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Review on Carbon Capture, Utilization, and Storage for Enhancing Gas Recovery

Ghamdan AL-khulaidi, Yankun Sun,* Ahmed.G. Alareqi, AL-Wesabi Ibrahim, Abubakar Magaji, and Xu Zhang

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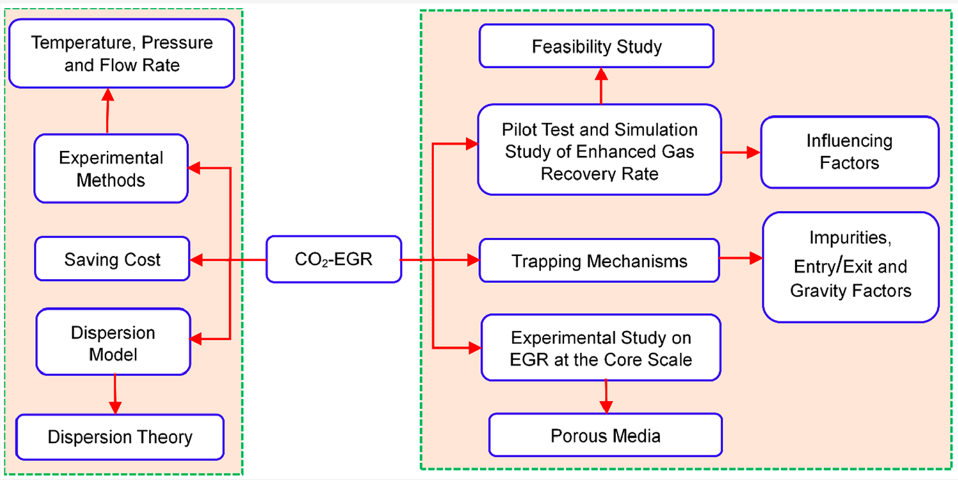

ABSTRACT: The escalating levels of carbon dioxide emissions (CO_2) and the detrimental consequences of global warming have spurred extensive research into identifying secure and reliable storage sites with ample capacity. Depleted gas reservoirs emerge as a promising option for CO_2 sequestration, solidifying their position as a viable carbon sink. These conventional or unconventional reservoirs retain substantial pore space after natural gas extraction and depressurization. Furthermore, their impermeable top layers ensure the long-term containment of hydrocarbons, enhancing the safety of this choice. Consequently, the cost of the process can be reduced through the incremental recapture of excess gas after carbon dioxide (CO_2) injection. This article is a comprehensive review of multiple published papers exploring the enhancement of shale gas recovery through CO_2 injection. It aims to present a thorough understanding of the concept of this technology, highlighting its benefits and drawbacks, comparing existing studies, and encouraging further research into the CO_2-EGR principle.

1. INTRODUCTION

High levels of CO_2 emissions from diverse industrial activities pose a significant risk to the Earth's ecosystem and climate system, which is already fragile.[1−4] Industrial use of fossil fuels like coal and oil has significantly grown due to evolving lifestyles and fast growth, leading to a continuous rise in CO_2 emissions annually.[5−9] Figure 1. It reported in 2022 that China released 11,472 million metric tons of CO_2 into the atmosphere. The US released 5,007 million metric tons, followed by India with 2,710 million metric tons, Russia with 1,756 million metric tons, and Japan with 1,067 million metric tons.[10] Nevertheless, it is worth noting that the atmospheric concentration of carbon dioxide (CO_2) has surpassed 410 ppm (ppm), a level that has not been observed in the past 800,000 years.[11,12] Table 1 presents a comprehensive overview of carbon dioxide (CO_2) emissions from diverse industrial sectors. To significantly reduce CO_2 emissions, the Chinese government has put forward a set of policies to tackle the current high emission levels. These include promoting coal consumption substitution and upgrading, actively developing the new energy industry, and implementing CCUS (Carbon Capture, Utilization, and Storage) to reach carbon peaking by 2030 and zero carbon emissions by 2060.[13−18] One effective solution to decrease CO_2 emissions involves storing CO_2 in

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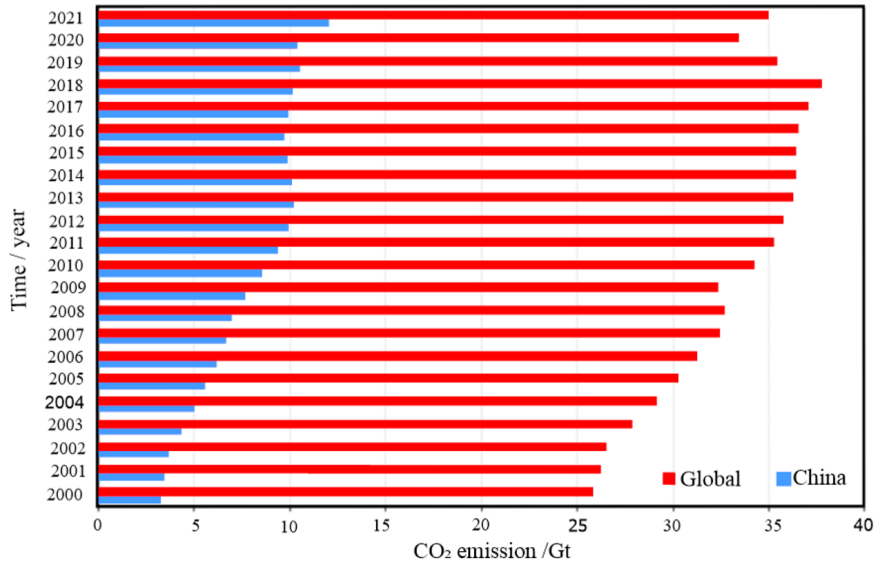

Figure 1. Yearly carbon dioxide emissions globally, specifically in China. Adapted from ref 28. Copyright 2023, with permission from Elsevier.

Table 1. Carbon Dioxide Emissions Originate from Diverse Industrial Sectors[a]

industrial sector	total emission (Mt CO_2/Yr)
Power plant	>10,000
Cement	>1000
Oil refineries	900
Iron and steel	900
Petrochemicals	155−300
Ammonia	160
LNG sweetening	160
Ethylene oxide	25−30

[a]Modified from ref 35.

depleted oil and gas reservoirs.[19−24] Compared to fossil fuels, shale gas is a renewable and practical energy resource that helps minimize the world's reliance on high-energy consumption and polluting output resources while providing additional alternatives for decreasing air pollutants and greenhouse emissions.[25] Shale is helpful in CO_2-EGR because it preferentially absorbs carbon dioxide over methane.[26,27]

The utilization of enhanced gas recovery (EGR) is an intriguing approach whereby carbon dioxide (CO_2) is injected into an exhausted oil or gas reservoir.[29] This injection augments the reservoir's pressure, facilitating the extraction of reserves that would otherwise remain unrecoverable.[30] Due to the depletion of conventional energy sources and the rising global energy demand caused by population growth and increased energy consumption rates. Petroleum and gas companies are increasingly exploring and exploiting unconventional gas resources like shale gas, tight gas, and coal seam gas.[31] These unconventional resources have poor permeability, requiring complex extraction processes to make them usable. Gas extraction from these reservoirs mainly involves using horizontal drilling and hydraulic fracturing to create artificial cracks, significantly improving gas recovery.[32] Many researchers have shown a strong interest in carbon dioxide (CO_2) because of its excellent performance in surface adsorption and its potential for reducing greenhouse gases. Carbon dioxide is more attractive to adsorption sites in the formation, allowing it to replace methane by efficiently adsorbing onto these sites and filling interstitial gaps. Figure 2 demonstrates the selective absorption of CO_2 in gas shale reservoirs, aiding in releasing

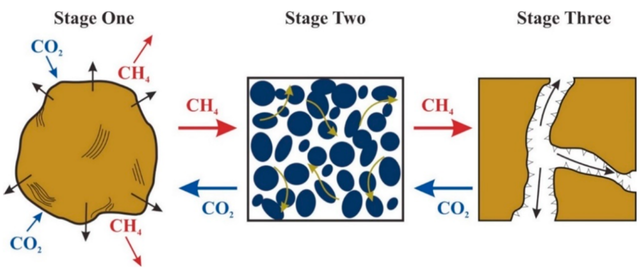

Figure 2. Diagram showing gas shale carbon dioxide and CH_4 flow dynamics. Adapted from ref 42. Copyright 2014, with permission from Elsevier.

methane and showcasing the active interchange of CO_2 and methane inside the shale matrix.[33] Furthermore, source rock capping identified as an effective measure to mitigate CO_2 leakage induced by capillary forces. Therefore, it is acceptable to store CO_2 in a depleted gas reservoir.[34]

The research carried out by ref 33 explores the effects of supercritical CO_2-EGR on shale gas reservoirs, placing its results within the broader context of scientific discussion on the subject. An analysis was conducted by ref 36 by measuring alterations in shale's splitting modulus and adsorbed energy, and this supports the theory of improved recovery via induced fractures. The systematic study conducted by refs 37−39 expands on the discussion by explaining the various mechanisms of CO_2-EGR in shale, such as swelling, viscosity reduction, and competitive adsorption also this provides a theoretical basis for the observations made by ref 33 regarding changes in shale properties. The findings from ref 36 regarding SC−CO_2-based drilling emphasis the practical implications of utilizing SC−CO_2 in underbalanced drilling situations, in line with ref 33, focus on the operational aspects of shale gas extraction. In addition, the economic analysis conducted by ref 40 presents a crucial viewpoint on the cost-effectiveness of SC−CO_2 applications, emphasizing the importance of evaluating economic feasibility in discussions by ref 33. Lastly,

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Table 2. Recent Reviewed Studies on ($CCUS/CO_2$-EGR)

number	publication year	topic reviewed	refs
1	2022	The significance of Carbon Capture and Storage (CCS)	Hameli et al.[47]
2	2021	Study methane (CH_4) and carbon dioxide (CO_2) adsorption properties on Longmaxi shale.	Xie et al.[48]
3	2020	Emphasizes enhancing the CO_2-EGR safety and cost-effectiveness.	Cao et al.[49]
4	2022	Explore CO_2 capture with South African coals, emphasizing adsorption mechanisms and efficiency.	Mabuza et al.[50]
5	2023	Examine the influence of coal pores on gas behavior and structure, emphasizing nitrogen use to enhance methane extraction.	Ji et al.[51]
6	2023	Explore the impact of coal's pore structure on methane storage, focusing on the importance of coal quality.	Li et al.[52]
7	2022	Injecting CO_2 into gas reservoirs to reduce emissions and boost output.	Jukić et al.[53]
8	2022	Enhance storage efficiency by optimizing the trapping of CO_2 in saline aquifers.	Mwakipunda et al.[54]
9	2023	Adsorption of gas in nanosized pores inside shale.	Zhang et al.[55]
10	2023	Hydraulic fracturing techniques to improve the efficiency of gas recovery.	Yu et al.[56]
11	2023	Enhance CCUS technology for carbon reduction objectives, including storage, retrieval, and hydrogen incorporation.	Li et al.[57]

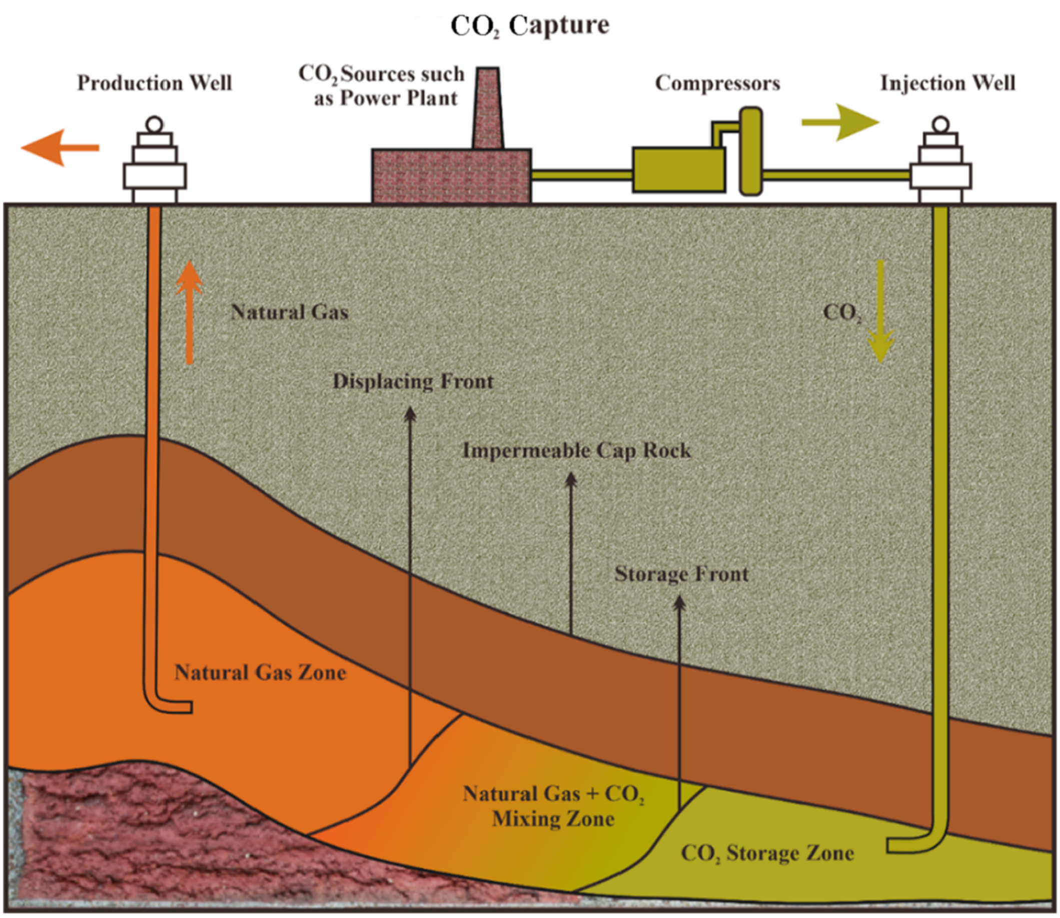

Figure 3. Contains the process schematic diagram. Adapted from ref 34. Copyright 2022, with permission from Elsevier.

the review by ref 41 regarding the competitive adsorption of CO_2 and CH_4 delves into the intricate molecular interactions within shale matrices, providing a thorough comprehension that aligns with ref 33 advocacy for combined experimental and simulation investigations. These discussions highlight the significant impact of $SC-CO_2$ in shale gas recovery and emphasize the importance of balancing technological progress, economic factors, and thorough research in managing shale reservoirs.

Insights from refs 33, 36−39, and 41 were used as examples in ref 33 work to show how supercritical CO_2 affects shale gas reservoirs, especially when it comes to enhanced gas recovery (EGR).

This engagement with external research underscores the multifaceted influences of supercritical CO_2, ranging from geochemical interactions altering reservoir properties to economic considerations of CO_2-EGR implementations. Through these discussions, the collective body of work not only underscores the transformative potential of CO_2-EGR in shale gas recovery but also delineates the intricate balance between technological advancement, economic considerations,

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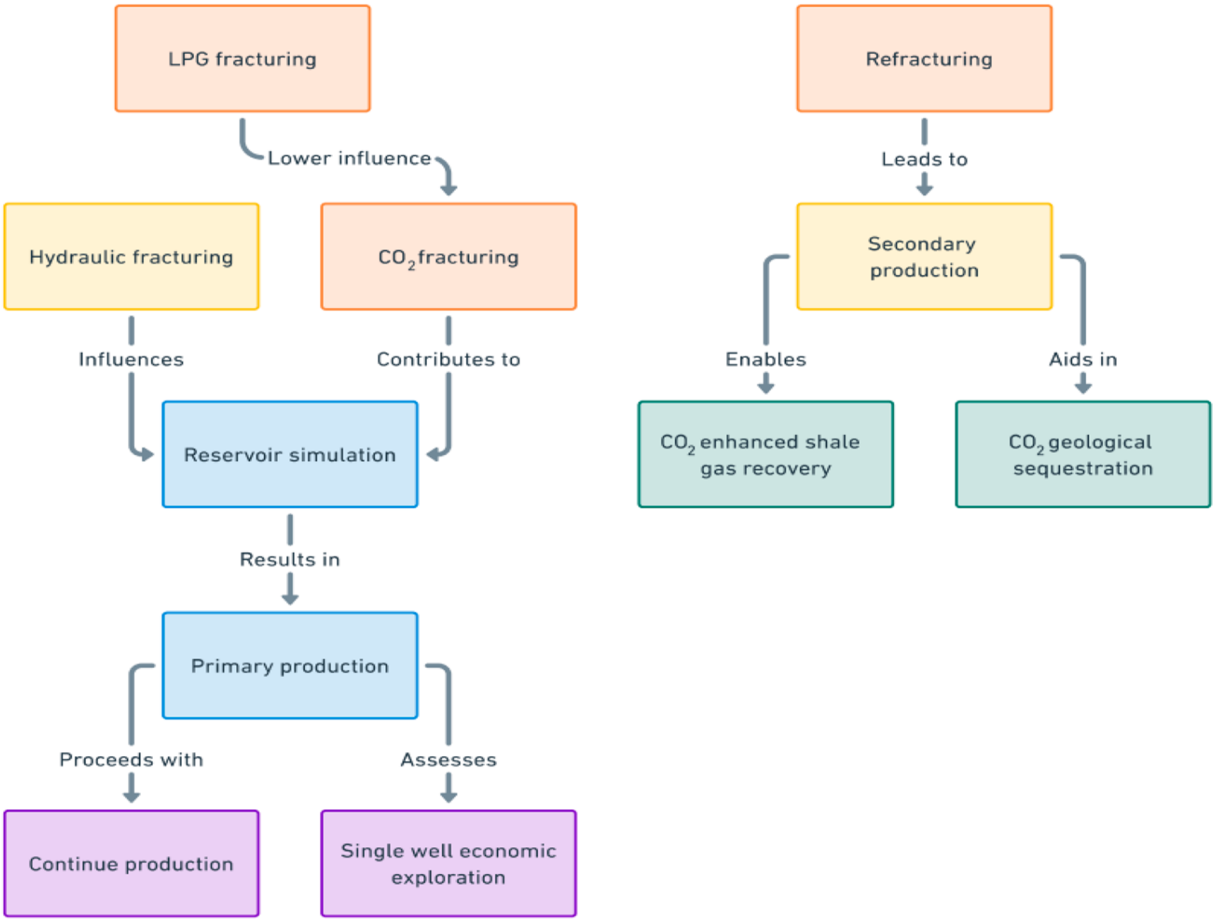

Figure 4. Technology's process diagram for CO_2-EGR.

and the need for comprehensive research methodologies to navigate the complexities of shale reservoir management.

