba et al. 2018; Hughes et al. 2012) for consolidated rocks.

Methane recovery

The methane produced was determined based on the total volume and composition of core flooding system effluents recorded by the gas flow meters and gas chromatography. The core flooding experiment was terminated when an insignificance composition (< 0.5%) of CH₄ or (> 99.5%) of CO₂ was noticed from the GC as shown in Fig. 13 with only CO₂ peak noticeable. These volumes are a fraction of the original gas in place (OGIP) in the Bandera grey core plug. The result is presented in Fig. 14. As can be observed, the worst CH₄ recovery was realized when pure CO₂ was injected; this was due to high diffusion volume and low minimum miscibility pressure (MMP) effect as it plumes transverses through the core plug during the displacement process. Considering the GAG injection, the run with 5 PV of cushion gas produced 44.39% CH₄ recovery. This poor sweep efficiency was a result of early N₂ detection by the GC due to the high volume of it injected and demonstrate similar property behaviour with CH₄ as discussed in Sect. 4.1.

Prior to the core flooding experiment, the OGIP of the Bandera grey core plug was evaluated using the well-known

![Fig. 10 Conventional N₂ and CO₂ injection effluent concentration profiles](image-url)
gas reservoir equation [Eq. (8)] as reported by (Abba et al. 2017, 2018).

\[
OGIP = \frac{P_v(1 - s_w)}{B_g}
\]  

(8)

where \( G \) is the original gas in place in \( \text{scm}^3 \), \( P_v \) is the pore volume in \( \text{cm}^3 \), \( s_w \) is the initial water of saturation fraction (\( s_w = 0 \) for a dry run), and \( B_g \) is gas formation volume factor in \( \text{cm}^3/\text{scm}^3 \) for this research. This was then used to determine the percentage of \( \text{CH}_4 \) recovery as shown in Fig. 14 and Table 3.

Furthermore, the \( \text{CH}_4 \) recovery was highest for conventional \( \text{N}_2 \) and 1 PV cushion gas experimental runs. However, not only the 1 PV cushion gas gives high recovery, but also the \( \text{CH}_4 \) recovered happens to experience the least impurity with 19% \( \text{N}_2 \) contamination. This signifies the potential application of \( \text{N}_2 \) gas during the enhanced gas recovery process.

**Carbon dioxide injection and storage**

In this study, the amount of \( \text{CO}_2 \) stored during the gas alternating gas injections was evaluated and recorded using Eq. (9) as reported by Xidong et al. (2019).

Fig. 11  Effluents concentration profiles with 1, 2, 3, and 5 PV cushion gas
where $V_{\text{injected CO}_2}$ is the volume of injected CO$_2$ recorded by the high-pressure syringe pump at time $t$ and $C_{t, CO_2}$ is the CO$_2$ mole percent in the effluent at time $t$ recorded by the gas chromatography (GC) analyser.

$$V_{\text{storage CO}_2} = V_{\text{injected CO}_2} - F_{\text{outlet}} \int_0^t C_{t, CO_2} \, dt \quad (9)$$

The exit effluents from the core holder were measured using the gas mass flow controller, while the displacing gases were introduced to the system via the ISCO pumps A/B and C/D through cells A and B as presented in Fig. 1. Both pumps were set at a constant flow rate of 0.4 ml/min, and the total injection time was recorded from the online core flooding logging data. For example, to inject 8 cm$^3$ (1 PV) of the cushion gas (N$_2$), the ISCO pump A/B was run for 20 min at the set flow rate, i.e. 0.4 cm$^3$/min × 20 min = 8 cm$^3$ or 1 PV. Thus, to measure the amount of the rest cushion volumes, the injection period was increased to 40, 60, and 90 min. To evaluate the total CO$_2$ injected, the total injection time at the end of each experimental test was obtained and the value was multiplied with the known injection rate after taken out the N$_2$ injection time before introducing the CO$_2$ into the Bandera grey core sample. For instant, at 1 PV cushion gas, the total injection time was 186.15 as shown in Table 5 in the ‘Appendix’ section. This means the actual CO$_2$ injection period is 166.15 min (186.15–20 min). Thus, the total volume of CO$_2$ injected was 66.46 cm$^3$ (0.4 cm$^3$/min × 166.15 min). This value was the same as 8.8 PV of total CO$_2$ injected. The same procedure is applied to the other experimental runs as shown in Table 4, more so, to evaluate the amount of CH$_4$ present in the core plug after saturation and before displacing gases injection. The OGIP technique in Eq. (8) was used since the pore volume of the Bandera grey core sample is known. The total volume of

