

# The progress of enhanced gas recovery (EGR) in shale gas reservoirs: A review of theory, experiments, and simulations

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## ARTICLE INFO

### Keywords:

Enhanced gas recovery  
Depleted reservoir  
Adsorption isotherm  
CO<sub>2</sub> sequestration  
Reservoir simulations

## ABSTRACT

Due to the combined advantages of injecting CO<sub>2</sub> for boosting natural gas recovery efficiency and sequestration of CO<sub>2</sub> in depleted shale gas reservoirs, the enhanced gas recovery (EGR) approach has recently attracted the attention of researchers. To analyze the viability of the increased gas recovery technique, many published studies were reviewed based on theoretical, experimental settings, and simulation models in this manuscript. The underlying link between geological and petrophysical factors is discussed, as well as how they affect CO<sub>2</sub> and CH<sub>4</sub> sorption. According to numerous studies, 30–55 percent of the CO<sub>2</sub> injected into the shales is adsorbed on the pores surface of the rock matrix, resulting in CH<sub>4</sub> desorption and additional natural gas recovery of 8–16 percent. For the application in diverse shales reservoir conditions, the best fit adsorption models (Langmuir, Ono Kondo, and D-A) were summarized. The theoretical findings of this work are anticipated to add to current studies on CO<sub>2</sub> adsorption and sequestration, as well as CH<sub>4</sub> desorption characteristics and the myriad simulation studies have revealed that well spacing, fracture permeability, injection pressure and strategies are key consideration for effective field demonstration for CO<sub>2</sub>-EGR projects. Despite the availability of theoretical explanations, experimental verification, and modeling findings, field-scale trials remain limited due to the risk of CO<sub>2</sub>-CH<sub>4</sub> mixing and the high cost of capturing, purifying, and re-injecting CO<sub>2</sub> into depleted reservoirs. Furthermore, the unpredictable heterogeneity of the shale formation still poses challenges on the gas recovery. The setbacks and limitations highlighted in this study will encourage academia and researchers to conduct more research into appropriate EGR technologies and their economic implications.

## 1. Introduction

The decline of conventional resources and growing global energy demand caused by increased population and energy consumption, petroleum companies are compelled to drill and produce gas from shale gas, tight gas, and coal seam gas (Wang et al., 2017). However, these unconventional are featured by poor permeability and their production is mainly enhanced by horizontal drilling and induced artificial fractures by hydraulic fracturing (Mojid et al., 2021). To combat CO<sub>2</sub> emissions and global warming concerns caused by the combustion of high carbon content fuel, the focus has switched to cheap, clean, and ecologically favorable resources (natural gas). Anthropogenic CO<sub>2</sub> can be sequestered/stored in a variety of geological formations, including

non-productive coal seams, deep saline aquifers, Basalt formations, and depleted oil/gas reservoirs, where gas storage takes many forms (Newell and Ilgen, 2018).

With the presence of an information about the sub-surface data such as porosity, permeability, water saturation, pre-existing hydraulically fractured network, surface infrastructure may help to partially offset cost associated with this storage option, and the broad spatial distribution of these wells may make them attractive targets for nearby industrial emitters (Hong et al., 2016).

To revive the production from the depleted or nearly depleted unconventional, repressurization (Ghazi et al., 2018) and re-fracturing must be employed. Gas injection into the reservoir largely boost the pressure and stimulate the gas flow from the pore spaces (Godec et al.,

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<https://doi.org/10.1016/j.petrol.2022.110461>

Received 11 December 2021; Received in revised form 16 February 2022; Accepted 21 March 2022

Available online 26 March 2022

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2013). Refracturing is an attempt to inject highly pressurized liquid to induce more fractures in the formation matrix so that gas flow from the matrix, fractures into the production well can occur. The gas injection into the reservoir is simultaneously accompanied by the dislodgment and movement of  $\text{CH}_4$  towards the production well (Hasan et al., 2017). The simultaneous injection of  $\text{CO}_2$  and the improved gas production have raised the researchers confidence on the feasibility of the projects due to previous cost that were impairing these dual processes (Hong et al., 2016). The gases that qualify for the injection into the reservoir are mainly  $\text{CO}_2$ ,  $\text{N}_2$ , or mixture of the two gases and they can substantially raise the yield of hydrocarbon production. Nevertheless, the whole EGR process is faced with gas-gas mixing, formation swelling caused by  $\text{CO}_2$  injection which renders the projects uneconomical and are prematurely aborted.

In contrast,  $\text{N}_2$  can be an alternative as could be easily obtained through cryogenic air separation. It requires less compression ratio than  $\text{CO}_2$ , which is why a lower amount of it was required to initiate much pressure in the  $\text{CH}_4$  reservoir. Also, the cost for sweetening process of  $\text{CH}_4$  contaminated with  $\text{N}_2$  is less than that of  $\text{CO}_2$ . This was why the fraction of produced  $\text{N}_2$  tolerance is higher than the  $\text{CO}_2$  limit during the natural gas exploration (Mohammed et al., 2020a). The suitability and selection of EGR must consider evaluation of precise technical, economic and sometimes political driven aspects in attaining the conflicting purposes of both improved recovery of natural gas and  $\text{CO}_2$  sequestration. The gas injected into the reservoir might be transformed to the hydrates due to the effect of pressure in the reservoir cooling the gas. The gas hydrates formed can have adverse effects in the pore spaces in proximity to the production well. In contrast, when  $\text{CO}_2$  is used as a drive gas, transition from the gas to liquid phase may occur when reservoir pressure is increased sufficiently, leading to more favorable gravity separation between the  $\text{CO}_2$  and the ambient gas. For example, the influence of the difference in  $\text{CO}_2$  and  $\text{N}_2$  solubility in water could affect the breakthrough at production wells, while the difference in interfacial tension (IFT) with water could affect the relative mobility of the gas in water and the effectiveness of low-permeability seals (Leeuwenburgh et al., 2014).

The intent application of EGR method is to recover the gas remaining after the failure of primary and secondary production methods by injecting  $\text{CO}_2$  or  $\text{N}_2$  or mixture that helps maintain pressure at a sufficiently high level for production to continue (Leeuwenburgh et al., 2014). Technically, when fluid is drawn from macro and micro fractures in unconventional reservoirs, the well is first fed at a high rate. The flow rate drops dramatically due to decreased pressure and the flow that takes over is from the matrix. At this time, an intervention using re-pressurization and re-fracturing methods are needed to boost recovery factor and extend the life of unconventional wells (Hasan et al., 2017).

Energy intensive facilities which depend on combustion of fossils fuels are the major contributor of the  $\text{CO}_2$  emission and these includes the power production sector, refinery plants, mining industry, metallurgy, the chemical industry, and the cement production plants (Abuov et al., 2020). The flue gas from these facilities can be collected, compressed and injected into the geological formations by using sophisticated methods. (Arora et al., 2019), defined  $\text{CO}_2$  capture and sequestration (CCS) as a collection of technical inventions that mitigate  $\text{CO}_2$  emissions from industrial plant sources and store it permanently into geologic structures such as saline aquifers, depleted or nearly depleted gas reservoirs and deep coal seams. Although the  $\text{CO}_2$  sequestration in saline aquifers which its potential as a sink is associated with doubts on possible leakage, loss of gas injectivity and structure (Tang et al., 2021). The CCS involves three key process that includes: Capture of  $\text{CO}_2$  from flue gas, compress it, transport in pipelines or trucks and finally injected underground for geologic sequestration. The formations are deeply seated consisting of rocks with numerous pore spaces and covered by non-permeable rock which prevent it from migrating upward.

The basics of EGR is based largely on the selective adsorption of injected gas and desorption of the indigenous natural gas from the organic matrix of the formation. The adsorption takes two forms namely physical adsorption (physisorption) maintained by the weak van der waal forces (Ashwani Kumar, 2020) and chemical adsorption (chemisorption) caused by the chemical bonds (Erkey, 2011). Physisorption in the porous media is attributed to the decreased randomness of gases (entropy) occurring due to increased intermolecular attractive forces between the pores organic matter and gas molecules forcing other parameters Gibbs free energy and enthalpy change to be negative so that the process can be spontaneous (Klewiah et al., 2020). Since the enthalpy change is  $< 0$ , the overall adsorption becomes exothermic and therefore less favoured at higher temperature according to Le Chatelier principle.

Previous studies have emphasized on the use of  $\text{CO}_2$ ,  $\text{N}_2$  (Ansari and Ghosh, 2013) and  $\text{CO}_2/\text{N}_2$  (Zhang et al., 2020b) to enhance gas recovery however, their results could not account to all parameters involved in theory, experiments and simulation models. (Biyanto et al., 2019), produced a model to simulate the EGR operating conditions that determines the overall cost of the projects. The parameters that were optimized during injection involves mass flow rate, temperature and pressure. This study reviewed the on-going methods of enhanced gas recovery with the emphasis on the theory, experimental and simulation studies and account for the limited field applications. The findings, setbacks and on-going limiting parameters presented herein are expected to motivate the academicians and researchers to effectively venture into further research associated with the feasible EGR methods and their economic implications.

