



Review

A Review of CO₂ Storage in View of Safety and Cost-Effectiveness

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Abstract: The emissions of greenhouse gases, especially CO_2 , have been identified as the main contributor for global warming and climate change. Carbon capture and storage (CCS) is considered to be the most promising strategy to mitigate the anthropogenic CO₂ emissions. This review aims to provide the latest developments of CO₂ storage from the perspective of improving safety and economics. The mechanisms and strategies of CO₂ storage, focusing on their characteristics and current status, are discussed firstly. In the second section, the strategies for assessing and ensuring the security of CO₂ storage operations, including the risks assessment approach and monitoring technology associated with CO₂ storage, are outlined. In addition, the engineering methods to accelerate CO₂ dissolution and mineral carbonation for fixing the mobile CO₂ are also compared within the second section. The third part focuses on the strategies for improving economics of CO₂ storage operations, namely enhanced industrial production with CO₂ storage to generate additional profit, and co-injection of CO₂ with impurities to reduce the cost. Moreover, the role of multiple CCS technologies and their distribution on the mitigation of CO₂ emissions in the future are summarized. This review demonstrates that CO₂ storage in depleted oil and gas reservoirs could play an important role in reducing CO₂ emission in the near future and CO₂ storage in saline aquifers may make the biggest contribution due to its huge storage capacity. Comparing the various available strategies, CO₂-enhanced oil recovery (CO₂-EOR) operations are supposed to play the most important role for CO₂ mitigation in the next few years, followed by CO₂-enhanced gas recovery (CO₂-EGR). The direct mineralization of flue gas by coal fly ash and the pH swing mineralization would be the most promising technology for the mineral sequestration of CO₂. Furthermore, by accelerating the deployment of CCS projects on large scale, the government can also play its role in reducing the CO₂ emissions.

Keywords: CO₂ storage; CO₂ utilization; security assessment; cost-effectiveness; CO₂ storage projects

1. Introduction

In the last few centuries, the CO_2 concentration in the atmosphere has already risen above 410 ppm from a level of below 300 ppm in pre-industrial times [1,2]. As shown in Figure 1, the continuous rise in the Earth's surface temperature appears to be strongly linked with atmospheric concentration of

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 CO_{2} , which suggests that CO_{2} may be the main contributor to global warming and climate change. In addition, it makes up an estimated 77% of greenhouse gases [3,4].

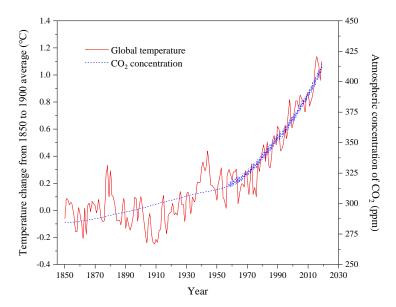


Figure 1. Correlation between atmospheric concentration of CO_2 and the global temperature since 1850s (Data from [1,2]).

In addition, the CO₂ emission may increase the frequency of extreme extratropical cyclones. Unless the greenhouse gas emissions are efficiently mitigated, the extratropical cyclones are projected to more than triple in number by the end of this century in both Europe and North America [5]. In response to such intense global climate change, the Intergovernmental Panel on Climate Change's (IPCC) assessment suggested that the average global warming should be limited less than 2 °C within this century on the basis of estimated results from integrated assessment models (IAMs) [6].

To achieve this goal, carbon capture and storage (CCS) needs to be promoted, as it is currently the most effective technology for slowing down atmospheric CO_2 enrichment and extenuating associated potential climate problems [7]. According to the estimation by the IEA [8], CCS alone could undertake almost a 19% reduction in global CO_2 emissions by 2050 (Figure 2). Furthermore, the overall cost of achieving the same emission reduction targets will increase by 70% without CCS [9], which highlights the importance of CCS on the mitigation of CO_2 emission from the economic point of view as well.

There are 51 CCS engineering projects across the world, mainly scheduled in North America, Australia, China, and Western Europe, although only 19 CCS projects are currently in operation (Figure 3) [10]. Although CCS has been proven to be technically feasible, the risks and economics associated with CCS challenges its large-scale application. Consequently, the contribution of CCS is still very limited in attenuating climate change because of the high cost and safety risk associated with CO₂ leakage [11,12]. More efforts on improving the safety and economics are required to develop the CCS technology, gaining support from government, and improving public acceptance to accelerate the application of large-scale CCS.

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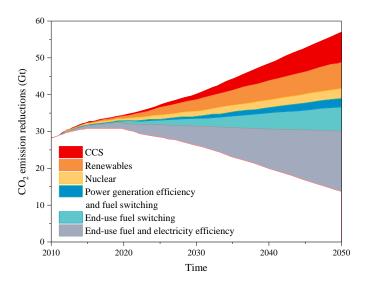


Figure 2. International Energy Agency (IEA) forecasts of key technologies for CO₂ emission reductions (modified from [8]).

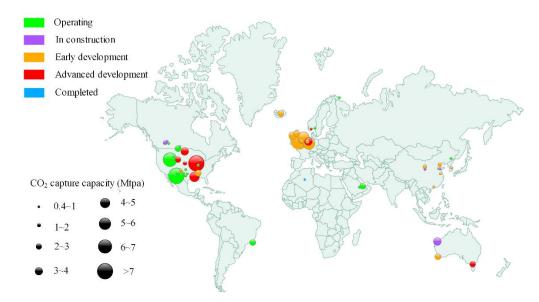


Figure 3. Commercial-scale integrated carbon capture and storage (CCS) projects around the world. Circle size is proportional to the CO_2 capture capacity, and the color indicates different stages of the lifecycle of the project (data from [10]).

In the last decade, nearly every aspect of CCS has been discussed extensively [13–44], see Table 1. However, the strategies for improving the safety and economics have not been reviewed in detail. In addition, the CCS technology is developing rapidly, and the recent progress needs to be reviewed and analyzed.

This review attempts to discuss the most recent developments on addressing challenges associated with assessing and decreasing the risks of leakage, cutting the cost of CO_2 storage and promoting the developments of commercial-scale CCS projects. Firstly, different mechanisms and strategies of CO_2 storage are summarized and discussed. Then, the risk assessment of CO_2 storage and strategies for accelerating CO_2 dissolution and mineral carbonation are reviewed. Finally, the strategies for improving cost-effectiveness, including enhanced industrial production with CO_2 storage and co-injection of CO_2 with impurities, are examined.

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Table 1. Summary of review literature on CCS technology.

Research Fields	Ref.	Parious Scane
Research Fields		Review Scope
	[13]	Review of the application of CO ₂ for enhanced oil and gas recovery
	[14]	Review of CO ₂ capture and reuse technologies, highlighting the strategies of CO ₂ capture in variety of scenarios, and the state of the art for CO ₂ utilization
CO ₂ capture and	[15]	Review of CO ₂ capture, utilization, and storage (CCUS) in Chinese Academy of Sciences,
utilization	[15]	highlighting the strategies for CCUS in China
	[16]	Review of the property impacts of CCS, highlighting the effect of uncertainties in
		thermal–physical properties on the design of components and processes in CCS Review of CCS highlighting the CO ₂ capture technologies, the pilot plants, and the economic
	[17]	and legal aspects of CCS
	[18]	Review of CO ₂ enhanced coal bed methane recovery, highlighting the CO ₂ storage trials in the
	[10]	San Juan Basin in USA, and the estimation of CO ₂ storage capacity in coal seams
	[19]	Review of CCUS technologies highlighting the engineering projects and their developments in China
	[20]	Review of CCS highlighting the findings obtained in CCS operational projects including the
	[20]	technologies of CO ₂ capture, separation, transport, and storage
	[21]	Review of CCS highlighting the options for CO ₂ storage, the evaluation criteria for CO ₂ storage
	[21]	sites, and the major CO ₂ storage projects
	[22]	Review of biomass with CCS (Bio-CCS), highlighting the economics and global status of Bio-CCS, and the role of Bio-CCS in the food–water–energy–climate nexus
Options for CO ₂		Review of CO ₂ storage in saline aquifers, highlighting the geological and operation parameters,
storage and CCS	[23]	and the monitoring technologies for existing saline aquifers storage operations
projects	[24]	Review of the CCS in a coal-fired plant in Malaysia, highlighting the choices of coal plants and
		the capture technologies Povious of CO storage in caling formations, highlighting the modeling of calculility transping
	[25]	Review of CO ₂ storage in saline formations, highlighting the modeling of solubility trapping Review of mineral carbonation (MC) technologies for CO ₂ sequestration, highlighting the
	[26]	mechanisms of MC technologies and their contribution in decreasing the cost of CCS
	[27]	Review of CCS projects and future opportunities, highlighting the technical details and business
	[]	plan for CCS projects
	[28]	Review of CO ₂ storage projects in China, highlighting the CO ₂ source, and CO ₂ storage strategies in China
	[20]	Review of CO ₂ mineralization product forms, highlighting the mineralization process for
	[29]	CO ₂ storage
	[30]	Review of CCS by using coal fly ash, highlighting the feasibility and prospects of CCS using coal fly ash
		Review of the relative permeability and residual trapping in CO ₂ storage systems, highlighting
CO ₂ -brine-rock	[31]	the estimating and measuring methods
systems		Review of the geochemical aspects of CO ₂ storage in saline aquifers, highlighting the advantages
-	[]	of CO ₂ storage in saline aquifers, and the CO ₂ -brine-rock interactions in the aquifers
	[33]	Review of geomechanical modeling of CO ₂ storage, highlighting the numerical methods and their application in the modeling of ground deformation, faults, and fracture propagation
	[24]	Review of CO ₂ sequestration highlighting the trapping mechanisms and the flow of CO ₂ brine
	[34]	in porous media system
	[35]	Review of the cement degradation in CO ₂ -rich conditions of CCS projects, highlighting the
	r1	degradation of Portland cement Povious of the rick assessment of COs storage, highlighting the regulations and strategies of rick
	[36]	Review of the risk assessment of CO ₂ storage, highlighting the regulations and strategies of risk assessment for CO ₂ storage
Well integrity and	[37]	Review of the isotopic composition of CO ₂ for leakage monitoring in CCS project, highlighting
risk assessment	[37]	the stable isotopes as a tracer for injected CO ₂
	[38]	Review of the integrity of existing wells for CCS, highlighting the mechanical well failure and chemical issue due to cement carbonation
	[20]	Review of well integrity of CCS, highlighting the corrosion of metallic and cement, and the
	[39]	remedial measures
	[40]	Review of caprock sealing mechanisms for CO ₂ storage, highlighting the problems associated
		with CO ₂ leakage, the leakage paths, and the factors that affect leakage Review of CO ₂ storage highlighting the capacity estimation of storage sites, the monitoring
	[41]	technologies, and the simulation tools for CCS
	[42]	Review of CO ₂ storage and caprock integrity, highlighting the major CCS project in operation
	[42]	and CO ₂ migration in the reservoirs
Storage efficiency	[43]	Review of CO ₂ storage efficiency in saline aquifers, highlighting the factors that affect CO ₂
and environmental	[10]	Plume migration and the methods to estimate the storage capacity
considerations	[44]	Review of environmental considerations for CO ₂ storage in a sub-seabed, highlighting the potential ecological impacts
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2. Mechanisms and Strategies of CO₂ Storage

2.1. Mechanisms of CO₂ Storage

As shown in Figure 4a, there are four main trapping mechanisms of CO_2 storage involving (1) structural and stratigraphic, (2) residual, (3) solubility, and (4) mineral trapping [42]. With structural and stratigraphic trapping, which is the most dominant trapping mechanism, once CO_2 is injected subsurface, it will rise to the top of geological structures due to the buoyancy effect but stay below the impermeable caprock. In residual trapping, the injected CO_2 displaces formation fluid when it flows through the formation rock. The displaced fluid disconnects and traps the remaining CO_2 within the pores of rocks due to the capillary force [45]. In the residual trapping mechanism, the saturation of trapped CO_2 is at least 10% and can reach more than 30% of the pore volume in some reservoir rocks [46,47]. In solubility trapping, CO_2 dissolves in formation fluids and becomes immobile, thereby decreasing the volume of free CO_2 [48]. This dissolved CO_2 will slightly increase the density of formation fluid by around 1%. It is sufficient to promote the convection flow with such a small density difference [49], which is also in favor of the trapping of CO_2 . The solubility of CO_2 in groundwater ranges from 2% to 6%, and it decreases with the rising temperature and salinity [47]. For the mineral trapping mechanism, CO_2 is trapped by geochemical reactions in reservoir, usually precipitating as carbonate, which can trap the CO_2 in immobile secondary phases effectively [50].