Researchers shows that storing carbon dioxide (CO_2) in exhausting gas reservoirs is a feasible method to reduce emissions and improve gas recovery.[43−46] However, many studies do not adequately investigate the technical challenges, economic impacts, environmental effects, and comparisons with other renewable energy alternatives. Current reviews of CCUS-CO_2/EGR given in Table 2. It is essential to fill these information gaps to fully evaluate current emission reduction methods' effectiveness and long-term viability. Although carbon capture, utilization, and storage (CCUS) technologies have advanced, concerns persist over their full potential for increased gas recovery. Otherwise, Worries about the economic sustainability, reservoir effects, and comparisons with other recovery techniques continue. To understand CO_2/CH_4 adsorption in coal and shale and the impact of supercritical CO_2, sophisticated prediction models and collaborative experimental-computational work are necessary. This article is an overviews carbon dioxide-enhanced gas recovery (CO_2-EGR), focusing on essential research and real-world uses. The study investigates the dynamics of CO_2 and CH_4 adsorption, necessary for enhancing CCS efficiency, and evaluates worldwide trends and research gaps in CO_2-EGR. By comparing many studies, this research offers insights into the efficiency of CO_2 displacement and adds to the broader conversation on CCS. It highlights the need for comprehensive academic and practical assessment in carbon management.

2. CONCEPT OF CO_2-EGR

The CO_2-EGR technique is a novel and promising technique that uses the increased affinity of carbon dioxide to methane in shale to boost methane production of desorption from the deep to stimulate the formation of the shale reservoirs simultaneously, long-term CO_2 storage in shale to contribute to CO_2 mitigation to address global warming. Whenever a shale gas extraction well approaches the economic potential consumption barrier, raised recovery measures, refracturing, and injecting nitrogen dioxide, carbon dioxide, or a mixture of gases must stimulate gas adsorption in the geological structure. It is worth noting that this CO_2-based technique is superior to other technologies because it absorbs CO_2 higher than methane. The idea of this technique mentioned in Figure 3. The flowchart of CO_2-EGR technology shown in Figure 4. CO_2 has diverse functions in various stages of CO_2-EGR.

Figure 4 illustrates the techniques of fracturing and their specific functions in the oil and gas production process. The process starts with LPG fracturing, which has little impact, then moves on to hydraulic fracturing, which considerably affects reservoir modeling. CO_2 fracturing aids in reservoir modeling and improves shale gas recovery. Reservoir modeling plays a crucial role in guiding primary production choices, determining whether production sustained or a well's economic feasibility evaluated. Refracturing seen as a forerunner to secondary production, facilitated by CO_2-improved recovery and supportive of CO_2 geological sequestration, demonstrating the interdependence of produc-

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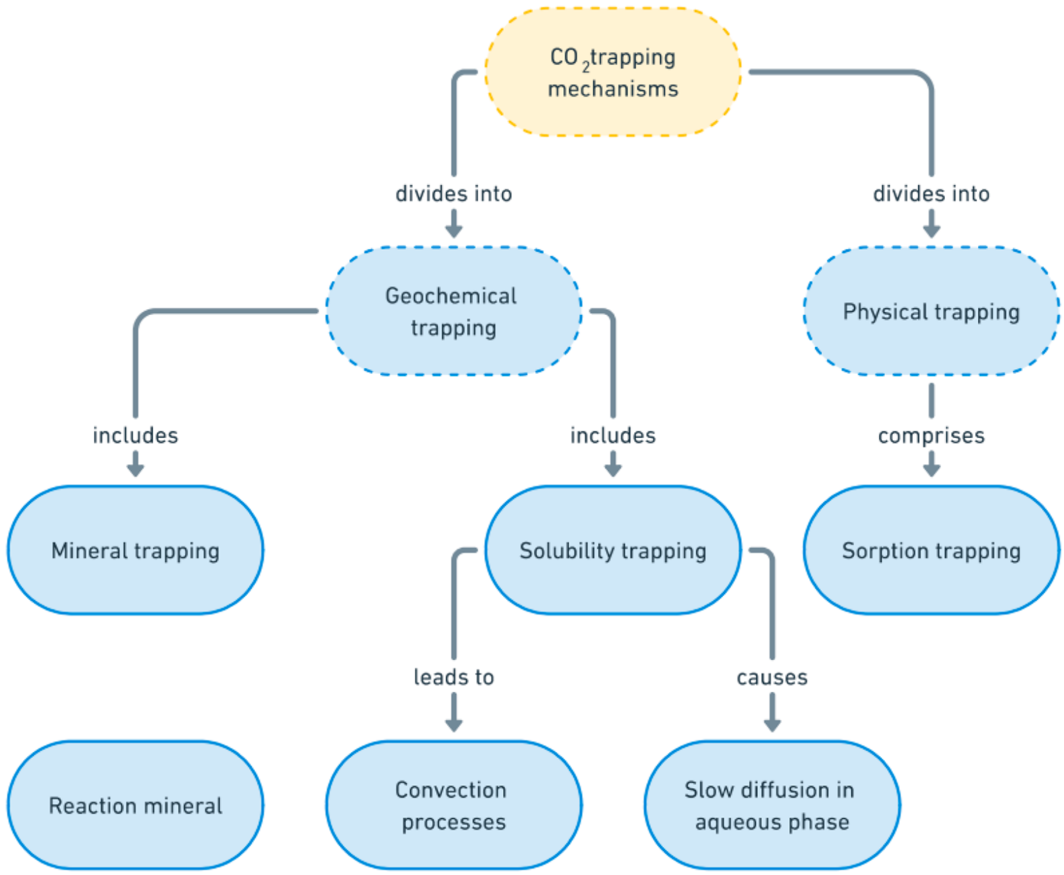

Figure 5. Different CO_2 trapping mechanisms during the geological storage processes.

tion improvement and environmental conservation in the business.

Supercritical CO_2 effectively sequestered in subterranean formations through two principal mechanisms: physical and geochemical trapping, as delineated in Figure 5.

This research explores the many CO_2 trapping mechanisms essential for the sequestration process, crucial for reducing atmospheric CO_2 levels and addressing climate change. Trapping methods classified into two main pathways: geochemical and physical trapping. Geochemical trapping involves the conversion of CO_2 by chemical interactions with minerals into stable carbonates, leading to the permanent storage of CO2 as reaction minerals. Physical trapping can divided into solubility trapping, where CO_2 dissolves in aqueous solutions, causing convection processes in the geological substrate, and sorption trapping, where CO_2 is adsorbed onto solid material surfaces, leading to slow diffusion in the aqueous phase. The processes explain the many techniques of CO_2 sequestration, each having specific consequences in terms of time and environment, which are crucial for developing efficient carbon capture and storage (CCS) technology.

The efficacy of the storage process is determined by a confluence of trapping mechanisms that are used to guarantee prolonged storage.[58] Compared to deeper saline aquifers, five primary methods for storing carbon dioxide in shale -reservoirs are as follows:

(1) Hydrodynamic entrapment, where carbon dioxide is buoyant, stays unable to move yet unable to rise to the surface because of shale's limited permeability.

(2) Adsorption trapping is when CO_2 is adsorbed onto the host rock's surface, causing the existing methane (CH4) to be desorbed.

(3) Dissolving carbon dioxide solutions trapped in formation water;

(4) Mineral entrapment occurs when CO_2 is geochemically bonded on the rock due to mineral precipitation.

(5) The remaining CO_2 phase is converted into an immobile portion, resulting in capillary entrapment.

Upon injecting CO_2 into shale formations, numerous CO_2 molecules vie for methane displacement from the adsorbent surfaces. Simultaneously, other CO_2 molecules seek out available surface sites. A pivotal mechanism that amplifies methane emissions and storage after CO_2 injection in shale is the adsorption process, notable for its intense competition between carbon dioxide and CH_4 for surface adsorption. This phenomenon arises from the adsorption phase, where a substantial portion of gas within the shale, as indicated by ref 59, is stored, typically ranging from 20% to 85%. It is worth noting that while elevating pressure within the natural gas reservoir can mitigate power losses, it may concurrently lead to diminished methane recovery and decreased CO_2 sequestration, as demonstrated by ref 60. Multiple research studies have consistently shown that shale formations exhibit a greater capacity for absorbing CO_2 than CH_4. In various reservoir conditions, it has been established that the absorption of CO_2 surpasses that of CH_4 by a significant margin, with absorption rates ranging from 2 to 10 times higher, as documented by refs 61 and 62 (Figure 6).

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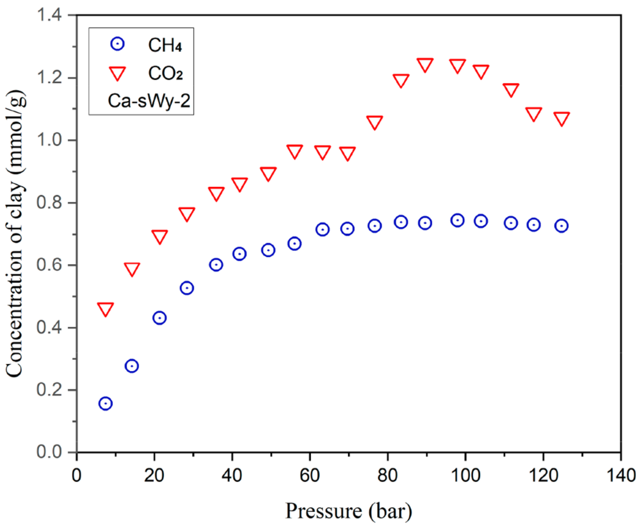

Figure 6. Sorption capacities for CO_2 and CH_4 on calcium montmorillonite (Ca-SWy-2) at 50C and 90 bar. Adapted from ref 63. Copyright 2020, with permission from Elsevier.

The concept of (CO_2-EGR) has been a topic of study and discussion for numerous years.[64,56] Nevertheless, the efficacy of this concept has yet to evaluate in practical applications. This phenomenon ascribed to two primary factors. Notably, CO_2 remains a costly resource, and the implementation of geological carbon storage techniques has not extensively adopted yet. Another factor of concern is the potential for excessive mixing between the carbon dioxide injected and the leading shale gas, which may diminish the value of natural gas resources.[65] Although shale gas and carbon dioxide can mixed at any pressure, the CH_4−CO_2 system exhibits noteworthy properties supporting the case for enhancing gas recovery. In brief, these characteristics are as follows:

(1) In reservoir circumstances, the density of CO_2 is much higher than that of methane (CH_4), with a difference factor between 2 and 10. This significant density disparity helps promote consistent displacement, influenced mainly by gravitational forces.

(2) The high solubility of CO_2 in formation water compared to CH_4 delays CO_2 breakthrough, improving the displacement process efficiency.

(3) Comparing the mobility ratios of CO_2 and CH_4 shows that due to CO_2's greater viscosity, it has a lower mobility ratio, indicating a more steady displacement process. The low viscosity of supercritical CO_2, similar to a gas, allows for easy injection into the geological formation, making it a strong candidate for improved gas recovery operations.

3. ADVANTAGES, DISADVANTAGES, AND FEASIBILITY OF CO_2-EGR

3.1. CO_2 Fracturing. Hydraulic fracturing has transformed the worldwide energy landscape, becoming the favored method for obtaining natural gas from unconventional shale deposits.[36] Within the context of low permeability reservoirs, it becomes apparent that only a few shales endowed with inherent fractures are readily employed for production. In contrast, the vast majority, constituting approximately 90% or even more, necessitates applying fracturing techniques to enhance the flow conductivity near the wellbore. Hydraulic fracturing is the predominant method in the oil and gas industry for bolstering the extraction of hydrocarbon resources. According to statistical findings, hydraulic fracturing was pivotal in rejuvenating nearly 60% of the world's oil and gas wells in 2010, as ref 66 reported. In addition to hydraulic fracturing, the utilization of liquefied petroleum gas fracturing (LPG) and carbon dioxide (CO_2) fracturing techniques has emerged as viable approaches to stimulate low-permeability reservoirs. For a comprehensive assessment of the advantages and disadvantages of these different fracturing methods, please refer to Table 3.

While widely utilized, hydraulic fracturing linked to considerable water consumption and substantial environmental contamination. Furthermore, the expense of LPG fracturing is prohibitively high. Moreover, the cost of LPG fracturing is exorbitantly high. However, the benefits of CO_2-fracturing seem to be comparatively more favorable. CO_2 fracturing primarily encompasses dry CO_2 fracturing and CO_2 foam fracturing. The process of CO_2 foam fracturing involves the combination of liquid carbon dioxide (CO_2) with a traditional water-based fracturing fluid in specific proportions. The result is creating a durable foam system characterized by the gas being enclosed within and liquid on the outside, specifically developed for use in reservoir stimulation projects.

Dry-CO_2 fracturing involves using supercritical or liquid CO_2 as the fracturing fluid to enhance the conductivity of the pool. Utilizing CO_2 fracturing has the potential to decrease the pressure required to achieve rock fragmentation and improve drilling efficiency. According to ref 67, pressure barrier of a CO_2 jet on marble is just 2/3 of a water jet, and it is much lower, at half or less, when applied to shale.

In the realm of wellbore operations, supercritical CO_2 presents a distinct advantage in its low viscosity. This property facilitates the efficient transmission of pressure within the wellbore, leading to a reduction in fracture pressure and the creation of intricate microfractures. Additionally, it effectively alleviates issues related to water sensitivity and water lock damage, often prevalent in reservoirs with high clay content. Moreover, supercritical CO_2 demonstrates a favorable cleaning effect on the supporting fracture diversion bed. Furthermore, CO_2 fracturing exhibits superior cost-effectiveness in oper-

Table 3. Comparative Examination of Various Techniques Used in the Stimulation of Shale Formations

shale stimulation technology	advantages	disadvantages
Hydro-fracturing	It has reached a high level of maturity technology and is extensively used.	The procedure involves high water use, possible harm to reservoir stability, probable fluid leakage, and generated earthquakes.
LPG fracturing	Enhance shale gas recovery without the need for water consumption or the occurrence of fracturing fluid leaks.	Combustible, explosive, and hazardous to health. The expense of storing before fracturing is rather high.
CO_2 fracturing	It enhances the recovery of shale gas while minimizing water usage and reservoir degradation.	Fracturing adoption is limited by substantial filtration losses of fracturing fluid and constraints on the size of operations.

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Table 4. Comparing Stimuli from Shale Cost Comparison between CO_2- and Hydro-Fracturing Methods

shale stimulation technology	price of hydro-fracturing fluid and CO_2	operating pressure (power: 100 kW, diameter: 50 mm)	equipment power requirement	environmental governance costs
Hydro-fracturing	116−166\$/t	≥124 MPa	Relatively high	High costs for back flow water disposal and treatment
CO_2 fracturing	13−51\$/t or even more	55 MPa	Relatively low	Friendly to the environment

Table 5. Effect of Injection Rates on CH_4 Recovery and Breakthrough[a]

core sample	Q (Ml\Min)	breakthrough (Min)	methane produced (CM^3)	RF = (CH_4 Produced × 100)/OGIP)[b]
Berea gray	0.2	9333	640.59	69.63
	0.4	73.32	819.09	89.63
	0.6	42.15	559.45	60.81
	0.8	40.15	476.28	51.77
	1	39.99	478.06	51.97
Bandera gray	0.2	76.32	550.53	63.37
	0.4	82.49	652.2	75.08
	0.6	35.65	495.76	57.07
	0.8	26.82	402.13	46.29
	1	35.32	313.69	36.11

[a]Adapted from ref 77. Copyright 2020, permission from Elsevier. [b]OGIP: Original gas in place.

ations, equipment requirements, and environmental responsibility. For a comprehensive cost analysis comparing hydraulic fracturing and CO_2-fracturing processes, please refer to Table 4.

