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**Numerical study of underground CO₂ storage and the
utilization in depleted gas reservoirs**

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1 Introduction

In this chapter, the background and the latest developments of CO₂ storage is introduced. Specifically, the background and the mechanisms and strategies of CO₂ storage, focusing on their characteristics and current status, are presented firstly. Then 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. Further, 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 are discussed. Based on the literature review, this thesis aims at reduce the risks related to CCS and increases the cost-effectiveness of CCS. The research objectives and outline of this thesis are also presented. The main contents of this chapter have been published in the following research paper (Cao et al. 2020): A review of CO₂ storage in view of safety and cost-effectiveness. *Energies*, 13(3), 600.

1.1 Introduction of underground CO₂ storage

The CO₂ concentration in the atmosphere locates at a level of below 300 ppm in pre-industrial times, whereas it has already risen above 410 ppm in the last few centuries (Met Office 2017; Scripps CO₂ Program 2019). Especially, the CO₂ concentration increases dramatically since 1960s as can be seen in Fig. 1.1. Fig. 1.1 also shows the correlation between the atmospheric concentration of CO₂ and the global temperature since 1850s. It can be seen that the continuous rise in global temperature is strongly related to the atmospheric concentration of CO₂, which indicates that CO₂ is the main contributor to global warming and climate change. More importantly, it is estimated that CO₂ makes up an 77% of greenhouse gases across the world (MacDowell et al. 2010; Rahman et al. 2017). Furthermore, the CO₂ emission may increase the frequency of extreme weather such as the extreme extratropical cyclones. Specifically, it is estimated that the number of extratropical cyclones will be more than triple by the end of this century in North America and Europe if the greenhouse gas emissions hasn't been efficiently mitigated (Hawcroft et al. 2018). To deal with such intense global climate problem, the Intergovernmental Panel on Climate Change's (IPCC) suggested that the increment of the average earth's surface temperature should be limited less than 2 °C within this century based on the Integrated Assessment Models (IAMs)'s estimation (Edenhofer et al. 2014).

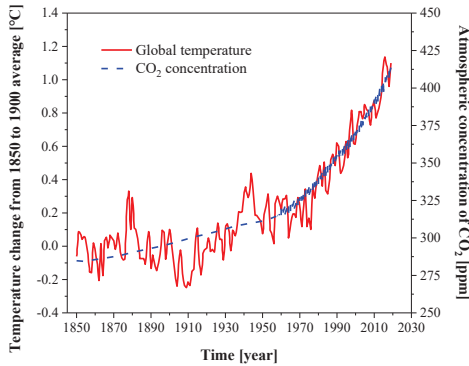


Figure 1.1 Correlation between atmospheric concentration of CO_2 and the global temperature since 1850s (Cao et al. 2020)

To achieve the IPCC's goal on global temperature control, carbon capture and storage (CCS) is supposed to be promoted. This is result from that CCS is currently regarded as the most effective strategy for slowing down the atmospheric CO_2 emissions and attenuating associated climate problems (Brinckerhoff 2011). As can be seen in Fig. 1.2, it is estimated that approximately 10.8 Gt CO_2 can be trapped through CCS alone by 2050, which undertakes almost 19% reduction in global CO_2 emissions (IEA 2010). Further, the overall cost of achieving the same targets of CO_2 emission reduction will increase by 70% without the application of CCS (IEA 2009), demonstrating the importance of CCS on the mitigation of atmospheric CO_2 emissions from the economic point of view as well. It should be mentioned that CCS is also beneficial for the circulation carbon economy, which offers a realistic and technology-neutral strategy that focusses on carbon management and will ultimately lead to a carbon-neutral energy future (IEF 2020).