![Fig. 12](image1.png) Concentration profiles of all the runs with cushion gas

![Fig. 13](image2.png) CO$_2$ and CH$_4$ composition recorded by the GC at the termination stage of the core flooding experiment
CH₄ in the core sample was measured to be 115 PV, and is presented in Table 2. The total CO₂ injected stored is presented in Table 4. From Table 4, it is adequate to know that the most significant amount of total CO₂ injected stored of 59.76% was recorded at 2 PV of cushion gas. This was characterized by the large pressure drop (dP) as shown in Fig. 15. The least storage was seen when conventional CO₂ injection was applied. This could be due to the high dispersion coefficient (5.02 × 10⁻⁸ m²/s) obtained during the pure CO₂ injection scenario since this parameter is a key for the economy of the enhanced CH₄ recovery projects (Du et al., 2019). Meanwhile, lower dispersion coefficient weakens the instantaneous mixing effect of CO₂ and CH₄ that inhibits the rapid breakthrough of the injected CO₂. As such, the injection of N₂ as cushion gas can effectively delay or prolonged the breakthrough point, and invariably and sequester more CO₂ over the long run (Xidong et al. 2019).

In consequence, to examine the displacement efficiency in terms of CH₄ recovery and CO₂ sequestration, Tables 3 and 4 were combined to generate Table 5. Looking at the later Table, there was an improvement in both recovery and storage when gas alternating gas injection technique was employed as against that of conventional CO₂ injection. This was so because, as the displacement process proceeds, N₂ does act as a barrier by creating a thin film layer between CO₂ and CH₄, making it difficult for carbon dioxide to penetrate and disperse into the methane due to the blanketing nature of nitrogen. This can be seen, where the least dispersion coefficient of 2.59 × 10⁻⁸ m²/s was recorded at the highest cushion gas volume of 5 PV.

Based on the literature consulted, it is quite difficult to achieve simultaneous CH₄ recovery and CO₂ storage using...
conventional injection. For example, Abba et al., (2019) in their work reported high CO$_2$ storage of 63.13% at 10 wt.% connate water. The same run yielded the poorest CH$_4$ recovery factor of 16.44% (combined of 79.57%). Their reason was due to the restrictive flow when CO$_2$ traverses the core sample as a result of higher salinity of the connate water sealing off the narrower pore spaces within the pore matrix due to its density. This means one must be compromised at the expense of the other. However, in this study both were improved compared to conventional single injection. A combination of 124.57% (59.76% CO$_2$ storage and 64.81% CH$_4$ recovery) was recorded at 2 PV of cushion gas. The same run gives the second least dispersion coefficient with a prolonged breakthrough point. With the results from the CH$_4$ recovery efficiency, CO$_2$ storage, and dispersion coefficient, it is apparent that the best and optimum cushion gas volumes for this study occur at 2 PV.

![Differential pressure (dP) changes during the experimental runs with cushion gas compared to conventional injections](image)

### Table 5 Efficiency of core flooding process in terms of CH$_4$ recovery and CO$_2$ sequestration for all runs

| S/N | Case study                   | Total CO$_2$ injected stored (% | CH$_4$ recovery (%) | Dispersion Coefficient (10$^{-8}$ m$^2$/s) |
|-----|------------------------------|---------------------------------|---------------------|------------------------------------------|
| 1   | Conventional N$_2$ Injection | N/A                             | 90.30               | 4.40                                     |
| 2   | Conventional CO$_2$ Injection| 23.55                           | 44.40               | 5.02                                     |
| 3   | 1 PV cushion gas             | 43.30                           | 89.17               | 3.59                                     |
| 4   | 2 PV cushion gas             | 59.76                           | 64.81               | 2.78                                     |
| 5   | 3 PV cushion gas             | 49.17                           | 75.95               | 3.27                                     |
| 6   | 5 PV cushion gas             | 56.41                           | 44.39               | 2.59                                     |