Nonetheless, it is essential to acknowledge that utilizing supercritical or liquid carbon dioxide fracturing technologies has its inherent drawbacks. These limitations stem from the notably low viscosity of CO_2, measuring just 0.1 mPa·s in its liquid state and 0.02 mPa·s in its gaseous and supercritical phases. Consequently, this reduced viscosity can lead to substantial filtration of the fracturing fluid, decreasing the sand-carrying capacity and sewing ability. Ultimately, these constraints may impose limitations on the extent of the fracturing operation.

3.2. CO_2 Storage in Depleted Gas Reservoirs. Numerous advantages are associated with the sequestration of CO_2 in depleted gas reservoirs. Initially, it is essential to recognize that these reservoirs come equipped with a substantial array of existing infrastructure, surface and subsurface, which can be readily repurposed for CO_2 storage with minimal modifications. Furthermore, this approach offers a high level of assurance regarding sealing integrity and cap-rock stability, as ref 68 emphasized. Additionally, it is noteworthy that the extent of pressure perturbations and consequent stress alterations in depleted oil and gas reservoirs is considerably lower than in aquifers. This distinction can be attributed to the extended history of oil and gas extraction from these reservoirs, a point underscored by ref 68.

A comparison between exhausted oil reservoirs and depleted gas reservoirs reveals that the latter presents distinct advantages for carbon capture and storage (CCS). The enhancement is mainly a result of the higher final recovery and enhanced gas compressibility, leading to a much bigger storage capacity per unit of pore volume. Observation aligns with the findings of ref 69 and 70. A comparative analysis of different reservoir types employed in this storage method reveals that condensate-gas reservoirs offer distinct advantages when contrasted with wet and dry-gas reservoirs. These advantages primarily stem from factors such as the constrained volume of remaining gas, the favorable phase behavior exhibited by the combination of condensate gas and CO_2, and its notable injectivity, as highlighted by research conducted by ref 71. Furthermore, it is essential to emphasize that the sequestration capacity of CO_2 per unit of pore volume in depleted condensate reservoirs substantially surpasses that of similar aquifers, approximately by a factor of 13, as demonstrated by ref 69. Nevertheless, it is crucial to acknowledge that phase transitions may manifest in depleted condensate reservoirs, a phenomenon not typically observed in dry and wet-gas reservoirs, as elucidated by ref 49.

3.3. Efficiency of shale gas recovery. Despite being in an early developmental phase and lacking commercialization, injecting CO_2 into shale formations has garnered attention from scholars. These academics have explored the potential of using CO_2 injection to improve shale gas recovery. The research by ref 72 showed the continuous CO_2 injection was more effective than the huff-and-puff approach in improving shale gas recovery. The highest recovery of shale gas achieved by this approach reported to be 15%. In a study conducted by,[73] a thorough examination was carried out on the use of CO_2-EGR in an exhausted shale gas resource. The findings of the study indicated that the highest recovery of shale gas from a single well reached 14%. In a study conducted by ref 74, it was shown that the injection of CO_2 may significantly improve the displacement of CH_4, with an enhancement of up to 7%. Implementation of a shale gas CO_2 injection displacement-desorption test achieved this. In a study conducted by ref 75, a model was developed that included multicomponent-transport and geo-mechanical impacts. The results of their analysis revealed that the recovery ratio of CH_4 might enhanced about 24% via continuous CO_2 flooding and by approximately 6% with huff-and-puff CO_2 injection. However, it is important to note that the infectivity of CO_2 and the potential efficiency of shale gas recovery exhibit spatial variability. In their research, ref 76 demonstrated that the incremental CH_4 recovery resulting from CO_2 injection was found to be very limited in both the CO_2 flood and huff-and-puff scenarios for the New Albany shale. It generally accepted that the early breakout of CO_2 in CH_4-producing wells, a result of gas−gas mixing, is one factor contributing to the low recovery ratio for shale gas. In the scenario of gas−gas mixing, the introduction of free-phase CO_2 leads to its migration toward the production wells. The

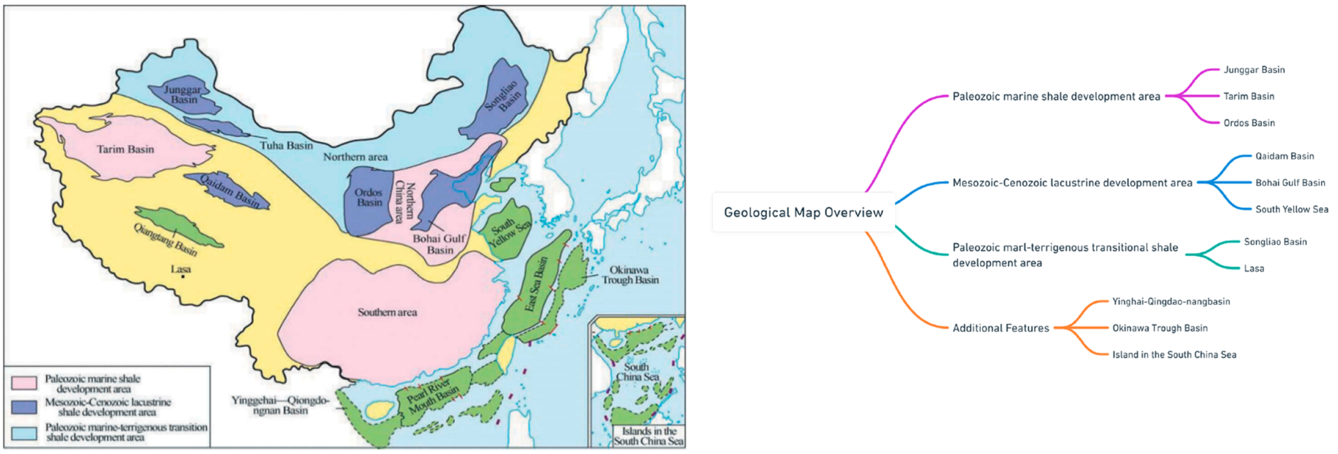

Figure 7. Illustrates the distribution of shale resources across China.

phenomenon occurs when there is a noticeable decline in shale gas production and a considerable rise in the rate of CO_2 generation. Table 5, illustrates the CH_4 production efficiency resulting from different injection rates on the rock sample cores.

The efficacy of injecting CO_2 and recovering CH_4 in a shale reservoir is contingent upon numerous engineering parameters, encompassing fracture conductivity, fracture half-length, fracture height, as well as operational factors like injection time, injection volume and geological conditions, such as porosity, permeability, layer thickness, and Total Organic Carbon (TOC) content, further, contribute to the intricate nature of this process.

Shale formations that occur naturally exhibit significant variations in (TOC) content, whereby even little fluctuations in TOC may substantially influence the efficacy of shale gas enhancement. Shales with higher TOC content have the potential to enhance shale gas recovery when combined with CO_2 injection, and this is due to their ability to sequester more CO_2 in an immobile phase, mitigating CO_2 breakthroughs in production. Deploying effective reservoir management and operational techniques is crucial, particularly in shale formations exhibiting varied geological, hydrological, and geochemical conditions. Optimizing CO_2 pressure or injection rates might employed to refine injection strategies for achieving optimal shale gas recovery and CO_2 storage. This objective can be accomplished by strategically delaying the CO_2 breakthrough, as ref 78 suggested.

3.4. Capacity for CO_2 Storage in Shale Formations.

Potential CO_2 storage in shale formations has evaluated using many methods, including analysis of CO_2 sorption-isotherms, geologic and volumetric-data, evaluations of gas-in-place, and estimations of eventual recovery of shale resources. According to ref 79 estimate, around 28 giga-tons of CO_2 have the potential to be sequestered in the most profound sections (equal to or more than 304 m or 1000 feet) and thickest parts (equal to or greater than 15 m or 50 feet) of the Ohio shale and New-Albany Shale formations. According to estimates, the Marcellus shale has the potential for CO_2 storage at depths above 915 m, with about 99 Gt of adsorbed-gas and 72 Gt of free-gas, resulting in a theoretical maximum storage capacity of 171 Gt. The projected quantity of technically available CO_2 storage is much smaller, with a value of 55 Gt, as ref 80 reported. Reference 81 proposed a production-based model to estimate the theoretical capacity of CO_2 storage in shale formations. This model relies on determining the ratio between the sorption and diffusivity of CO_2 and CH_4. Their findings indicated that the Marcellus Shale can store around 10.4−18.4 rigatonis (Gt) CO_2 by 2030.

China has significant shale reserves and resources, as seen in Figure 7. China's total recoverable shale gas resources are approximately 25 trillion cubic meters. Paleozoic Marine Shale in the Junggar, Tarim, and Ordos Basins recognized for its organic-rich marine shale, suggesting the presence of oil reserves. Mesozoic-Cenozoic lacustrine shale found in Qaidam, Bohai Gulf, and South Yellow Sea basins has fine-grained sediments from ancient lakes, making it favorable for shale oil and gas development. Moreover, The Paleozoic Transitional Shale in the Songliao Basin and Lasa combines marine and terrigenous deposits, reflecting a diversified depositional environment with different shale properties. In addition, it mentions additional areas such as the Yinghai-Qingdao-nang basin, Okinawa Trough Basin, and an island in the South China Sea that might hold shale resources.

The process of evaluating shale's potential CO_2 storage capacity, as outlined by the U.S. Department of Energy's National Energy Technology Laboratory, described by eq 1. In a study conducted by ref 82, we estimated the overall capacity for CO_2 storage, which amounted to 4177 (Gt). This estimation was further broken down into three categories: marine shale, which accounted for 1388 Gt; marine-terrigenous transitional coal bed carbonaceous shale, which accounted for 1684 Gt; and lacustrine shale, which accounted for 1105 Gt.

$$G_{CO_2} = AE_A h_g E_h \left[\phi_\rho CO_2 + (1 - \phi) \rho_{sCO_2} \right] \tag{1}$$

The theoretical carbon dioxide storage capacity (CO_2) in shale reservoirs denoted as GCO_2 in Equation 1. Variables A and hg represent the shale reservoir's area and thickness, respectively. The porosity of the shale reservoir, denoted as S, established at a value of 0.35, the porosity of the shale reservoir, marked as ϕ, is found at a value of 0.35, the parameter ρCO_2 represents the density of supercritical CO_2 or liquid under reservoir conditions. Specifically, it defined as 628.61 kg/m^3, matching a temperature of 40 °C and a pressure of 10 MPa. Parameter ρsCO_2 represents the quantity of CO_2 adsorbed per unit volume of solid rock. This value has been determined to be 68.3 kg/m^3 based on experimental investigations of CO_2-adsorption on Longmaxi-shales in the

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Table 6. CO_2-EGR Process Regular Displacing Experiments

the sort of rock	concentrated fluid	temperature (°C)	pressure (MPa)	important findings	refs
Carbonate cores	methane	20−60	3.55−20.79	Carbon dioxide might improve the extraction of CH_4, whether it is within the gas, liquid, or supercritical phase.	86
Carbonates core	whether wet or dry, it was filled with gas.	20−80	3.55−20.79	The CO_2 ratio rises with temperature and falls with pressure.	92
Berea's sandstone core	dry the core, 10% water first-saturation, and 10% brine initially saturated (20 wt %).	40	8.96	The saltiness of pooled water reduces carbon dioxide dispersion in methane.	93
Sandstone and carbonate's core	CH_4	60−80	10−12	The remaining water causes the pores to become smaller, which enhances the dispersion of supercritical CO_2 and CH_4.	94
Sandstone's core	90% CH_4 + 10% carbon dioxide simulating natural gas, respectively.	40−55	10−14	Carbon dioxide has a higher dispersion coefficient in Synthetic natural gas than methane.	95
Sandstone's core	nitrogen (N_2) with H_2O from the formation.	50	21	The variability effect predominates in the impermeable core, whereas the effect of gravity segregating influence is notable in the absorbent core and porous.	96
Bandera sandstone core	CH_4	50	8.96	At lower flow rates, gravitation substantially affects CO_2 flow habits.	93

Sichuan basin of China efficiency factors for area and thickness denoted as E_A and E_h, respectively, assigned one value.

In contrast to deep saline aquifers, CO_2 has competitive adsorption capabilities with in-place methane. It may effectively trap as an adsorbed phase on the surface of the host rock in shale, in addition to other trapping mechanisms such as dissolution trapping, structural trapping, residual trapping, and minerals trapping. Gas adsorption is a significant factor in CO_2 storage inside shale reservoirs. The CO_2 adsorption process onto the shale matrix might serve to stabilize the reservoir pressure. However, this adsorption can also affect the solubility trapping mechanism, potentially disrupting the CO_2-water-rock processes.

Moreover, this interference may heightened when the concentration of CO_2 decreases over time. Furthermore, the alterations in permeability and porosity resulting from geochemical processes over a period lead to modifications in reservoir pressure, influencing gas adsorption onto the host rock.

Storage of CO_2 in shale reservoirs is influenced by several characteristics, such as the mineral composition of the shale, the pressure and temperature of the pool, which in turn affect the properties of CO_2, the wettability and conductivity of the reservoir, as well as the capacity for gas-adsorption. The favorable nature of high carbon dioxide (CO_2) adsorption capacity in shale for CO_2 injection into shale reservoirs has been shown by empirical evidence.[83] The quantity of adsorbed CO_2 is directly proportional to the CO_2 adsorption isotherm, but the presence of CO_2 in other phases negatively correlates with it.

4. CO_2-EGR PROCESS

Due to the CO_2-EGR's advanced technology, the extraction of gases from CO_2 injection is enhanced, and the remaining gas in a decreasing or exhausted reservoir is both displaced and repressurize to achieve EGR,[65] particularly for reservoirs of soured gas where carbon dioxide is generated along with gas.

Separated carbon dioxide from generated gases can be reintroduced into the reservoir for better gas recovery. Furthermore, CO_2 may lower the reservoir fluids' dew point pressure in wet gas reservoirs, making it advantageous for removing blockage condensate and enhancing methane production.[84] According to estimates in ref 65, carbon dioxide displacement may raise gas recovery by as much as 11%. The feasibility of CO_2-enhanced gas recovery has investigated in many studies that used experiments and simulations.[85,86] Table 6 overviews a few standard displacement tests conducted under various temperature and pressure circumstances that revealed the CO_2-EGR process and offered recommendations for using this technology.

The release of carbon dioxide from the reservoir that produces CO_2-contaminated gas is the biggest problem with CO_2-Enhanced gas recovery.[87,85] CO_2-EGR geologic structures, notably the microstructures, should be studied in depth since the favored route substantially influences the breakthrough of carbon dioxide and the complete recouping of methane.[88] Irreducible water in reservoirs also affects CO_2 and CH_4 exchange. Dispersion rises since the holes occupied by fundamental water lead to smaller pores and convoluted flow routes. Mitigation of carbon dioxide breakthrough at the production well may potentially be achieved by the use of CO_2 injection techniques, using a horizontal-well configuration in the lower sections of the reservoir, while simultaneously conducting methane production in the higher regions.[85,89] To guarantee supercritical phase displacement and attain an excellent CH_4 recovery, injecting CO_2 at the beginning of natural gas production has positive effects.[85] CO_2 injection in the latter stages may increase CCS performance, making it more appealing when CO_2 sequestration is discussed.[85,90] Overall, the moment of carbon dioxide injection is very important for getting the best gradual recoveries by permissible generated carbon dioxide emphasis at the production well, governed by the CCS projects' economics.[91]

Due to CO_2 being significantly heavier than CH_4, it takes up less area and moves around at less speed, perhaps mitigating carbon dioxide breakthrough. Nevertheless, If the reservoir contains CO_2 injected into its gas phase, it will take up lots of space and mix with methane rapidly more quickly; perhaps it may cause early carbon dioxide (CO_2) breakthroughs.[97]

Geological characteristics have a significant impact on CO_2-enhanced gas recovery performance. Viscous and gravitational forces affecting permeability, formation dip, and thickness are critical to displacement stability. Fluid parameters include the salinity of the water, and the diffusion coefficient comes in second when influencing CO_2 breakthrough.[91] In particular, connate water in the reservoir positively influences CO_2-

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enhanced gas reservoir performance. As a result, CO_2 fluids in reservoirs dissolve, which is advantageous for increasing the capacity for storing and reducing CO_2 breakthrough at the wellhead.[65,98,99] In general, CO_2-EGR technology remains in its infancy. It requires further work to solve the issues, such as reducing CO_2 breakthrough and attaining good results in improving CH_4 recovery and CO_2 sequestration. Political events in upcoming years and decades may significantly impact the economy.[100]

4.1. Adsorption of CO_2/CH_4. With enormous environmental advantages for geological CO_2 storage and financial benefits for shale gas extraction, carbon dioxide Hydraulic Fracturing Improved Recovery (CO_2-EGR) has gained international interest recently. Throughout this work, case experiments and modeling studies on competing carbon dioxide adsorption, methane adsorption, and CO_2−CH_4 displacement in shales in the CO_2-EGR cycle reviewed. Shale has a higher potential to bind carbon dioxide than gas (CH_4) when subsurface circumstances and the thermal maturity of the organic elements in the shale are present, particularly in the primary (or micro-) pore volume. Like coalbed methane, shale gas maintains a separate location with a different gas trapping by physical adsorption.[101]

CO_2 technology significantly enhances gas recovery and offers a distinctive opportunity to facilitate the extraction of more CH_4 from underground reserves. This achievement is realized through the liberation of trapped CH_4 within the reservoir matrix and its migration to fractures, facilitated by competitive adsorption with CO_2, as demonstrated by.[102] In this process, simultaneous binding of CO_2 and CH_4 is imperative. CO_2 and CH_4 exhibit an affinity for adhering to the reservoir material's inner surfaces after injection. Due to competitive adsorption, the removal of CH_4 and the absorption of CO_2 occur concurrently. Figure 2, illustrates this process's flow and mass transfer dynamics within a shale matrix.