### 1.1. Mechanism for $\text{CO}_2$ trapping for geological storage

The geologic storage method has been proved to be relatively superior for long-term storage of  $\text{CO}_2$  because of easy site convenience and economic aspects. The assessment of reservoirs storage capacity and trapping mechanisms for  $\text{CO}_2$  plays major roles in the verification of storage potentials, the evaluation of storage security, and the monitoring arrangement for the same type of projects (Anwar et al., 2018). The injected  $\text{CO}_2$  into the geological media can be retained by four different mechanism which are meant for storage and storage security. These are (i) structural trapping caused by an overlying impermeable cap rock and stratigraphic trapping due to the presence of anticlines or faults that prevent upward movement (Fig. 1a). It is the main mechanism that store injected  $\text{CO}_2$ . (ii) Capillary/residual trapping, involves tiny spots of  $\text{CO}_2$  in the pores being immobilized by capillary forces and remain as separate phase. Residual trapping contributes significantly to the immobilization of  $\text{CO}_2$  on short time scales and is very secure when compared to the stratigraphic trapping (Fig. 1b) (Zhao et al., 2014) (iii) solubility trapping, the injected  $\text{CO}_2$  dissolves in the fluid (brine) present in the reservoir to form carbonic acid which later dissociates into carbonates and hydroxyl ion. The latter descends down the porous storage medium and rendering it unavailable as a separate phase (Fig. 1c) and (iv) mineral trapping, occurs when  $\text{CO}_2$  dissolves in reservoir fluid to form carbonic acid which interact with the rock minerals in the geological media to form the stable carbonate mineral as precipitates, making it a suitable long-term storage feature although not popular (Fig. 1d) (Raza et al., 2016).

## 2. Enhanced gas recovery in conventional reservoirs

So far, the only EGR field demonstrations are sandstone reservoirs in Netherlands and Canada. Because of their intrinsic heterogeneity and fluid flow and transport complexity, carbonate reserves have not yet been considered for any EGR project. The only EGR field demonstrations so far have taken place in sandstone reservoirs in the Netherlands and Canada. Carbonate reserves have not yet been considered for any EGR project due to their inherent heterogeneity, fluid flow, and transport

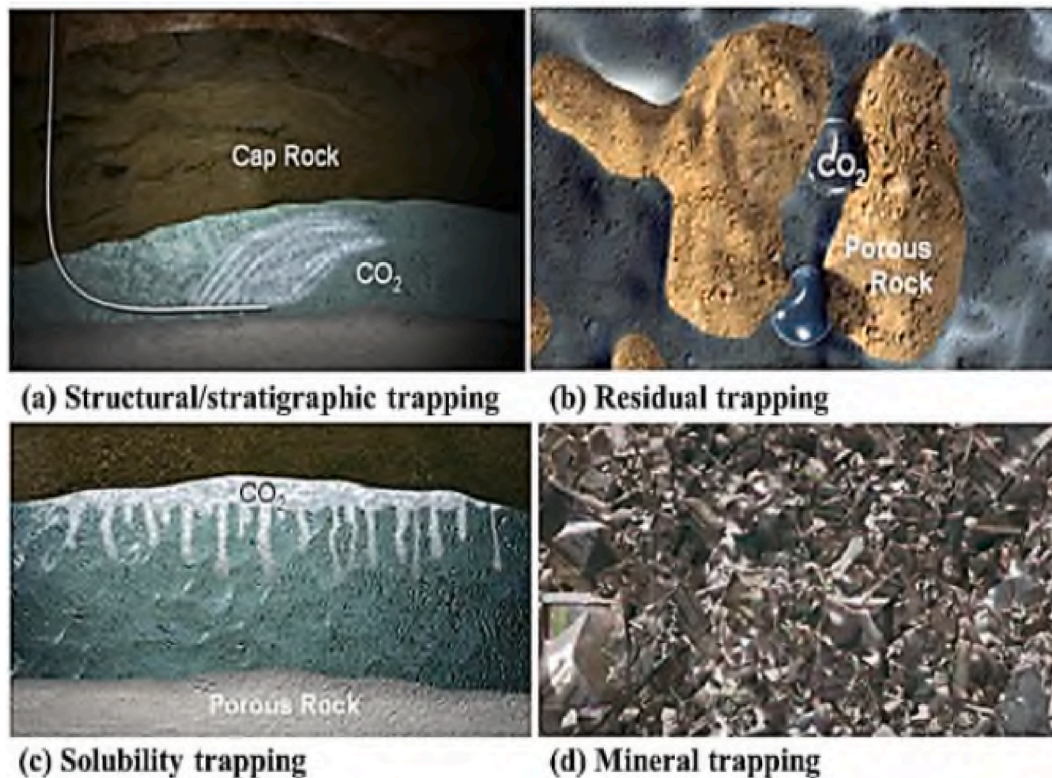


Fig. 1. Illustration of geological trapping mechanism of CO<sub>2</sub> (Aminu et al., 2017).

complexity. The CO<sub>2</sub>-EGR in sandstone reservoirs is characterized by the high permeability high porosity which determine the volume of stored gas and the flow speed of the gas (Hamza et al., 2021). The reservoirs are heterogenous with varying amount of clay, silica minerals. Table 2 represent the methane production efficiency due to varied injection rate on the sandstone core samples.

The transition from conventional to unconventional resources such as shale and coal seams has been fraught with difficulties such as extremely low porosity and permeability, complex heterogeneity and extreme conditions. Intensive characterization of shales and coal seams has revealed some similarities and differences exist among the two and some experimental facts regarding coal seam can be applied in shales (Ao et al., 2020). For example, the pore structure of shale is more complex than that of coal because it contains both organic and inorganic pores. Because of the material's diversity, investigating the organic and inorganic pores of shale is difficult (Liu and Zhu, 2016).

### 3. Selection of gas injection for EGR

#### 3.1. Gas types

The popular gases injected for the EGR processes are mainly CO<sub>2</sub>, CH<sub>4</sub> and N<sub>2</sub> with each one exerting different mechanism in the formation pores and matrix. CO<sub>2</sub> competitive advantage over other gases in terms of the formation surface adsorption and environmental reduction of CO<sub>2</sub>, many researchers have been attracted to it. Due to the higher attraction of CO<sub>2</sub> to the formation pore surfaces, it competitively adsorbs and easily induce desorption of methane and occupy the spaces by itself. The potentials of CO<sub>2</sub> are summarized in Table 1. Apart from the former advantage of storage, the injected CO<sub>2</sub> has a tendency of triggering bulge thereby affecting the permeable channels in the formation (Lu et al., 2016; Lin et al., 2019; Ao et al., 2020) while the injection of N<sub>2</sub> adsorption is very poor but causes the shrinkage which is favorable for the injectivity of gas (Zhang et al., 2020a).

Table 1

The summary of CO<sub>2</sub> injection and storage estimates in depleted gas reservoirs (Hamza et al., 2020).

Feature	Potential output
CO <sub>2</sub> storage capacity (Gigatons)	390–750
Recovery mechanism	CH <sub>4</sub> desorption due to CO <sub>2</sub> adsorption
Miscibility	Completely miscible
Recovery efficiency	10–35% of OGIP

#### 3.2. CO<sub>2</sub> injection

The popularity of CO<sub>2</sub>-EGR method is associated with the coupling of CH<sub>4</sub> recovery enhancement and geological storage of CO<sub>2</sub> in the depleted or nearly depleted reservoirs undoubtedly having more economic benefits (Siwei et al., 2019). Due to the reservoir temperature and pressures, the production of CH<sub>4</sub> and the sequestration of CO<sub>2</sub> processes must be accomplished in the form of sc-CO<sub>2</sub> (Mohammed et al., 2020b). The promising outcomes of the increased recovery factor by the injection of CO<sub>2</sub> are attributed to firstly, the preferential adsorption tendency of the shale formation due to stronger attraction between injected CO<sub>2</sub> molecules and the organic matter in the shale formations (Wang et al., 2016a, b; Iddphonce et al., 2020). This is possible because of the availability of large clay content that provides the adsorption and storage sites for CO<sub>2</sub> (Sun et al., 2020). The surface area of different clay minerals influences their CH<sub>4</sub> sorption capacity, and montmorillonite has the highest CH<sub>4</sub> adsorption capacity attributed to the largest micropores volumes and large surface area (Wang et al., 2020b), Hwang and Pini (2019). Clay minerals have an impact on shale's methane adsorption capacity, particularly in organic-lean shale deposits with low TOC. Micropores volume and structures make up montmorillonite clay minerals, gives them bigger surface areas and more possible adsorption sites than other minerals. The governing factors of the sorption are the cation exchange and pressure where the effects of the latter is more



pronounced (Hu et al., 2019). With different clay minerals, methane adsorption capacity varies significantly, with CH<sub>4</sub> sorption capacity decreasing in the order montmorillonite > I-S mixed layer > kaolinite > chlorite > illite (Liang et al., 2021). Secondly, the shale reservoirs tend not to allow leakage of any gas because of its caprock strength as they have been able to hold the natural gas for years (Eshkalak et al., 2014; Liu et al., 2021; Edvardsen et al., 2021). The caprock is made up of layers with extremely low permeability that cover the storage formation and keep hydrocarbons from leaking out without risking gas injectivity (Shales et al., 2013; Raza et al., 2015). It is meant to hold the natural gas or injected gas from leakage (Newell et al., 2017). The injectivity of gases within the shales are not affected by the caprock strength. In shales, injectivity is very poor however, it can be improved by the use of horizontal drilling and hydraulic fracturing to unlock and allow gas flow to the fractures and then wellbore (Edvardsen et al., 2021).