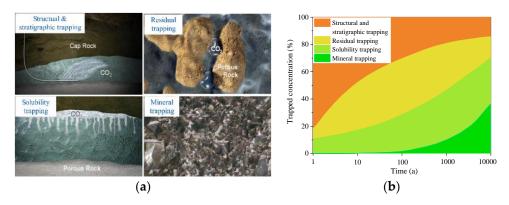


Figure 4. (a) The four main CO₂ trapping mechanisms [51]; (b) the contribution of four CO₂ trapping mechanisms with time (modified from [52]).

As shown in Figure 4b, these trapping mechanisms play different roles on CO_2 storage in the time scale from 1 to 10,000 years. Clearly, structural trapping plays a vital role in the initial stage of CO_2 storage, and its effect weakens gradually. The residual trapping and solubility trapping show a significant impact in tens of years and lock up a certain amount of CO_2 for thousands of years. With respect to mineral trapping, it begins to show a significant effect almost around a hundred years and plays a key role at a geological timescale.

2.2. Strategies of CO₂ Storage

2.2.1. CO₂ Storage in Saline Aquifers

 CO_2 storage in saline aquifers is one of the most important options due to the huge amount of storage capacity, which is estimated to be sufficient for the sequestration of 10,000 Gt of CO_2 , namely the emissions from large stationary sources for more than 100 years [32,53,54]. Compared with the other storage sites, the saline aquifers usually possess more wide distribution and greater regional coverage. Therefore, it has a better chance to be located near the CO_2 emission sources, which could reduce the cost of CO_2 transportation [47,55]. The most crucial issue brought by the sequestration of CO_2 in saline aquifers is pressure build up and CO_2 plume migration in formation, which has the potential to lead to the fracturing of formation and reactivation of faults and leakage of CO_2 ,

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which should receive more attention [56]. Birkholzer et al. [57] conducted a numerical simulation to determine the influence of large-scale CO_2 storage with an injection rate of 1.52 million tons per year (Mtpa) in an open saline aquifer. Their results indicated that there is significant pressure build up in the formation more than 100 km away from the injection zone, but the CO_2 plume migration is rather small—that is, around 2 km—and is concentrated on the top of saline aquifer due to the buoyancy effect. They also showed that the pressure perturbation may reach shallow groundwater formation when there is a caprock with relatively high permeability (higher than 10^{-18} m²) between the saline aquifer and the shallow layers. However, the migration of reservoir fluids into groundwater formation is extremely unlikely. This demonstrates the safety of large-scale CO_2 storage in saline aquifers.

There are mainly five commercial-scale CCS projects in saline aquifers, including the Sleipner project [58–60], the Snøhvit project [61], the In Salah project [62,63], the Gorgon project [64], and the Quest project [65]. Detailed geological and engineering data are shown in Table 2.

Num.	Project	Injection Rate (t/d)	Permeability (mD)	Depth (m)	Thickness of Reservoir (m)	Thickness of Caprock (m)	Reservoir Temperature (°C)	Reservoir Pressure (MPa)	Ref.
1	Snøhvit	2000	450	2550	60	30	95	28.5	[23,61]
2	Sleipner	2700	3000	1000	250	75	37	10.3	[23,58, 66]
3	In Salah	3500	13	1800	20	900	90	17.9	[23,63]
4	Gorgon	10,410	25	2300	280	250	100	22	[23,64]
5	Quest	2960	100	2000	40	70	55	18.9	[65–68]

Table 2. Large-scale CCS project in saline aquifers.

Regarding the Sleipner project, CO_2 was separated from the methane produced at the Sleipner field in the North Sea. Then, the CO_2 was injected into a regional saline aquifer within the Utsira Sand formation, and a total of 18 million tons were injected by 2018 since its initiation in 1996 [60]. Based on the engineering experiences from the Sleipner project, the CO_2 separated from the liquefied natural gas (LNG) project was injected to the deeper Tubåen Formation at a rate of 2000 tons per day in the Snøhvit CCS project, which is located in the Barents Sea. The project was launched in 2008 with a total amount of 1600 ktons of CO_2 injected until August 2012, and it is expected that about 23 million tons of CO_2 will be stored there based on the projected lifetime of the Snøhvit LNG project [61,69].

The CCS project at In Salah, Algeria, is one of the world's pioneering CCS projects. More than 3.8 million tons of CO_2 have been injected to the Carboniferous sandstone at the Krechba field since 2004 [62]. This CCS project is unique due to the diversity of monitoring methods, including satellite monitoring and 4D seismic data, which have been used to monitor the response of formation to CO_2 injection. Meanwhile, the accessibility of these monitoring data to the public is very high [62,63,70–77], so it could be a commendable case to study the CCS in saline aquifers.

The Gorgon CCS project located in the northwest of Australia, and it owns a Jurassic saline reservoir in the Dupuy Formation. In the lifetime of the Gorgon project, more than 120 million tons of CO₂ is planned to be injected at a rate of approximately 3.8 Mtpa [64].

The Quest CCS project began in 2015 and is designed to store the CO_2 from an existing facility for upgrading heavy oil in Scotford of Alberta, Canada. It is expected that approximately 27 million tons of CO_2 can be injected to the Basal Cambrian Sands formation at an injection rate of 1.08 Mtpa through three to eight vertical wells [65].

Aside from the previously mentioned large-scale CCS projects, there are some small-scale projects as well. These include the Ketzin pilot site [78,79], the Illinois Basin-Decatur Project [80], and the Shenhua CCS demonstration project [81]. These projects have been conducted with detailed modeling and monitoring during operation, which demonstrates the safety of this technology and helps increase the public acceptance.

Although the storage capacity of saline aquifers is huge, the overall progress of CO₂ storage in such aquifers throughout the world is still slow due to the lack of financial incentives. Therefore, some

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policies related to the taxes on carbon emission with a higher price might need to be formulated, which highlights the important role of the government on the application of CCS at a large scale.

2.2.2. CO₂ Storage in Depleted Oil and Gas Reservoirs

There are many advantages of CO_2 storage in depleted oil and gas reservoirs. Firstly, oil and gas reservoirs have a large amount of existing equipment installed on the surface and underground, which could be reused for CO_2 storage with only minor modification. Secondly, the seal quality and integrity of the caprock are guaranteed and have been comprehensively characterized during the exploration and production process [56]. Thirdly, the extent of pressure perturbations and induced stress changes is much smaller in depleted oil and gas reservoirs compared with aquifers because of the long-term extraction of oil and gas [56]. Compared with depleted oil reservoirs, the depleted gas reservoirs are more favorable for CCS due to higher ultimate recovery and compressibility of gas, a larger storage capacity per pore volume is available [82–84]. Comparing the types of reservoirs used in this form of storage, condensate gas reservoirs are more advantageous over wet and dry gas reservoirs resulting from the little remaining gas, the phase behavior of the mixture of condensate gas and CO_2 , as well as the good injectivity of it [85]. Furthermore, the sequestrated CO_2 per pore volume in depleted condensate reservoirs is very high: approximately 13 times higher than that of the equivalent aquifer [82]. However, attention should be paid as the phase change may occur in depleted condensate reservoirs, while not in dry and wet gas reservoirs.

There are some traits associated with the long-term trapping mechanisms of CO_2 in natural gas fields. The noble gas and carbon isotope traces results show that solubility trapping in formation water is dominant, while mineral trapping is limited in the natural gas reservoirs with siliciclastic or carbonate lithologies [86]. It should be mentioned that the residual gas in the depleted reservoirs has an effect on the CO_2 sequestration. Generally, capillary trapping capacity exhibits a positive relation with the remaining gas, while structural trapping capacity, dissolution trapping capacity, and the total storage capacity are inversely related with it [87].

The most important issue related to CO₂ storage in depleted gas reservoirs is the low reservoir pressure, it's sometimes below 20 bar at the initial stage of injection, which may lead to a strong Joule-Thomson cooling effect, probably reducing the reservoir temperature, further forming hydrate, freezing the residual water, and even compromising the well injectivity, especially when cold CO2 is injected [88–90]. When the temperature of the reservoir is over 40 °C, the Joule–Thomson cooling effect is not noticeable in permeable reservoirs, even though the initial formation pressure is as low as 2 MPa. However, the Joule-Thomson cooling may lead to the formation of hydrate, as the initial formation pressure is 6 MPa and the reservoir temperature is less than 20 °C [88]. In order to avoid the Joule-Thomson cooling and ensure a relatively high temperature at a low reservoir pressure, a high temperature of injected CO₂ or a high mass flow rate should be applied. It should be noted that the high mass flow rate may lead to other problems at the beginning and shut-in of the injection. The system in the ROAD project connects a CO₂ capture system at the Masvlakte Power Plant with an offshore depleted gas field that has a depletion pressure below 2 MPa [91]. It is a single source and sink system that allows pressure and temperature control at the shoreline inlet of the offshore pipeline by adjusting the level of after cooling at the compressor. It is used to ensure a high downhole temperature and ease the Joule-Thomson cooling effect. That is, a high temperature of injected CO2 is applied in the reservoir with low pressure, whereas a low temperature of injected fluids can be acceptable in the reservoir with higher pressure, to keep the injection pressure requirement at a low level [90]. Furthermore, the co-injection of SO₂ and CO₂ is an alternative method to suppress Joule–Thomson cooling and shows a beneficial thermal consequence [92]. In addition, the presence of methane can potentially reduce the Joule–Thomson cooling effect [93].

A few projects dedicated to CO₂ storage have been implemented in depleted gas reservoirs. The first demonstration project in Australia named the CO2CRC Otway Project is well-known [94], in which the CO₂ was injected into the Waarre C Formation at a depth of about 2050 m. This project

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commenced in March 2008 and ended on August 2009, with a total storage capacity of 65,445 tons [95]. It is worth mentioning that a community led "stakeholder reference group" has been set up in this project to communicate with the public and help increase their acceptance about CCS technology, which could be a demonstration for other CCS projects. Overall, the CO2CRC Otway Project demonstrates that the sequestration of CO₂ in depleted gas fields can be achieved safely [95], and it provides a basis for the large-scale CO₂ sequestration in depleted oil and gas fields. According to the experience gained in this project, the suitability and storage capacity of similar depleted gas reservoirs has been evaluated. For example, the depleted P18-4 gas field on the offshore of Netherlands [96] and the DF-1 South China Sea Gas field [97], owning a potential capacity of 1 Gt and 8 Mt respectively, are identified as suitable sites for the sequestration of CO₂.

Generally, due to the advantages of low risk and cost-effectiveness, CCS in depleted reservoirs can play an important role in the mitigation of global warming, before the wide application of large-scale CCS in saline aquifers [98].

2.2.3. CO₂ Storage in Coal Beds

 CO_2 injection into coal beds is another attractive strategy for CO_2 storage. Most of the suitable coal beds for CO_2 storage are located at a depth ranging from 300 to 900 m [99]. The sequestration of CO_2 in coal beds possesses the major advantage that the potential coal beds are usually located nearby the existing or planned coal-fired power plants. Therefore, the transportation cost could be reduced significantly. However, CO_2 storage in coal beds is still an immature technology, and only some pilot studies have been conducted on its suitability and storage capacity. The evaluated effective storage capacity of Cretaceous–Tertiary coal beds in Alberta, Canada is 6.4 Gt [99], and the potential storage capacity for the coal beds in China is about 142.67 Gt [100], which signifies the potential contribution of coal beds on the mitigation of CO_2 emissions.

2.2.4. CO₂ Storage in Deep Ocean

The CO_2 can also be directly injected into the deep ocean at water depth of more than 2700 m [101,102], where the liquid CO_2 can sink downward to the seafloor, because the CO_2 is denser than seawater under high pressure and low temperature [102,103]. The storage capacity is extremely large due to the enormous volume of the ocean. However, this CCS technology cannot be applied widely because it may affect the marine environment.