Several studies have investigated the adsorption characteristics of gases, consistently finding that pure CO_2 shows a higher adsorption capacity than CH_4. This trend holds across a diverse range of shale and coal types, as indicated by the research conducted by refs 103 and 104. Furthermore, the desorption process of carbon dioxide exhibits a more significant hysteresis effect compared to that of CH_4. This characteristic proves advantageous when considering the long-term sequestration of CO_2 in deep underground storage. During a trial, CO_2 and CH_4 mixed; shale sometimes absorbed CH_4 more than CO_2, but its ability to absorb CO_2 was more substantial than its ability to absorb CH_4. Adsorption capacity measurements show that CO_2 is enhanced in the presence of CH_4, suggesting that competitive adsorption helps bring about CO_2-EGR. There could be less carbon dioxide and CH_4 competitive adsorption in a CO_2/CH_4 hybrid system than in a system with either gas present alone since the adsorption qualities depend on the relative pressures of each component in the mixing gas.[105] Along with inert gas adsorption tests with single-component gases and CO_2−CH_4 mixtures, some experts have also done dynamic gas adsorption studies with CO_2−CH_4 substitution features. They found out that injecting CO_2 has the excellent impact of competing with CH_4 to take it up, which helps improve the enhancement of methane gas.[106]

Adsorption sites compete for carbon dioxide and methane, which plays a significant role in the competitive adsorption processes. Competition for carbon dioxide and methane adsorption depends on the thermal force among gases and coal/shale matrices, gas molecule size, and matrix micropore network accessibility.[107] The molecular characteristics of CO_2 and CH_4 are different, as seen in Table 7. Molecule methane

Table 7. Physical Characteristics and Molecular Parameters of CO_2 and CH_4[a]

property	CO_2	CH_4
Molecular mass, m (/moll)	44	16
Critical temperature, Tc (K)	304.2	190.5
Critical pressure, *Pc* (MPa)	7.4	4.6
Critical density, ρc (kg/m^3)	467.6	162.7
Phase density, kg/m^3	1028	372
αM (cm^3/moll) polarizability	7.34	6.54
Kinetic diameter, σk (Ao)	3.3	3.8
Collision diameter, σk (Ao)	4.00	3.82
Molecule diameter, eff (Ao)	3.63	3.81

[a]Reproduced from ref 107.

has a diameter of 3.80Ao. Meanwhile, the carbon dioxide molecule has an active diameter of 3.30Ao, which is relatively modest.[108] As a result, when comparing CO_2 and CH_4 molecules, the former is easier to pass through micronano holes, making it possible for CO_2 to interact with more adsorption sites.[102,107]

Kerogen's functional molecules exhibit a more robust interaction with CO_2 due to its higher quadrupole moment, leading to a greater likelihood of physisorption through van der Waals forces than methane. The affinity of carbon dioxide for organic surface materials surpasses that of CH_4. CO_2 competes with CH_4 for adsorption sites on the matrix's surface.[109] Lower heat required for CH_4 desorption than CO_2 makes simultaneous CH_4 desorption and CO_2 adsorption feasible. Unique supercritical properties of CO_2 facilitate enhanced adsorption and contact with rock surfaces, ultimately improving CH_4 adsorption.[110] Despite these insights, the underlying mechanics of competitive adsorption remain a subject of ongoing investigation.

Clays exhibit compatibility with CO_2 due to alterations in their crystalline structure and the breakdown of their organic molecules under heat. This phenomenon explains the peak of CO_2 uptake occurring at 100 °C and 4.5 MPa, as observed in the study by ref 111. Researchers conducted experiments involving a range of CO_2 concentration proportions and gas mixtures. The results of these investigations highlighted that the rate of adsorption in shale is intricately dependent on the specific composition of the absorbed gas. Predominant research in CO_2 injection methods centered on evaluating shale reservoirs' dual capability to recover CH_4 and sequester CO_2. The intricacies of shale's porous structure, coupled with its exceedingly low permeability, pose a formidable challenge in ascertaining the feasibility of these methodologies at a field-scale trial. While numerous studies have endeavored to explore CO_2 injection through

In simulation-based approaches, the absence of comprehensive models that can effectively elucidate the intricate mechanisms inherent to CO_2-EGR is predominantly attributed to the computational intricacies that are both intricate and time-consuming. Augmented affinitive nature of CO_2, surpassing that of CH_4, coupled with broader spreading characteristics of additional natural gas within micropores, as empirically evidenced by ref 112. Reservoir analyses and modeling investigations have substantiated the capacity of shales to sequester CO_2 through adsorption onto organic substrates, akin to coal, and placement within existing shale fractures. This fundamental concept underpins the feasibility of employing shales as a potential medium for CO_2 storage.

Furthermore, select financial analyses have been conducted, founded upon the technical viability of CO_2-EGR methodologies, offering insights into the economic considerations accompanying this approach. A study conducted by ref 113 revealed that the benefits derived from the produced methane could potentially outweigh the costs associated with CO_2 storage, presenting a compelling case for the overall utility of the CO_2-EGR technique, as corroborated by ref 34. Consequently, forthcoming research endeavors should encompass a comprehensive analysis of the variances among shale gas reservoirs, specifically focusing on their influence over the intricate fluid dynamics governing gas production, and this pertains notably to the competitive interplay of kinematic adsorption between carbon dioxide and CH_4 and the composite gas flow behaviors.

Consequently, further pertinent studies and model experiments are required to measure competitive adsorption precisely and estimate the adsorption ratios between CO_2–CH_4, particularly under mixed circumstances. When looking at the different adsorption behaviors of CO_2 and CH_4 in coal and shale, it is important to compare the two, and one essential metric to consider is the selective adsorption coefficient, abbreviated as ($\alpha CO_2/CH_4$). This value is provided:[114,115]

$$\alpha_{CO_2/CH_4} = \frac{x_{CO_2} y_{CH_4}}{x_{CH_4} y_{CO_2}} = \frac{V_{LCO_2} P_{LCH_4}}{V_{LCH_4} P_{LCO_2}} \tag{2}$$

In evaluating gas blends, the molar ratios of gases in the free and adsorbed phases are represented by x and y, respectively. The Langmuir factors, *VL* and *PL*, are critical parameters in this context. Higher values of these factors indicate a more pronounced displacement capacity of CO_2 over CH_4, which is essential for the efficiency of the process. Typically, the CO_2 to CH_4 ratio in both coal and shale substrates exceeds unity, facilitating the displacement of adsorbed CH_4 by CO_2. This characteristic is pivotal because it underpins the feasibility of employing CO_2 Enhanced Gas Recovery (CO_2-EGR) and CO_2 storage techniques, as evidenced by research from refs 115–117. The ratio of CO_2 to CH_4 has a value that varies depending on the kind of coal and shale, as well as the temperatures and pressures of the reservoir, the mineral composition, and the pore structure of either coal or fracking. Reservoirs' geological state varies by area, and coal and shale are heterogeneous in the CO_2-ESGR processes. Therefore, to perform case-based research, it is necessary to determine the CO_2/CH_4 value in various reservoirs.

The study by ref 51 thoroughly examined the pore structure and deformation characteristics of low-permeability coal during CH_4/N_2 adsorption–desorption processes. The study revealed important advancements in micropores and transition pores in the coal samples, highlighting a prevalent occurrence of semiopen pores in the pore structure. This research delves into the complex and varied characteristics of coal pore structures, highlighting their significant impact on the effectiveness of methane extraction and nitrogen injection. Moreover, developing a strain model for coal adsorption–desorption represents a considerable advancement, investigating the efficacy of N_2 injection in displacing CH_4 in low-permeability coal, and this suggests a practical approach to increase gas flow and improve the efficiency of coal seam gas control. Reference 52 used modern methods such as HPMI, LPGA-N_2, and SEM to analyses the intricate pore architectures of coals of different ranks from Australia and China in the study. The study used FHH fractal theory and the MENGER SPONGE model to calculate fractal dimensions associated with pore volume, surface area, and distribution, investigating their impact on gas adsorption dynamics. Coal metamorphism plays a crucial role in determining the fractal characteristics of coal pores, which affects their gas adsorption qualities and influences coal's gas storage capacity, and this highlights the relationship between coal's fractal structure and gas storage efficiency.

4.1.1. Under Geological Circumstances. Understanding the adsorption process of shale gas is of paramount importance in directing fossil fuel exploration and production and in facilitating numerical modeling for a quantitative assessment of the quantity of adsorbed gas under specific geological conditions.[118,66] A well-established approach in this domain involves conducting adsorption isotherm measurements in laboratory settings, which garnered significant attention in research endeavors. These experiments are instrumental in evaluating the gas adsorption properties of shale, shedding light on the interaction dynamics between gases and source rock samples under controlled temperature and pressure conditions. Such studies have been conducted by refs 61 and 119.

Carbon dioxide (CO_2) has a greater propensity for sorption onto shale reservoirs than methane (CH_4). With the same pressures, Devonian's shale has 5.3 times the capacity to adsorb carbon dioxide than methane.[120] In comparison to CH_4, which ranged within the range of 0.03 to 0.47 mmol/g, according to a study conducted by ref 61 carbon-containing shale found in the Parana's Basin of Brazil exhibited a range of extra adsorption capacities for carbon dioxide, ranging from 0.14 to 0.81 mmol/g.

In a comprehensive study, researchers meticulously assessed carbon dioxide and methane adsorption capacity within the Montney, Eagle, Barnett, Ford, and Marcellus shale formations in the United States and Yanchang shale from China's Ordos Basin. Furthermore, their investigation of organic-rich shale within the Fort Worth Basin revealed a notable discrepancy in the shale's predilection for carbon dioxide over methane. Notably, findings were reported by refs 116, 121, and 122 corroborating this observation. It is worth noting, however, that the gas adsorption capacity of shale exhibits considerable variability across different geographical locations, even among shale formations within the same structural context, as highlighted by ref 29. Drawing from an extensive body of scholarly research, it is evident that the injection of carbon dioxide (CO_2) can significantly enhance methane adsorption (CH_4). However, it is imperative to underscore that the efficacy of this approach is contingent upon geological conditions and various technical parameters, thus highlighting the nuanced interplay between these factors in the context of methane recovery. Rich organic shale has an enormous potential to store carbon dioxide, which occurs during CO_2 flooding and pure CO_2 storage stages. Shale gas, a natural gas, is a significant clean energy source with considerable deposits worldwide. CO_2-EGR is a viable approach for accessing shale gas reserves concurrently with the permanent sequestration of CO_2 in subsurface formations. Reference 123 showed

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substantial potential. CO_2-EGR has not yet been tried in a gas reservoir despite being debated for over 20 years.[124,125] Primary factors contributing to these challenges include the limited utilization of geologic sequestration techniques and the ongoing high cost associated with carbon dioxide. Additionally, concerns regarding the potential rapid comingling of injected CO_2 with existing CH_4, leading to a degradation of organic gas supply, serve as valid justifications for the opposition to carbon storage and enhanced gas recovery (CSEGR). A comparable investigation was also seen by refs 64 and 126. The study's findings suggest that including methane in carbon dioxide injection could contaminate the resulting natural gas, reducing its market value and necessitating additional expenditures for impurity removal. Conversely, the research indicated that the integrity of the reservoir maintained over geological time spans in the case of depleted gas reservoirs, alleviating concerns related to potential seepage into groundwater and surrounding land areas.

Improving simulation capabilities by integrating non-isothermal factors and undertaking extensive reservoir characterization is crucial. Augmenting these initiatives with a field pilot experiment focused on (CSEGR) during the interim period is indispensable. This holistic approach will validate the simulation results' accuracy and provide robust evidence to support the concept's viability.

4.1.2. Molecular Simulation. A molecular simulation is an effective tool for studying configuration settings in terms of molecular mobility and response.[123,127] Thus far, much research has conducted to examine carbon dioxide (CO_2) and methane (CH_4) adsorption characteristics in shale formations, employing a diverse range of experimental techniques and numerical and molecular simulations.

Using numerical and molecular simulations proves advantageous in evaluating enhanced gas recovery (EGR) efficiency and carbon dioxide (CO_2) storage capacity in shale formations. Additionally, these simulations are valuable in analyzing the impact of reservoir parameters on the adsorption behavior of both CO_2 and methane (CH_4).[128,129,127] According to certain simulations, an additional gas production of 7% observed concerning the Marcellus shale located in the eastern region of the United States. Based on the Langmuir volume derived from the adsorption isotherm, this area's theoretical maximum capacity for carbon dioxide (CO_2) storage is estimated to be 1.6 Mt/km^2.[130] Molecular simulations suggest optimal operating conditions at a depth of 1 km for the displacement of CH_4 by CO_2 in shale.[131] Kerogen generated from higher plants has superior characteristics as the organic type for shale-based carbon capture and gas storage (CCGS).[132] Additionally, it has observed that the presence of reservoir moisture enhances the efficiency of enhanced gas recovery (EGR) in shale formations.

Research on CO_2/CH_4 competitive adsorption using actual kerogen modeling has been undertaken.[133] delved into the volumetric straining of kerogen and modeled the CO_2/CH_4 adsorption behaviors within Nanopores. Their findings indicated carbon dioxide adsorption leads to more significant swelling in volumetric strain than CH_4. Meanwhile, ref 127 explored how kerogen reacts to CO_2/CH_4 competitive adsorption to enhance shale gas extraction through CO_2 injection. Their comparative adsorption tests using pure gases and specific CO_2-CH_4 mixtures in shale demonstrated that CO_2 and CH_4 competitively adsorbed. Competitive adsorption exhibits distinct behaviors when examined in adsorption tests employing single-component gas systems versus those utilizing CO_2-CH_4 mixtures. Using the Grand Canonical Monte Carlo (GCMC) methodology, ref 127 rigorously investigated the competitive adsorption mechanisms of CO_2 and CH_4 within the nanopores of shale kerogen. An intricate analysis has undertaken regarding the moisture content, deploying scanning electron microscopy (SEM) and UNGER has advanced molecular modeling techniques. Also, ref 127 deliberated on multiple parameters, including the effects of temperature, the distribution dynamics of carbon dioxide and methane, the quantification of moisture, the adsorption selectivity metrics, and the optimal conditions for injections. Empirical findings elucidated that methane adsorption onto kerogen exhibited a direct proportionality with pressure; however, an inverse correlation with temperature was observed. The adsorption selectivity of CO_2 over CH_4 was determined to be 2.53 to 7.25, signifying preferential adsorption of CO_2 over CH_4 across varying temperature conditions. Similar observations and conclusions were echoed in subsequent studies.[134−136] In addition, they have found the same results. Nevertheless, the interaction between CO_2 and CH_4 in shale is accurately determined using numerical and molecular simulations and experimental measurements. These methods provide direct insights into the interplay between the two gases and establish widely accepted adsorption isotherms.[105,137] Reference 120 conducted a study on Devonian shale from Kentucky focusing on adsorption-isotherms. Their findings revealed that the adsorbed carbon dioxide (CO_2) ratios to methane (CH_4) were approximately 5 to 1. In contrast, ref 138 reported that adsorbed CO_2/CH_4 ratios were almost 3 to 1 when examining the New Albany shale Illinois basin under pressures of approximately 7 MPa. The sorption isotherms of pure CH_4, CO_2, and mixed CO_2/CH_4 indicate that the preferred carbon dioxide (CO_2) sorption is enhanced by increased temperature and CO_2 concentration within the CO_2/CH_4 mixture.[105] Regrettably, laboratory assessments often involve conducting adsorption isotherms of CO_2, CH_4, or CO_2/CH_4 mixtures individually using volumetric or gravimetric techniques. However, these approaches cannot accurately determine the competitive adsorption between carbon dioxide (CO_2) and CH_4.[139,140] Nevertheless, it expected to use simplified reservoir models or make certain numerical and molecular research assumptions. Consequently, the findings obtained from these studies provide only general limitations on the actual interplay between carbon dioxide (CO_2) and carbon monoxide (CH_4) in shale formations.