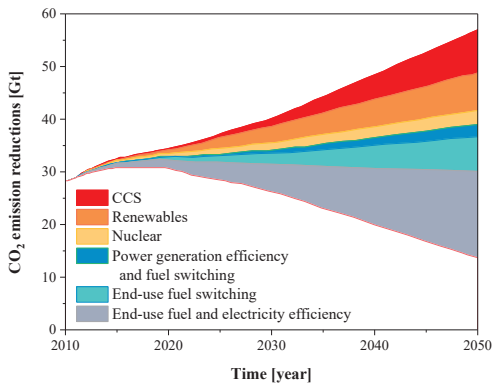


Figure 1.2 IEA forecasts of key technologies for CO_2 emission reductions (Cao et al. 2020; IEA 2010)

As can be seen in Fig. 1.3, a total of 51 CCS engineering projects are projected across the world, which are mainly scheduled in North America, Australia, Western Europe, and China. It should be mentioned that only 19 CCS projects are currently in operation (Global CCS Institute 2019). The main factors challenge the large-scale application of CCS are the high cost and safety risk associated with CO₂ leakage, even though CCS has been proven to be technically feasible. As a result, the contribution of CCS is still very limited in mitigating climate change (Gislason and Oelkers 2014; Pawar et al. 2015). Therefore, more research efforts on improving the safety and economics of CCS are required to develop this kind of technology, improving public acceptance, gaining support from government, and to accelerate the application of CCS in large-scale.

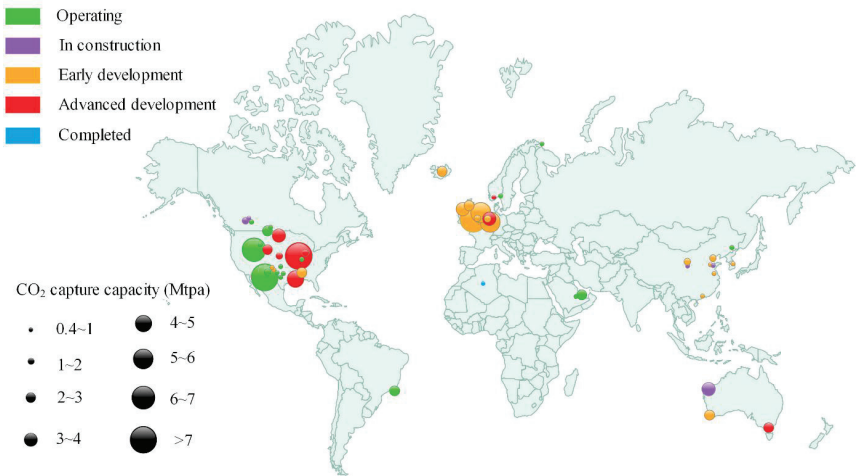


Figure 1.3 Commercial-scale integrated CCS projects around the world. Circle size is proportional to the CO₂ capture capacity and the color indicates different stages of the lifecycle of the project (Cao et al. 2020)

The review literatures in the past ten years on CCS technology are summarized in Tab. 1.1. It can be seen that almost every aspect of CCS technology including CO₂ capture and utilization, options for CO₂ storage and CCS projects, CO₂-brine-rock systems, well integrity and risk assessment, and storage efficiency and environmental considerations have been discussed extensively in the last decade (Abid et al. 2015; Abidoye et al. 2015; Aminu et al. 2017; Atia and Mohammadi 2018; Bachu 2015; Bai et al. 2016; Boot-Handford et al. 2014; Burnside and Naylor 2014; Carroll et al. 2014; De Silva et al. 2015; Godec et al. 2014; Kemper 2015; Koytsoumpa et al. 2018; Li et al. 2013; Li and Liu 2016; Liu et al. 2017; Mayer et al. 2015; Michael et al. 2010; Oh 2010; Pan et al. 2016; Pires et al. 2011; Riaz and Cinar 2014; Sanna et al. 2014; Shukla et al. 2010; Singh and Haines 2014; Song and Zhang 2013; Tan et al. 2016; Tang et al. 2014; Verduyn et al. 2011; Wee 2013; Zahid et al. 2011; Zhang and Bachu 2011).