Thirdly, Selection of depleted reservoirs as a sink to anthropogenic CO<sub>2</sub> is more economical than injection CO<sub>2</sub> into saline aquifers. This is attributed to the simultaneous injection of CO<sub>2</sub> to the recovery of hydrocarbons (oil/gas) in which their market prices are attractive. Also, the preliminary costs of saline aquifer characterization (permeability, thickness and extent of storage reservoir, tightness of caprock, geological structure, lithology) and infrastructure building (well drilling, monitoring site) are minimized (Eshkalak et al., 2014). This is contrary to the depleted reservoirs which already have the infrastructures in place such as injection well, geological informations are all known and the hydrocarbon can simultaneously be recovered in return (Benson et al., 2005). Deep saline aquifers, on the other hand, are regarded as the best of all storage options due to their global availability, accessibility, and large storage potential (Mkema and Bin, 2020).

Due to the shale reservoir conditions of high temperature (370–500 K) and pressure (15–20 MPa), the CO<sub>2</sub> injected as liquid instantly exist in supercritical state (above 31 °C and 7.38 MP) (Shi et al., 2017). It has been suggested that CO<sub>2</sub> be injected in a liquid state rather than a supercritical state since it is more energy efficient and exerts less pressure at the wellhead (Jurewicz and Thompson, 2010). Supercritical CO<sub>2</sub> density is close to the liquid making it easier for huge CO<sub>2</sub> deposits and its gas-like viscosity which makes it easy to diffuse in the reservoir and induced natural gas movement (Gupta and Peter, 2020b). The field monitoring data generated from the fracturing process using CO<sub>2</sub> waterless fluid show that CO<sub>2</sub> in supercritical state can last longer in soaking and flow back processes. The blending of methane and the injected gas significantly compromise the quality of the produced natural gas since they are miscible in all proportions at reservoir conditions (Ahn et al., 2020). The premature CO<sub>2</sub> breakthrough is likely to occur due to the dispersion effects under the reservoir conditions. (Patel et al., 2016a). Despite the fact that CO<sub>2</sub> and natural gas were mixable, their physical properties were potential favorable for reservoir repressurization without extensive mixing which was beneficial process of CO<sub>2</sub>-EGR. For instance, CO<sub>2</sub> had density higher than methane by 2–6 times higher at all relevant reservoir conditions. In addition, the lower mobility of CO<sub>2</sub> as compared to that methane, makes it a high viscosity component in the reservoir, and due to this feature, the natural gas can easily be displaced by the injected CO<sub>2</sub> gas. Furthermore, the mixing up of natural gas with the injected CO<sub>2</sub> in the reservoir is largely minimized due to its high solubility (Khan et al., 2013).

(Vandeweyer et al., 2011), reported the field applications where CO<sub>2</sub> sequestration in nearly depleted gas fields at offshore have been performed. As part of monitoring, the gamma ray, electromagnetic imaging tool, cement bond log and multi-finger imaging tools provided confidence that well integrity in leakage prevention can be guaranteed for more years of CO<sub>2</sub> injection to come (Kühn et al., 2013). also reported the Altmark gas field well integrity, geological processes for the possible pilot application of EGR using CO<sub>2</sub>. Similar to other CO<sub>2</sub>-EGR methods, this project was mainly focused on simultaneous sequestration of CO<sub>2</sub> and CH<sub>4</sub> recovery processes from the depleted reservoir. Fig. 2 illustrates the CO<sub>2</sub> injection that induces the dislodge and flow of residual CH<sub>4</sub>

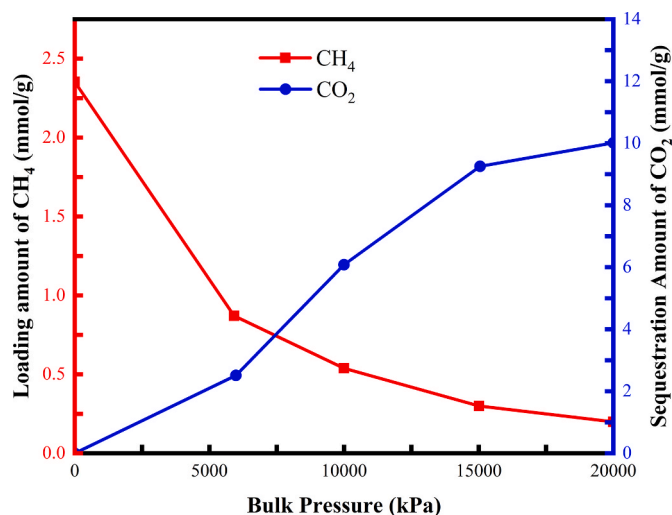


Fig. 2. Sorption trend for the CO<sub>2</sub>/CH<sub>4</sub> respectively over the reservoir bulk pressure (Sun et al., 2017).

from the micropores of the matrix.

The comparative study of adsorption and breakthrough of CO<sub>2</sub> and the mixture of CO<sub>2</sub>-N<sub>2</sub> on shale sample is illustrated in Fig. 3. Different concentration ratios of CO<sub>2</sub> and the mixtures were studied and the results revealed that shales adsorption capacity depends on the nature of the gas adsorbed. The adsorption and sequestration of 100% CO<sub>2</sub> was extremely higher than the mixtures and the breakthrough occurs at very high adsorption volume meaning that the residence time is very long. For effective use of CO<sub>2</sub>-N<sub>2</sub> mixtures, higher concentration ratio (80%:20%) was recommended because of high displacement efficiency and lower swelling of the shales.

(Sun et al., 2016) revealed that the higher CO<sub>2</sub> adsorption tendency over CH<sub>4</sub> and higher diffusing rate for the residual natural gas in nanopores indicating that the displacement efficiency is very high such that sequestration and the production of natural gas are assured as seen in Fig. 4.

### 3.3. N<sub>2</sub> injection

The injection of nitrogen for EGR is favoured by reservoirs

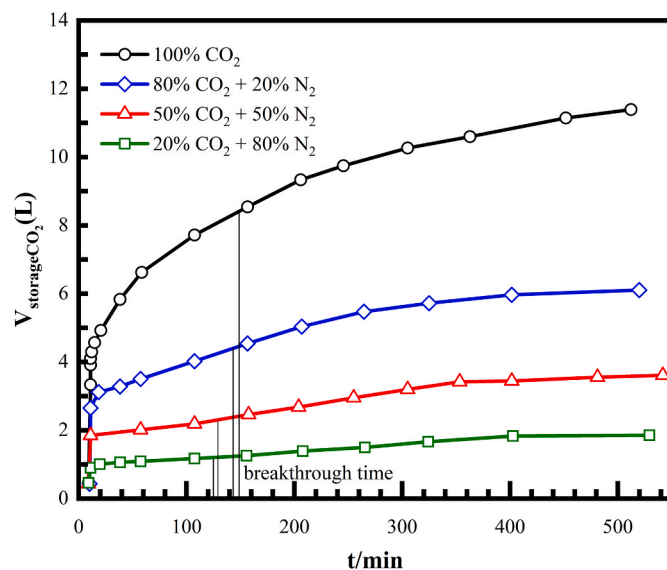


Fig. 3. Comparative adsorption and breakthrough of CO<sub>2</sub> and mixture of CO<sub>2</sub>-N<sub>2</sub> on shale sample (Ashwani Kumar, 2020).