### Conclusion

In this study, the Bandera grey sandstone core plug was used as the standard porous media. An empirical and experimental core flooding runs were carried out to investigate the production of methane and carbon dioxide during EGR displacement scenarios in the presence of N$_2$ as cushion gas, to register the effects of its existence. A significant recovery and storage of CH$_4$ and CO$_2$ were recorded and analysed where the cushion gas volume was 2 PV; this was attributed to the inhibitory flow of the injected CO$_2$ to disperse itself into the CH$_4$ and was characterized by low dispersion coefficient. The worst result was obtained at the conventional CO$_2$ injection scenario. This was due to high CO$_2$ diffusion volume and low conductivity of CO$_2$ as it plumes traverses through the core plug during the displacement runs. Methane recovery and carbon dioxide storage can both be influenced by the addition of N$_2$ as cushion gas prior to CO$_2$ injection into the reservoir. Thus, in turn, the displacement efficiency of the current research exhibits better results than that of conventional CO$_2$ injection. However, the presence
of N₂ tends to increase CH₄ recovery by reducing the partial pressure of methane, and at the same time act as a barrier between CO₂ and CH₄ creating a thin barrier making it difficult for early CO₂ breakthroughs as a result of its blanketing effect. This work shows that N₂ as cushion gas influences both CH₄ recovery and CO₂ sequestration. Further work will entail an examination of the effect of connate water salinity on this novel method. Also, the phase behaviour of mixed gases post breakthrough would be investigated.

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**Appendix**

See Tables 6, 7, 8 and 9.

**Table 6** Effluent concentrations at 1 PV of cushion gas volume

| PVI  | CO₂ Conc. (%) | yCO₂ | N₂ Conc. (%) | yN₂ | CH₄ Conc. (%) | yCH₄ |
|------|---------------|------|--------------|-----|--------------|------|
| 0.01 | 0.421         | 0.0042| 0.037        | 0.0004| 99.539       | 0.9954|
| 0.32 | 0.449         | 0.0045| 0.04         | 0.0004| 99.514       | 0.9951|
| 0.61 | 0.465         | 0.0047| 0.042        | 0.0004| 99.493       | 0.9949|
| 0.92 | 0.482         | 0.0048| 0.042        | 0.0004| 99.476       | 0.9948|