However, the strategies for improving the safety and economics of CCS have not been discussed in detail. In addition, the technology of CCS is developing rapidly so that the recent development needs to be reviewed and discussed.

Table 1.1 Summary of review literature on CCS technology (Cao et al. 2020)

Research fields	Source	Review scope
CO ₂ capture and utilization	Atia and Mohammadi 2018	Review of the application of CO ₂ for enhanced oil and gas recovery
	Koytsoumpa et al. 2018	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
	Li et al. 2013	Review of CO ₂ capture, utilization, and storage (CCUS) in Chinese Academy of Sciences, highlighting the strategies for CCUS in China
	Tan et al. 2016	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
	Boot-Handford et al. 2014	Review of CCS highlighting the CO ₂ capture technologies, the pilot plants, and the economic and legal aspects of CCS
	Godec et al. 2014	Review of CO ₂ enhanced coalbed methane recovery, highlighting the CO ₂ storage trials in the San Juan Basin in USA, and the estimation of CO ₂ storage capacity in coal seams
	Liu et al. 2017	Review of CCUS technologies highlighting the engineering projects and their developments in China
Options for CO ₂ storage and CCS projects	Pires et al. 2011	Review of CCS highlighting the findings obtained in CCS operational projects including the technologies of CO ₂ capture, separate, transport, and storage
	Aminu et al. 2017	Review of CCS highlighting the options for CO ₂ storage, the evaluation criteria for CO ₂ storage site, and the major CO ₂ storage projects
	Kemper 2015	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
	Michael et al. 2010	Review of CO ₂ storage in saline aquifers, highlighting the geological and operation parameters, and the monitoring technologies for existing saline aquifers storage operations
	Oh 2010	Review of the CCS in coal-fired plant in Malaysia, highlighting the choices of coal plants and the capture technologies
	Riaz and Cinar 2014	Review of CO ₂ storage in saline formations, highlighting the modeling of solubility trapping
	Sanna et al. 2014	Review of mineral carbonation (MC) technologies for CO ₂ sequestration, highlighting the mechanisms of MC technologies and their contribution in decreasing the cost of CCS
	Singh and Haines 2014	Review of CCS projects and future opportunities, highlighting the technical details and business plan for CCS projects
Tang et al. 2014	Review of CO ₂ storage projects in China, highlighting the CO ₂ source, and CO ₂ storage strategies in China	

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	Verduyn et al. 2011	Review of CO ₂ mineralization product forms, highlighting the mineralization process for CO ₂ storage
	Wee 2013	Review of CCS by using coal fly ash, highlighting the feasibility and prospects of CCS using coal fly ash
CO ₂ -brine-rock systems	Burnside and Naylor 2014	Review of the relative permeability and residual trapping in CO ₂ storage systems, highlighting the estimating and measuring methods
	De Silva et al. 2015	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
	Pan et al. 2016	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
	Abidoye et al. 2015	Review of CO ₂ sequestration highlighting the trapping mechanisms and the flow of CO ₂ -brine in porous media system
Well integrity and risk assessment	Abid et al. 2015	Review of the cement degradation in CO ₂ -rich condition of CCS projects highlighting the degradation of Portland cement
	Li and Liu 2016	Review of the risk assessment of CO ₂ storage, highlighting the regulations and strategies of risk assessment for CO ₂ storage
	Mayer et al. 2015	Review of the isotopic composition of CO ₂ for leakage monitoring in CCS project, highlighting the stable isotopes as a tracer for injected CO ₂
	Zhang and Bachu 2011	Review of the integrity of existing wells for CCS, highlighting the mechanical well failure and chemical issue due to cement carbonation
	Bai et al. 2016	Review of well integrity of CCS highlighting the corrosion of metallic and cement, and the remedial measures
Storage efficiency and environmental considerations	Song and Zhang 2013	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
	Zahid et al. 2011	Review of CO ₂ storage highlighting the capacity estimation of storage sites, the monitoring technologies and simulation tools for CCS
	Shukla et al. 2010	Review of CO ₂ storage and caprock integrity, highlighting the major CCS project in operation and CO ₂ migration in the reservoirs
	Bachu 2015	Review of CO ₂ storage efficiency in saline aquifers, highlighting the factors that affect CO ₂ plume migration and the methods to estimate the storage capacity
	Carroll et al. 2014	Review of environmental considerations for CO ₂ storage in sub-seabed, highlighting the potential ecological impacts

