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Perspective

Carbon Circular Utilization and Partially Geological Sequestration: Potentialities, Challenges, and Trends

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Abstract: Enhancing carbon emission mitigation and carbon utilization have become necessary for the world to respond to climate change caused by the increase of greenhouse gas concentrations. As a result, carbon capture, utilization, and storage (CCUS) technologies have attracted considerable attention worldwide, especially in China, which plans to achieve a carbon peak before 2030 and carbon neutrality before 2060. This paper proposed six priorities for China, the current world's largest carbon emitter, to achieve its dual carbon strategy in the green energy transition process. We analyzed and summarized the challenges and potentialities of conventional carbon utilization (CU), carbon capture utilization (CCU), and CCUS. Based on the current development trend, carbon dioxide capture, circular utilization, and storage (CCCUS) technology that integrates carbon *circular* utilization and partial sequestration, with large-scale underground energy storage were proposed, namely biomethanation. Technically and economically, biomethanation was believed to have an essential contribution to China's renewable energy utilization and storage, as well as the carbon circular economy. The preliminary investigation reveals significant potential, with a corresponding carbon storage capacity of 5.94×10^8 t~7.98×10^8 t and energy storage of 3.29×10^{12} kWh~4.42×10^{12} kWh. Therefore, we believe that in addition to vigorously developing classical CCUS technology, technical research and pilot projects of CCCUS technology that combined large-scale underground energy storage also need to be carried out to complete the technical reserve and the dual-carbon target.

Keywords: carbon neutrality; carbon circular utilization; partially geological sequestration; renewable energy; underground biomethanation

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1. Introduction

Greenhouse gases (GHG) are gases that trap heat in the Earth's atmosphere, of which carbon dioxide (CO_2) is a major component and is emitted through human activities such as burning fossil fuels (coal, natural gas, and oil), biological respiration, and specific industrial reactions (e.g., manufacture of cement) [1]. In 2020, CO_2 accounted for about 76% of all greenhouse gas emissions from human activities [2]. Moreover, the concentration of atmospheric CO_2 has significantly increased from a preindustrial baseline of 280 ppm, reaching 415 ppm in 2022 [3,4]. Particularly, it has maintained an annual growth of 2.4 ppm over the past decade, which has aggravated the greenhouse effect and led to an increase in global temperatures. To limit these impacts, the Paris Agreement, which aimed to constrain warming to 1.5–2 °C, was proposed in 2015 [5]. Apparently, this goal related to the whole global effort requires drastic reductions in GHG emissions. According to the IPCC's report, limiting temperature rise to 1.5 °C by the end of the century, global annual emissions need to be reduced from 50 $GtCO_2e$ (billion metric tons of CO_2 equivalent) to 25–30 $GtCO_2e$ by 2030 [6,7]. Evidently, various efforts have been undertaken globally, to date, more than 130 countries worldwide have committed to carbon neutrality, with the developed world's universal label proposing to achieve carbon neutrality by 2050 and Germany by 2045. As the world's largest emitter of CO_2 with a population of 1.3 billion, China has also put forward the target of carbon peaking and carbon neutrality before 2030 and 2060, respectively. Regarding China's external dependence on oil and gas has exceeded 70% and 40% [8], respectively, and coal remains China's main fossil energy source [9], achieving carbon neutrality is of critical strategic importance to ensuring China's energy security, improving ecological management, and enhancing national competitiveness. Furthermore, among the dual carbon strategies, significantly improving energy efficiency, increasing the proportion of renewable energy, promoting a green and low-carbon energy transition, and building a new type of power system mainly based on renewable energy are necessary measures and essential approaches.