| 1.20 | 0.485         | 0.0049| 0.044        | 0.0004| 99.471       | 0.9947|
| 1.50 | 0.501         | 0.005 | 0.044        | 0.0004| 99.455       | 0.9946|
| 1.81 | 0.501         | 0.005 | 0.044        | 0.0004| 99.455       | 0.9946|
| 2.11 | 0.510         | 0.005 | 0.045        | 0.0005| 99.445       | 0.9945|
| 2.40 | 0.560         | 0.0056| 0.045        | 0.0005| 99.395       | 0.9940|
| 2.68 | 0.587         | 0.0059| 0.046        | 0.0005| 99.367       | 0.9937|
| 2.97 | 0.614         | 0.0061| 0.046        | 0.0005| 99.340       | 0.9934|
| 3.27 | 0.615         | 0.0062| 0.046        | 0.0005| 99.339       | 0.9934|
| 3.57 | 0.674         | 0.0067| 0.046        | 0.0005| 99.280       | 0.9928|
| 3.87 | 0.71          | 0.0071| 0.046        | 0.0005| 99.244       | 0.9924|
| 4.16 | 0.739         | 0.0074| 0.046        | 0.0005| 99.215       | 0.9922|
| 4.46 | 0.764         | 0.0076| 0.047        | 0.0005| 99.189       | 0.9919|
| 4.76 | 0.79          | 0.0079| 0.047        | 0.0005| 99.163       | 0.9916|
| 5.08 | 0.819         | 0.0082| 0.047        | 0.0005| 99.134       | 0.9913|
| 5.37 | 0.841         | 0.0084| 0.050        | 0.0005| 99.109       | 0.9911|
| 5.67 | 0.856         | 0.0086| 0.050        | 0.0005| 99.094       | 0.9909|
| 5.96 | 0.856         | 0.0086| 0.051        | 0.0005| 99.093       | 0.9909|
| 6.26 | 0.857         | 0.0086| 0.249        | 0.0025| 98.894       | 0.9889|
| 6.55 | 0.876         | 0.0088| 1.908        | 0.0191| 97.216       | 0.9722|
| 6.85 | 1.166         | 0.0117| 7.961        | 0.0796| 90.873       | 0.9087|
| 7.14 | 7.107         | 0.0711| 16.814       | 0.1681| 76.079       | 0.7608|
| 7.44 | 27.371        | 0.2737| 19.025       | 0.1903| 53.604       | 0.536 |
| 7.75 | 56.115        | 0.5612| 13.234       | 0.1323| 30.651       | 0.3065|
| 8.04 | 75.681        | 0.7568| 7.141        | 0.0714| 17.178       | 0.1718|
| 8.34 | 87.422        | 0.8742| 3.143        | 0.0314| 9.435        | 0.0944|
| 8.64 | 94.339        | 0.9434| 0.950        | 0.0095| 4.711        | 0.0471|
| 8.94 | 96.827        | 0.9683| 0.408        | 0.0041| 2.765        | 0.0277|
| 9.24 | 98.946        | 0.9895| 0.105        | 0.0011| 0.949        | 0.0095|
| 9.58 | 99.226        | 0.9923| 0.069        | 0.0007| 0.484        | 0.0048|
| 9.89 | 99.317        | 0.9932| 0.060        | 0.0006| 0.404        | 0.0040|