2.2.5. CO₂ Storage in Deep-Sea Sediments

The option of CO₂ storage in deep-sea sediments not only combines the merits of geologic storage and ocean storage, but it also avoids many shortcomings [104–107]. For example, it is free from the potential harm to the ocean ecosystems as the CO₂ is injected into the sediment deep beneath the ocean rather than directly into ocean. The storage mechanisms in terrestrial sequestration such as dissolution trapping, residual trapping, and mineral trapping still play a positive role. In addition, new storage mechanisms, including gravitational trapping and hydrate trapping, also work in the sequestration. The gravitational trapping comes from the fact that the higher density drives the CO₂ into the deep sea [103] to the so-called negative buoyancy zone (NBZ). The depth at which the density of CO_2 is identical to the salinity and temperature-dependent density of seawater is approximately 2700 m [52]. The hydrate trapping works because of the formation of CO₂ hydrate under the condition of high pressure and low temperature [104]. Figure 5 shows the long-term evolution of injected CO₂ in the deep-sea sediments. At the initial stage of injection, a little of hydrates form at the bottom of hydrate formation zone (HFZ), which is beneficial to reduce the permeability of the caprock. The area of hydrate caprock expands along with more CO₂, reaching the bottom of the HFZ and limiting the CO₂ below it. Meanwhile, the aqueous saturated with CO₂ will sink downward because of the buoyancy-driven advection. Finally, the hydrate CO₂ and liquid CO₂ will dissolve in seawater and change into CO₂ aqueous solution through diffusion, and permanent storage occurs.

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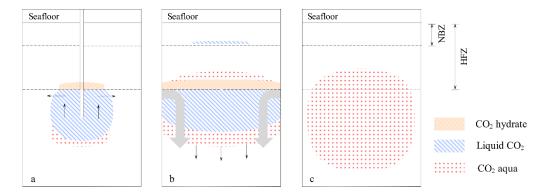


Figure 5. The long-term evolution of the injected CO₂ in deep-sea sediments (modified from [104]).

Despite the enormous capacity and feasibility this technology shares, it is still in the formulation technology readiness level. In addition, CO_2 storage in deep-sea sediments is far more expensive than onshore methods. In addition, it may take a long time to increase the public acceptance of this method [106,108].

In a summary of this part, there are several options for the underground CO_2 storage, including the saline aquifers, depleted oil and gas reservoirs, coal beds, deep ocean, and the deep-sea sediments. The pros and cons of these storage strategies are summarized in Table 3.

Table 3. The pros and cons of	a variety of storage strategies.

Option	Pros	Cons
Saline aquifers	Huge amount of storage capacity, wide distribution, commercial technology readiness level	No economic benefit
Depleted oil and gas reservoirs	Existing installed equipment, guaranteed caprock integrity, characterized geological conditions, small pressure perturbations and induced stress changes, additional oil and gas recovery	Demonstration technology readiness level
Coal beds	Low transportation cost due to its potential location near the coal-fired power plants, additional coal bed methane recovery	Pilot plant technology readiness level
Deep ocean	Large storage capacity	Formulation technology readiness level, no economic benefit, may affect the marine environment
Deep-sea sediments	Enormous storage capacity, free from the potential harm to the ocean ecosystems	Formulation technology readiness level, no economic benefit, far more expensive than onshore methods

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3. Security Assessment of Underground CO₂ Storage

3.1. Numerical Methods for the Security Assessment of CO₂ Storage

The brine migration caused by CO_2 injection may affect the ground water resources [57]. In addition, the chemical reaction between CO_2 with the cement and well string [79,109,110], and the formation response such as the reactivation of faults and the shear failure of caprock, may lead to the failure of well integrity and caprock integrity, resulting in the leakage of CO_2 [11,42]. Therefore, before the implementation of a CCS project, it is very important to assess risks through predicting the CO_2 injection-induced formation responses, including the formation pressure change, formation deformation, and migration of CO_2 plume, etc. Generally, such temporal and spatial responses of formation can be predicted both analytically and numerically. However, due to the physical and geological complexities in CCS projects, only a few semi-analytical models have been developed to estimate risks related with the migration of CO_2 plume and leakage along abandoned wells [111–115]. For example, Nordbotten et al. [112] derived the solution for CO_2 plume evolution during injection in dimensionless form as follows:

$$\left(Q_{\text{well}} \left(p(r_{\text{well}}, t) - p_{\text{init}}\right) + \Delta E_{\text{p}}\right) \left(\frac{2\pi\lambda_{w}kB}{Q_{\text{well}}^{2}}\right) \\
= -\int_{0}^{1} \frac{(\lambda - 1)\ln r'(b')}{((\lambda - 1)b' + 1)^{2}} db' + \Gamma \int_{0}^{1} b' [r'(b')]^{2} db' + \frac{1}{2} \frac{\lambda - 1}{\lambda} \ln \left(\frac{\pi B\varphi}{V(t)}\right) \tag{1}$$

where Q_{well} presents the volumetric injection rate; $p(r_{\text{well}},t)$ denotes the fluid pressure at the injection well; p_{init} is the initial pressure; ΔE_{p} presents the potential energy required to submerge the CO₂ into the denser water; λ_{w} presents the mobility of water; k is the permeability; k is the reservoir thickness; k denotes the total mobility; k is the porosity; k denotes the total injected volume; and k presents the thickness in the CO₂ plume profile, which is a function of radial distance from the injection well k0 and time k1.

The numerical simulation methods are more popular in assessment of the risks associated with CO₂ storage. Many thermal–hydraulic (TH) coupled simulators have been developed for multi-component and multi-phase flow in CO₂ storage, such as TOUGH [57,116,117], ECLIPSE [118,119], CMG-GEM [120–123], STOMP [124,125], MRST [126], COMET3 [127], IPARS [128], MUFTE-UG [129], FLUENT [130], and Tempest [131,132]. The hydro–thermal–mechanical (THM) coupled simulators are mostly constructed based on the coupling framework of fluid flow simulator (TH) and mechanical simulator (M). Specifically, the THM simulators used in CCS mainly include TOUGH-FLAC3D [63,133,134], TOUGH2-RDCA [135], TOUGH-RBSN [136], Sierra Arpeggio (Aria-Adagio) [73], OpenGeoSys-ECLIPSE [137], ABAQUS-ECLIPSE [138], and ECLIPSE-VISAGE [76]. Among them, the TOUGH-FLAC has been well tested and applied in many simulations of CCS [63,139], and it was developed by the Lawrence Berkeley National Laboratory [140,141].

In recent years, some software has been developed for the risk assessment in CCS, such as the National Risk Assessment Partnership (NRAP) Toolset and Leakage Assessment and Cost Estimation (PyLACE). The NRAP is designed to evaluate the environmental risks associated with CCS operation by the U.S. Department of Energy's National Energy Technology Laboratory [142]. In NARP, the geological system of CCS is divided into several subsystems firstly, and each subsystem is characterized by a reduced-order model [143]. Then, the reduced-order models are linked by an integrated assessment model based on the system modeling approach [142]. Finally, the whole system model can be used to evaluate the risk performance. The framework of the NARP is shown in Figure 6.

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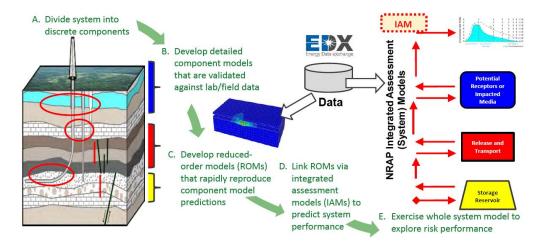


Figure 6. The framework of the National Risk Assessment Partnership (NARP) [144].

By using NARP, two major types of risk, including CO₂ leakage and induced seismicity, can be simulated. Additionally, the behavior of several important components in the CCS systems, including reservoirs, seals, wells, and ground water aquifers, could be modeled using corresponding tools. For example, the Wellbore Leakage Analysis Tool (WLAT) could be used for the assessment of the leakage potential of existing wells [145], and the Design for Risk Evaluation and Monitoring (DREAM) is developed for the evaluation and optimization of monitoring designs for long-term CO₂ storage operation.

The Python-based web application PyLACE is designed to quantify the financial risks associated with potential CO_2 leakage in a CCS system [146]. There are two major functional blocks in PyLACE: one of them is metamodel development, and the other one is metamodel-based decision support. It can convert the process-level risk assessment models into high-fidelity metamodels for the purpose of online assessment by using the high-performance computing and cloud computing infrastructures.

Recently, a deep neural network inversion was applied on 4D seismic data for estimating saturation and pressure [147], proving the availability of deep neutral network on the application of data-based inversion. To make the assessment of CCS more efficient and effective, the machine learning technology is encouraged to be used. It can be forecasted that the evaluation of CCS will be more and more intelligent with the development of machine learning technology.

3.2. Monitoring Technologies in the Assessment of the CCS Risks

The injected CO₂ will be retained in underground for a long period, and the CO₂ plume may affect the surrounding environment and the groundwater in particular [148]. Since it is difficult to predict with reasonable accuracy the key issues or risks in CCS by utilizing only simulation tools [149], the monitoring and history matching is very important in CCS assessment. The monitoring is used in most of the field development plans and routine field operations [62]. The most common monitoring technology used in CCS includes 3D seismic, 4D seismic, microseismic, vertical seismic profiling, gravimetry, cross-hole electromagnetic, pressure and temperature monitoring, geochemical sampling, soil and gas sampling analysis, tracers, atmospheric monitoring, microbiology, core analysis, satellite monitoring, and distributed temperature sensing technology.

The 3D seismic can provide a tri-dimensional image of the formation structures and the CO_2 plume. The quality of the 3D seismic is affected by the medium. In off-shore monitoring, the 3D seismic monitoring data with high quality could be obtained, and the CO_2 bodies above 106 kg at the depth of 1–2 km could identified due to the enhanced penetration of seismic waves in water [148].

The 4D seismic involves repeating 3D seismic in time-lapse mode to image the CO₂ plume in the reservoir over time, which is beneficial for the monitoring of the migration process of CO₂. A major challenge for the 4D signal to reflect the field data with high accuracy is the non-repeatable noise level

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in the data. This is on account of that the seismic imaging experiment is difficult to be repeated from one survey to the next due to the variations in the sources' receiver positioning and geometry, the soil moisture content, and the formation water properties [150].

The microseismic activity levels show a correlation with the CO_2 injection periods. Thus, it is beneficial for the understanding of the subsurface CO_2 injection and migration process. In this procedure, the one-dimensional array that consists of several three-component downhole geophones will be deployed in the vertical well. Afterwards, the waveform data will be detected by the geophones and then transferred to the digitizers for recording [151].

Apart from conventional surface seismic acquisition, the hydrophone arrays, buried geophone, and the fiber optic cables are permanently installed in the vertical seismic profiling systems to achieve long-term monitoring and obtain the geological structure details [152]. This monitoring technology has been successfully used in the Ketzin pilot site and the MRCSP project.

Gravimetry testing can detect the variations of fluid density due to CO_2 injection and thus provide the information of the location of CO_2 . The limit of this method is that the testing result is affected by the shape of the CO_2 plume [148,153].

Cross-hole electromagnetic is a non-invasive method on the determination of the subsurface physical and chemical properties. It can also provide the information for the detection and monitoring of the location of CO_2 . In this domain, the electrical conductivity before and after the CO_2 injection is obtained firstly. Then, it can be converted to the CO_2 saturation with the help of inversion algorithms and appropriate rock-physics models [154,155]. It should be mentioned that this monitoring technology is only suitable for small-scale areas such as the area between wells [154].

Monitoring the formation fluid pressure and the variation of temperature can help in evaluating the risks associated with the failure of the caprock integrity and identifying the flow path of the injected CO_2 , respectively. It is important to point out that the wellhead pressure and temperature cannot provide enough information for the CO_2 injection process, which has been certified at the Ketzin pilot site [156]. Therefore, the downhole pressure and temperature monitoring are recommended in the CO_2 storage operation.