In the following section, the most recent progress on addressing the challenges related to assessing and decreasing the risks of CO₂ leakage, cutting the cost of CO₂ storage, and promoting the developments of commercial scale CCS projects will be reviewed and analyzed. Firstly, the mechanisms of CO₂ storage and the strategies of CO₂ storage are reviewed and discussed. Then the risk assessment of CO₂ storage and strategies for decreasing the risks of CO₂ leakage, including accelerating CO₂ dissolution and mineral carbonation, are summarized. Finally, the strategies for cutting the cost and acquiring

additional benefits of CO₂ storage to improve its cost-effectiveness, including co-injection of CO₂ with impurities and enhanced industrial production with CO₂ storage, are discussed.

1.2 Mechanisms of CO₂ storage

Figure 1.4 shows the phase diagram of CO₂. Considering that the pressure and temperature in the process of CO₂ storage is range from approximately 5 to 60 MPa and 20 to 150 °C respectively, thus the CO₂ may in the gaseous and supercritical state. For instance, when the pressure and temperature reach to the critical pressure and critical temperature, i.e., 7.38 MPa and 31.04 °C, CO₂ will exit in supercritical state and owns the characters of both gaseous CO₂ and liquid CO₂. On the one hand, the supercritical CO₂ has a low viscosity like gas, which is beneficial for improving the injectivity. On the other hand, the supercritical CO₂ has a high density like liquid, which is beneficial for improving the storage capacity in CCS systems.

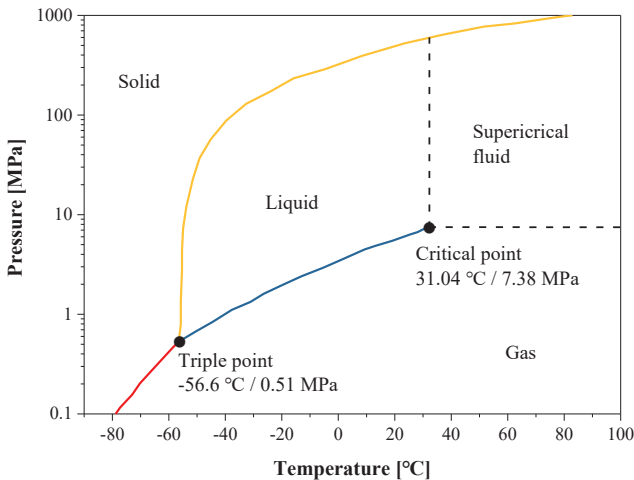
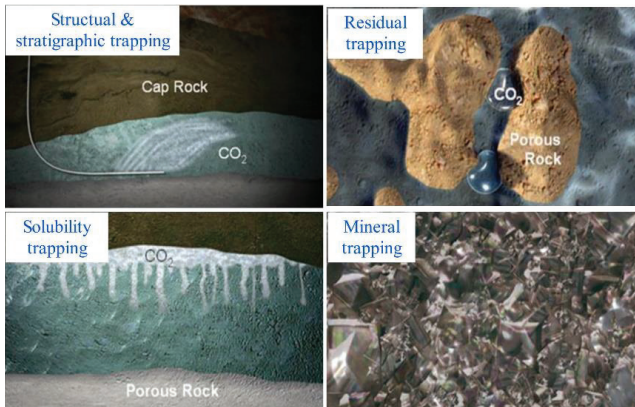