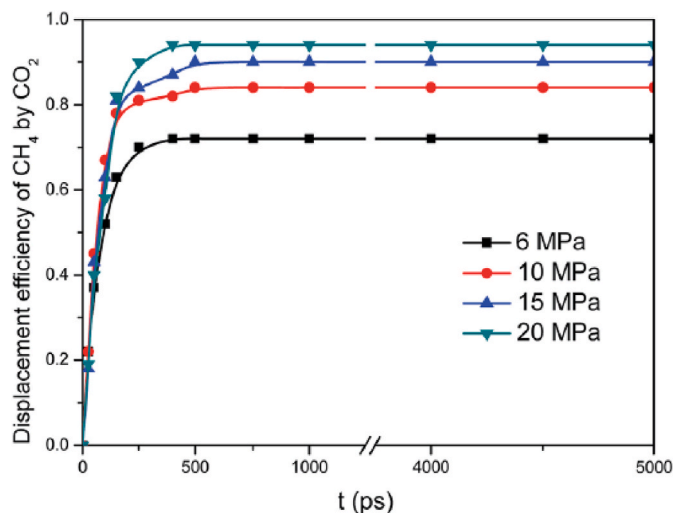


Fig. 4. The efficacy of CO<sub>2</sub> in adsorbed CH<sub>4</sub> displacement at different bulk pressures and temperature (Sun et al., 2016).

characterized by low permeable zones. Its injection does not reduce the permeability of the pores and fractures which can occur due to swelling induced by the gas (Kang et al., 2019). It causes the desorption of methane by lowering the partial pressure allowing it to flow towards the production well (Du et al., 2019). It has been observed that injecting a CO<sub>2</sub>-N<sub>2</sub> mixture for EGR has various benefits, including reduced pore swelling, improved CH<sub>4</sub> recovery, and CO<sub>2</sub> burial. The gas recovery impact, on the other hand, was determined by the injected gas formulation and the variation of pore size. The CH<sub>4</sub> recovery rate of more than 90% can be achieved when the total bulk fraction of CO<sub>2</sub> and N<sub>2</sub> is about 90% (Zhang et al., 2020c). Others have reported that, injection of CO<sub>2</sub>-N<sub>2</sub> mixture at the ratio of 1:1 enhance the displacement of natural gas by the almost 89% efficiency as compared to the same for N<sub>2</sub> or CO<sub>2</sub> alone. This was attributed to the synergistic effect of the two gases (Ashwani Kumar, 2020).

As illustrated on Fig. 5, which exhibits Langmuir isotherms (gas concentration as a function of pressure) for CH<sub>4</sub>, CO<sub>2</sub> and N<sub>2</sub>, the adsorption is largely affected by the pressure. It is positively correlated with the injection pressure. However, coal/shale matrix swelling is likely to happen due to the influx of the more CO<sub>2</sub> molecules with respect to the natural gas. This in turn reduces the porosity and effective permeability and indeed limit the field scale application of the EGR-CO<sub>2</sub> method (Oudinot et al., 2017a).

The inherent challenges of gas-gas mixing, high compression ratio

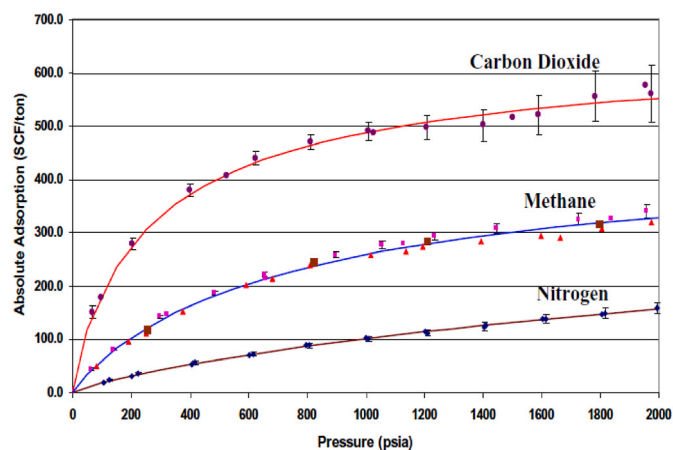


Fig. 5. Langmuir adsorption isotherm for CO<sub>2</sub>, CH<sub>4</sub>, and N<sub>2</sub> (Oudinot et al., 2017b; Zhou et al., 2013).

and formation swelling associated with the CO<sub>2</sub> injection, have influenced the use of N<sub>2</sub> injection as a feasible alternative approach for EGR. The advantages of N<sub>2</sub> injection over CO<sub>2</sub> are lower compression ratio, which means that a smaller volume of it is needed to generate significant pressure in the natural gas reservoir. In addition, natural gas tainted with N<sub>2</sub> has a lower cost of sweetening than natural gas contaminated with CO<sub>2</sub> making it economical (Mohammed et al., 2020a).

At constant pressure and temperature, the heat of adsorption for CO<sub>2</sub> is greater than for CH<sub>4</sub> in shales and hence favours its adsorption. Elevated temperature impairs the adsorption behaviours of either CO<sub>2</sub> or CH<sub>4</sub>. Furthermore, the adsorption trend is linked to the physical constants of the corresponding gases as shown in Table 3. Due to the increase of kinetic diameter in this order CO<sub>2</sub> < N<sub>2</sub> < CH<sub>4</sub>, the adsorption of these gases also increases. The trend for the adsorption increased in the order of increasing kinetic diameter for the gas molecules concerned.

However Zhang et al. (2020b) reported the injection of CO<sub>2</sub>/N<sub>2</sub> mixture to accommodate the dual advantages of the two gases in the formation. Due to higher affinity between the CO<sub>2</sub> and the pore surfaces, injected gas can readily adsorb itself and compel the desorption of CH<sub>4</sub> which facilitate sequestration and production (Carchini et al., 2020). Nevertheless, the injected CO<sub>2</sub> adversely affects formation permeability due to swelling that impairs gas production and injectivity of CO<sub>2</sub>. In contrast, the injection of N<sub>2</sub> causes the formation shrinkage that enhance the gas injectivity and gas production (Lu et al., 2016).

#### 4. Enhanced gas recovery methods

##### 4.1. Re-fracturing by waterless fracturing technology

The benefits of using CO<sub>2</sub> as a fracturing fluid additive might include the following; 1) CO<sub>2</sub> is miscible with CH<sub>4</sub>, where significant CO<sub>2</sub> diffusion into the matrix can occur; 2) CO<sub>2</sub> has a higher adsorption capability within the matrix in comparison to CH<sub>4</sub>, facilitating the production of CH<sub>4</sub> post fracturing work; 3) CO<sub>2</sub> can work as an energizing fluid as it has a relatively high solubility in water, which assists the flow back of water due to a solution gas drive mechanism; 4) Use of CO<sub>2</sub> can mitigate the clay swelling problem to some extent, and 5) The use of CO<sub>2</sub>-based energized fluid reduces or even prevents the swabbing operations (Gao and Li, 2016).

##### 4.2. Formation heat treatment (FHT)

The invention of FHT technology was meant to overcome the problem of aqueous phase trapping which was severely reducing the gas production in the poorly permeable formations (Kang et al., 2016). The underlying mechanism of FHT in shale and tight gas reservoirs triggers the following remarkable changes; it evaporates water which blocked the gas channels and limit gas flow, it stimulate desorption of gas in the matrix due to the rise of temperature, facilitate formation thermal

Table 2

The variation of injection rates and its effect on methane production (Mohammed et al., 2020c).

Core Samples	Q (ml/min)	Breakthrough (min)	CH <sub>4</sub> Produced (cm <sup>3</sup> )	RF = $\frac{CH_4 \text{ Produced}}{OGIP} \times 100$
Berea gray	0.2	93.33	640.59	69.63
	0.4	73.32	819.09	89.04
	0.6	42.15	559.45	60.81
	0.8	40.15	476.28	51.77
	1.0	39.99	478.06	51.97
Bandera gray	0.2	76.32	550.53	63.37
	0.4	82.49	652.20	75.08
	0.6	35.65	495.76	57.07
	0.8	26.82	402.13	46.29
	1.0	35.32	313.69	36.11

**Table 3**  
Comparison of physical constants of CH<sub>4</sub>, CO<sub>2</sub> and N<sub>2</sub>(Hamza et al., 2020).

Parameter	Gas			Reference
	CO <sub>2</sub>	CH <sub>4</sub>	N <sub>2</sub>	
Molar polarizability, αM (cm <sup>3</sup> /mol)	7.34	6.54	4.39	Liu et al. (2020b)
Collision diameter, (Å)	4.00	3.82	3.68	Liu et al. (2020b)
Kinetic diameter, (Å)	3.30	3.80	3.64	(Liu et al., 2020b; Mehio et al., 2014)
Effective molecule diameter, (Å)	3.63	3.81	3.66	Liu et al. (2020b)

expansion and cracking which increases permeability and gas flow into the production wellbore (Liu et al., 2020a).