The geochemical sampling analysis could reveal the chemical variations, such as the drop of pH and natural variations in water chemistry, which is crucial for establishing a useful baseline for groundwater hydrology [157]. In addition, it can provide the information of the variation of the concentration of minerals that may be induced by the dissolution of carbonates and precipitation of anhydrite. It should be mentioned that the chemical reaction is a slow process in the sandstone formation [158], which is difficult to be detected in a short-term CO_2 storage process.

Soil and gas monitoring can provide the data of CO_2 concentration, which is beneficial for the definition of the baseline before the injection [148]. In addition, it can provide more data on natural CO_2 variations in different environments and associated seasonal fluctuations.

Tracers monitoring is a cost-effective method for monitoring the origin of CO_2 observations at wells and in the storage complex. The mechanism involves the co-injection of some specific compounds that can be detected in a very small concentration such as SF_6 , SF_5CF_3 , and the isotope ¹⁴C together with CO_2 [95,159]. Thus, the trail of the injected CO_2 could be reflected by the traces.

Atmospheric monitoring means detecting the atmospheric concentration of CO_2 that may be changed by the leakage of CO_2 from the underground, which is beneficial for the identification of the anomalies above the natural base line [160]. The reliability of this technology may be affected by the significant natural variation of atmospheric concentration of CO_2 induced by the organic matter decomposition and the soil respiration [148].

Microbiology monitoring can be conducted on the samples of reservoir fluids and minerals before and after the CO_2 injection, which could be defined as a baseline and the modification caused by the presence of CO_2 , respectively [161]. Specifically, the biocenosis such as the sulfate-reducing bacteria (SRB) in the rock substrate and fluid samples could be analyzed by the molecular biological method, polymerase chain reaction—single strand conformation polymorphism (PCR-SSCP) method,

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and fluorescent in situ hybridization method [162]. This information related to the microbiology is valuable for the identification of biogeochemical process that affect the diffusion of CO_2 in the reservoirs.

Core analysis is essential for the acquisition of the petrophysical and rock mechanical properties. Some measuring methods including the SEM imaging, XRD, and X-ray elemental analysis are usually included to obtain the micro morphological and mineralogical properties of the core [62].

The satellite-borne synthetic aperture radar (SAR) monitoring can provide the information related to the change of the ground surface caused by CO₂ injection operation, which can be used to modify the model of CO₂ distribution in the underground. By using SAR, the amplitude and the phase can be obtained firstly. Then, the phase difference between two observations can be converted into the displacement of the surface through the platform altitude and look angle [163]. Comparing the geophysical surveys methods, the satellite-borne SAR monitoring is considered as a cost-effective monitoring tool, and it has been successfully used in the In Salah project [163].

The distributed temperature sensing (DTS) technology is developed to overcome the limitations of conventional temperature monitoring that cannot provide the high vertical spatial resolution and real-time data. The fiber optic cable is used as a distributed sensor in DTS, which offers the possibility to measure the temperature from the surface to the bottomhole along the extension of the fiber [164,165].

The advantages and applications of the monitoring technologies in the CCS demonstration projects are summarized in Tables 4 and 5, respectively. It shows that the geochemical sampling analyses, 3D seismic, microseismic, and pressure and temperature logs have gained the most popularity among the monitoring technologies due to their high performance in acquiring the characteristics of geological structures and formation fluids.

Table 4. Main monitoring technologies in CCS.

Monitoring	Advantages	Ref.
Technology	<u> </u>	
3D seismic	Provides a tridimensional image of geological structures and the plume migration of CO_2 .	[62]
4D seismic	Significant benefits for overburden imaging and time-lapse responses with improved acquisition plan.	[62]
Microseismic	It is very useful for monitoring geomechanical response to injection.	[151]
Vertical seismic profiling	Valuable information on the geological structure details.	[152]
Gravimetry	Beneficial for the evaluation of formation fluids density and CO ₂ plume.	[153]
Cross-hole electromagnetic Pressure and	Advantageous for the detection and monitoring of the location of CO_2 .	[155]
temperature monitoring	Direct information for the evaluation of the stability of the reservoir.	[156]
Geochemical sampling	Natural variations in water chemistry are crucial for establishing a useful baseline for groundwater hydrology.	[157]
Soil and gas sampling	More data on natural CO_2 variations in different environments and associated seasonal fluctuations is needed.	[62]
Tracers	Valuable and cost-effective method for monitoring the origin of CO ₂ observations at wells and in the storage complex.	[62]
Atmospheric monitoring	Useful data to identity the anomalies above the natural baseline.	[160]
Microbiology	Valuable data to identify biogeochemical process that affect the diffusion of CO_2 in the reservoirs.	[161]
Core analysis	Good petrophysical data and rock mechanical properties are essential.	[62]
Satellite monitoring	Valuable and cost-effective monitoring data for onshore CO ₂ injection operation.	[163]
Distributed temperature sensing technology	It can provide high-resolution information on the migration of CO_2 in the reservoir.	[164]

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Table 5. Application of the main monitoring technologies in some CCS demonstration projects (modified
from [148]).

Monitoring Technology	Sleipner	Frio	Nagaoka	Ketzin	In-Salah	Otway	Weyburn	MRCSP
3D seismic	×		×	×	×	×		
4D seismic				×	×			
Microseismic	×		×		×		×	×
Vertical seismic profiling		×						×
Gravimetry	×				×		×	×
Cross-hole electromagnetic		×		×	×			
Pressure and temperature logs		×	×	×	×			×
Geochemical sampling		×	×		×	×	×	×
Soil and gas sampling		×			×		×	
Tracers		×			×	×		
Atmospheric monitoring						×		
Microbiology				×				
Core analysis					×		×	
Satellite monitoring					×			×
Distributed temperature sensing technology								×

3.3. Generating CO₂-in-Water Foams

The injected CO₂ exhibits much lower viscosity and density compared with oil and brine in the reservoir conditions and leads to a poor displacement efficiency. Generating high viscosity CO₂-in-water foams with large gas volume fraction would be an effective strategy to address this issue [166], which has some advantages for both oil reservoirs and saline aquifers. For oil reservoirs, CO₂-in-water foams can improve oil recovery and the economics of CCUS in petroleum systems [167]. Guo and Aryana [168] used a glass microfluidic device to investigate the flow behavior of foam in oil-saturated heterogeneous porous medium, and it resulted that the foam injection can improve the oil recovery through the improvement of the sweep efficiency. A field test of CO₂ foam flooding was conducted in the North Ward-Estes field in Texas, demonstrating that the foam can notably improve the sweep efficiency and be economically successful [169]. For saline aquifers, CO₂-in-water foams can also improve the sweep efficiency, storage capacity, and economics. Guo et al. [170] used a glass-fabricated microfluidic device to investigate the effect of various factors on the CO₂ storage capacity in aquifers. Their results showed that the CO₂ storage capacity would be increased by over 30% with the foam injection compared with CO₂ injection cases, demonstrating the superiority of CO₂ foam on the improvement of CO₂ storage capacity. It should be mentioned that the foam can also reduce the risk of leakage in the underground CO₂ storage unit result from the reduction of fluid mobility. For instance, due to the significant shear rates differences between the flowing in the reservoir rock and leakage pathway, the foam may become gel inside the leak and reduce the leakage through the shear-induced gelation, i.e., the particles break the interaction barrier of forming clusters in high shear stress condition [171].

To achieve stable CO_2 foams, the surfactants are used traditionally. The hydrophilic/ CO_2 -philic balance (HCB) is considered as the principle for the designing of surfactants for CO_2 foams, which can characterize the balance of surfactant and solvent interactions [172]. In recent years, the nanoparticles have been used combining with the surfactant solutions to improve the stability of CO_2 foams, and Worthen et al. developed the concept of HCB to be nanoparticle HCB [173]. Worthen et al. [174] generated viscous and stable CO_2 -in-water foams with the mixture of nanoparticles (bare colloidal silica) and surfactant (caprylamidopropyl betaine). They suggested that the formation of foams was caused by the reduction of interfacial tension through the surfactant, while the stability of foams may be improved by the adsorption of nanoparticles at the CO_2 -water interface. The behavior of silica nanoparticles on the reduction of carbon footprint was also demonstrated by Rognmo et al. [167]. In addition, some other nanoparticles such as nano lauramidopropyl betaine with alpha-olefin sulfonate also have been demonstrated to perform well in maintaining a high foam quality [170], showing the superiority of nanoparticles on the stabilizing of CO_2 foams. Overall, the generating of CO_2 -in-water

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foams especially combining with the nanoparticles are supposed to be a candidate in the designing of CO₂ storage, while the overall cost should be considered.

3.4. Accelerating CO₂ Dissolution Process

As mentioned in above sections, the free CO₂ can remain more than 1000 years underneath the caprock, which increases the uncertainties in the long-term fate of injected CO₂ and increases the cost of long-term monitoring operation. To address this issue, accelerating the dissolution process of CO₂ and minimizing the free CO₂ in the underground is an effective strategy [175]. Cameron and Durlofsky [176] used the Hooke-Jeeves direct search algorithm to optimize the locations of CO₂ injection wells and the injection rate to minimize the mobile CO₂ in the CCS system within 1000 years, and the results showed that the fraction of mobile CO₂ would decrease from 0.220 to 0.072 in the optimal case, highlighting the importance of well location optimization. Anchliya et al. [175] proposed an engineered injection method to promote the dissolution and trapping of CO₂. Figure 7 shows the schematic of the engineered injection process. Compared with conventional CO₂ injection scenarios, there is another brine injection well located exactly over the horizontal CO₂ injection well and near the top of reservoir. In the engineered injection system, another two brine production wells are placed at either side of the CO₂ injection well. The brine injection well is used to limit the upward movement and impel the horizontal flows of CO₂ under a lateral pressure gradient provided by brine injection, which increases the sweep efficiency and enhances the CO₂ dissolution and trapping [175]. Numerical simulation studies show that about 90% of the injected CO₂ could be immobilized within 20 years after CO₂ injection ceases due to the fast dissolution trapping and residual trapping. It should be pointed out that the potential risk of pressurization due to CO₂ injection could also be addressed by the engineered injection method through controlling the brine injection and production rates. In addition, it can enhance the storage capacity of CO₂ in a bounded aquifer formation. Additional drilled wells are needed for the engineered injection system, which increases the cost of this CCS operation. Consequently, the adaptability of the engineered injection method should be quantified and site specific.

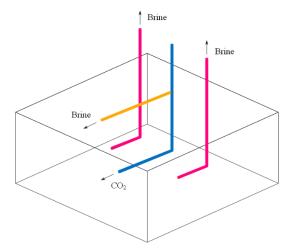


Figure 7. Engineered injection method to accelerate CO₂ dissolution and trapping (adapted from [175]).

Another injection scheme called water-alternating-gas (WAG) injection was firstly proposed in the petroleum industry in the late 1950s to improve the sweep efficiency of reservoirs. The different types of WAG injection are illustrated in Figure 8. Zhang and Agarwal investigated the potential of the WAG injection scheme with the goal of improving the efficiency of CO_2 storage [177,178]. The results showed that the optimized WAG scheme could accelerate CO_2 dissolution and decrease the impact zone up to 14% comparing with that of the constant gas injection scheme. However, the WAG injection scheme may decrease the total storage capacity of the reservoirs due to the large amount of injected

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water. In addition, it may increase the cost in the injection process, thus it has not been applied widely in the CCS operation.

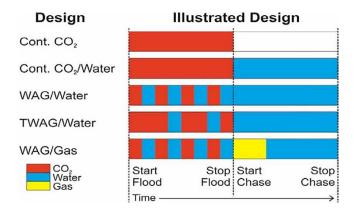


Figure 8. Schematic of various schemes of water-alternating-gas (WAG) injection [179].

To eliminate the adverse effects of WAG injection schemes on storage capacity, an injection scheme combining the intermittent injection method and brine production was proposed by Tanaka et al. [180]. The schematic diagram of intermittent injection is shown in Figure 9, which suggests that a diagonal pair of wells were used for CO₂ injection alternately. In this injection scheme, a diagonal pair of wells (Well 1 and Well 3) are used for CO₂ injection, while another pair of wells (Well 2 and Well 4) are used for producing brine, which will be re-injected to the reservoir through Well 1 and Well 3. Numerical simulation results show that both the dissolved and residual CO₂ are increased. Specifically, the ratio of the trapped CO₂ increases by 20% compared with the base case, which has only one well with continuous injection. Another advantage of this injection scheme is that it could mitigate the pressure buildup through intermittent injection and water production, which highlights the importance of the CO₂ injection design and brine management.