Figure 1.4 Phase diagram of fluid carbon dioxide (Data from Vargaftik 1975)

After the CO₂ has been injected into underground reservoirs, four main CO₂ trapping mechanisms may play a role on the trapping of CO₂ storage. As shown in Fig. 1.5a, the CO₂ trapping mechanisms consist of structural and stratigraphic trapping, residual trapping, solubility trapping, and mineral trapping (Shukla et al. 2010). The structural and stratigraphic trapping is regarded as the most dominant trapping mechanism. Once CO₂ is injected into subsurface reservoir formations, it will migrate upward to the top of geological structures owing to the buoyancy effect. Then the CO₂ will stay below the impermeable caprock. Regarding the residual trapping, the injected CO₂ will displace formation fluids when it migrates through the reservoir rock. Further, the displaced fluid disconnects and traps the remaining CO₂ within the pores of rocks due to the capillary force (Bradshaw et al. 2007). In the residual trapping,

the CO_2 is trapped by capillary force. It can achieve trapped CO_2 at a saturation of at least 10% and even reach more than 30% of the pore volume in some formation rocks (Krevor et al. 2015; Zhang and Huisigh 2017). Regarding solubility trapping, CO_2 will dissolve in formation fluids and become immobile, thus decreasing the mole fraction of free CO_2 (Bian et al. 2019). It should be mentioned that the dissolved CO_2 will slightly increase the density of formation fluids by around 1%, which is sufficient to promote the convection flow with the help of such a small density difference (Zhang et al. 2008). This convection flowing is also in favor of the trapping of CO_2 . Under the temperature, pressure, and salinity conditions of conventional CCS reservoirs, the solubility of CO_2 in groundwater ranges from 2% to 6%. It should be pointed that the solubility of CO_2 decreases with the growing temperature and salinity (Zhang and Huisigh 2017). In mineral trapping mechanism, CO_2 is trapped by the geochemical reactions with the rocks in reservoir. The CO_2 usually precipitates as carbonate so that it can be trapped in immobile secondary phases effectively (Sundal et al. 2014).

As shown in Fig. 1.5b, different trapping mechanism plays different role on CO_2 storage in the time scale between 1 and 10,000 years. It can be seen that the structural trapping plays an important role in the initial stage of CO_2 storage. However, the effect of structural trapping becomes weak gradually. Fig. 1.5a also shows that the residual trapping and solubility trapping have a significant impact in the time scale of tens of years. Further, the residual trapping and solubility trapping would lock up a certain amount of CO_2 for thousands of years. Regarding the mineral trapping, it begins to work at almost around one hundred years and its effect would increase gradually. Finally, the mineral trapping can play a key role in a geological timescale.



(a)

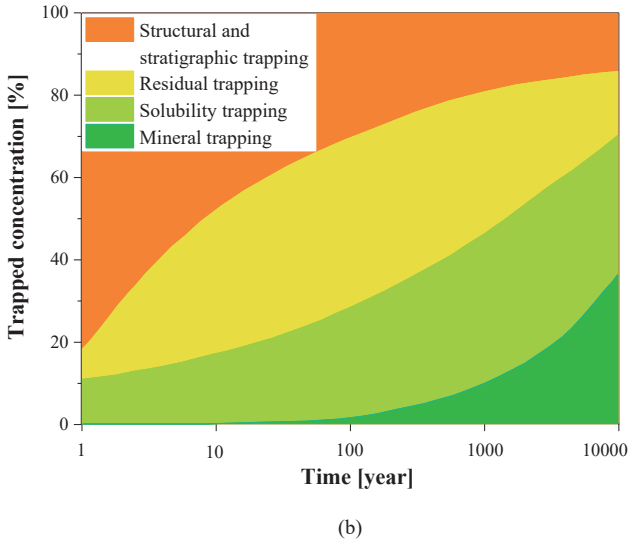


Figure 1.5 (a) The four main CO₂ trapping mechanisms (Zhao et al. 2014); (b) the contribution of four CO₂ trapping mechanisms with time (Cao et al. 2020; Metz et al. 2005)