4.1.3. Adsorption Isotherm Models for Gases. Adsorption isotherms delineate the behavior of ad-sorbate gases within porous media under isothermal conditions as external pressures are varied.[119] Additionally, models of adsorption isotherms, which incorporate equilibrium data and delineate adsorption traits, offer insights into the intricate interactions between contaminants and adsorbent substrates.[141] Over the past decade, many isotherm models have been introduced, encompassing the Langmuir, Freundlich, Dubinin−Radushkevich, Temkin, and Toth frameworks, among others.[142] The popular and straightforward Freundlich and Langmuir isotherms have used in many types of research to simulate gas adsorption in a porous medium inside shale. Still, Freundlich never demonstrated a better match, and ref 143 conducted experiments to account for sorption in shale with low and high pressures using Langmuir isotherm and the Ono-Kondo model. Reference 139 ascertained that the Ono-Kondo

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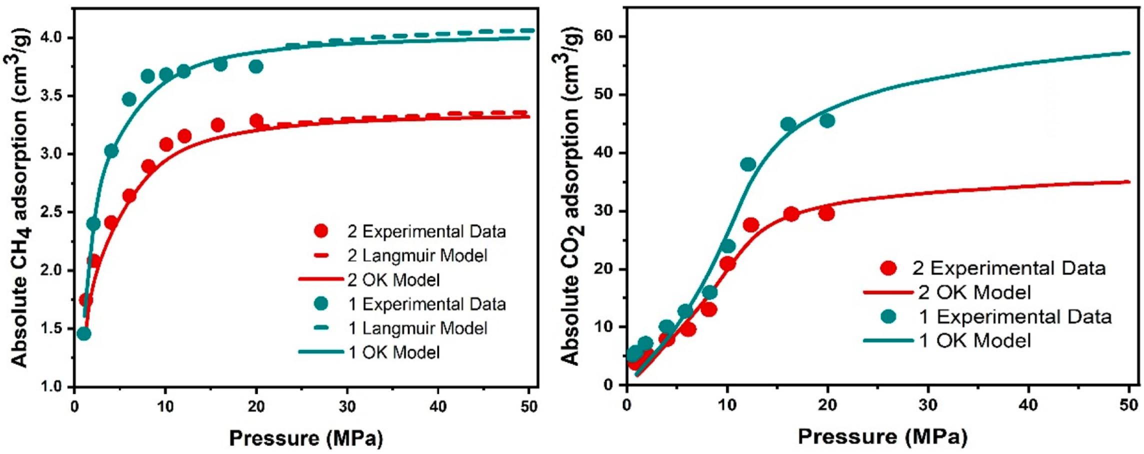

Figure 8. Ono-Kondo and Langmuir's isotherm models for carbon dioxide and methane on Longmaxi samples. Adapted from ref 148. Copyright 2021, with permission from Elsevier.

model provided a superior fit for interpreting the adsorption data from experiments under both high and low-pressure conditions. It discerned from Figure 8. That, given the specific reservoir conditions, the Langmuir model was not an optimal representation. When experimental data were matched into several isotherm models, ref 136 also showed the adsorption propensity of samples obtained from the Ordos-Basins and Sichuan in China. In forecasting carbon dioxide and CH_4 adsorption isotherm, the Ono-Kondo model provided an excellent match concerning Langmuir, Dubibin-Astakhov, and others.

Shale gas is a possible alternative power source for the world's energy.[144,145] Shale gas reservoirs may now be productively produced thanks to cutting-edge horizontal drilling and hydraulic fracturing techniques, which increases the need for a deeper comprehension of shale gas adsorption behavior.[146,147]

When examining the sorption properties of CO_2 and flue gases on South African coals,[50] research reveals that the empirical data for pure CO_2 and flue gases align with the theoretical predictions of three traditional adsorption isotherm models (Langmuir, Freundlich, and Temkin) for CO_2, and the Extended-Langmuir model for flue gases. The study outlines a relationship between temperature and sorption capacity, indicating that the sorption process releases heat and decreases the sorbed phase's volume as temperature increases. In addition, the study outlines a preferential sorption hierarchy for flue gas constituents, with CO_2 having the highest sorption capacity, followed by N_2, O_2, SO_2, and NO_2. According to the findings, the Langmuir isotherm model best represents the CO_2 sorption mechanism, suggesting a monolayer coverage phenomenon. On the other hand, the dynamics of flue gas sorption most accurately depicted by the Extended Langmuir model. However, there is a decrease in maximum adsorption capacity as the temperature increases. This study highlights the intricate balance of physical and chemical interactions involved in the CO_2 adsorption process on coal, confirming its practical feasibility, spontaneous nature, and exothermic nature.

4.2. Gas Displacement during CO_2-EGR Summarized.

Many factors, encompassing rock properties, inherent gas attributes, and operational conditions, play a pivotal role in determining the ultimate recovery of shale gas via carbon dioxide injected. Based on scholarly research, it posited that the displacement of CH_4 by CO_2 is miscible under reservoir conditions. Primary factors in replacing CO_2−CH_4 are the competing sorption and transport characteristics of both carbon dioxide and CH_4 in shale. An amount of carbon dioxide on the shale surface has a higher adsorption affinity than CH_4. Moreover, compared to CH_4 molecules, CO_2 molecules are straight and have a lower kinetic diameter (0.33 nm) (0.38 nm) consequently.

Carbon dioxide may displace competitively preabsorbed CH_4 out of micropores and enter a more constrained pore system.[149] Numerous studies have conducted to elucidate the phenomenon of insoluble displacement by assessing the interfacial tension (IFT) between supercritical carbon dioxide and CH4. However, the empirical results from these efforts lacked consistent validation, and variations in IFT across the interface of the two distinct gases were attributed to Cortège's pressures.[150,151]

This section succinctly encapsulates the principal factors influencing displacement efficiency within the CO_2-EGR methodology.

4.2.1. Temperature. An elevation in temperature induces the migration of carbon dioxide and CH_4 molecules, thereby augmenting the experimental condition coefficient due to intensified collisions among the gas molecules.[27] Most of the carbon dioxide gas is stored as free gas, which increases the danger of leaking in the event of a mechanical breakdown or imbalance. Hence, higher aquifer temperatures are not advised for CO_2 gas storage.[54]

At elevated temperatures, minimal carbon dioxide gas remains sequestered in the immobile phase, rendering CO_2 sequestration in aquifers with ascending temperatures inadvisible. Nonetheless, the aquifer temperature must surpass the critical point at which carbon dioxide transitions to its vapor phase, identified as 31.05 °C.[54] Moreover, the storage site should subjected to a cyclical regimen of injection and cessation; thermal fluctuations influencing the adjacent rock strata and casing might transpire. Such variations could precipitate CO_2 gas permeation from cap-rock, case, and defunct wells.[152]

4.2.2. Injection Rate. Increasing the injection rates led to an early breakthrough of the injected carbon dioxide (CO_2) in Berea rock samples at temperatures between 40 to 50 °C and pressures of 8.96 MPa. However, excess gas in depleted gas wells adversely affects the flow rates.[153] Geologically, the carbon dioxide (CO_2) injection into the 3650 m-deep aquifers

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significantly impacted methane generation and storage of CO_2.[85] The impact of different injection rates on methane generation shown above in Table 4. It posited that elevated injection rates during the initial phases result in superior gas recovery. However, from an economic perspective, the cost associated with CO_2 capture and storage escalates with increased injection rates.

4.2.3. Density and Viscosity. Unique properties of CO_2, such as its lower viscosity, enhanced infusibility, increased density, elevated liquid solubility, and minimal surface tension, facilitate the extraction of shale gas.[154,110] Residual methane in porous structures alters the density and viscosity of carbon dioxide.[155] The deposition of substantial amounts of carbon dioxide facilitated by the near-liquid properties of supercritical CO_2 combined with its gas-like viscosity. These characteristics promote easy diffusion within the reservoir and stimulate the displacement of natural gas.[156]

4.2.4. Effect of variability. Influence variations in rock and carbonate samples on supercritical CO_2 and CH4 dispersion were studied. It was found that carbonates correlate with rapid responses and extended tails in pulse breakthrough curves.[157] The displacement of nitrogen gas by carbon dioxide in the test section, with an average porosity of 15% and a permeability of 55 mD, has significantly affected by heterogeneity. Gravity segregation, on the other hand, is dominating with lower flow rates (2 Cm^3/min) for core samples with lowly (1.5 mD) and high (270 mD) permeability.[158,96] Due to water filling a part of a pore volume, the gases' dispersion factor among the gases as the general area for interaction among both reduces. As a result, the mixture of carbon dioxide and methane increased.

Additionally, the increased density of saline water means that elevating the salinity of the solution boosts the dispersion coefficients between CH_4 and CO_2. While CH_4 constitutes over 85% of natural gas, the gas also comprises other light hydrocarbons, such as ethane and propane, and contaminants like CO_2, H_2S, and N_2.

Researchers generally use CH_4 for natural gas in their studies instead of hazardous gases like H_2S for ease and safety. Modeling methods showed that shale and carbonate preferentially absorb H_2S, which is advantageous to CH_4 recovery but may marginally limit CO_2 sequester.[159] Since H_2S is more soluble in water than CO_2, the mixture's breakthrough period would be delayed, improving gas recovery.[160]

5. CO_2-EGR MECHANISMS

Besides gas migration via organic pores or fracture networks, a shale gas reservoir's storage and transport mechanisms are profoundly influenced by organic content, pore architecture, and thermal conditions.[161] Through competitive adsorption, CO_2 supersedes adsorbed CH_4 within the shale and further displaces free CH_4 residing in fractured pores, enhancing shale gas extraction efficiency.[63] Refer to Figure 9 for a schematic representation elucidating the CO_2 displacement/replacement mechanism of CH_4 in shale during CO_2 fracturing to optimize shale gas recovery.

This paper details the hydraulic fracturing process used to extract natural gas from shale formations. The diagram explains the complex steps of hydraulic fracturing and the resulting routes for gas extraction. Fracturing fluid is first pumping into the wellbore to create significant cracks in the surrounding rock. Proppant particles keep the fissures open to aid in the extraction of hydrocarbons. Three types of gas are showing:

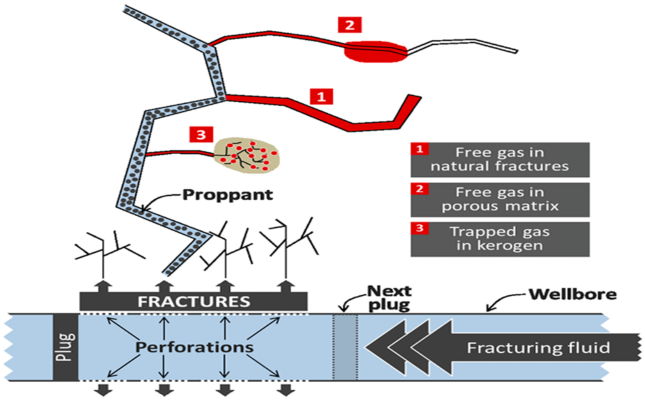

Figure 9. Schematic representation demonstrating how CO_2 may displace or replace methane in shale during the process of carbon dioxide fracturing to boost the recovery of the gas. Reproduced from ref 162. Copyright 2015, with permission from Elsevier.

(1) free gas in natural fractures, (2) free gas in the porous matrix, and (3) trapped gas in kerogen. The diagram shows the strategic placement of plugs and perforations to control the flow of fracturing fluid and enhance the rock's exposure to high-pressure fluid, ultimately improving gas extraction and recovery. This diagram illustrates the technical details of hydraulic fracturing, which is crucial for modern energy production by extracting unconventional gas.

The gas movement is controlled by the gas's slipping impact or Knudsen diffusing in the case of free gas, which is present inside macrospores or cracks; it observed that less pressure and higher temperature conditions facilitate its occurrence, considering the average molecular path is more important than the effective pore throat diameter. Surface diffusion primarily removes the molecules adsorbed from the microspore surfaces to absorb gas in organic-rich microspores. Once the strength of intermolecular collisions reaches a critical point in the context (of a low Knudsen number), a gas's migration process is often characterized by a viscous flow, commonly referred to as (Darcy flow).

This research evaluates the integration of CO_2 sequestration and EGR processes, aiming for an optimal method that ensures superior CO_2 storage and effective gas recovery. A central consideration for the viability of this method is the economic profitability of EGR and CO_2 storage. Figure 3 above illustrates the intertwined processes of CO_2 sequestration and enhanced gas recovery, suggesting a potential for economical CH_4 recovery alongside CO_2 storage.[42] By addressing the complexities associated with CO_2 capture through EGR and sequestration, this study aids in determining the most advantageous parameters for the operation.[163]

The salinity of connate water can potentially hinder EGR's efficiency.[93] Furthermore, the intrinsic heterogeneity of shale rock typically compromises gas production during primary depletion.[164,165] However, this heterogeneity expected to offer advantages during EGR's huff-and-puff gas injection phase. Consequently, the dual benefits of environmental CO_2 reduction and enhanced shale gas extraction position CO_2-EGR as a promising technology with extensive potential applications.

5.1. EGR Simulation Modeling. Many numerical models of liquid flow through porous media must be performed for precise storage tank forecasting and improvements to pump CO_2 for the EGR cycle successfully[166] by injecting carbon

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dioxide into the gas fields that have been exhausted. In the 1990s, ref 124 mimicked proper disposal of carbon dioxide, wherein carbon dioxide was caught from the fossil power plant and sequestered further into an underground structure with increased shale gas first from a reservoir. Reference 125 suggested combining H_2 generation from natural gas with capturing CO_2 by condensing and infusing the separated carbon dioxide toward exhausted gas reservoirs for CO_2 storage and improved shale gas recoveries. When they originally suggested the basic concept of CSEGR.[124,125] They preliminarily evaluated the viability of CSEGR using simulation. The first research emphasis of the CSEGR study is on the technological and financial viability of CSEGR. To examine the technical and commercial viability of CSEGR, refs 64 and 167−170 performed several simulations and modeling.

References 171 and 169 described the practicability of CO_2-EGR in the Rio Vista gas reserves in the Midwest of America, US, alongside a qualitative analysis of the dissimilarities between CO_2 and natural gas concerning physicochemical characteristics (viscosity and density). The dual model mimicked the Rio Vista gas, proving the CO_2-EGR's technical viability.[34] In contrast, natural gas may have attained its present prominence lately, indicating that efforts to enhance natural gas output are a relatively recent phenomenon. Initial gas shale simulations[172] show that it is best to depress the gas reservoir before injecting carbon dioxide, regardless of whether the aim is to optimize gas output or store extra CO_2. The results of CO_2-EGR calculations and tests on shales and coals have shown that natural gas output rapidly declines after reaching a peak in just a few years. This is because the clay swells, causing gas flow to be impaired by permeability losses and infectivity.[33]

Simulation investigations demonstrated how tight rock (0.1 mD) slowed CO_2 breakout; hence, CO_2 breakthrough time increases when permeability drops. Homogeneity has a detrimental influence on CO_2 breakthrough curves as well. The ideal environment for beginning CO_2 injection in the field is one where the temperature and pressure are in the range of CO_2 subcritical conditions.[173]

5.2. EGR Experimental Studies. As global concerns regarding climate change intensify, coupled with a growing worldwide population and escalating demand for fossil fuels, researchers have undertaken numerous experimental studies to address these issues. These studies aim to demonstrate effective strategies for reducing greenhouse gas emissions. While enhanced gas recovery through CO_2-EGR offers promise, its practical application remains limited. EGR experimental studies are multidimensional, yielding data for isotherm models, kinetics, and thermodynamics modeling. Most studies' settings consider several characteristics that aid simulation and modeling.[174] Findings indicate that due to the improved competitive adsorbed factors, gaseous CH_4 is easily replaced by CO_2 injection.[175] According to the findings, as CH_4 adsorption in sandstone formations decreases, carbon dioxide. Reference 176 did an experimental analysis of carbon dioxide injection into a rock filled with gas. Increased injection pressures improve displacement efficiency, as shown in studies conducted on a sand pack filled with water and methane. Measurements taken at the pack's output show that injecting exhaust gas (15% CO_2, 85% N_2), pure N_2, and pure CO_2 corroborate the importance of pressure in maximizing gas recovery.