The FHT experimental results by (Kang et al., 2016) revealed that permeability and desorption of natural gas can be enhanced by the injection of heat at a temperature range of 400–500 °C although is extremely difficult to achieve such high temperature in the reservoir. It was further reported that gas displacement and the flow rates were improved due to large fracture network induced and the remove of water blockage effect. Furthermore, numerical simulation by (Zhu et al., 2016) suggested that gas recovery can be enhanced by the application of heat during fracturing. The effect of high temperature to reverse the sorption behaviour of methane and induce more fractures significantly contributes to the total gas produced in a prolonged lifespan of the reservoir as shown in Fig. 6. The investigation of thermal effect on gas production on coal seams provided similar findings that the rise in temperature stimulate the desorption of indigenous gas (Wojtacha-rychter and Smoli, 2018).

## 5. Theoretical description

### 5.1. Fluid storage in unconventional reservoirs

The unconventional reservoirs store the natural gas in two major forms namely the adsorbed state in the complex organic matter (kerogen) and free state in the pore spaces and fractures (Zhou et al., 2019b; Kang et al., 2011). The adsorbed states on the internal surfaces of nanopore/micropore shale organic materials and clay minerals such as illite account for huge storage potential of about 20–80% (Edwards et al., 2015; Perera, 2017; Gupta and Peter, 2020a, b). The parameters governing that increase storage capacity in the unconventional are the total organic carbon, elevated pressure, lowered temperature, source rock maturity and clay minerals content (Zhou et al., 2019a). Some

amount of gas is also stored in the dissolved form in the organic matter of the matrix and the lowering of pressure can induce its release into the pores and fractures (Mohagheghian et al., 2019; Zhou et al., 2019c). In shales, gas storage occurs largely by physisorption where Van der waal forces and electrostatic interactions are involved. Depending on the pressure in the formation, the physical adsorption may lead to the formation of single (low pressure) or double layers (higher pressure)(Rani et al., 2019a).

### 5.2. Fluid flow and dispersion in porous media

Due to the complexity of the flow path geometry, the fluid flow in porous media is contrary to the normal capillaries and therefore measurements regarding the extent of dispersion should be executed. The velocity difference at nanopores scale and tortuosity account for mechanical dispersion (Fig. 7). The transport of solutes in a porous medium is affected by hydrodynamic dispersion, which originates from the concurrent action of molecular diffusion (resulting from concentration gradients) and advection (resulting from fluid flow velocities) (Hughes et al., 2012). This process is typically quantified by means of a dispersion coefficient with both longitudinal (L) and transverse (T) components as shown in equations (1) and (2) (Nguyen and Papavassiliou, 2020). However, the transverse dispersion ( $D_T$ ) coefficient is extremely small when compared to longitudinal dispersion coefficient and also difficult to measure in the laboratory and therefore ignored by some authors (Abba, 2020).

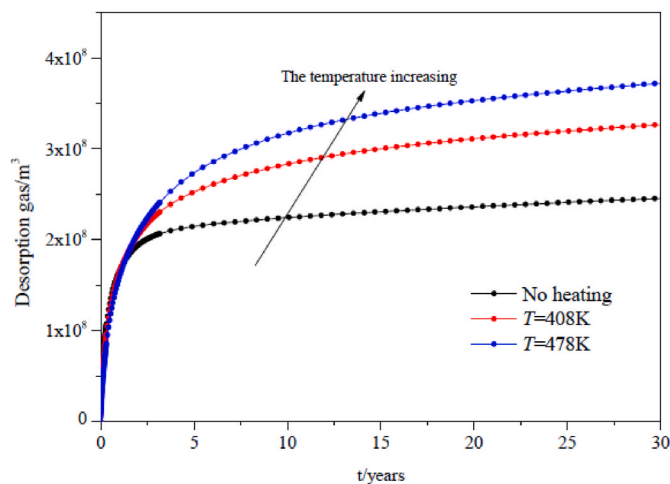
$$D_L = D'_m + \alpha_L u^n \quad \text{eqn 1}$$

$$D_T = D'_m + \alpha_T u^n \quad \text{eqn 2}$$

where  $D'_m$  is an effective molecular diffusion coefficient in the porous medium,  $u$  is the interstitial velocity in the direction of flow, and  $\alpha$  is the so-called dynamic dispersivity, which can be regarded as the intensive property of the porous medium. The variation in fluid flow velocity in pores networks are primarily caused by differences in solute path length, different velocities due to friction in the pores, and pore sizes (Eskandari and Science, 2019).

The extent of dispersion for the injected CO<sub>2</sub> and indigenous CH<sub>4</sub> can be alleviated by considering certain critical factors such as strategies for the gas injection and production and the fluids behaviour (Patel et al., 2016b). The flow of injected CO<sub>2</sub> in the reservoir pore spaces is likely to blend with the indigenous natural gas (CH<sub>4</sub>) and minimizes the effectiveness of CO<sub>2</sub> storage and discharge of CH<sub>4</sub>. The tendency of the gases to mix in the porous media called hydrodynamic dispersion will occur due to the advection and diffusion (Abba, 2020; Nguyen and Papavassiliou, 2020). It is attributed to the gas slippage and diffusion at molecular level (Kabir et al., 2018). Due to the miscibility effect of these gases, intensive practical experience is required to make the process of storage and recovery of gases economical (Mohammed et al., 2020b; Honari, 2016). Displacement of indigenous methane from the matrix is highly favoured by the supercritical nature of CO<sub>2</sub> where it is characterized by higher viscosity relative to the CH<sub>4</sub> allowing the easy flow of the gas to the production well (Kabir et al., 2018). The CO<sub>2</sub> preferential adsorption on the shale and coals matrix pore surfaces induce desorption of natural gas is depicted in Fig. 8.

Due to the huge disparity of the simulated and field observed EGR processes, the formulation of novel models is necessary. Several models have been invented to simulate the EGR process to account for the dispersion where advection and dispersion have been considered as the only compelling mechanism for the mixing of the CO<sub>2</sub> and CH<sub>4</sub>. This has led to the failure of the EGR at the field scale and therefore numerical dispersion accounting for more parameters are inevitable (Kumar et al., 2010; Honari et al., 2013).



**Fig. 6.** The effect of temperature on gas desorption and production for adsorbed gas (Zhu et al., 2016).



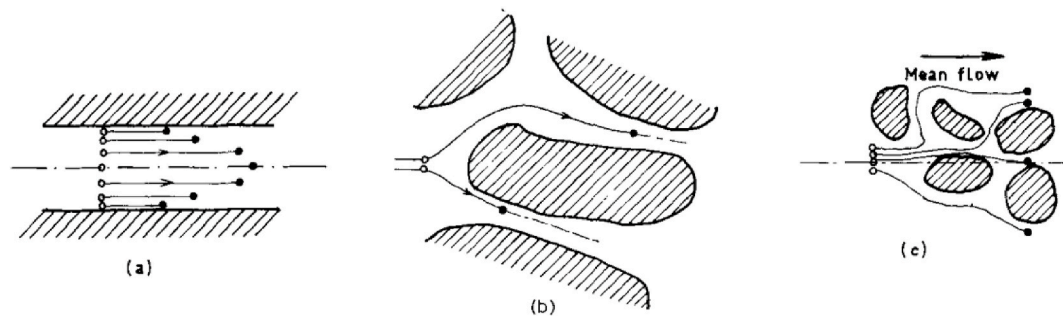


Fig. 7. Dispersion due to (a) friction in the pore (b) solute path (c) pore sizes (Dentz et al., 2018).

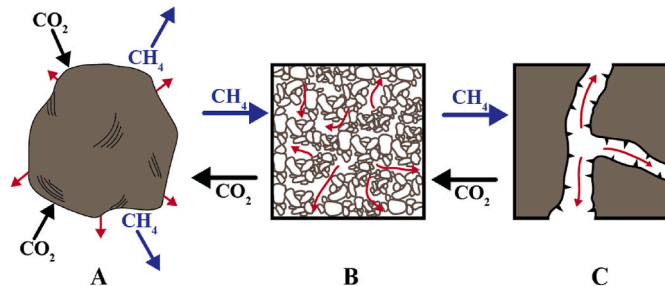


Fig. 8. The scheme of influx/outflux flow of  $\text{CO}_2/\text{CH}_4$  respectively from shale matrix to the fractures (Godec et al., 2014).