1.3 Geologic storage options of CO₂

1.3.1 Saline aquifers

CO₂ storage in saline aquifers is one of the most important strategies because of the huge amount of storage capacity. It is estimated that approximately 10,000 Gt of CO₂ could be sequestered by the saline aquifers around the world. In other words, the saline aquifers are sufficiently store the CO₂ emissions from large stationary sources for more than 100 years (Celia et al. 2015; Davison et al. 2001; De Silva et al. 2015). Furthermore, the saline aquifers usually have a greater regional coverage and more wide distribution compared with the other storage options. Therefore, the saline aquifers have a better chance to be located nearby the sources of CO₂ emission, which could reduce the cost of CO₂ transportation (Cooper 2009; Zhang and Huisingh 2017). There are two crucial problem brought by CO₂ storage in saline aquifers. The first one is the pressure build up, which has the potential to lead to the fracturing of formation and the reactivation of faults. The second one is the CO₂ plume migration in formation, which may lead to the leakage of CO₂ that should be paid more attention (Orlic 2016). Birkholzer et al. (2009) conducted a numerical simulation to investigate the impact of large-scale CO₂ sequestration with an injection rate of 1.52 million tons per year (Mtpa) in a saline aquifer open boundary. The results showed that there is significant pressure build up in the reservoir formation at the zone even more than 100 km away from the injection zone, whereas the CO₂ plume migration is rather small that is approximately 2 km and is concentrated on the top of saline aquifer caused by the buoyancy effect. Their results also

showed that the pressure perturbation could affect the shallow groundwater formation if there is a caprock with relatively high permeability (higher than 10^{-18} m²) between the shallow layers and the saline aquifer. Fortunately, it should be mentioned that the migration of reservoir fluids, i.e., the CO₂ and formation water, into groundwater formation is extremely unlikely. This demonstrates the safety and suitability of large-scale CO₂ sequestration in saline aquifers.

A total of five commercial-scale CCS projects across the world have been launched in saline aquifers, including the Sleipner project (Audigane et al. 2007; Audigane et al. 2006; Williams and Chadwick 2018), the Snøhvit project (Hansen et al. 2013), the In Salah project (Ringrose et al. 2013; Rutqvist et al. 2010), the Gorgon project (Flett et al. 2008), and the Quest project (Bourne et al. 2014). Regarding the Sleipner project, the CO₂ was injected into a saline aquifer within the Utsira Sand formation. The injected CO₂ was separated from the produced natural gas at the Sleipner field in the North Sea. Generally, a total of 18 million tons of CO₂ has been injected by 2018 since the initiation in 1996 (Williams and Chadwick 2018). Based on the engineering experiences of the Sleipner project, the Snøhvit CCS project that is located in the Barents Sea was launched in 2008 with a total amount of 1600 ktons of CO₂ injected till August 2012. In this project, the CO₂ separated from the LNG project was injected into the deeper Tubåen Formation. It is scheduled that around 23 million tons of CO₂ would be sequestered in the reservoir based on the projected lifetime of the Snøhvit LNG project (Hansen et al. 2013; Simmenes et al. 2013).

The project located at In Salah, Algeria, is a pioneering CCS project across the world. A total of more than 3.8 million tons of CO₂ have been injected into the Krechba field since 2004 (Ringrose et al. 2013). It's worth to be mentioned that the diversity of monitoring methods including satellite monitoring and 4D seismic have been used in this CCS project to monitor the response of formation to CO₂ injection. Meanwhile, the accessibility of the monitoring data to the public is very high (Bjørnarå et al. 2018; Eiken et al., 2011; Gemmer et al. 2012; Newell et al. 2017; Rinaldi and Rutqvist 2013; Rinaldi et al. 2017; Ringrose et al. 2013; Rutqvist et al. 2010; Shi et al. 2013; Stork et al. 2015), so it could be served as a commendable case to investigate the CCS in saline aquifers.