They examined recovery at various degrees of generated gas contaminants, Using multiple injection pressures and gases, and the findings revealed that CO_2 was always the optimum choice for recovery.[53]

5.3. Laboratory Experiments of Gas/Gas Displacement. The first operational experiments of gas/gas displacement for enhanced gas recovery were conducted in Hungary.[177] The investigation employed horizontally oriented, long-milled rock packs (lower permeability, which 100−300 mD) saturated with connate water at a pressure of 2500 kPa, showed methane recovery rates were in the 70−90% range. The experiment conducted at a temperature around 630Co, where carbon dioxide was always a gas. Initially, pure CO_2 and N_2 evaluated for their ability to displace methane. Additionally, it was found that the gas recovery increased, and some of the injected stream's gas had been retrieved when methane was replaced using a mixture of CO_2 and 20% methane.

Laboratory tests assessing the feasibility of replacing methane with CO_2 (liquid and supercritical states) have yielded promising results.[86] When using 1 × 1 ft., dry carbonate cores (with no connate water) arranged in a horizontal position and subjected to pressures ranging from 500 to 3,000 psi (3,448−20,685 kPa) and temperatures between 70 and 140 °F (21−60 °C), methane recovery at the CO_2 breakthrough point ranged from 73% to 87%, observed recovery rates improved at higher pressures.

Enhanced Gas Recovery (EGR) via gas−gas displacement offers a potential approach to prolonging the operational life and economic viability of many depleting volumetric gas reservoirs. Paramount Resources implemented a field trial of this concept in Alberta's Athabasca region, targeting a methane-rich layer above an oil sands interval conducted as a component of the GRIPE Project.

Reference 178 investigated enhancing shale gas recovery using CO_2, N_2, and CO_2/N_2 mixtures, focusing on how these gases influence methane recovery and gas flow and found that CO_2 injections lead to the highest methane recovery with a sharp displacement front, whereas N_2 injections result in the lowest. The study underscores the importance of selecting the right displacing fluid and gas mixture ratios to optimize gas recovery and CO_2 sequestration. Figure 10 illustrates the repeatability of the CO_2 and CH_4 displacement studies in the study's graphical representations. The consistent findings from several iterations confirm the dependability of the experimental

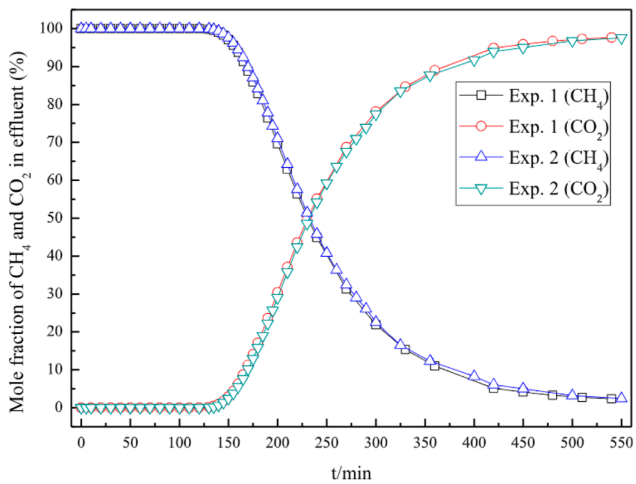

Figure 10. Reproducibility of CO_2−CH_4 displacement experiment. Reproduced from ref 178.

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data and the effectiveness of the approaches used. Figure 11 illustrates the composition fluctuations of the generated gas

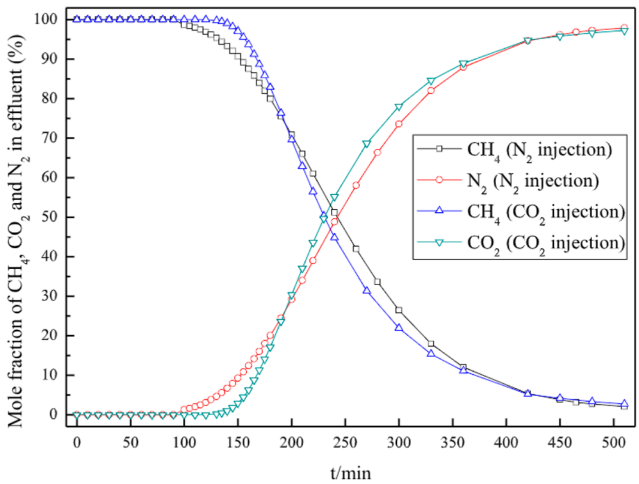

Figure 11. Illustrates the variations in the gas composition produced during CO_2 injection compared to N_2 injection. Reproduced from ref 178.

after injecting CO_2 and N_2. This study highlights the variations in gas recovery between two gases and reveals the flow dynamics that control displacement processes in shale deposits. Comparative insights are crucial for understanding the operational efficiency and effectiveness of using CO_2 and N_2 to enhance shale gas recovery in unconventional gas extraction for contemporary energy production strategies.

6. COST ANALYSIS AND ECONOMIC VIABILITY OF CCS AND CO_2-EGR

Although CCS technologies promise to help reduce carbon emissions, their economic feasibility is hindered by expensive initial and ongoing expenditures.[179,180] The economic impact of carbon capture and storage (CCS) and enhanced gas recovery (EGR) technologies is intricate and impacted by several variables beyond technological aspects.[40] (CCS) efforts experience significant variations in the economic environment due to a combination of factors. The factors involved include the size and location of the facility, inherent characteristics of the CO_2 emission source, and the particular CCS technology used. A field without access to infrastructure may be highly expensive. Therefore, for a CO2 injection and storage project to be implemented, the necessary infrastructure and a well-defined supply chain must first be built.[181] For example, it was difficult to launch a large-scale project in West Texas since the cost of starting CO_2 injection and storage operations needed $40 per ton of CO_2 with 18 billion tones.[182] Furthermore, it has been stated that CO_2 storage costs vary from $40 to $60 per ton.[183] A comprehensive understanding of cost engineering ideas, meticulous equipment design evaluations, and recognition of external variables such as legislative changes and market fluctuations are essential to comprehend CCS projects' many cost aspects and financial sustainability.[184,185]

The primary reason for the restricted use of Enhanced Gas Recovery (EGR) methods in real-world applications is the high costs associated with CO_2 collection, purification, and injection processes. In addition, economic difficulties are worsened by the potential dilution of methane quality due to its thorough mixing with injected CO_2, which requires advanced and costly gas separation methods to guarantee the output quality.[33] This section delves into the issues and the urgent need for more cost-effective and dependable CO_2 collecting and purification devices. The calculation for the CO_2-EGR and Sequestration project involves the capturing cost, compression cost, transportation cost, storage cost, and injection cost modules. The NPV computed for the study conducted by ref 163. Utilizing the following equations:

$$(w_{GP}F_{GP} + w_{OP}F_{OP})(1 - r_{tax}) + w_{SCO_2}F_{SCO_2} \tag{3}$$

$$w_{WP}F_{WP} + w_{1CO_2}F_{1CO_2} + C_{cap}F_{cap} + C_{CO_2-SEP}F_{PCO_2} + C_{capfacility} + C_{c-p} + C_{trans} + C_{storage} + C_{INJ} \tag{4}$$

$$NPV = \sum_{t=1}^{t_{dep}} \frac{\text{Revenue}_t\text{-Cost}_t}{(1 + IRR)^t} \tag{5}$$

Where W_{GP}, represents gas price per MSCF, $W_{OP,}$ represents oil price per STB, W_{SCO2}, indicates the CO_2 tax credit per metric ton, $W_{WP,}$ indicates the cost of disposing of water per stock tank barrel, W_{1CO2}, denotes the cost of injecting CO_2 per thousand standard cubic feet (MSCF), $C_{CO2,}$ represents the cost of separating CO_2 per thousand standard cubic feet of CO_2 separated, C_{CAP}, denotes the cost of capturing CO_2 per thousand standard cubic feet of collected CO_2, F_{GP}, depicts gas output during a certain duration t, F_{OP}, depicts oil output over a certain time frame t, reflects the amount of carbon dioxide stored during a certain time period t, F_{WP}, depicts water output within a certain time frame *t*, $F1_{CO2,}$ depict the amount of CO_2 supplied during a certain time frame *t*, F_{PCO2}, depict isolated CO2 from the overall field output within a certain time frame *t*, $t_{dep,}$ Is the cumulative production and accounting time for a field., F_{cap}, depict CO_2 sequestered over a certain time frame *t*, *IRR*, denotes the interest rate, $C_{capacitlity}$, is the yearly cost for capturing CO_2 from electricity generation, C_{C-P}, Is the annual expense for compressing and pumping CO_2 to the injection location., C_{trans}, is the yearly cost of carrying CO_2 to the capturing location, $C_{storage}$ is the annual cost for CO2 storage, C_{INJ} is the yearly cost for CO_2 injectino, $Revenue_t$ Is the cumulative income for a certain duration. *t*, Is the incremental cost for a certain duration *t*, and *NPV* is the project's net present value.

Reference 163 demonstrates the economic viability of CO_2 -EGR and Sequestration using cost analysis and objective functions such as Net Present Value (NPV). Reference 118 conducted a basic economic evaluation using EGR simulation data to determine the revenue and expenses associated with various optimization scenarios. They set a natural gas price of $9.15/MMBtu, a carbon tax price of $11/t, and a horizontal injection well drilling cost of $2211/m. The computation was as Economic gain was calculated by adding the money from natural gas to the savings in carbon tax and then subtracting the drilling cost. Seven examples were analyzed. Case 1 served as the baseline. Examples 2−5 used vertical injection wells without drilling expenditures, whereas Cases 6 and 7 included horizontal injection wells with drilling costs taken into account. As seen in Figure 12, all optimized scenarios resulted in considerably increased gas revenues and extra-economic advantages compared to the baseline scenario. Case 7 yielded the highest benefit from natural gas, amounting to $2,269.46 × 10∧3, while Case 3 secured the greatest economic benefit, reaching $2,427.86 × 10^3. Enhancing CO_2 injection techniques

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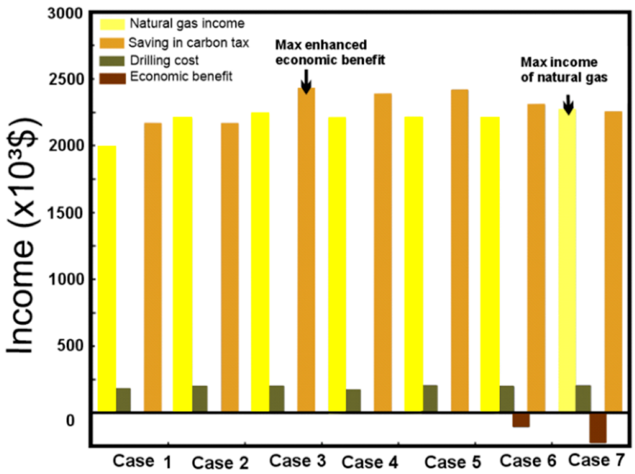

Figure 12. Economic analysis of baseline case (Case 1) and optimized cases. Reproduced from ref 118. Copyright 2021, with permission from Elsevier.

and refining simulation models, as outlined by ref 119 may lower economic obstacles by reducing gas dispersion; refs 186 and 187 emphasize the need for clear reporting and updating cost data to guarantee precise economic evaluations in Carbon Capture and Storage (CCS) projects. Further studies have confirmed that the economics of EGR are sensitive to factors like gas market prices, CO_2 supply costs, and injection-to-production volume ratios, as discussed by refs 85, 169, and 188.

7. FIELD APPLICATIONS AND A TEST PILOT

In general, CO_2-EGR development at depleting gas recoveries is still at the pilot stage, with little field testing having been conducted, summarized in Table 8.

This section provides a comprehensive overview of field applications and pilot studies in CO_2-EGR, highlighting various projects worldwide. However, it appears to lack a critical analysis of the gaps and limitations encountered in these field studies. For instance, it might benefit from discussing the scalability of these projects, long-term viability, environmental impacts, regulatory challenges, and the transferability of results from pilot to commercial scale. Additionally, comparative analysis between different geological settings or technological approaches could provide insights into optimization strategies for future projects. Addressing these aspects would offer a more rounded perspective on the state of CO_2-EGR field applications

Field-testing indicated the likelihood of enhancing gas recovery by CO_2 storage to boost gas recovery and capture significant carbon dioxide despite technical and financial obstacles. One of the fields' efforts to capture carbon dioxide in a depleting gas reserve has been the Otway Field in Australia. This facility held approximately 65,445 tons of carbon dioxide melded with methane.[189] The Cooperative Research Center for Greenhouse Gas Technologies (CO_2CRC) Otway field is a closely watched showcase pilot of carbon dioxide storage in a depleted gas field.[190,191]

To reduce the effects of climate change, we must create areas that need significantly greater processing and power, such as highly sour gases or heavy oils, while minimizing greenhouse gas (GHG) emissions. CCS is one of the solutions that used to reduce carbon emissions. This choice is by the (IPCC) as one of the most massive step strategies and technologies that are now in use during the 2007 report for governments on the Reduction of Weather Change.[192]

CO_2CRC site's monitoring showed that none of the atmosphere samples, soil gases, or groundwater aquifers had any trace components. The CO_2CRC Otway filed helped clarify the scientific process of CO_2 storage in exhausted shale gas, proving that CO_2 storage in depleted gas fields may be secure and efficient.[34] A simulation analysis as a pilot project in Taiwan's gas condensate depletion reservoir discovered that CO_2 removal and revaporization of condensate improved gas recoveries.[193] In a Nigerian area, an exhausted gas reserve in shale structure for carbon sequestrations with an expected total storage space of 147 MM tones of CO_2. A limestone-exhausted gas reservoir in southwest France was the site of the 2008 CO_2-capturing operation with a target carbon storage capacity of 75,000 tons annually over several years.[194] Two experiments conducted to pump CO_2 into an exhausted gas field (K12−B, Netherlands-ORC project). Natural gas that generated contained 13% CO_2, which separated and reintroduced once again into the gas storage tank.[195] Around 3.8 billion tons of carbon dioxide were estimated to be stored in another exhausted limestone storage tank off the coast of UK.[196]

The CO_2 injection revealed that CO_2 injection following gas exhaustion seemed to have enormous increments of recovered gas, which was about 10% of the original remaining gas. Contrarily, initial CO_2 injection reduced CH_4 generation due to the influence of the semisealing flaw, which lowers the effectiveness of carbon dioxide replacement.[172] In Germany's Altmark exhausted storage tank, a CO_2-EGR project titled CLEAN, including an anticipated injecting of 100,000 t of

Table 8. Some CO_2-EGR Field Applications

description	results	refs
Australia's Southeast: Otway Project	Sixty-five thousand four hundred forty-five tones of CO_2 and CH_4 combination have been stored at this location.	189
A field in Nigeria's Niger Delta.	It is around 3.32 and 3.54 km deep, which changes thermally around 33.66 and 13.46 km/h. There 147 MM ton potential storage capacity for CO_2.	197
South-Western France, specifically the depleted gas field of Lacq-Rousse	A carbon dioxide (CO_2) 75,000-ton-capacity storage facility annually is wanted.	194, 198, 199
The K12−B gas-reservoir is located in The Netherlands.	About 13% of the natural gas industry's CO_2 emissions captured and reinjected into the reservoir.	195
SNS offshore gas field in the United Kingdom.	3.8 billion tons of carbon dioxide stored, with a 10% increase in gas recovery from the original gas.	196, 200
German Altmark Field CLEAN project.	About 100,000 tons of (CO_2) stored in gas reservoirs.	201, 202
A Dutch gas field.	Capacity projected at 0.4 million tons of CO_2, with additional gas recovery limited to 1%.	203