PVI is the total amount of injected gas in pore volumes.
| PVI  | CO₂ Conc. (%) | yCO₂ | N₂ Conc. (%) | yN₂ | CH₄ Conc. (%) | yCH₄ |
|------|---------------|------|--------------|-----|---------------|------|
| 0.01 | 0.018         | 0.0002 | 0.027       | 0.0003 | 99.955       | 0.9996 |
| 0.32 | 0.018         | 0.0002 | 0.033       | 0.0003 | 99.949       | 0.9995 |
| 0.64 | 0.025         | 0.0003 | 0.039       | 0.0004 | 99.936       | 0.9994 |
| 0.94 | 0.030         | 0.0003 | 0.042       | 0.0004 | 99.928       | 0.9993 |
| 1.22 | 0.034         | 0.0003 | 0.046       | 0.0005 | 99.920       | 0.9992 |
| 1.52 | 0.038         | 0.0004 | 0.049       | 0.0005 | 99.913       | 0.9991 |
| 1.81 | 0.040         | 0.0004 | 0.051       | 0.0005 | 99.909       | 0.9991 |
| 2.11 | 0.042         | 0.0004 | 0.053       | 0.0005 | 99.905       | 0.9991 |
| 2.41 | 0.044         | 0.0004 | 0.054       | 0.0005 | 99.902       | 0.9990 |
| 2.70 | 0.046         | 0.0005 | 0.055       | 0.0006 | 99.899       | 0.9990 |
| 2.98 | 0.048         | 0.0005 | 0.057       | 0.0006 | 99.895       | 0.9990 |
| 3.27 | 0.049         | 0.0005 | 0.056       | 0.0006 | 99.895       | 0.9990 |
| 3.57 | 0.051         | 0.0005 | 0.057       | 0.0006 | 99.892       | 0.9989 |
| 3.85 | 0.052         | 0.0005 | 0.057       | 0.0006 | 99.891       | 0.9989 |
| 4.13 | 0.053         | 0.0005 | 0.058       | 0.0006 | 99.889       | 0.9989 |
| 4.43 | 0.056         | 0.0006 | 0.059       | 0.0006 | 99.885       | 0.9989 |
| 4.71 | 0.056         | 0.0006 | 0.059       | 0.0006 | 99.885       | 0.9989 |
| 5.00 | 0.057         | 0.0006 | 0.060       | 0.0006 | 99.883       | 0.9988 |
| 5.28 | 0.057         | 0.0006 | 0.061       | 0.0006 | 99.882       | 0.9988 |
| 5.58 | 0.058         | 0.0006 | 0.061       | 0.0006 | 99.881       | 0.9988 |
| 5.86 | 0.059         | 0.0006 | 0.062       | 0.0006 | 99.879       | 0.9988 |
| 6.15 | 0.060         | 0.0006 | 0.062       | 0.0006 | 99.878       | 0.9988 |
| 6.46 | 0.061         | 0.0006 | 0.063       | 0.0006 | 99.876       | 0.9988 |
| 6.75 | 0.063         | 0.0006 | 0.065       | 0.0007 | 99.872       | 0.9987 |
| 7.05 | 0.064         | 0.0006 | 0.066       | 0.0007 | 99.870       | 0.9987 |
| 7.33 | 0.067         | 0.0007 | 0.067       | 0.0007 | 99.866       | 0.9987 |
| 7.62 | 0.067         | 0.0007 | 0.080       | 0.0008 | 99.925       | 0.9993 |
| 7.91 | 0.069         | 0.0007 | 0.091       | 0.0009 | 99.013       | 0.9901 |
| 8.21 | 0.070         | 0.0007 | 0.7307      | 0.0731 | 92.623       | 0.9262 |
| 8.49 | 2.1450        | 0.0215 | 21.087      | 0.2109 | 76.768       | 0.7677 |
| 8.77 | 26.814        | 0.2681 | 22.894      | 0.2289 | 50.292       | 0.5029 |
| 9.07 | 72.473        | 0.7247 | 9.315       | 0.0932 | 18.212       | 0.1821 |
| 9.35 | 90.375        | 0.9038 | 2.685       | 0.0269 | 6.940        | 0.0694 |
| 9.64 | 96.228        | 0.9623 | 0.814       | 0.0081 | 2.958        | 0.0296 |
| 9.92 | 98.660        | 0.9866 | 0.243       | 0.0024 | 1.097        | 0.0110 |
| 10.20 | 99.152       | 0.9915 | 0.152       | 0.0015 | 0.696        | 0.0070 |
| 10.50 | 99.335       | 0.9934 | 0.121       | 0.0012 | 0.479        | 0.0048 |