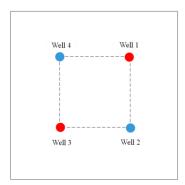


Figure 9. Schematic of the intermittent injection method (modified from [180]).

In terms of the above methods for accelerating the dissolution of CO_2 , they may increase the cost in the injection process due to water injection. In addition, additional wells are needed for the engineered injection and intermittent injection with four wells, which results high costs on the operation. However, the cost on the monitoring would be reduced due to the relative low leakage risk caused by the rapid dissolution of CO_2 . The overall economic costs of the CCS unit and the storage capacity are supposed to be taken into account for optimization when considering the utilization of these technologies.

3.5. Accelerating Mineral Carbonation Process

The mineralization of CO_2 is an effective way to fix the injected CO_2 and guarantee its security permanently. However, for conventional CCS in saline aquifers, it takes tens of thousands of years

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for the mineral trapping because of the low reactivity of silicate minerals in sedimentary rocks with CO_2 [52]. To accelerate the mineral trapping of CO_2 , some novel methods have been developed, such as CO_2 storage in basalt rock formation [12], CO_2 storage in peridotite formation [181], direct mineralization of flue gas by coal fly ash [182], direct aqueous mineral carbonation [29], and pH swing mineralization [183].

3.5.1. CO₂ Storage in Basalt Rock Formation

Applying the sequestration of CO₂ in basalt formations is proposed by Gislason and Oelkers [12]. Basalt contains approximately 25% of magnesium, calcium, and iron oxides, and it is far more reactive with carbonic water compared with sedimentary silicate rock [184]. Another merit of this method is that abounding basaltic rocks exist on the Earth's surface [185], which offers the possibility of large application of CCS in basalt formation. The field test of CCS in basalt was conducted in the CarbFix pilot project in Iceland in 2012 [159]. In this project, 175 tons of pure CO₂ were injected with water firstly. Then, 73 tons of CO₂–H₂S mixture (55 tons CO₂) were fully dissolved in water and injected. The measured dissolved inorganic carbon of ¹⁴C and the monitoring well shows that more than 95% of injected CO₂ was mineralized to carbonate minerals within two years, demonstrating the efficiency of mineral trapping of CO₂ in basalt. However, large-scale application of this technology requires substantial quantities of water during the CO₂ injection process. In addition, the cost of storing and transporting CO₂ for the CarbFix project is about twice that of storage in typical sedimentary basins. Therefore, the application prospect of this technology is not very promising in the near future.

3.5.2. CO₂ Storage in Peridotite Formation

The mantle peridotite is mainly composed of olivine and pyroxene, which can react with CO_2 and H_2O to form hydrous silicate, Fe-oxides, and carbonates. Isotope analysis and reconnaissance mapping indicate that about 10^4 to 10^5 tons of CO_2 per year are trapped to solid minerals via peridotite weathering effect in Oman [181]. The main reaction can be formulated as:

$$2Mg_2SiO_4 + Mg_2Si_2O_6 + 4H_2O = 2Mg_3Si_2O_5(OH)_4$$
 (2)

$$Mg_2SiO_4 + 2CO_2 = 2MgCO_3 + SiO_2$$
(3)

$$Mg_2SiO_4 + CaMgSi_2O_6 + 2CO_2 + 2H_2O = Mg_3Si_2O_5(OH)_4 + CaCO_3 + MgCO_3.$$
 (4)

As shown in Figure 10, the carbonation rate could be enhanced more than 1 million times compared with the natural rate with the three-step operation process, beginning with drilling and fracturing, followed by injection of hot CO_2 (approximately 185 °C) at a rapid rate to heat the fractured peridotite, and then injecting CO_2 with normal temperature. In this case, the system maintains a temperature of 185 °C and high carbonation rate due to the exothermic carbonation of peridotites. It is estimated that the peridotite in Oman alone could trap more than 1 billion tons of CO_2 per year into carbonate minerals, showing a huge amount of capacity.

However, except for the peridotite exposing through large thrust faults, the mantle peridotite is generally more than 6 km below the seafloor and 40 km below the land surface [181], making it difficult for the sequestration of CO_2 in these formations. Thus, the peridotite in shallow areas could be used effectively for the sequestration of CO_2 .

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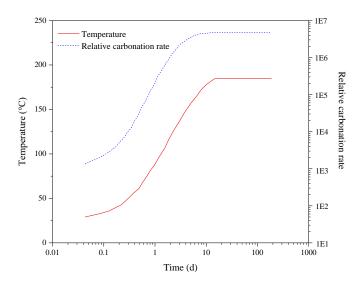


Figure 10. Calculated temperature and the carbonation rate relative to the rate for CO_2 in surface water at 25 °C and 0.1 MPa in the three-step injection operation (adapted from [181]).

3.5.3. Direct Mineralization of Flue Gas by Coal Fly Ash

Reddy et al. [182] conducted a preliminary experiment to study the reaction between flue gas and coal fly ash in a fluidized bed reactor. This experimental setup is shown in Figure 11. The results show that the concentrations of CO_2 and SO_2 in flue gas dropped from 13.0% to 9.6%, from 107.8 to 15.1 ppmv, respectively in 2 min. In addition, the Hg in flue gas was also mineralized by fly ash particles. Reddy et al. [182] conducted a pilot-scale study with a fly ash content of 100–300 kg to investigate the feasibility of this technology. In the pilot studies, the fly ash particles were fluidized by the flowing of flue gas in a fluidized bed reactor to ensure sufficient mixing and contact between them, and the reaction occurred under a fixed pressure of 115.1 kPa. Experimental results show that the content of $CaCO_3$ produced by the reaction of flue gas and fly ash ranged from 2.5% to 4% in 10 min. Meanwhile, the contents of S and Hg in the fly ash increased from non-detectable to 0.45 and 0.5 mg/kg, respectively. This confirmed that the flue gas components can be captured without separation and mineralized by fly ash particles using an accelerated mineral carbonation process. However, the treatment of the carbonated fly ash produced by using this method is still an important problem that needs be addressed [30].

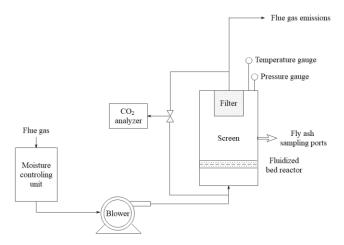


Figure 11. Preliminary experimental setup for CO₂ capture and mineralization (modified from [182]).

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3.5.4. Direct Aqueous Mineral Carbonation

Direct aqueous mineral carbonation is a process that uses a bicarbonate-bearing solution mixed with reactant minerals such as magnesium and calcium silicate rocks to convert gaseous CO_2 into solid form [186]. The magnesium and calcium silicate rocks are distributed all over the world. For instance, the magnesium silicate rock in Eastern Finland has the ability to store 10 million tons of CO_2 per year for 200 to 300 years [187].

The overall chemical reaction of the carbonation of serpentine can be described as:

$$Mg_3Si_2O_5(OH)_4 + 3CO_2 = 3MgCO_3 + 2SiO_2 + 2H_2O.$$
 (5)

In the process, the serpentine was treated at 630 °C to attain an active mineral. Then, the stoichiometric conversion of 78% of the silicate to carbonate was observed by using the bicarbonate-bearing solution at the conditions of 150 °C and 18.5 MPa, with 15% solids. This occurred within 30 min, showing an extremely high reaction rate for the carbon mineralization. To reduce the energy consumption on the heat reactivation and increase the effective reaction area, mechanical activation was proposed by adding an attrition grinding step. In this case, the reaction condition was optimized to 25 °C and 1 MPa, which still led to up to 65% carbonation within 1 h [186]. For instance, the combination of grinding and heat activation was used to attain a better carbonation mineralization performance, as shown in Figure 12 [29]. Based on this concept, the feasibility of this technology in the mineralization of flue gas was investigated by Verduyn et al. [29]. As shown in Figure 12, the relative high pH due to the lower solubility of flue gas makes it difficult for the leaching of cations. To eliminate this, the contact of the mineral and flue gas is done in a slurry mill and a leaching basin.

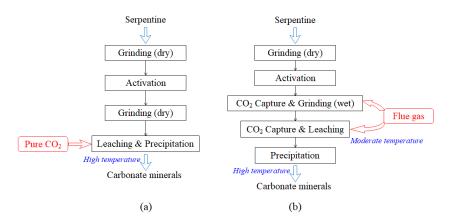


Figure 12. Mineralization process concept for (a). pure CO₂, (b). flue gas (modified from [29]).

3.5.5. pH Swing Mineralization

To increase the conversion efficiency in the CO_2 mineral trapping process, the pH swing approach was proposed by Park and Fan [188]. They combined the internal grinding in acidic solvent for a rapid dissolution of a serpentine sample. Three solid products including SiO_2 -rich solids, iron oxide, and magnesium carbonate were produced by controlling the pH. Teir et al. [187] used HCl and HNO $_3$ to dissolute serpentinite and then transformed the serpentinite to hydromagnesite with the help of CO_2 . In their results, the pure hydromagnesite, which is thermally stable at 300 °C, was produced. However, the additional amount of chemicals used in this work increase the costs and make it infeasible for CCS operations. To reduce the cost, a pH swing CO_2 mineralization method with a recyclable reaction solution was proposed by Kodama et al. [189]. They selectively extracted the alkaline-earth metal from steelmaking slag in an ammonium chloride solution. The reacted solution was used for CO_2 absorbent and produced ammonium carbonate. Then, the calcium carbonate was precipitated in another reactor with the recovery of ammonium chloride. The results showed that the selectivity of the calcium extraction reaction reached 60%, and pure $CaCO_3$ was produced with an energy consumption

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of 300 kWh/t-CO₂. Wang and Maroto-Valer [183] proposed a modified carbon mineralization process as shown in Figure 13. Through experimental studies, they concluded that $(NH_4)_2CO_3$ is more beneficial for increasing the efficiency of carbon fixation compared with NH_4HCO_3 , and the optimal efficiency of CO_2 mineralization reaches 46.6%. Furthermore, the pH swing mineralization process was optimized by Sanna et al. [190] under different temperature conditions. The results showed that the total CO_2 trapping efficiency was 62.6% at the condition of 80 °C, with the molar ratio of 1:4:3 for Mg:NH₄ salts:NH₃. However, the energy consumption and overall economic cost need to be lowered before any large-scale application.

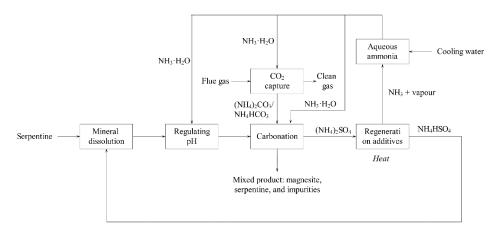


Figure 13. The schematic of carbon mineralization process using recyclable ammonium salts (modified from [183]).

To summarize this section, CO_2 storage in basalt rock formation and peridotite formation is limited by the distribution of the particular rock. In addition, the substantial quantities of water and energy consumption for heating increase the cost a lot. The direct mineralization of flue gas by coal fly ash would be a promising technology result since the gas could be mineralized without separation, which decreases the overall cost a lot. The pH swing mineralization may be another promising technology for the sequestration of CO_2 due to the sustainability. The recyclable and cheap chemical reagents ought to be introduced into the mineralization process to make this technology more cost-effective.