The Quest CCS project launched in 2015, which is designed to store the CO₂ from an existing facility for upgrading heavy oil in Scotford of Alberta, Canada. It is expected that around 27 million tons of CO₂ could be injected into the Basal Cambrian Sands formation through 3 to 8 vertical wells with an injection rate of 1.08 Mtpa (Bourne et al., 2014).

The Gorgon CCS project is located in the northwest of Australia. There is a Jurassic saline reservoir in the Dupuy Formation that can be served as reservoirs for CO₂ storage. During the lifetime of the Gorgon project, a total of more than 120 million tons of CO₂ is planned to be injected into the Dupuy Formation at an injection rate of 3.8 Mtpa (Flett et al. 2008).

Aside from the forementioned large-scale CCS projects, there are some small-scale projects as well, including the Illinois Basin-Decatur Project (Finley et al. 2013), Ketzin pilot site (Martens et al. 2012; Opedal 2018), and Shenhua CCS demonstration project (Yang et al. 2017). Generally, these CCS projects have been conducted with detailed modeling and monitoring during operation, which demonstrates the safety and suitability of this technology. At the same time, it helps increase the public acceptance about CCS technology.

However, although the CO₂ storage capacity of saline aquifers is huge, the overall application of CO₂ storage in saline aquifers across the world is still at a small-scale because of the lack of financial incentives. Therefore, the policies related to the taxes on carbon emission may need to be formulated, which demonstrates the important role on the application of CCS should be played by the government.

1.3.2 Depleted oil and gas reservoirs

There are many merits for CO₂ storage in depleted oil and gas reservoirs. Firstly, there are many existing equipment installed on the surface and underground in depleted oil and gas reservoirs, thus it can be reused for CO₂ sequestration with only minor modification. Secondly, the seal quality and the integrity of the caprock are guaranteed. The geological conditions of the depleted oil and gas reservoirs have also been comprehensively characterized during the exploration and production process (Orlic 2016). Thirdly, the change of induced stress and the extent of pressure perturbations is much smaller compared with saline aquifers due to the long-term extraction of oil and gas from the reservoirs (Orlic 2016). It should be mentioned that the depleted gas reservoirs are more favorable for CCS compared with depleted oil reservoirs. This is result from that a larger CO₂ storage capacity per pore volume is available due to the higher compressibility of gas and ultimate recovery (Barrufet et al. 2010; Mamora and Seo 2002; Stein et al. 2010). Regarding the types of gas reservoirs used in this form of storage, the condensate gas reservoirs are more advantageous over the wet and dry gas reservoirs. There are several reasons account for it. Firstly, there is little gas remained in the condensate gas reservoirs thus more effective volume could be used for CO₂ sequestration. Secondly, the phase behavior of the mixture of condensate gas and CO₂ is favourable for CO₂ sequestration. Thirdly, the good gas injectivity is accompanied with the condensate gas reservoirs (Raza et al. 2018). Furthermore, the stored CO₂ per pore volume in depleted condensate reservoirs is very high. Specifically, it is approximately 13 times higher than that of the equivalent aquifer (Barrufet et al. 2010). However, it should be mentioned that the phase change may occur in depleted condensate reservoirs that should be paid for attention.

There are some characteristics associated with the long-term trapping mechanisms of CO₂ in natural gas fields. It is reported that the solubility trapping in formation water is dominated while the mineral trapping is limited in the natural gas reservoirs with siliciclastic or carbonate lithologies. This is verified by the results of noble gas and carbon isotope traces (Gilfillan et al. 2009). It is worth to mention that the residual gas saturation in the depleted reservoirs has an impact on the CO₂ storage capacity.