PVI is the total amount of injected gas in pore volumes
Table 8  Effluent concentrations at 3 PV of cushion gas volume

| PVI | CO$_2$ Conc. (%) | yCO$_2$ | N$_2$ Conc. (%) | yN$_2$ | CH$_4$ Conc. (%) | yCH$_4$ |
|-----|------------------|---------|-----------------|--------|-----------------|--------|
| 0.01| 0.118            | 0.0012  | 0.041           | 0.0004 | 99.841          | 0.9984 |
| 0.31| 0.131            | 0.0013  | 0.041           | 0.0004 | 99.828          | 0.9983 |
| 0.60| 0.137            | 0.0014  | 0.413           | 0.0041 | 99.815          | 0.9982 |
| 0.94| 0.142            | 0.0014  | 0.043           | 0.0004 | 99.799          | 0.998  |
| 1.23| 0.157            | 0.0016  | 0.044           | 0.0004 | 99.792          | 0.9979 |
| 1.52| 0.162            | 0.0016  | 0.046           | 0.0005 | 99.792          | 0.9979 |
| 1.81| 0.162            | 0.0016  | 0.046           | 0.0005 | 99.789          | 0.9979 |
| 2.09| 0.164            | 0.0016  | 0.047           | 0.0005 | 99.782          | 0.9978 |
| 2.38| 0.169            | 0.0017  | 0.049           | 0.0005 | 99.782          | 0.9978 |
| 2.67| 0.169            | 0.0017  | 0.049           | 0.0005 | 99.777          | 0.9978 |
| 2.97| 0.173            | 0.0017  | 0.050           | 0.0005 | 99.771          | 0.9977 |
| 3.24| 0.179            | 0.0018  | 0.050           | 0.0005 | 99.765          | 0.9977 |
| 3.53| 0.185            | 0.0019  | 0.050           | 0.0005 | 99.763          | 0.9976 |
| 3.82| 0.186            | 0.0019  | 0.051           | 0.0005 | 99.758          | 0.9976 |
| 4.12| 0.191            | 0.0019  | 0.051           | 0.0005 | 99.754          | 0.9975 |
| 4.40| 0.193            | 0.0019  | 0.053           | 0.0005 | 99.749          | 0.9975 |
| 4.71| 0.197            | 0.002   | 0.054           | 0.0005 | 99.748          | 0.9975 |
| 4.99| 0.197            | 0.002   | 0.055           | 0.0006 | 99.745          | 0.9975 |
| 5.30| 0.199            | 0.002   | 0.056           | 0.0006 | 99.741          | 0.9974 |
| 5.59| 0.200            | 0.002   | 0.059           | 0.0006 | 99.741          | 0.9974 |
| 5.88| 0.200            | 0.002   | 0.059           | 0.0006 | 99.737          | 0.9974 |
| 6.17| 0.204            | 0.002   | 0.059           | 0.0006 | 99.716          | 0.9972 |
| 6.45| 0.205            | 0.0021  | 0.079           | 0.0008 | 99.450          | 0.9945 |
| 6.78| 0.205            | 0.0021  | 2.694           | 0.0269 | 97.101          | 0.9710 |
| 7.06| 0.205            | 0.0021  | 10.214          | 0.1021 | 89.581          | 0.8958 |
| 7.35| 0.387            | 0.0039  | 25.009          | 0.2501 | 74.604          | 0.7460 |
| 7.68| 1.141            | 0.0114  | 44.953          | 0.4495 | 53.906          | 0.5391 |
| 7.98| 3.269            | 0.0327  | 59.989          | 0.5999 | 36.742          | 0.3674 |
| 8.26| 21.793           | 0.2179  | 53.931          | 0.5393 | 24.276          | 0.2428 |
| 8.56| 60.864           | 0.6086  | 26.340          | 0.2634 | 12.796          | 0.1280 |
| 8.85| 82.816           | 0.8282  | 10.089          | 0.1009 | 7.095           | 0.0710 |
| 9.14| 91.405           | 0.9141  | 4.088           | 0.0409 | 4.507           | 0.0451 |
| 9.43| 94.774           | 0.9477  | 2.017           | 0.0202 | 3.209           | 0.0321 |
| 9.73| 96.533           | 0.9653  | 1.111           | 0.0111 | 2.356           | 0.0236 |
| 10.00| 97.503          | 0.975   | 0.727           | 0.0073 | 1.770           | 0.0177 |
| 10.30| 97.995          | 0.98    | 0.559           | 0.0056 | 1.446           | 0.0145 |
| 10.60| 98.154          | 0.9815  | 0.509           | 0.0051 | 1.337           | 0.0134 |
| 10.90| 98.247          | 0.9825  | 0.486           | 0.0049 | 1.267           | 0.0127 |
| 11.20| 98.378          | 0.9838  | 0.455           | 0.0046 | 1.167           | 0.0117 |
| 11.60| 98.533          | 0.9853  | 0.413           | 0.0041 | 1.054           | 0.0105 |

PVI is the total amount of injected gas in pore volumes
Abba MK, Abbas AJ, Athari A, Martin M, Bello S, Salihu MS, Nasr GG (2019) Solubility trapping as a potential secondary mechanism for CO₂ sequestration during enhanced gas recovery by CO₂ injection in conventional natural gas reservoirs: an experimental approach. J Nat Gas Sci Eng 71:1–12. https://doi.org/10.1016/j.jngse.2019.103002

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### Table 9 Efluent concentrations at 5 PV of cushion gas volume