4. Strategies for Improving the Cost-Effectiveness of CO₂ Storage

4.1. Enhanced Industrial Production with CO₂ Storage

Resources production during CO₂ storage is an effective method to partly cover the cost of CCS and obtain additional economic benefits, which is called CO₂ capture, utilization, and storage (CCUS) [19]. In the process of CCUS, CO₂ usually works as a working fluid to enhance the recovery of underground resources through displacement, dissolution, thermal conductivity, and reactive transport. The potential geological formations for CCUS include oil reservoirs, gas reservoirs, saline aquifers, shale formation, un-mineable coal seams, hot dry rock, uranium deposit formation, and natural gas hydrate reservoirs [191]. The corresponding CCUS technologies are CO₂-enhanced oil recovery (CO₂-EOR), CO₂-enhanced gas recovery (CO₂-EGR), CO₂-enhanced water recovery (CO₂-ECBM), CO₂-enhanced shale gas recovery (CO₂-ESGR), CO₂-enhanced coal bed methane recovery (CO₂-ECBM), CO₂-enhanced geothermal systems (CO₂-EGS), CO₂-enhanced in situ uranium leaching (CO₂-IUL), and CH₄-CO₂ replacement from natural gas hydrates, respectively [191,192].

4.1.1. CO₂-EOR

In CO₂-EOR technology, the CO₂ is injected into oil reservoirs to enhance the recovery of crude oil, which is the most successful and promising technology combining the utilization and

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sequestration of CO_2 [27,193,194]. The displacement of oil by CO_2 can be classified as multicontact miscible and immiscible processes, depending on the properties of CO_2 and the reservoir fluids at the condition of reservoir pressure and temperature [195,196]. In the multicontact miscible displacement procedure, the minimum miscibility pressure (MMP) is required for multicontact miscible displacement. The immiscible displacement occurs when the pressure is lower than the MMP, with less components exchange between CO_2 and oil in the reservoir [197].

There are three CO₂ injection schemes for the operation of CO₂-EOR, including continuous injection, water-alternating-gas (WAG) injection, and cyclic injection. In the continuous injection scheme, CO₂ injection and oil production are running continuously, which has been applied in the North Cross Devonian Unit for enhanced oil recovery [198]. The multicontact process can be achieved through vaporizing and condensing [199]. However, this injection scheme has not gained much popularity in the field application compared with the WAG injection and cyclic injection. WAG injection is the widely used form, because it can decrease the mobility ratio between the injection fluids with oil and lead to late gas breakthrough and high oil recovery. In the design of WAG injection, the optimization algorithm such as the Lagrangian and stochastic simplex approximate gradient algorithm could be used to obtain the maximum net present value [200]. Although the WAG injection is beneficial for improving the oil recovery, it may cause the gas to flow upward, while the water and oil flow downward due to the large density differences, resulting in early gas breakthrough, especially in the reservoirs with highly permeable channels and large vertical heterogeneity [199]. To address this issue, the cyclic injection process, i.e., gas huff-n-puff process was proposed, which is composed of three stages, including the gas injection stage, well shutting state, and the oil production stage.

For conventional oil reservoirs, CO₂ flow dominated throng the rock matrix. The mechanism of CO₂-EOR is due to the solubility of CO₂ in oil under the supercritical phase condition, which can decrease the oil density and viscosity, leading to enhancing the oil recovery [199]. For the unconventional tight oil reservoirs such as shale oil reservoirs, fracturing is an essential technic for the exploitation. In this scenario, CO₂ flowing is dominated by fracture flow instead of rock matrix flow. The process of the CO₂-EOR in fractured oil reservoirs can be divided into four steps, as shown in Figure 14. In the initial stage (Step 1), the injected CO₂ flows rapidly through the fracture. Then, the CO₂ starts to permeate into the rock matrix under the displacement effect (Step 2). During this stage, the permeating CO₂ may carry oil into the rock and decrease the oil production. Simultaneously, the permeated CO₂ would lead to the swelling of oil and then mitigating out of the matrix, which is beneficial for the oil production. The oil continues to swell and lower viscosity by the permeated CO₂, and moves to the fracture in the follow stage (Step 3), which corresponds to the well shutting stage in the huff-n-puff process. Finally, the pressure equilibrium inside of the matrix is approached, thus the migrating of the miscible or immiscible oil from the matrix to the fracture is dominated by diffusion effects. The oil in the bulk CO₂ is swept through the fractures to the production well by the production pressure [201]. The cyclic injection scheme can also promote the propagating of reservoir pressure due to CO₂ injection near the injection well, especially for the reservoir with ultralow permeability. Specifically, the CO₂ huff-n-puff performs better on the oil recovery when the reservoir permeability is lower than 0.03 mD [202]. Characterization of the flow behavior of CO₂ and oil in the low permeability formation with complex natural and hydraulically created fractures under the in situ conditions are supposed to be emphasized, which is beneficial for improving the efficiency of CO₂-EOR.

Apart from increasing oil recovery, CO₂-EOR provides an additional advantage of CO₂ sequestration, which could be an important economic incentive for early CO₂ storage projects [203]. Typically, 3 tons of CO₂ injection can produce approximately 1 bbl of incremental oil. It is shown that about a 5% to 15% enhancement in oil production can be achieved by using CO₂-EOR [204]. In the largest discovered fields over the world, it is estimated that approximately 470 billion barrels of incremental oil could be produced simultaneously with 140 billion metric tons of stored CO₂ by using CO₂-EOR [205].

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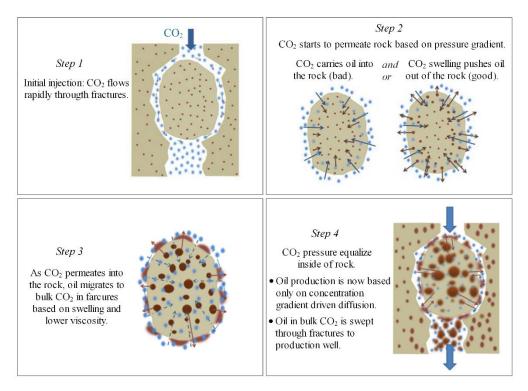


Figure 14. Conceptual steps of CO₂-enhanced oil recovery (CO₂-EOR) in fractured tight oil reservoirs ([201]).

The first CO₂-EOR pilot project was implemented at the SACROC oil field in 1972 [206], in which CO₂ foam was implemented to alter the mobility and improve the sweep efficiency [207,208]. At present, CO₂-EOR is a relatively mature technology that has been widely used in the petroleum industry to enhance oil recovery for tens of years, with the capacity of more than 1000 million tons of CO₂ stored subsurface [209,210]. This technology has gained great success in North America. In the USA, more than 260,000 bb/d are produced due to the application of CO₂-EOR technology [211]. In the Weyburn oilfield in Canada, the CO₂-EOR project was conducted to extend the life of an oilfield. About 20 million tons of CO_2 is planned to be sequestrated in the oil reservoir [212,213]. In recent years, the feasibility of CO₂-EOR in China has been massively studied. The first CO₂-EOR project in China, i.e., the Jilin Oilfield, injected nearly 217,000 tons of CO₂ with a storage efficiency of 96% by April 2013 [214], and the CO₂ capacity is about 600,000 tons [10]. The technology of CO₂-EOR has application prospects in the Shengli Oilfield and Bohai Bay Basin, with 6.7% incremental oil recovery and 683 million tons of incremental oil production, respectively [215,216]. CO₂-EOR also attracted much attention in Europe. In Poland, the potential utilization of anthropogenic CO₂ for CO₂-EOR was studied in the B8 oilfields in Baltic Sea and Brage on the Norwegian Continental Shelf [217], which is a part of the ongoing PRO_CCS project funded by Norway Grants. The simulation results showed that the total storage capacity of Brage and B8 oilfields are 33 and 4.8 million tons in 17 years of injection, with an expected incremental oil production of 98 and 14.6 million bbls, respectively.

To co-optimize the CO_2 storage and enhanced oil recovery, Ampomah et al. [218] proposed an objective function (Equation (6)) considering both CO_2 storage and oil production, which can be optimized by the neural network and genetic algorithm. In the optimal case of the Farnsworth field unit, more than 94% of CO_2 could be sequestrated with approximately 80% of the oil produced, which provides a guideline for the co-optimization of CO_2 storage and EOR.

$$f = w_1 \times FOPT + w_2 \times FGIT \tag{6}$$

where *w* is weight assigned to vector, *FOPT* is the cumulative produced gas, and *FGIT* is the cumulative injected gas.

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Similarly, a framework to co-optimize the oil production and CO₂ storage was developed by Jahangiri and Zhang [219]. In the framework, the net present value (NPV) is treated as the optimization objection function, which was solved by the ensemble-based optimization algorithm.

$$NPV = \sum_{t=1}^{T} \frac{C}{(1+r)^{t}} - C_{0}$$
 (7)

where t is the time step, T is the operation period, r is the periodic discount rate, C is the cash flow in the time step that is determined by the price, injection, and production volume of CO_2 and oil, and C_0 is initial investment.

By using this method, the well injection patterns and injection rates for the maximum NPV can be determined. In addition, the discrete time optimization model could be used for maximizing the total profit in CO_2 -EOR operations, with consideration of both enhanced oil recovery and geological CO_2 sequestration [220].

Artificial neural network models can be used to predict and optimize the performances of CO₂-EOR, as shown in Figure 15 during the multi-cycled water-alternating-gas process [221]. There are four neurons in the input layer corresponding to water-to-gas injection time ratios (WAG), temperature, permeability ratio, and initial water saturation, respectively. Subsequently, the oil recovery, oil production rate, gas and oil ratio (GOR), and net CO₂ storage amounts are set as the targets, which correspond with four neurons in the output layer and with 10 neurons in the hidden layer. The oil recovery and net CO₂ storage can be accurately predicted in this framework. For instance, the optimal injection scheme could be obtained for a maximum economic profit in various reservoir conditions by using this method.

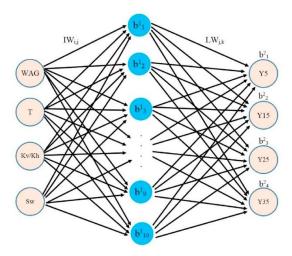


Figure 15. Artificial neural network structure of the models, Y represents oil recovery, oil production rate, gas and oil ratio (GOR), and net CO₂ storage amount [221].

The machine learning approach can also be applied to optimize oil recovery as well as CO_2 storage [222]. As shown in Figure 16, a history matching model was developed based on production history data in the CO_2 -EOR process under this optimization framework. Then, the hybridized multi-layer and radial basis function neural network method were utilized to train a proxy model, which is beneficial for increasing computational effectiveness in the optimization process. After a proxy model with reliable accuracy was realized, the machine learning optimization algorithm was used to obtain the optimal solution of the objective function that incorporates the role of parameters such as oil recovery and CO_2 storage. This work highlights the adaptability of a robust machine learning approach for optimizing the CO_2 -EOR process. Considering the mature technology and huge market demand, it can be concluded that CO_2 -EOR may play a more important role on the mitigation of CO_2 than other strategies of CO_2 utilization in the next few years.

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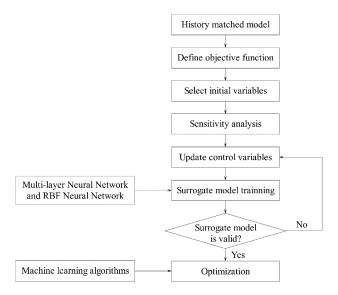


Figure 16. Flowchart of the optimization framework (modified from [222]).

4.1.2. CO₂-EGR

The technology of CO_2 -EGR means that it enhances the gas recovery by CO_2 injection. Enhanced gas recovery is realized by both the displacement and re-pressurization of the remaining gas in a depleting or depleted reservoir [223], especially for sour gas reservoirs in which CO_2 is produced mixed with the natural gas. The separated CO_2 from the produced gas could be injected back into the reservoir to enhance gas recovery. Additionally, CO_2 has the potential to reduce the dew point pressure of reservoir fluids in wet gas reservoirs, which is favorable for eliminating condensate blockage and improving CH_4 production [224,225]. It is estimated that up to 11% incremental gas recovery can be achieved by CO_2 displacement [223].

The feasibility of CO₂-EGR has been investigated by many experimental and numerical simulation studies [83,131,132,211,224,226–230]. Some typical displacement experiments in a variety of temperature and pressure conditions, revealing the mechanism of CO₂-EGR and providing a guide line for the application of this technology, are summarized in Table 6.

Table 6. Typical displacement experiments on the CO ₂ -enhanced gas recovery (CO ₂ -EGR)	process
Table of Typical displacement experiments on the CO2 children gas recovery (CO2 EGIV)	process.