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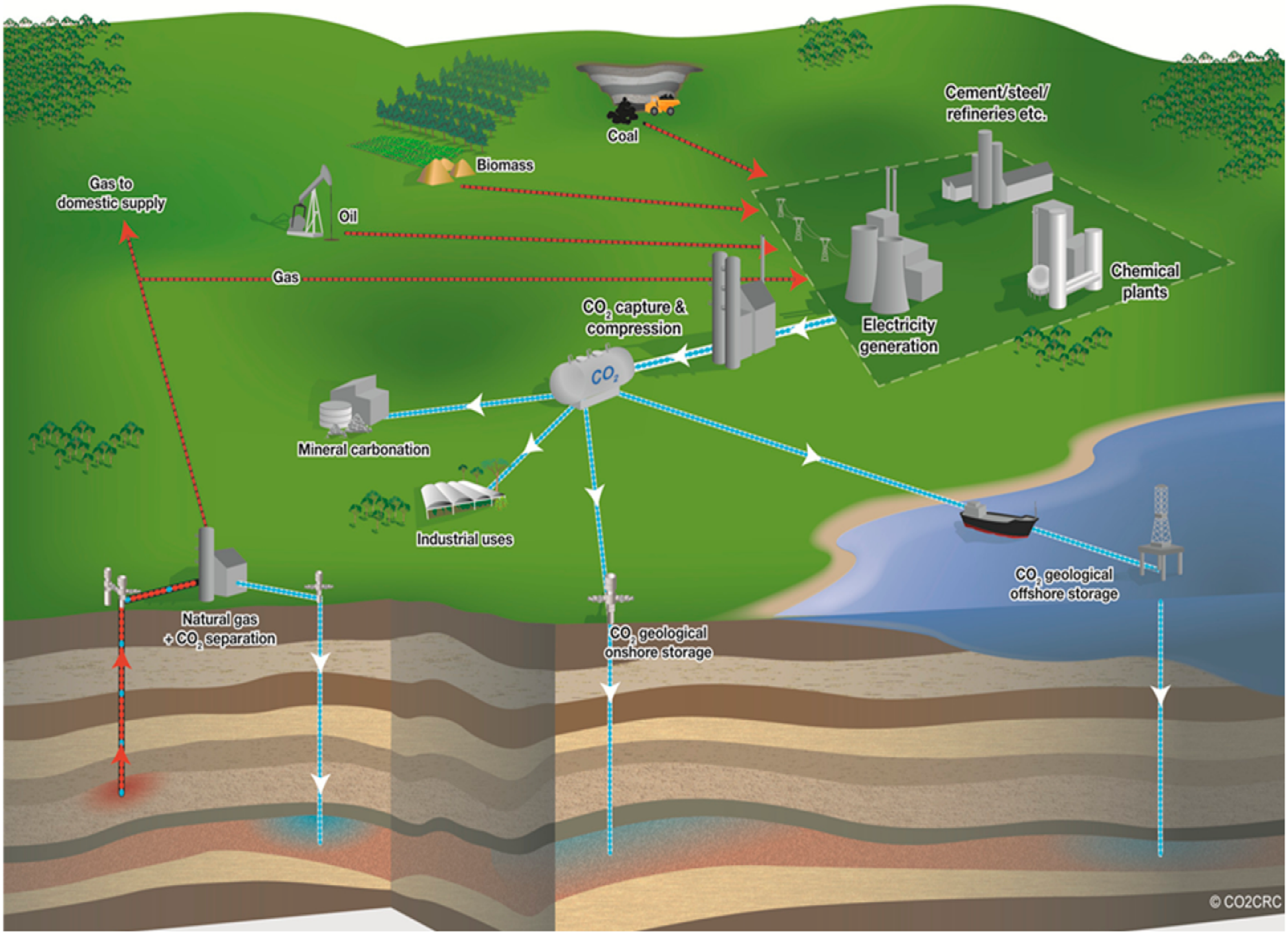

Figure 13. A schematic diagram of possible CCS projects. Adapted from ref 209. Copyright 2013, with permission from Elsevier.

carbon dioxide, underwent a feasibility assessment.[204] Experiments on the viability of storing 0.4 M tons of carbon dioxide in an exhausted storage tank in Holland found that just 1% more gas could be recovered.[203] Limited CO_2-EGR field pilot instances in shale rock have been recorded.[205] Although the Devonian Ohio Rocks have shown high capacities for storage of close to 28 Giga-tons, the site trial has been stalled due to the discovery of a packer's equipment problem.[120] An additional project test employing huff and puff CO_2 carried out in the Chattanooga shale formations. The findings demonstrated that typical hot CO_2 boosts methane recovery and eliminates condensate block.[206]

Previous research and application initiatives are significant for furthering the establishment of CSEGR technologies. CSEGR's fieldwork expertise and experience from such studies used to develop eligibility requirements for screening exhausted gas reservoirs for safe, dependable, and long-term CO_2 storage. Although improving methane output via CO_2 injection in rock reservoirs looked promising, only a few field pilots have been documented.[120,206]

Referenced research and example demonstration projects are crucial in advancing the future technical growth of (CO_2-EGR). The projects have significantly enhanced the field by providing experience and information, which has helped progress CO_2-EGR. Increasing the execution of these initiatives will enhance the transition to commercial-scale operations.

Based on field pilot experiments, it is evident that the 2009 EIA criteria for selecting CO_2 storage sites need revision. Reservoirs with permeability below 200 mD appropriate for CO_2 sequestration, despite previous recommendations suggesting otherwise. Revised criteria should include key elements recommended by ref 207, such as

(1) Evaluating cap-rock integrity after CO_2 injection at higher-pressure levels.

(2) Developing comprehensive abandonment protocols to mitigate CO_2 leakage poststorage.

(3) Studying the flow dynamics and paths of injected CO_2, which may vary from natural hydrocarbon motions.

Evaluating storage capacity involves considering the kind of reservoir and comparing the injection duration with the historical production period.

8. CARBON CAPTURE, SEQUESTRATION, AND STORAGE (CCS)

Sequestering CO_2 in geological formations emerges as a pivotal approach to mitigate anthropogenic carbon dioxide emissions.[47] The principle of CCS is clear-cut, encompassing three main stages: capturing CO_2 (typically from fossil fuel power plants), transporting the CO_2, and subsequently sequestering it in geological formations such as saline aquifers or oil reservoirs to prevent its release into the atmosphere in Figure 13.

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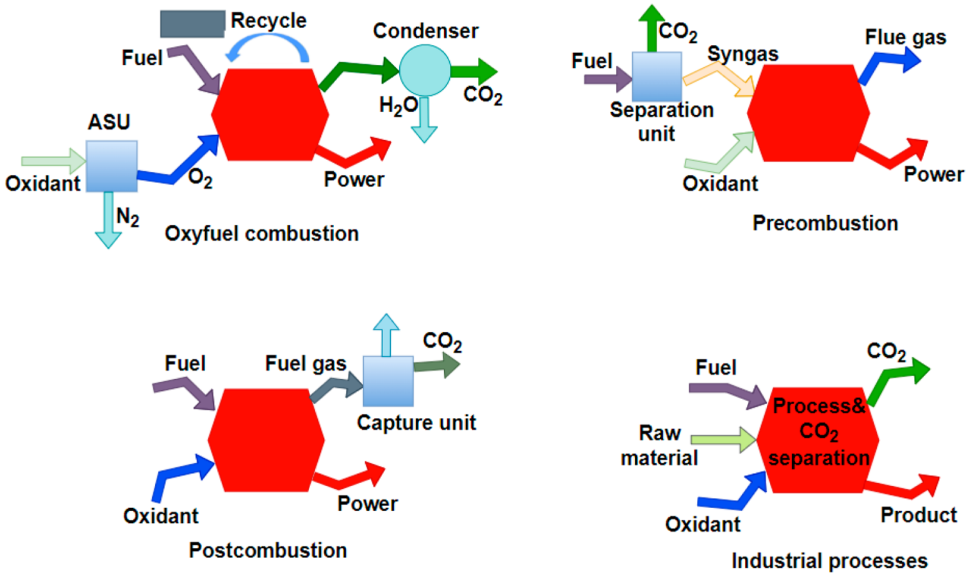

Figure 14. CO_2 processes. Adapted from ref 208. Copyright 2020, with permission from Elsevier.

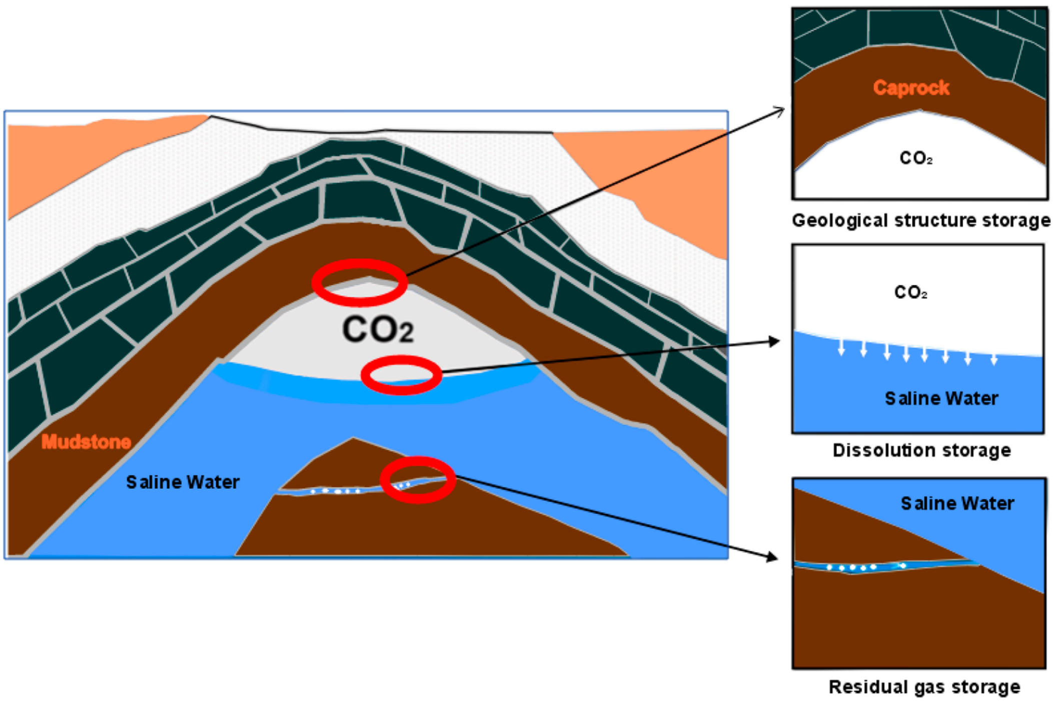

Figure 15. Formation structure physically traps injected CO_2. Adapted from ref 28. Copyright 2023, with permission from Elsevier.

The Figure 14, shows the schematic representations of CO_2 capture processes demonstrating oxyfuel combustion, pre-combustion, and postcombustion applied to the energy sector. Final diagram shows a generic carbon dioxide separation procedure a net-zero business uses.[208]

The paper describes four specific carbon capture and storage (CCS) methods used in industrial and power production settings to reduce CO_2 emissions. The oxyfuel combustion technique uses 100% oxygen for burning, resulting in an exhaust gas with a high CO_2 content, making it simpler to collect. In the precombustion method, fuel is transformed into a blend of hydrogen and carbon monoxide known as syngas, with CO_2 being removed before combustion. Postburning capture is the process of extracting CO_2 from flue gases after the combustion of fuel, providing a feasible solution for upgrading current power plants. Finally, certain industrial processes use CO_2 separation as a component of their operating procedures. Every method is created to effectively absorb and isolate CO_2, reducing its environmental footprint.

Reference 210 noted that after the cessation of injection, the supercritical CO_2 tends to migrate upward through the porous and permeable rock. This migration primarily driven by the buoyancy effect, which arises from the density disparity between the CO_2 and other fluids present in the reservoir. CO_2 may move horizontally along certain routes until it reaches a cap rock, fault, or another sealed gap. This technique will successfully reduce the continuous movement of carbon dioxide, as shown in Figure 15.

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Evaluating rock integrity is crucial for minimizing CO_2 leakage from depleted gas reservoirs. As noted by ref 157, depleted oil and gas reservoirs exhibit superior rock integrity compared to other CO_2 storage options, like saline aquifers. CO_2 reacts with water and produces carbonic acid, which can dissolve mineral structures in rocks. As the pH drops following the acid's consumption, calcium precipitates as calcium carbonate or calcium sulfate, depending on the available in refs 211 and 212. Therefore, CO_2 may either increase or decrease permeability and porosity after dissolving or precipitating minerals.

The CO_2 attains a supercritical state when subjected to temperatures beyond 31.04 C° and pressures surpassing 7.38 MPa. Reference 213 observed a significant increase in shale permeability, ranging from 3 to 8 times, after implementing a core flooding experiment. This increase attributed to the interaction between the shale and CO_2. Furthermore, the researchers observed a decrease in capillary breakthrough pressure, with reductions from 33% to 46%, attributed to this reaction. Reference 214 also documented the alteration in mineral composition, decrease in capillary-breakthrough pressure, and increase in porosity attributed to the exposure of shale samples to supercritical carbon dioxide ($scCO_2$). According to ref 215, the adsorption capacity of CO_2 and CH_4 exhibited a reduction after exposure to $scCO_2$. Several mineral oxides are included in shale, sandstone, and carbonate formation. In this category include the elements SiO_2, K_2O, NaO, CaO, MgO, Mg_2SiO_4, $Mg_3Si_2O_5$, and $CaSiO_3$. These mineral oxides react with CO_2 and water vapor under high pressures and temperatures. According to ref 216 research, various metal carbonates are primary products.

The reaction with water in monovalent metals like sodium and potassium is

$$M_2O + H_2O \rightarrow 2MOH \tag{6}$$

The reaction with water in divalent metals like (Ca_2^+) and (Mg_2^+) is

$$MO + H_2O \rightarrow M(OH)_2 \tag{7}$$

For each alkali or alkaline earth element, M denotes its metal ion. These metals undergo the same reactions when exposed to CO_2:

The alkali metals are

$$M_2O + CO_2 \rightarrow M_2CO_3 \tag{8}$$

When it comes to alkali earth metals:

$$MO + CO_2 \rightarrow MCO_3 \tag{9}$$

When metal hydroxide reacts with CO_2, the result is metal carbonate and water. Na_2O, a metal oxide, reacts with carbon dioxide in the following way:

$$Na_2O + H_2O \rightarrow 2NaOH \tag{10}$$

$$2NaOH + CO_2 \rightarrow Na_2CO_3 + H_2O \tag{11}$$

Other minerals, including $Mg_3Si_2O_5(OH)_4$, $CaSiO_3$, and Mg_2SiO_4, react with CO_2 to form metal carbonates.[216]

$$Mg_2Si_{2+2}MgCO_3 + SiO_2 \tag{12}$$

$$Mg_3Si_2O_5(OH)_4 +_3 CO_2 \rightarrow_3 MgCO_3 +_2 SiO_2 +_2 H_2O \tag{13}$$

$$CaSiO_3 + CO_2 \rightarrow CaCO_3 + SiO_2 \tag{14}$$

The amount of CO_2 that can dissolve in water decreases when the salinity, temperature, or pressure increases. Nevertheless, it found that the influence of temperature and pressure becomes negligible at 50 °C and 28 MPa.

According to ref 212, the composition of minerals and the surface area of the cap-rock affect the rate at which CO_2 reacts with the minerals in the cap-rock. Mechanical properties, including Young's modulus, Poisson's ratio, and compressive strength, may explain how carbon dioxide reactivity in cap-rock minerals weakens the cap-rock.[217]

In the context of exhausted oil and gas fields, CO_2 migration may effectively impeded by using abandoned wells that securely sealed with solid cement plugs. One of the potential hazards related to trapping is the occurrence of leakages that may occur behind the casing. Numerous investigations have been undertaken to examine the phenomenon of CO_2 leakage through geological formations and pre-existing wells.[218−222]

The potential of CCS to mitigate emissions is contingent upon several factors. These include the capacity of CCS to absorb, transport, and store CO_2, the occurrence of any leakage during transit, and the long-term storage capacity of CO_2.[58] International Energy Agency (IEA) has reported that, CCS initiatives should account for roughly 15−20% of total emissions of greenhouse gas reduction by 2050. Without CCS, the entire cost to halve CO_2 emissions by 2050 will climb by 70%.[223]

There are five CCS projects of commercial size operating in salty aquifers: the Sleipner project,[224,225] Snhvit-project,[226] In-Salah project,[227,228] Gorgon-project,[229] and Quest-project.[230]

According to estimates, geological formations may hold over 10,000 gigatones of CO_2, a significant amount compared to total human greenhouse gas emissions.[231] While saline aquifers have massive storage capacity, global progress in storing CO_2 in such aquifers remains sluggish due to the absence of economic benefits. As a result, various regulations relating to higher-priced greenhouse gas-emission levies may need to be developed, emphasizing the government's critical role in the widespread adoption of CCS.[49]

9. GLOBAL STORAGE CAPACITY AND CCS READINESS

Out of the 21 currently, active projects focused on CO_2 capturing, only 6 are committed explicitly to geologic storage. These projects are Snøhvit, Sleipner, Illinois Basin, Quest, Gorgon, and Qatar LNG. Currently, the combined storage capacity of these facilities is around 7 million metric tons per year (Mtpa), a fraction of the total captured amount of 40 Mtpa. Despite the consistent increase in CO_2 storage tonnage since 1972,[232] the sluggish pace of expansion continues to be a significant obstacle to achieving net-zero aspirations.

According to an analysis of historical and proposed projects up to 2025, the estimated storage rate in 2050 predicted to be 718 Mtpa. This figure falls far short of the 6,000−7,000 Mtpa rate.[233] A comprehensive historical storage high rate analysis explicitly focusing on dedicated storage projects only reveals a somewhat less optimiztic outlook. In a scenario with an average dedicated storage rate of 7 million tonnes per year (Mtpa) in 2020, it is anticipated that by 2050, assuming a consistent growth rate, the storage rate may reach 75 Mtpa.