| Time (PV) | CO₂ Conc. (%) | yCO₂ | CO₂ Conc. (%) | yN₂ | CH₂ Conc. (%) | yCH₄ |
|-----------|---------------|------|---------------|------|---------------|------|
| 0.01      | 0.047         | 0.0005 | 0.039         | 0.0004 | 99.914       | 0.9991 |
| 0.31      | 0.055         | 0.0006 | 0.039         | 0.0004 | 99.906       | 0.9991 |
| 0.66      | 0.071         | 0.0007 | 0.039         | 0.0004 | 99.890       | 0.9989 |
| 0.96      | 0.085         | 0.0009 | 0.039         | 0.0004 | 99.876       | 0.9988 |
| 1.25      | 0.099         | 0.0010 | 0.040         | 0.0004 | 99.861       | 0.9986 |
| 1.53      | 0.108         | 0.0011 | 0.041         | 0.0004 | 99.851       | 0.9985 |
| 1.83      | 0.115         | 0.0012 | 0.041         | 0.0004 | 99.844       | 0.9984 |
| 2.15      | 0.122         | 0.0012 | 0.042         | 0.0004 | 99.836       | 0.9984 |
| 2.44      | 0.125         | 0.0013 | 0.042         | 0.0004 | 99.833       | 0.9983 |
| 2.73      | 0.128         | 0.0013 | 0.043         | 0.0004 | 99.829       | 0.9983 |
| 3.02      | 0.134         | 0.0013 | 0.044         | 0.0004 | 99.822       | 0.9982 |
| 3.31      | 0.135         | 0.0014 | 0.044         | 0.0004 | 99.821       | 0.9982 |
| 3.59      | 0.136         | 0.0014 | 0.044         | 0.0004 | 99.820       | 0.9982 |
| 3.88      | 0.138         | 0.0014 | 0.044         | 0.0004 | 99.818       | 0.9982 |
| 4.17      | 0.141         | 0.0014 | 0.045         | 0.0005 | 99.814       | 0.9981 |
| 4.46      | 0.143         | 0.0014 | 0.046         | 0.0005 | 99.811       | 0.9981 |
| 4.74      | 0.143         | 0.0014 | 0.053         | 0.0005 | 99.804       | 0.9980 |
| 5.05      | 0.149         | 0.0015 | 0.06          | 0.0006 | 99.791       | 0.9979 |
| 5.35      | 0.151         | 0.0015 | 0.06          | 0.0006 | 99.789       | 0.9979 |
| 5.63      | 0.156         | 0.0016 | 0.068         | 0.0007 | 99.776       | 0.9978 |
| 5.92      | 0.160         | 0.0016 | 0.079         | 0.0008 | 99.761       | 0.9976 |
| 6.21      | 0.164         | 0.0016 | 0.094         | 0.0009 | 99.742       | 0.9974 |
| 6.50      | 0.167         | 0.0017 | 0.164         | 0.0016 | 99.669       | 0.9967 |
| 6.79      | 0.171         | 0.0017 | 0.709         | 0.0071 | 99.120       | 0.9912 |
| 7.07      | 0.174         | 0.0017 | 2.063         | 0.0206 | 97.763       | 0.9776 |
| 7.36      | 0.177         | 0.0018 | 5.162         | 0.0516 | 94.661       | 0.9466 |
| 7.65      | 0.180         | 0.0018 | 10.918        | 0.1092 | 88.902       | 0.889 |
| 7.95      | 0.181         | 0.0018 | 19.534        | 0.1953 | 80.285       | 0.8029 |
| 8.23      | 0.186         | 0.0019 | 30.643        | 0.3064 | 69.171       | 0.6917 |
| 8.53      | 0.190         | 0.0019 | 44.933        | 0.4493 | 54.877       | 0.5488 |
| 8.83      | 0.192         | 0.0019 | 60.055        | 0.6006 | 39.753       | 0.3975 |
| 9.12      | 0.200         | 0.0020 | 70.416        | 0.7042 | 29.384       | 0.2938 |
| 9.41      | 3.406         | 0.0341 | 75.774        | 0.7577 | 20.820       | 0.2082 |
| 9.72      | 60.648        | 0.6065 | 30.691        | 0.3069 | 8.661        | 0.0866 |
| 10.00     | 88.588        | 0.8859 | 7.601         | 0.076  | 3.811        | 0.0381 |
| 10.30     | 97.136        | 0.9714 | 1.593         | 0.0159 | 1.271        | 0.0127 |
| 10.60     | 98.501        | 0.985  | 0.716         | 0.0072 | 0.783        | 0.0078 |
| 10.90     | 99.045        | 0.9905 | 0.413         | 0.0041 | 0.501        | 0.0050 |
| 11.20     | 99.237        | 0.9924 | 0.303         | 0.0030 | 0.399        | 0.0040 |

PVI is the total amount of injected gas in pore volumes

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