## 6. Recovery mechanism and adsorption isotherms in unconventional reservoirs

### 6.1. Recovery mechanism

The injection of  $\text{CO}_2$  and the success of gas recovery is based on the two plausible mechanisms which are (1) pressure enhancement where gas injected induces buoyancy and competitive adsorption with the natural gas in situ (2) displacement where methane is swept away by the arrival of  $\text{CO}_2$ . The injected  $\text{CO}_2$  in the reservoir exist in supercritical conditions and the fluid-fluid and fluid-rock interactions follow different mechanism (Honari et al., 2016). However, the tendency of gas fingering and  $\text{CO}_2$ - $\text{CH}_4$  mixing can be lowered due to the higher density and viscosity of pure  $\text{CO}_2$  that allows the heavier one to sink below the two gas phases. For the output recovery of OGIP at 73–85%, the  $\text{CO}_2$  breakthrough is inevitable (Ghazi et al., 2018).

### 6.2. Gas adsorption isotherms

In an attempt to model and simulate the sorption and displacement of  $\text{CO}_2$  and  $\text{CH}_4$  respectively, numerous studies have been published. In the porous media, gas flow is influenced by the adsorption tendency and therefore accurate measurement of the adsorption property play a crucial role in the modeling (Eliebid et al., 2018b). The measure of natural gas that is adsorbed on the reservoir rock surfaces is crucial to determine the adsorption and desorption processes. The retention of gas which largely depends on pressure occurs physically due to the electrostatic interactions and van der Waals forces, while the chemical bonds can lead to the chemical adsorption. The gas adsorption to the rock surfaces surge as the pressure increases while decrease in pressure intensifies the gas desorption. Several studies have reported the popular and simple isotherms which are Freundlich and Langmuir isotherms to model the gas adsorption in porous media in shales, however Freundlich has never shown the best fit (Mahmoud, 2019). Failure to select the appropriate adsorption isotherm can lead to serious errors and mistakes and therefore it is advocated to thoroughly evaluate the models (Fianu et al., 2019). Mixed models superiority have been reported and

suggested for modeling in mono and dual gas phases, Langmuir model, BET model (Duan et al., 2016), Ono-kondo model (Chi et al., 2019). The laboratory experimental data generates the adsorption isotherms which are compared with the findings from the best fitting models so that the adsorption capacity and rates can be determined.

(Merey and Sinayuc, 2016) experimented on the Langmuir isotherm and Ono-Kondo model to account the adsorption at high and low pressures in shales. He found that Ono-Kondo model could best fit the adsorption data derived from the experiment at both pressure conditions (high/low) in comparison to the well-known Langmuir model as shown in Fig. 10. Actually, the unconventional reservoirs are very deep such that the reservoir pressure and temperature are also high. Therefore, prediction of the gas adsorption can best be explained by Ono-Kondo model than Langmuir (isotherm) model which cannot hold true at this condition (Xiong et al., 2016; Bi et al., 2017). The reservoir conditions made the Langmuir model not the best option as shown in Fig. 9, although under low pressure, both models may provide the best fit. Furthermore, temperature dependent adsorption model was proposed to account for the effect of temperature in shale adsorption process (Fianu et al., 2019). Similar findings were reported by (Chi et al., 2019) where adsorption was well predicted by the Langmuir + k model. However, the high affinity of  $\text{CO}_2$  and the shales was confirmed by the high interaction energy derived from the Ono-Kondo model.

(Zhou et al., 2019a) also reported the adsorption tendency of samples collected from the Sichuan and Ordos Basins in China where experimental data were fitted in various isotherm models. However, for the supercritical  $\text{CO}_2$  which is obviously the common reservoir condition, the Ono-Kondo model produced a best fit as compared to Langmuir, Dubinin-Astakhov and Dubinin-Redushkevich models in predicting the adsorption isotherm of  $\text{CO}_2$  and  $\text{CH}_4$ . The results were in consistency with other research findings reported elsewhere (Bi et al., 2017; Chi et al., 2019).

## 7. Experimental study

The experimental studies of EGR are based on multi-disciplinary settings that generates data for adsorption isotherm, kinetic and thermodynamic models. The settings of these experiments considers different parameters that facilitate the modeling and simulations (Wang et al., 2020a). Huo P et al. (Huo et al., 2017) experimented on the Pore structure analysis, adsorption tests and displacement experiment of natural gas due to the injection of  $\text{CO}_2$ . The results suggest that the  $\text{CH}_4$  could easily be displaced by the injection of  $\text{CO}_2$  due to superior competitive adsorption behaviors (Berawala and Andersen, 2019). Monitoring of injection pressure among other parameters such as pore structures, minerals composition, TOC content, ensures that adsorption of  $\text{CO}_2$  for sequestration and  $\text{CH}_4$  desorption for gas recovery can simultaneously occur with maximum certainty (Du et al., 2020b).

Liu et al. (2017b) reported the experimental set up investigating fluid interactions involving the indigenous methane adsorbed on the pore surfaces and the injected  $\text{CO}_2$  using sophisticated NMR method. They

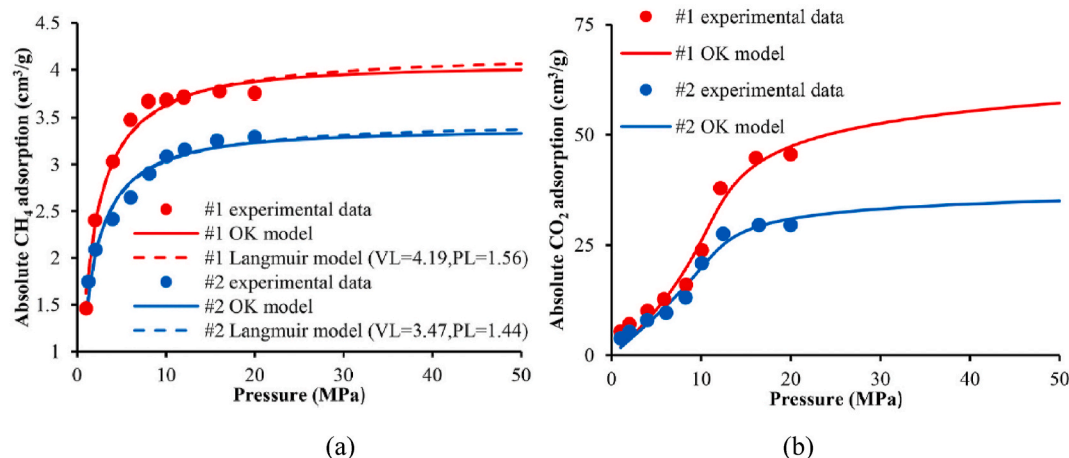


Fig. 9. Langmuir isotherm and Ono-Kondo models for (a) CH<sub>4</sub> and (b) CO<sub>2</sub> on Longmaxi samples (Zhang et al., 2021).

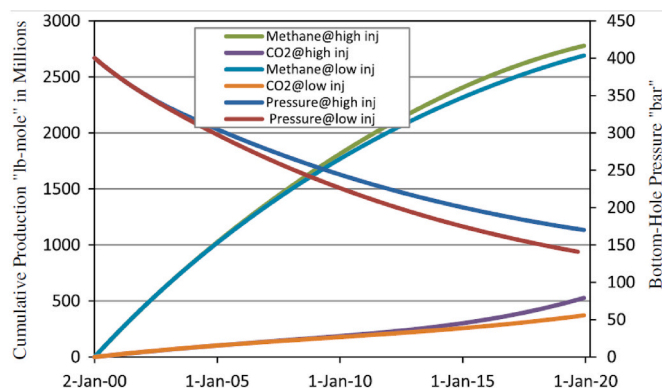


Fig. 10. Effects of CO<sub>2</sub> injection rate to the BHP and total gas produced (Khan et al., 2013).

were based on the low-field NMR measurement method that principally involves the use of magnetic behaviour of <sup>1</sup>H in the molecule that resonate at a specific frequency. The method allows the structural elucidation of a molecule based on the different chemical environment experienced by the hydrogen atoms. In the reservoir, water and methane are only molecules that will exhibit NMR response by flipping upon exposure to appropriate radio frequency, while CO<sub>2</sub> is unaffected making it easier for the study of CO<sub>2</sub> induced displacement of CH<sub>4</sub>. It was concluded that the recovery efficiency of methane increases with the injection of CO<sub>2</sub> and the NMR method is effective in the analysis of CH<sub>4</sub> displacement induced by the CO<sub>2</sub> injection.

Eliebid et al. (2018a) investigated the effect of mono-phased CO<sub>2</sub> injection and a binary phase of CO<sub>2</sub> (10 vol%) and CH<sub>4</sub> on CH<sub>4</sub> adsorption tendency on Pink Desert carbonate reservoir. According to the analysis of several adsorption isotherms, pure CO<sub>2</sub> injection overcomes CH<sub>4</sub> adsorption and contributes significantly to total gas recovery. Zhang et al. (Zhang and Ranjith, 2019). conducted an experimental on core flooding tests for CO<sub>2</sub>/CH<sub>4</sub> and volumetric strain measurement. Their research study attempted to determine not only the variance in CO<sub>2</sub> injection pressures and CH<sub>4</sub> production, but also the variation in coal seam swelling and shrinkage behaviour during CO<sub>2</sub> injection and CH<sub>4</sub> production. The results in Table 4, revealed that CH<sub>4</sub> recovery rate increase with the rise in CO<sub>2</sub> injection pressure through competitive adsorption. Similar isotherm adsorption experiments were conducted by (Du et al., 2020a) and found that CO<sub>2</sub> adsorption in shale formation increases while the adsorption of CH<sub>4</sub> decreases.