The distinction between project types, precisely capture and storage, can be ambiguous, leading to a discernible trend of overestimating the volumes of CO_2 injected. The exact CO_2 amounts while comprehensive databases often detail the

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capture capacity of specific facilities, it is imperative to recognize that this information does not always align with the volume of sequestered material. The commonly used approach in published reports calculates the total storage capacity by multiplying the annual storage capacity by the number of years the facility has been operational. However, it is essential to note that this method oversimplifies the actual situation since it does not account for lower injection rates or instances when facilities cease injection before the stated duration.

Recent research endeavors have examined the adequacy of possible CCS capacity and aimed to determine the required quantity of CCS capacity based on specific emission paths.[234] Therefore, there is an increasing comprehension of the worldwide capacity for CO_2 storage, and the estimations of theoretical resources (including prospective unknown and found storage sites) are vast, reaching over 17,000 Gt.

Nevertheless, converting storage estimations into creating storage sites for CCS is sometimes intricate. This complexity arises from several factors, including the fluctuating growth rates of CCS projects and the challenge of accurately measuring the portion of a storage resource considered a bankable reserve. A bankable reserve refers to a storage resource with a confirmed size and pore space that may be effectively used. Moreover, the cost extent of available storage capacity is undetermined worldwide.

The levels of regional identification of capacity for storage exhibit variability and are at varying stages globally basis, including modest evaluations and comprehensive estimations.[235,236] In contemporary times, there has been a growing recognition of the significance of using historical well-development scenarios in offshore and mature hydrocarbon basins to comprehend bankable resources.[237] Offshore basins used by oil and gas corporations for a considerable period and have shown substantial volumetric importance can meet and surpass the resource demands on a (Gt) scale. The efficacy of CCS initiatives in certain nations gauged by their proficiency in executing projects, from the early design phase to the commencement of operations. CCS readiness evaluations[238] of various nations provide a comprehensive analysis of their position along the deployment spectrum. This includes countries that have made little progress or have low potential for CCS implementation, as well as those that are well prepared and actively engaged in exploring innovative approaches.

10. MONITORING TECHNIQUES FOR THE RISK EVALUATION OF CCS

When well integrity compromised in carbon capture, storage, and Enhanced Gas Recovery (CO_2-EGR) operations, it leads to several pathways for the unintentional release of CO_2. As shown in Figure 16, these leakage paths may occur via or near old wells, as well as in presently active wells that do not meet the necessary construction criteria. Wells that have not been properly sealed after being abandoned are likely to be major pathways for CO_2 movement, particularly when CO_2 is being injected and natural gas is being produced simultaneously.[239,240] This situation highlights the need to strictly follow well construction and abandonment standards to reduce the possibility of CO_2 leakage in CO_2-EGR projects. Monitoring and analyzing geotechnical responses is a significant challenge in implementing field-scale carbon dioxide (CO_2) storage projects. These responses, including deformation (strain) and temperature changes induced by CO_2 injection, are subsurface and ground-level.[241−243]

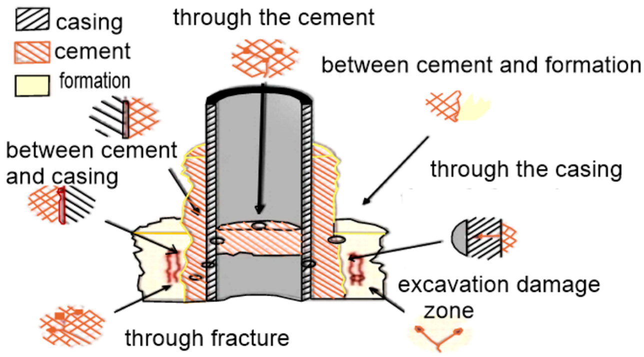

Figure 16. Leakage pathways for carbon dioxide (CO_2) ascent result from a loss of good integrity in the CSEGR. Adapted from ref 244. Copyright 2016, with permission from Elsevier.

Most often utilized monitoring techniques in CCS are cross-hole electromagnetic, gravimetry, 4D seismic, 3D seismic, microseismic, vertical seismic profiling, and others. Table 9, summarizes the benefits of technology for monitoring in CCS experiments.[244]

Distributed fiber optic sensing (DFOS), a fast-developing fiber-optic technique allowing long-term geophysical monitoring for CO_2 geological storage (CGS), has drawn greater interest to be investigated at various scales.[245−249] In addition, DFOS offers many inherent benefits, including its compact size, resistance to corrosion, resistance to high pressures and high temperatures (HPHT), immunity to electromagnetic interference (EMI), and more.[250,251,241]

During CO_2 injection and natural gas production, leakage along these channels may happen through or along abandoned wells and inadequately built operative wells.[244] The candidate technique for developing underground carbon dioxide (CO_2) storage must have a minimum residence length of 1000 years and a leakage rate of less than 0.1% per year.[252] Reference 241 had a field experiment, using DFOS technology in a 300-m well casing in Japan to monitor carbon dioxide as it injected into the surface. The results showed the minor stress caused by the gaseous injection of CO_2 could constitute a compressive deformation of the reservoir formation. In addition, the DFOS tool provides deformation information, such as depth and amount the target layer distorted in the subsurface, which may be utilized in geotechnical monitoring of cap-rock and wellbore integrity.[253] Monitoring and record matching are crucial in CCS evaluation since it is challenging to forecast the significant problems or dangers in CCS using simple simulation techniques.[254]

11. CHALLENGES AND PERSPECTIVES

The paper provides a detailed analysis of the challenges and perspectives associated with carbon capture, utilization, and storage (CCUS) for enhancing gas recovery, specifically focusing on CO_2-EGR in shale gas reservoirs. Here is a summary of the key points:

(1) Undertaking a CO_2 injection and storage project in a region without pre-existing infrastructure, such as the West Texas plan, might result in substantial expenses. Thus, setting up the necessary infrastructure and an efficient supply chain is essential for the effective implementation of such initiatives.

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Table 9. Outlines Advantages of Technology for CCS Experiment Monitoring

technology	description	advantages	limitations	applications	refs
Distributed fiber optic sensing (DFOS)	Enable comprehensive long-term geophysical surveillance for carbon dioxide geological storage (CGS)	provides deformation information	utilizing in geotechnical monitoring of cap rock and wellbore	Long-term tracking of the CCS in subsurface.	241
3D Seismic	Offers a three-dimensional representation of formation structures and the CO_2 plume.	High-quality data used for off-shore monitoring to detect CO_2 reservoirs exceeding 106 kg at depths of 1−2 km.	Quality influenced by the medium.	Monitoring carbon capture and storage activities in offshore locations.	30
4D Seismic	Utilizing time-lapse 3D seismic imaging to track the movement of a CO_2 plume over a period of time.	Useful for tracking the movement of CO_2.	Inconsistent noise level due to changes in survey circumstances.	Continuous monitoring of CCS over an extended period.	255
Microseismic	Utilizes downhole geophones to detect waveform data for analyzing the injection and movement of CO_2 in the subsurface.	Correlates with the duration of CO_2 injection.	Deployment is necessary in vertical wells.	Monitoring of subsurface injection.	256
Vertical Seismic Profiling	Utilizes permanently placed hydrophone arrays, subsurface geophones, and fiber optic cables for extended monitoring purposes.	Acquires intricate geological formations.	Needs installed permanently.	Long-term tracking of the CCS.	257
Gravimetry	Finds CO_2 by detecting changes in fluid density caused by CO_2 input.	Gives information on where CO_2 is.	The form of the CO2 stream changed the result.	Finding the position of CO_2.	258
Cross-hole Electromagnetic	A simple way to find out about the qualities of the ground and monitor where the CO_2 is.	Gives detailed information about the properties of the ground.	This only works for small spaces.	Monitoring in between wells.	259

(2) Enhancing the efficiency of CO_2 sequestration in shale formations faces challenges, including the intricate CO_2-shale interactions, refining injection methodologies, and advancing the forecasting of sequestration results. Enhancing simulation capabilities through the integration of nonisothermal factors and thorough reservoir characterization is crucial. Conducting field pilot experiments that focus on CO_2-EGR during the interim period is crucial for validating simulation results and offering evidence to back the concept's feasibility.

(3) Utilizing geologic sequestration techniques to store carbon dioxide poses significant challenges due to its limited use and high associated costs. Issues regarding the quick blending of injected CO_2 with existing methane (CH_4) impact the quality of organic gas supply, creating a major challenge for the implementation of CO_2 storage and enhanced gas recovery. Research in molecular simulations and experimental studies has demonstrated potential in assessing the EGR efficiency and CO_2 storage capacity in shale formations. These studies contribute to analyzing the impact of reservoir parameters on the adsorption behavior of both CO_2 and methane, which could enhance gas recovery and sequestration techniques.

(4) Adding methane to carbon dioxide injection may lead to contamination of the resulting natural gas, lowering its market value and requiring extra costs for impurity removal. Ensuring the reservoir remains intact over long geological periods is essential to avoid any leakage into groundwater and nearby land.

(5) Loss of well integrity in Carbon Storage and Enhanced Gas Recovery (CSEGR) may provide pathways for CO2 to escape. This might occur either during the CO2 injection phase or the natural gas extraction process. It is essential to design and maintain wells correctly to avoid leaks, which might impede the performance of the Carbon Storage and Enhanced Gas Recovery (CSEGR) process and create environmental hazards. Enhanced management of well structures is crucial to reduce the risk of CO2 leakage and preserve the stability of the storage site.

(6) The diversity found in gas reservoirs, with differences in permeability, porosity, and rock properties, adds complexity to CO_2-EGR operations, necessitating customized strategies for each reservoir.

12. CONCLUSION

The integration of Carbon Dioxide Enhanced Gas Recovery (CO_2-EGR) into depleted gas reservoirs for carbon capture and storage (CCS) purposes presents a strategic approach to achieving carbon neutrality and enhancing sustainable energy production. Leveraging the extensive knowledge obtained during the exploration and development phases of gas reservoirs, which includes understanding reservoir capacity, permeability, porosity, and the integrity of cap rocks and seals, CO_2 injection into these formations emerges as a highly viable method. This viability is further underscored by the existence of essential infrastructure such as pipelines and wells, which can be readily adapted for CCS, thereby minimizing the need for extensive modifications.

Empirical evidence from field applications and pilot tests across diverse geographic regions, including the Otway field in

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Australia, the Niger Delta, South-Western France, The Netherlands, the UK, and Germany, attests to the feasibility and efficacy of CO_2-EGR in both augmenting gas recovery and securely storing CO_2. Such projects highlight the dual benefits of utilizing exhausted gas fields for CO_2 storage, while simultaneously enhancing gas production. Comparative studies further reveal that CO_2 injections are more effective than nitrogen (N_2) injections in improving shale gas recovery, show casing the superior capability of CO_2-EGR technologies.

Moreover, advancements in monitoring techniques for CO_2 injection processes have significantly mitigated environmental and resource risks, bolstering confidence in CCS projects. While the costs associated with CCS not entirely negated by enhanced gas recovery, the economic benefits of CO_2-EGR, including potential cost reductions, render CCS a more viable option for widespread adoption.

The critical role of government policies and incentives in ensuring the economic viability of CCS initiatives cannot be overstated. Regulatory support, alongside continuous research into gas composition changes and flow dynamics within shale formations, is imperative for optimizing gas recovery and CO_2 sequestration strategies. Investing in CO_2-EGR not only furthers environmental sustainability efforts but also improves gas recovery efficiencies, overcoming challenges to pave the way for a more sustainable and environmentally friendly future.

In conclusion, CO_2-EGR presents a compelling case for both environmental and economic sustainability in the context of CCS. Through leveraging existing infrastructure, demonstrating effectiveness via global projects, and supported by advancements in monitoring and regulatory frameworks, CO_2-EGR stands as a pivotal strategy in the quest for carbon neutrality and enhanced gas recovery. The continuous evolution of CO_2-EGR technologies, backed by robust policy frameworks and research, is essential for overcoming barriers and realizing the full potential of CCS in contributing to a sustainable energy future.

■ AUTHOR INFORMATION

Corresponding Author

Yankun Sun − *Hubei Provincial Key Laboratory of Oil and Gas Exploration and Development Theory and Technology, China University of Geosciences, Wuhan 430074, China; Key Laboratory of Tectonics and Petroleum Resources, China University of Geosciences, Ministry of Education, Hubei, Wuhan 430074, China;* orcid.org/0000-0001-7233-7858; Email: sunyankun@cug.edu.cn

Authors

Ghamdan AL-khulaidi − *Hubei Provincial Key Laboratory of Oil and Gas Exploration and Development Theory and Technology, China University of Geosciences, Wuhan 430074, China; Key Laboratory of Tectonics and Petroleum Resources, China University of Geosciences, Ministry of Education, Hubei, Wuhan 430074, China;* orcid.org/0009-0001-1347-1878

Ahmed.G. Alareqi − *Hubei Provincial Key Laboratory of Oil and Gas Exploration and Development Theory and Technology, China University of Geosciences, Wuhan 430074, China; Key Laboratory of Tectonics and Petroleum Resources, China University of Geosciences, Ministry of Education, Hubei, Wuhan 430074, China;* orcid.org/0000-0002-1200-6843

AL-Wesabi Ibrahim − *School of Automation, China University of Geosciences, Wuhan 430074, China;* orcid.org/0000-0003-1848-427X

Abubakar Magaji − *Key Laboratory of Deep Geothermal Resources, MNR,, China University of Geoscience, Wuhan 430074, China;* orcid.org/0009-0007-5912-4314

Xu Zhang − *Energy Exploration Center, Geological Exploration Technology Institute of Anhui Province, Hefei 230041, China;* orcid.org/0000-0002-2849-5792

Complete contact information is available at: https://pubs.acs.org/10.1021/acs.energyfuels.3c05211

Author Contributions

Ghamdan AL-khulaidi: Writing the original draft, Methodology, Material preparation, Data collection and analysis. Yankun Sun: Supervision, Project administration, Methodology, Formal Analysis, Software, Reviewing and Editing. Ahmed G. Alareqi: Methodology, Conceptualization, Reviewing and Editing. AL-Wesabi Ibrahim: Reviewing and Editing. Abubakar Magaji: Software, Investigation, Writing and Editing. Xu Zhang: Funding Support, Reviewing and Editing. All authors read and approved the final manuscript.

Notes

The authors declare no competing financial interest.

Biographies

Ghamdan AL-khulaidi, born in 1994 in Sana'a, Yemen, earned his B.S. in Oil and Natural Gas Engineering from China University of Geosciences, Wuhan, in 2018. He is now a master. Scholar at the same university, focusing on carbon capture storage techniques and technologies to enhance gas recovery aiming to reduce greenhouse gases, AI applications in engineering, machine learning algorithms, and numerical simulations using COMSOL Multiphasics.

Yankun Sun, Distinguished Professor at China University of Geosciences, specializes in unconventional oil and gas, geostorage of CO2, and intelligent subsurface monitoring. With advanced degrees from Fuzhou University and the Chinese Academy of Sciences, he joined many international societies like AGU, ACS, SPE, JpGU and ARMS and he is also the editorial member of Sustainability, Data in Brief and Advances in Geo-energy Research.

Ahmed G. Alareqi, born in 1989 in Taiz, Yemen, earned his B.S. in Oil and Natural Gas Engineering from Aden University in 2012 and his Master's from China University of Geosciences, Wuhan, in 2019. He is now a Ph.D. scholar at the same university, focusing on drilling cementing techniques and technologies to improve cement and drilling efficiency, aiming for optimal compressive and shear bond strength and enhancing penetration rates in basement rock drilling.

AL-Wesabi Ibrahim earned his B.S. in Electrical Power and Machines Engineering from Sana'a University, Yemen, in 2014 and his Master's in Control Science and Engineering from China University of Geosciences, Wuhan, in 2020. Currently, he is a Ph.D. candidate at the same university, focusing on power electronics, and artificial intelligence in engineering, innovative grid technologies, control methods for PV systems, hybrid renewable energy, and optimization through machine learning algorithms.

Abubakar Magaji earned his B.S. in Civil Engineering from Abubakar Tafawa Balewa University, Nigeria 2011, and a Master's in Civil Engineering from Cyprus International University in 2018. He is pursuing his Ph.D. at the China University of Geosciences, focusing on enhanced geothermal systems, AI applications in engineering, deep closed-loop systems, hybrid renewable energies, machine learning algorithms, and numerical simulations using COMSOL Multiphasics.

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Xu Zhang, born in 1990 in Chaohu City, Anhui Province, China. He received his Ph.D. in Chinese Academy of Geological Science and a postdoctor of University of Science and Technology of China. He is currently a senior engineer of Geological Exploration Technology Institute of Anhui Province.

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