Rock Type	Saturated Fluids	T (°C)	P (MPa)	Key Observations	Ref.
Carbonate core	CH ₄	20–60	3.55–20.79	Whether CO_2 is in the gas, liquid, or supercritical phase, it could enhance the recovery of CH_4 .	[83]
Carbonate core	Saturated with methane with or without water	20–80	3.55-20.79	The coefficient of CO ₂ increases with temperature and decreases with pressure.	[231]
Berea sandstone core	Dry core, initial saturation of 10% water, and initial saturation of 10% brine (20 wt %), respectively	40	8.96	The salinity of connate water will decrease the dispersion of CO_2 in CH_4 .	[232]
Sandstone and carbonate core	CH ₄	60–80	10–12	The residual water narrows the pore and consequently increases the dispersion of supercritical CO_2 and CH_4 .	[233]
Sandstone core	CH ₄ and simulate natural gas (90% CH ₄ + 10% CO ₂) respectively	40–55	10–14	The dispersion coefficient of CO_2 in the simulate natural gas is larger than that of CH_4 .	[234]
Sandstone core	Formation water and N_2	50	21	The gravity segregation effect is notable in the porous and permeable core, while the heterogeneity effect becomes dominant in the low permeability of the core.	[235]
Bandera sandstone core	CH ₄	50	8.96	The gravity has significant effects on the flow behavior of SCO ₂ at lower flow rates.	[236]

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The most critical hurdle in CO_2 -EGR is the breakthrough of CO_2 in the reservoir producing CO_2 -contaminated gas [131,237]. Actually, the preferential pathway has a significant impact on CO_2 breakthrough and ultimate CH_4 recovery [238], so the geological formations for CO_2 -EGR, especially the microstructures, are supposed to be characterized in detail. Irreducible water in reservoirs also has an impact on the mixing of CO_2 and CH_4 [239]. The dispersion increases with irreducible water, because the pores occupied by irreducible water lead to much narrower pores and more tortuous flow paths.

In addition to the above-mentioned geological parameters, engineering parameters also have a significant influence on gas mixing and CO_2 -EGR performance [131,240,241]. CO_2 injection with a horizontal well in the lower parts and CH_4 production in the upper parts of reservoirs could mitigate the breakthrough of CO_2 at the production well [131,242]. CO_2 injection during the early decline phase of natural gas production is beneficial for ensuring the displacement in supercritical phase and achieving a high CH_4 recovery [131]. On the contrary, it may cause the trapping of CH_4 in unswept areas under high pressure. CO_2 injection in the late phase could avert this shortcoming and improve the performance of CCS, which is more attractive when the CO_2 sequestration is considered [131,243]. On the whole, it can be concluded that the time of CO_2 injection to obtain a maximum incremental recovery is highly affected by the allowable produced CO_2 concentration at the production well, which is determined by the economics of CCS projects [244].

Whether the CO_2 is injected in the early or later stage, it is recommended to inject CO_2 at a relatively high pressure to ensure the supercritical phase in the displacement process. In this case, the distribution of CO_2 is dominated by gravity forces [245]. As the CO_2 is much more dense than CH_4 , CO_2 will occupy the smaller space and spread at a slower rate, which could mitigate CO_2 breakthrough. However, if the injected CO_2 is in gas phase in the reservoir, the CO_2 will occupy a large volume and mix with the CH_4 more easily, which can lead to an early CO_2 breakthrough [245].

Regarding the injection rate, of course, a high injection rate can increase the gas recovery [241]. However, a high injection rate also brings excessive gas mixing, which is harmful for methane production. It is suggested that the injection rate should be lower than the production rate to avoid an early breakthrough of CO₂ [237]. In Al-Hasami's study [223], 9% incremental methane recovery can be achieved when the CO₂ injection rate is only 13% of the production rate. Instead of the constant injection rate, a constant pressure injection scheme was proposed to avoid potential risks related to high pressure [246]. The optimal injection strategy could be achieved by an optimization code based on genetic algorithm and multi-phase simulator TOUGH2 (GA-TOUGH2).

Geological parameters also greatly affect the performance of CO₂-EGR. The viscous and gravity force affecting parameters e.g., permeability, formation dip, and thickness, play a vital role in the stability of displacement. The fluid properties such as the diffusion coefficient and water salinity take second place on affecting the CO₂ breakthrough [244]. Specifically, the connate water in reservoirs has a positive impact on CO₂-EGR performance. As a result, the dissolution of CO₂ in reservoir fluids is favorable for enhancing the storage capacity and mitigating the CO₂ breakthrough in the production well [223,237,247].

There are several CO₂-EGR projects around the world, including the Alberta gas field project, the K12-B field project, and the CLEAN project. The Alberta gas field project is located in Canada. In this project, impure CO₂ with less than 2% of H₂S has been injected into the depleted Long Coulee Glauconite F gas Pool in southeastern Alberta since 2002, but the operation was terminated in 2005 because of the breakthrough of acid gas [245]. The K12-B gas field is located in the Dutch continental shelf in the North Sea, with a reservoir depth of about 3800 m below the sea level. The reservoir pressure has dropped from 40 MPa to 4 MPa with a production of 90% of the initial gas in place. The initial reservoir temperature is 128 °C [248]. Over 0.1 million tons of CO₂, which is separated from the produced gas directly at the offshore platform, has been injected over a period of 13 years since 2004. Monitoring data shows that the well integrity has remained stable [249]. Furthermore, no major complications occurred in the lifetime of this project, which proves that the safety of CO₂-EGR can

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be ensured [250]. The CLEAN project was conducted between 2008 and 2011 to inject CO₂ into the Altmark natural gas field in Germany. The risk assessment of this project has been conducted based on digital databases. The results showed that the safety and efficiency of EGR technology based on CO₂ injection could be achieved. Meanwhile, the borehole integrity could be achieved without any intervention, providing a guideline on CO₂-EGR [251].

Generally, the technology of CO₂-EGR is still immature and needs more efforts to address the problems, such as mitigating the CO₂ breakthrough and achieving a favorable performance in both enhancing CH₄ recovery and CO₂ sequestration. The economic success is largely dependent on the political developments in the next years and decades [251].

4.1.3. CO₂-EWR

Similar to CO₂-EOR and CO₂-EGR, CO₂-EWR is a methodology combining CO₂ sequestration and saline water production [252–254], which is developed from the technology of CCS in saline aquifers. Figure 17 shows a diagram of CO₂-EWR technology. The operation of CO₂-EWR could decrease formation pressure and avoid potential leakage through extracting formation water, thus it could further improve the storage efficiency and achieve higher security and stability of large-scale geological CO₂ sequestration [255]. Besides, the produced saline water could be used for drinking, industrial, and agricultural utilization after desalination treatment such as using a high-efficiency reverse osmosis system [252]. Meanwhile, the deep brine resources obtained through the cascade extraction may create economic profit and fill the cap of cost in the operation of CCS technology [253]. Kobos et al. [252] proposed a numerical simulation model to investigate the feasibility of CO₂-EWR based on a hypothetical case study from a representative power plant and saline formation in the southwestern part of the United States. In their work, the extracted saline water was treated with a high-efficiency reverse osmosis system and then used as power plant cooling water. The results showed that the coupled technology of CO₂ storage and saline water extraction and treatment is feasible for tens to hundreds of years.

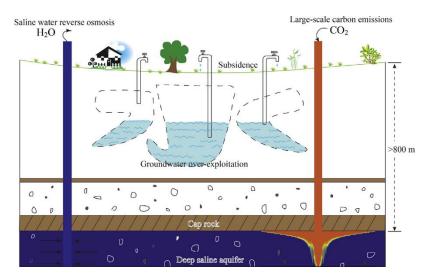


Figure 17. Depiction of the CO₂-enhanced water recovery (CO₂-EWR) technology [253].

Unfortunately, the added cost of extraction wells is considered a shortcoming of CO₂-EWR [252]. Furthermore, the production of brine must be ceased once the breakthrough of CO₂ occurs [256]. In general, under effective engineering design, the CO₂-EWR technology has application prospects.

4.1.4. CO₂-ESGR

In regard to CO₂-ESGR technology, the CO₂ is injected into shale gas reservoirs to replace and displace shale gas, for the ultimate goal of enhancing the shale gas recovery, with a side benefit of

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 CO_2 sequestration synchronously [257]. The dominate mechanism of CO_2 -ESGR is the competitive absorption of CO_2 by shale matrix [199], such as with a CO_2 sorption capacity up to 1 mmol per gram for the Muderong Shale [258]. In addition, the pressure gradient displacement plays an important role [259]. Liu et al. [259] conducted a numerical simulation to evaluate the feasibility of CO_2 -ESGR, and results showed that over 95% of the injected CO_2 was instantaneously adsorbed and sequestrated in the reservoirs. However, only limited ESGR performance was detected due to the limited communication between the wells in this study. The feasibility of CO_2 -ESGR on the Devonian Gas Shale Play of eastern Kentucky was investigated by Schepers et al. [127]. They found that the huff-n-puff scenario was not suitable, while the full-field continuous CO_2 injection was a good option. About 300 tons of CO_2 were injected within one and half months. A significantly increased recovery was attained, and approximately half of the injected CO_2 was sequestrated. Therefore, it can be concluded that there remains a long way before the application of CO_2 -ESGR, and the contribution to the mitigation of CO_2 emission is still limited.

4.1.5. CO₂-ECBM

In CO₂-ECBM, the CO₂ is injected into un-mineable coal seams to displace and replace coal bed methane, simultaneously achieving CO₂ sequestration in the coal seams. Similar to the CO₂-ESGR, CO₂ works as displacing fluid and is competitive in the process of CO₂-ECBM [260,261]. The potential ECBM recovery in China is estimated to be over 3.751 Tm³ [100], highlighting the superiority of this technology. However, the injected CO₂ in CO₂-ECBM projects is usually less than 1 million tons per year, and many coal seams usually with low permeability such as those in Western Europe are not suitable for the application of this technology [148]. Herein, the role of CO₂-ECBM on mitigating the emission of CO₂ is limited.

4.1.6. CO₂-EGS

Geothermal energy is regarded as a clean, renewable, and reliable energy for its advantages of sustainability and environment-friendly characteristics [262]. It is extracted through water traditionally. Brown [263] firstly proposed the concept of using supercritical CO_2 instead of water as the heat exchange fluid in an EGS. It has been proven that the heat extraction efficiency of CO_2 -based systems is superior to water-based systems. If this concept is popularized, more regions worldwide with relatively low temperature can be used for electricity production in an economically beneficial manner [254,262,264]. In addition, the mobility of CO_2 is better than that of water, which is beneficial for the production of fluids and the extraction of geothermal energy.

In recent years, the technology combining geothermal extraction and CO_2 storage has gained more attention [264,265], which can achieve an efficient geothermal energy extraction as well as mitigating CO_2 emission. For example, the regional energy deficit could decrease by 22.1% and the CO_2 emissions could decrease by 31.3% in the Latium Region in Central Italy if the CO_2 -EGS was applied [266]. However, the technology of the CO_2 plume geothermal system is still at the conceptual stage and pre-feasibility studies phase, and many efforts need be devoted toward its study before its application.

4.1.7. CO₂-IUL

 CO_2 -IUL is a technology that injects CO_2 and leaches uranium ore out of geological formation through reaction with ore and minerals in the ore deposits [191]. CO_2 -IUL could increase the recovery of uranium and simultaneously be favorable for CO_2 storage, especially for sandstone-type uranium mining [191]. However, the global annual demand of natural uranium is only around 0.1 million tons [267], thus it may be difficult for CO_2 -IUL to reduce CO_2 emission significantly due to its limits and demands.