Table 4

The rate of CH<sub>4</sub> production and CO<sub>2</sub> sequestered (Zhang and Ranjith, 2019).

CO <sub>2</sub> injection pressure (MPa)	Initial CH <sub>4</sub> available (m <sup>3</sup> /t)	CH <sub>4</sub> remained after CO <sub>2</sub> flooding (m <sup>3</sup> /t)	CH <sub>4</sub> recovery rate (%)	Sequestered CO <sub>2</sub> after flooding (m <sup>3</sup> /t)
-	18.53	8.94	51.73	-
6	18.51	0.63	96.57	37.25
7	18.37	0.41	97.79	38.02
8	18.46	0.06	99.68	39.88
9	18.46	0	100	40.89
10	18.50	0	100	41.62

## 8. Modeling and simulation of EGR

For successful injection of CO<sub>2</sub> for EGR process, numerous detailed numerical modeling describing the fluid flow in porous media have to be conducted for the accurate reservoirs prediction and optimizations (Ajayi et al., 2019). Several researchers have reported their findings by using various simulations software to create 3-dimensional models to study the dual processes of CO<sub>2</sub> injection and CH<sub>4</sub> production.

(Liu et al., 2017a) employed a dual permeability model to account for the fluid flow in the shale reservoir with fractured and the matrix pathways. The constant pressure injection and constant rate injection were considered during optimization to minimize the CO<sub>2</sub>-breakthrough and improve CH<sub>4</sub> production (Fig. 10).

In his model (Liu et al., 2020a), studied the gas recovery mechanism by thermal stimulation where increased heat induced gas desorption and loss of water through evaporation. The model was meant to study the multiphase flow in shales and how heat can enhance the gas recovery. Furthermore (Liu et al., 2016), studied long-lasting physicochemical aspects of CO<sub>2</sub> injected into the Yanchang shale of the Ordos basin in China using a GEM simulator as a simplified 2-D model. In this model, the interactions between CO<sub>2</sub>-water-rock reactions and gas adsorption were simulated to find out the mechanisms at which the CO<sub>2</sub> are trapped in shale gas reservoirs and the displacement of indigenous CH<sub>4</sub>. They concluded that two trapped mechanisms, namely supercritical and adsorbed phases account for short to medium term and mineral trapping account for long-term storage. Furthermore, gas displacement follows two stages depending on the concentration of injected CO<sub>2</sub>. Stage one allows only the build-up of pressure caused by the injection of CO<sub>2</sub> followed by stage two by which desorption of the CH<sub>4</sub> from the pore's surface occurs due to the competitive advantage of the CO<sub>2</sub> over CH<sub>4</sub> in adsorption process.

(Li and Elsworth, 2015) applied a dual porosity dual permeability model and the two injection modes of continuous and pulsed were



investigated. The model assumed that 1) An isothermal system exists where shale reservoir is characterized by a homogeneity, isotropy and elastic continuum. 2) An ideal gas in the reservoir with its viscosity being constant at isothermal conditions. 3) Darcy's law and Fick's law account for the flow of gases in the fractures and transport of gases in the shale matrix respectively, 4) Gas sorption only occurs within the matrix. It was found that the production mechanism and flow occur either in fracture or matrix permeable channels. Initially the CH<sub>4</sub> production is dominated by the fracture flow and later by matrix flow due to the exhaustion of the free and adsorbed gas on the fractures. Also, the continuous injection mode results into more CO<sub>2</sub> being sequestered in comparison to the pulsed injection mode (Sun et al., 2013). developed a new dual-porosity model and used COMSOL to simulate the effects of CO<sub>2</sub> injection in the shale gas reservoir and investigate the transport mechanism dominating the flow of binary components in porous media. Due to the variation of pore diameter, different transport mechanisms ranging from Knudsen diffusion, viscous and normal diffusion were observed. As illustrated in Fig. 11 (a), the CH<sub>4</sub> is continually being desorbed and displaced, and the continual sequester of CO<sub>2</sub> in the shale reservoir. On the other hand, Fig. 11 (b) indicates that variation in injection pressure for CO<sub>2</sub> produces tremendous change in the methane gas recovery. The increase in the CO<sub>2</sub> injection pressure in the reservoir elevates the concentration of CO<sub>2</sub> making the ratio of the rates of CO<sub>2</sub> injected to that rate of CH<sub>4</sub> production. The rise in injection pressure results into simultaneous increase in the CO<sub>2</sub> storage and the production of more CH<sub>4</sub> from the formation.

The application of dual porosity/dual permeability models to simulate the gas flow in the fractures was reportedly encountered by some setbacks where (1) they could not accurately model disconnected fractures and complex fractures, (2) transfer function between matrix and fractures cannot be assessed accurately. In an endeavor to overcome these limitations, a discrete fracture model (DFM) was invented where individual fractures are assigned a size, an orientation and a permeability to accurately monitor the effect of each one.

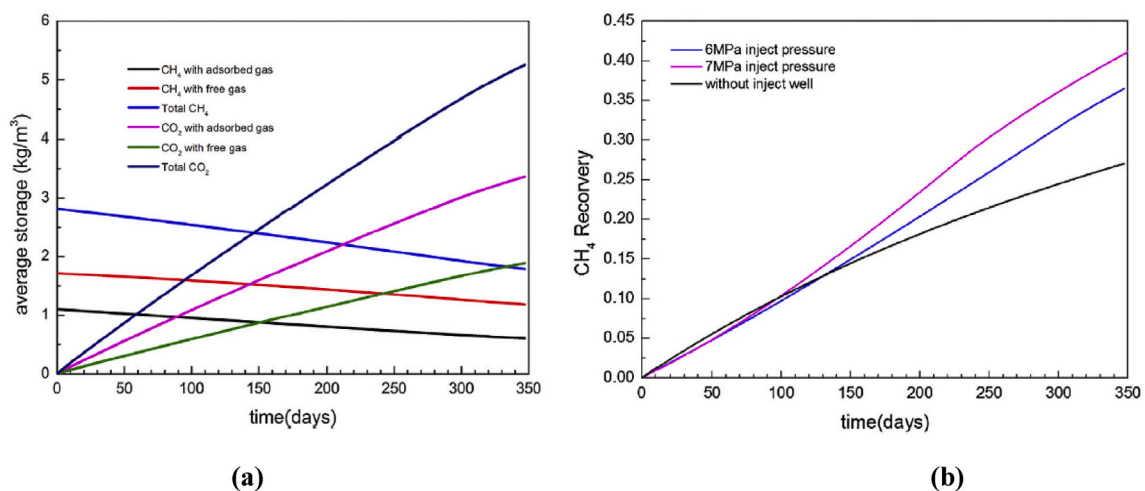
In his work (Zhan et al., 2021), the, CMG-GEM simulator was used to create a numerical model for assessing the feasibility of CO<sub>2</sub> sequestration in shales. The data from New Albany Shale and his consideration of continuous and pulse injection strategies were used to assess the reservoir and found that CO<sub>2</sub> sequestration in shale gas reservoirs can be executed. This is possible due to effective design of well spacing and effective hydraulic stimulation. (Zhang et al., 2021), applied a coupled method involving multiple interacting continua (MINC) model and discrete fracture model (DFM) model to describe the gas flow in the matrix and complex hydraulic fractures respectively. The use of these

unstructured models were also suggested elsewhere (Cheng et al., 2020). Furthermore, they investigated the adsorption isotherm of CO<sub>2</sub> and CH<sub>4</sub> on the longmaxi shale reservoir by gathering experimental data and then implicated the Langmuir and Ono-Kondo models where the Ono-Kondo exhibited a best fit for the adsorption prediction in shale reservoirs. However, due to the complexity of the grid and high computational cost of the DFN method, its wide application in the field is still restricted. Therefore, (Wan et al., 2020), used a modified method embedded discrete fracture model (EDFM) to simulate association of the dual processes of multistage hydraulic fracturing and production waning in the Marcellus shale gas reservoir. In this model, the effect of each fracture is incorporated and formulated to embed complex fracture geometry which was ignored in other models into a third-party reservoir simulator to simulate multi-lateral well production. Its computational proficiency and accuracy to model flow of gas in a reservoir characterized by intricate artificial and natural fractures has put it on the edge over other models. To represent gas adsorption on shales, several adsorption models have been applied, with varied degrees of success (Table 5). Due to the high reservoir pressure and temperature, multi-phase gas flow, the adsorption tendency excludes some of the common

**Table 5**

The comparison of adsorption models for EGR in shales.