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4.1.8. CH₄-CO₂ Replacement from Natural Gas Hydrates

The technology of CH₄-CO₂ replacement from natural gas hydrates (NGH) is regarded as a win–win method for exploring NGH and simultaneously storing CO₂ in the form of CO₂ hydrates formation [268–271]. As shown in Figure 18, the mechanisms of this technology can be divided into four steps. Firstly, the CO₂ molecule diffuses into the surface of CH₄ hydrate and decreases the stability of CH₄ hydrate structure (Figure 18a). Secondly, due to CH₄ hydrate dissociation, the CH₄ molecule escapes from the hydrate cage (Figure 18b). In the next stage, the hydrate is re-formed. As shown in Figure 18c, the CO₂ molecules mainly occupy the large cage, while CH₄ molecules occupy the small cage. Finally, the CH₄ molecules diffuse from the surface of hydrate and change into gas, while the CO₂ molecule diffuse into deeper hydrate layer to continue replacing the CH₄ in hydrate (Figure 18d) [270]. To improve the performance of CH₄–CO₂ replacement from natural gas hydrates, a thermal stimulation approach was proposed [271]. By using this approach, the CH₄ replacement exhibits an upper limit of 64.63%, and the maximum CO₂ storage efficiency can reach up to 78.40%–96.73% [271].

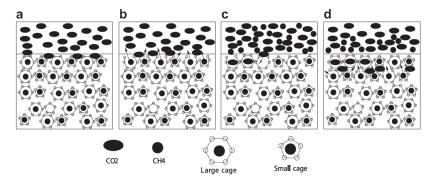


Figure 18. Schematic diagram for CH₄–CO₂ replacement in hydrates [270].

Based on the concept of thermal stimulation to CH_4 – CO_2 replacement, a geothermal-assisted CO_2 replacement method (GACR) was proposed by Liu et al. [272]. As described in Figure 19, the CO_2 with ambient temperature was injected into geothermal reservoir for heating, then the heated CO_2 flows upward into the hydrate-bearing layer (HBL) to enhance the NGH dissociation. Numerical simulation results showed that the GACR method can significantly accelerate NGH dissociation and increase CH_4 recovery. However, the application of this method is limited by a strict precondition that a thermal reservoir exists below the methane hydrate reservoir.

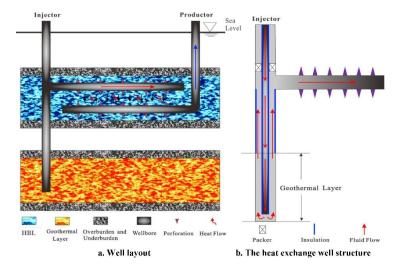


Figure 19. Schematic well group configuration diagram of the geothermal-assisted CO₂ replacement method (GACR) [272].

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The research on CH_4 – CO_2 replacement from natural gas hydrates is still in the preliminary experimental and numerical study phase [271,273,274]. However, it is expected that great progress will be made in the near future under the stimulation of methane hydrate production and CO_2 sequestration.

In a short summary of this part, the utilization of CO_2 for resources production and CO_2 storage are encouraged to be designed for the whole process of engineering operation to optimize its performance. To achieve this goal, artificial intelligence may play an important role. Although CO_2 -EOR has been used commercially, the other technologies are in the pilot plant phase in terms of technology readiness level [204]. CO_2 -EOR would be the most promising technology combining the utilization and sequestration of CO_2 in the next few years. CO_2 -EGR is another promising technology, whose application is limited by the mixing of CO_2 and CH_4 in the reservoir. Considering that the mixing behavior is affected by the geological parameters, i.e., porosity, permeability, residual water saturation, and engineering parameters, i.e., injection rate, injection pressure, production rate, a site selection system for CO_2 -EGR project is encouraged to be developed.

4.2. Co-Injection of CO₂ with Impurities

The biggest obstacle for large-scale CO_2 storage is the lack of financial incentives [12]. Most strategies of CO_2 storage could not generate profit, so the measures to decrease the cost of CO_2 storage is very important and beneficial for the large-scale application of this technology. It is estimated that the cost of carbon capture and storage is dominated by the process of capture and gas separation, which costs \$55 to \$112 per ton of CO_2 [12]. Therefore, co-injection of CO_2 with impurities can be a cost-effective option for CO_2 sequestration [275].

The impurities may be co-injected with CO_2 including CH_4 , H_2S , SO_2 , N_2 , and O_2 . Among them, H_2S and CH_4 are usually mixed with CO_2 in produced acid gas, which can be used in CO_2 storage. Other gases are the main components of flue gas, which is captured from the major CO_2 emission sources such as power plants [276].

The impure CO_2 can also be used for CCUS especially for CO_2 -EOR with multicontact miscible CO_2 flooding [195]. In the multicontact miscible displacement procedure, the MMP is a key control variable due to it having a notable impact on the design and development of assets and being closely related to the economically feasibility, which is affected by the impurities in CO_2 a lot. In general, the presence of H_2S and SO_2 in CO_2 can reduce the MMP [195,277], while the presence of CH_4 , CO_2 can increase the MMP of CO_2 [278,279], which is disadvantageous for the CO_2 -EOR operation and may increase the risks for the fracturing of the formation due to the higher injection pressure required.

The presence of impurities may change the other CO_2 thermophysical properties and phase behavior [280], and affect the performance of CCS. N_2 would lead to a delay for CO_2 breakthrough when it is co-injected, because the solubility of CO_2 in irreducible water is much higher than that of N_2 [281]. However, the N_2 would decrease the density of the dissolved phase and increase the risk in the long term [282]. Generally, the storage capacity of reservoirs decreases proportionally to the concentration and the compressibility factor of impurities when N_2 is co-injected with CO_2 [82]. For instance, the reduced storage capacity may be even higher than the volume fraction of impurities when O_2 is included. However, the negative impact of impurities on capacity could be alleviated by storing the impure CO_2 in a reservoir with high temperature [276].

There is no significant effect of H_2S , with a fraction of less than 30%, as impurity on the dissolution of CO_2 [275]. However, when it was co-injected with CO_2 under the condition of 20 MPa and 45 °C, the H_2S with a concentration over 20% has a potential to decrease the interfacial tension and increase the contact angle, leading to a low capillary force [283]. This means that H_2S may increase the risks of gas leakage, which should receive attention. The impure CO_2 with H_2S can be trapped by hematite even in a dry system driven by the reduction of ferric iron in hematite by sulfide species, verifying the feasibility of co-injection of CO_2 with CO_2 with CO_2 with CO_3 with CO_4 with C

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 CH_4 produced from an acid gas reservoir may also serve as impure gas and be used in CCS projects. There is no significant negative influence of CH_4 on the interfacial tension and wettability even with the concentration of up to 20% [283]. The storage capacity of reservoirs also decreases proportionally to the concentration and compressibility factor of CH_4 [82]. As the concentration of CH_4 in injected CO_2 is very low, its effect on the CCS is minor and can be neglected.

The presence of SO_2 usually controls the acid-induced reactions with calcium-rich minerals when it is co-injected into reservoirs as an impurity, but the quantitative effect is very minor and could be negligible based on the German Bunter Sandstone [285]. Generally, the porosity in sandstone increases under the impact of SO_2 , while the porosity of the intermediate shale layer decreases because of the conversion of dominant calcite to anhydrite [286]. For instance, the conversion of Ca^{2+} bearing carbonate to anhydrite is observed when the SO_2 was co-injected with CO_2 into the German Bunter Sandstone [287]. A field study was conducted to investigate the geochemical impacts of SO_2 and SO_2 as impurities on the reactions of minerals and fluids in a siliciclastic reservoir [288]. The SO_2 -saturated water with impurities was injected into reservoirs and allowed to interact with minerals for three weeks. The results showed that the pyrite dissolved due to the SO_2 acting as an oxidizing agent. However, the concentrations of SO_2 and SO_2 are 67 ppm and 6150 ppm respectively, which is too low to lead to a significant effect on fluid-rock interaction. It could be inferred that the impact of impurities on the interaction with formation rock is highly dependent on the composition of minerals, which should be analyzed site specifically. It should be mentioned that the co-injection of SO_2 and SO_2 could suppress Joule-Thomson cooling, which is a beneficial thermal consequence for SO_2 .

In short, co-injection of CO_2 with impurities is an effective strategy to reduce the cost of CO_2 storage. However, the interaction of the impurities with formation rock, and the effect of impurities on thermophysical properties of reservoir fluids need to be further studied to reduce the uncertainties in CO_2 storage process.

4.3. Prospects of CCS/CCUS Technologies

The economic factor for the CCS projects is believed to be one of the most important incentives for the industry. The price for CO_2 emissions at the first major carbon market and also the biggest one, i.e., the European Union Emission Trading System, is approximately \$7 per ton of CO_2 , which is much lower than the cost of CCS [12]. Therefore, there are no financial incentives of CCS for industries unless a higher price of carbon emission is set, demonstrating the important role that should be played by the government in the mitigation of CO_2 emission.

Table 7 shows the large-scale CCS projects (more than $0.4 \,\mathrm{Mtpa}$) throughout the world from 1972 until the end of the 2020s. It can be seen that the CCUS for EOR and CCS in saline formations have made major contributions toward CO_2 storage, which is in accordance with the prediction model formulated by Mac Dowell et al. [203]. Nearly half of 51 large-scale CCS projects scheduled by the end of the 2020s are designed for EOR, which shows its economic viability in EOR operations. In addition, 21 projects of CCS in saline formations are also planned, since its CO_2 storage capacity may up to 4 Mtpa. Moreover, the average CO_2 capture capacity of CCS in depleted gas fields is much greater than that of EOR and may reach to 2.8 Mtpa, showing the potential of the mitigating of CO_2 emissions. However, on account of the great extent of the mixing between CO_2 and CH_4 , which is an obstacle for enhancing additional recovery of CH_4 , there are only three large-scale CCS projects in depleted gas fields in the near future. With the development of technology to address this issue, CCS in depleted gas fields can play a more important role in CO_2 storage.

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Strategy	Under Evaluation	EOR	Saline Formation	Depleted Gas Fields	Total
Quality of project	24	3	21	3	51
Capture capacity (Mtpa)	42.11-43.41	8.1 - 8.6	40.35-85.1	7.5–8.5	98.06-145.61
Average capture capacity (Mtpa)	1.75–1.81	2.7–2.87	1.92-4.05	2.5–2.83	1.92-2.86

Table 7. Large-scale CCS projects (more than 0.4 Mtpa) from 1972 to the end of the 2020s (data from [10]).

5. Conclusions

The status of the strategies for CO_2 storage has been discussed in view of assessing the security as well as improving the cost-effectiveness. In addition, the role of CCS technologies and their potential contribution on the mitigation of CO_2 emissions in future are summarized. Based on the studies carried out in this review, the following conclusions have been obtained.

Firstly, the sequestration of CO_2 in depleted oil and gas reservoirs could play an important role in reducing CO_2 emissions in the near future, because the existing installed equipment and comprehensively characterized reservoir integrity will significantly reduce the cost of CCS. The leakage of CO_2 through abandoned wells is an obstacle for the application of this technology. To address this issue, the long-term experiments and molecular dynamic simulations are needed to figure out the kinetics between CO_2 with the well string, cement, as well as formation minerals under the relevant conditions. Secondly, if implemented on a large scale, CO_2 storage in saline aquifers may make the biggest contribution in reducing CO_2 emissions due to its huge storage capacity. Moreover, the scientifically proven technologies such as CO_2 storage in coal beds, deep ocean, and deep-sea sediments are still immature technologies and do not appear to be capable of making a great contribution to the mitigation of CO_2 emissions in the foreseeable future.

Another point is the need to investigate accurate risk assessment associated with CO₂ storage and provide a guideline for the design of CCS projects. Attempting to make the CCS assessment more intelligential, the machine learning technology ought to be used.

It has also been demonstrated that the direct mineralization of flue gas by coal fly ash would be a promising technology result since the gas could be mineralized without separation. In addition, the pH swing mineralization may be another promising technology for the sequestration of CO_2 due to the sustainability. The recyclable and cheap chemical reagents ought to be introduced into the mineralization process to make this technology more cost-effective.

Among the variety of CCUS strategies, CO_2 -EOR followed by CO_2 -EGR is supposed to play the most important role in the mitigation of CO_2 in the next few years. The utilization of other strategies seems to be negligible in the near future. The co-injection of impurities with CO_2 is an effective methodology to decrease the overall cost of CO_2 storage. The physical and chemical effects of the impurities on reservoir fluids and formation rock should be studied site specific, to reduce the uncertainties in CO_2 storage.

The government is supposed to play a major role in mitigating CO_2 emission, a higher tax on CO_2 emissions and financial subsidy on CO_2 storage is encouraged to accelerate the deployment of CCS projects at a large-scale.

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