Model used	Merits	Limitations	References
Langmuir	<ul style="list-style-type: none"> <li>• It's easy to use</li> <li>• Best fit for monolayer adsorption</li> </ul>	<ul style="list-style-type: none"> <li>• Inapplicable in multicomponent mixtures</li> <li>• Not suitable for adsorption at high pressures and temperature</li> </ul>	(Alafnan et al., 2021), (Merey and Sinayuc, 2016)
Ono-Kondo	<ul style="list-style-type: none"> <li>• Explain adsorption in multilayers.</li> <li>• Apply in adsorption data across a wide temperatures and pressures ranges</li> </ul>	<ul style="list-style-type: none"> <li>• Not the best model for subcritical CO<sub>2</sub></li> </ul>	(Zhou et al., 2019b), (Rani et al., 2019b), (Merey and Sinayuc, 2016)
Dubinini-Astakhov (D-A) Dubinin-Radushkevich D-R	<ul style="list-style-type: none"> <li>• Best model for subcritical CO<sub>2</sub></li> <li>• Best for adsorption in microporous solids</li> </ul>	<ul style="list-style-type: none"> <li>• Does not provide the best fit for supercritical CO<sub>2</sub></li> <li>Its equation cannot revert to Henry's law when the pressure drops to zero.</li> </ul>	(Zhou et al., 2019a), (Rani et al., 2019b)



**Fig. 11.** (a) Rate of storage and production disparity of CH<sub>4</sub> and CO<sub>2</sub> per unit volume, (b) CH<sub>4</sub> recovery over time for different production scenarios (Sun et al., 2013).

isotherms such as Langmuir. Detailed strengths and limitations can be covered in fundamental work of suggested references on Table 5.

## 9. Current non-shales-EGR field tests

Although numerous theoretical, laboratory experimental data and simulations have been collected, there are limited field applications of CO<sub>2</sub>-EGR method. The CO<sub>2</sub> has been injected in (1) a partially depleted gas reservoir offshore of the Netherlands (Vandeweyer et al., 2011) and (2) a Canadian onshore depleted gas reservoir. Some findings have suggested the injection of CO<sub>2</sub> from the onset of the gas field operation for efficient gas recovery. Vandeweyer et al. (2011) applied modern tools to assess well quality using multi-finger imaging tools, cement bond log, down hole video log, electromagnetic imaging tool and gamma rays. The inner radii and the fault of the casing was determined by a combination of multi-finger imaging (MFI) tools and the electromagnetic imaging (EMI) tool where the MFI tool impaired by the deposition of salts and EMI can overcome that barrier thereby producing accurate data. The data generated from these measurements and the CO<sub>2</sub> flow exhibited no reservoir failure to store the gas.

In another endeavor, Urosevic et al. (2011) in Otway Basin Pilot Project reported the CO<sub>2</sub> sequestration monitoring technique using seismic data. Their intents were to ascertain the permanent storage of CO<sub>2</sub> and the potential risk of leakage over years. The land seismic 3D survey and borehole seismic surveys were employed to produce time lapse data which indicated very minor changes which cannot jeopardize the storage of gas.

Connell et al. (2014) conducted a CO<sub>2</sub>-ECBM field test in China's Shanxi basin where several horizontal wells were used for injection, production and monitoring processes of the CO<sub>2</sub>. The amount of CO<sub>2</sub> injected into the reservoir for this trial was about to 460 tonnes for the duration of 196 days. The gas migration in the reservoir was observed using the installed u-tube and water sampling system where reservoir fluids (displaced methane and injected CO<sub>2</sub>) were analysed to determine their extent of flow in the poor permeable coal seams. The monitoring was supported by the tracers injected at the onset and end of the time of CO<sub>2</sub> injection. In contrast to the other studies, it was revealed that the rate of injectivity did not show the decreasing trend presumably because of gas behaviour when flowing in the lengthy horizontal well and insufficient injection time for CO<sub>2</sub>.

(Kiel, 2011) investigated the Altmark gas field project in Germany for the potential enhancement of gas recovery and sequestration of CO<sub>2</sub>. Wellbore integrity, geological processes, and reservoir monitoring activities were conducted to ensure the gas storage doesn't risk the environment and the natural gas can be recovered. Similarly (Jenkins et al., 2012), used CO<sub>2</sub>-CRC Otway Project, in Australia for assessing the storage safety and effective monitoring of the processes to ensure compliance and generate enough data for other new projects. The findings were targeting to unlock the social, political and scientific barriers over the sequestration processes. By applying test models and simulations, they were able to justify that the CO<sub>2</sub> storage in depleted gas reservoirs can be safe and effective. The underground contamination of the soil and water were monitored by time-lapse seismic survey where CO<sub>2</sub> plume before and after the injection were recorded any analysed. Their interpretation did not reveal any caprock leakage that can put the project at suspicious (Hannis et al., 2017). The EGR field trials presented are from sandstone and CBM reservoirs, however, the field data for shales are still undisclosed. The available trails are meant to encourage the researchers and other stakeholders that CO<sub>2</sub>-EGR are technically possible and more researches on the unconventional are needed.

## 10. Conclusion and recommendations for future study

The combination of theoretical data, laboratory discoveries, and reservoir modeling results has demonstrated that the simultaneous CO<sub>2</sub> injection and CH<sub>4</sub> recovery operations are technically and economically

feasible. That is possible if dispersion can be avoided by good reservoir management methods and production control measures are followed. Field tested EGR processes are very scarce due to the high cost associated with the capture, purification and injection of CO<sub>2</sub>. On top of that, the fear of excessive mixing of methane with the injected gas which potentially minimize its quality demanding an expensive separation technique which inflates the cost of CH<sub>4</sub> production. More rigorous study into CO<sub>2</sub> capture and purification systems that are both inexpensive and reliable must be developed. Membrane technology, adsorption, cryogenic capture, chemical, and physical approaches are all still prohibitively expensive. On the other hand, dispersion of injected CO<sub>2</sub> and resident CH<sub>4</sub> in the reservoir can be partly be minimized by optimizing the injection rate, pressure, strategy and detailed understanding of the flow regimes. Also, the improved modeling and simulations methods can mitigate some of the limitations.

1. It was shown that the injection of CO<sub>2</sub>-N<sub>2</sub> mixture at the ratio of 1:1 enhance the displacement of natural gas by the almost 89% efficiency as compared to the same for N<sub>2</sub> or CO<sub>2</sub> alone. This was attributed to the synergistic effect of the two gases.
2. The experimental findings derived from the separate injection of CO<sub>2</sub> and N<sub>2</sub> into the shale reservoirs followed by the pressure reduction increases the CH<sub>4</sub> recovery. However, CO<sub>2</sub> injection produce more methane due to its ability to lower partial pressure and inducing desorption and the ability of the CO<sub>2</sub> to outweigh the adsorption of CH<sub>4</sub> from the pore surface.
3. The experimental findings on CO<sub>2</sub> injection in shales have shown that, the higher CO<sub>2</sub>/CH<sub>4</sub> fraction during the injection, the huge displacement of CH<sub>4</sub> is expected. The higher concentration of injected gas is expected to interact with the reservoir during the early stages and free gas in the pores and fractures is immediately displaced. The recovery efficiency of about 25% will be achieved but with the increased fracture half-length. Based on experimental data and adsorption isotherms, preferential adsorption of CO<sub>2</sub> over CH<sub>4</sub> in shales surfaces allows faster and simple adsorption which displace the indigenous natural gas from the pore spaces. Desorption of methane occurs as a result of competitive adsorption/desorption mechanism making certain for the sequestration and EGR.
4. The findings from shales and coals experiments and simulations for CO<sub>2</sub>-EGR have indicated that there is a rapid decline in the natural gas production after its peak production within few years. This is attributed to the clay swelling that impairs gas flow due to permeability loss and injectivity.
5. Regardless of the promising experimental and modeling results, the EGR field trials are still very scarce presumably because of uncertainties of the reservoir parameters, injection parameters and the costs associated with the capture, purification and re-injection of the CO<sub>2</sub>. The injected gas must be pure enough to induce significant effects in the phase change to supercritical and allow smooth interaction with the formation organic and inorganic components. The success of the EGR on the field demonstration largely depends on the invented techniques to lower the formation swelling and injectivity induced by CO<sub>2</sub> and ensure to safeguard the environmental issues.

## Data availability statement

Few data were analysed in the manuscript and no datasets were generated during this study.

## 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.

## Acknowledgement

The authors would like to acknowledge the support from the China University of Geosciences, Wuhan, China.

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