Based on the analysis of the practical experience overseas, as well as the consideration of China's status quo, "Six Priorities" are proposed by the authors as follows: (1) Energy saving and energy efficiency prior to renewable energy; (2) Local renewable energy prior to remote energy; (3) Renewable energy prior to a carbon sink; (4) Natural carbon sink prior to anthropogenic carbon sink like CCUS; (5) Carbon reduction prior to a carbon sink; (6) Electrification prior to hydrogenation. However, China's land natural carbon sink capacity is restricted, with an estimated around 1.1 billion tons, and it is expected to grow steadily to nearly 1.5 billion tons in the future by expanding forestry, planting trees, etc. [10]. Comparatively, in 2019, China's CO_2 emissions from fossil fuel combustion reached 9.8×10^9 t, and the total carbon emissions exceeded 1×10^{10} t in 2018 [11]. Therefore, achieving a balance of carbon emissions and carbon sink also requires anthropogenic carbon uptake, like CCUS, which is an important part of China's technology portfolio for achieving its carbon neutrality target. According to the current technologies development, in 2050 and 2060, the emission reductions that need to be realized through CCUS technologies are $0.6{\sim}1.4 \times 10^9$ t and $1.0{\sim}1.8 \times 10^9$ t of CO_2, respectively [12]. In addition, considering the compatibility of sources and sinks in China, the emission reduction potential offered by CCUS technology can basically fulfill the requirements for realizing the carbon neutrality goal [13]. Meanwhile, the proportion of wind and solar power in total electricity production is expected to reach 85% by 2060 from 11.5% in 2021. At that time, wind and solar power will generate nearly 2.6×10^{13} kWh resulting in high demand for large-scale energy storage due to the fluctuation and instability. Comparatively, the underground energy storage technologies (e.g., underground hydrogen storage, methane storage, etc.) are more favorable due to their large capacity (more than 10^{12} kWh) and long-term (a few months) storage characteristics. Consequently, CCUS technologies combined with underground energy storage will experience unprecedented development opportunities, especially those that incorporate underground energy storage capacity, and will play a key role in carbon neutrality processes.

This paper briefly reviews and analyses the technologies of CU, CCU, and CCUS first. Moreover, based on CO_2 circular utilization, geological storage, and underground large-scale energy storage, the concept of carbon dioxide capture, circular utilization, and storage (CCCUS) is proposed. As a carbon circular utilization technology for realizing underground artificial natural gas production, hydrogen, and methane storage, biomethanation's technical mechanism, principles, and site selection criteria are described in detail, and the potential and development challenges are analyzed as well. Finally, development suggestions are made combined with China's actual situation and strategy in the hope of providing a reference for the development of China's CCUS industry and carbon neutrality targets.

2. CU/CCU

For several decades, CO_2 has been utilized directly in various activities with/without chemical or biological conversion. These include, but are not limited to, carbonated beverages, inerting agents, and extractants in chemical industries (e.g., for the decaffeination of coffee and drinking water abstraction). Additionally, CO_2 also can be used for refrigeration, fire suppression, and plastics production. It is also used as a supplement injected into metal castings, respiratory stimulant (added to medical O_2), aerosol can propellant, etc. Moreover, through chemical or biological conversion, fuels, biomass, or bioproducts such as aquaculture feed can be achieved. The above-mentioned applications are already relatively mature technologies with complete supply, production, and sales chains. However, the above often have a small scope. CO_2 utilization is not expected to deliver emissions reductions on the same scale as carbon capture and storage (CCS) but is still a part of an "all technologies" approach. Particularly, carbon-based energy sources and chemicals will play an important role in the future defossilized industrial society. Based on CO_2 capture (industrial or direct air capture) and power-to-X technologies, collected CO_2 and electrolytically generated hydrogen can be used to produce possible products and materials. Theoretically, these technologies are available where carbon carriers are required for material use, especially in transportation, basic chemicals, and industries. Among them, are organic base materials such as methanol and high-value chemicals ethylene, propylene, butane, and butadiene as well as the aromatics benzene, toluene, and xylene. In addition, synthetic methane can be produced both surface and subsurface, as detailed in Chapter 4. The future relevance of producing synthetic fuels from carbon dioxide and hydrogen, namely power-to-fuels, is primarily in aviation and maritime transport, and to a lesser extent heavy-duty transport.

Currently, nearly 2.3×10^8 t of CO_2 is used per year globally contains CO_2 enhanced oil recovery (CO_2-EOR) and is still increasing rapidly, with preliminary estimates of over 2.7×10^8 t by 2025 [14]. The primary customer is fertilizer production, which annually consumes around 1.3×10^8 t CO_2 for urea synthesis. Subsequently, the food and beverage industry each consumed 3% of the total, corresponding to 6.9×10^6 t. In addition, Metal fabrication and other uses accounted for 2% and 4%, corresponding to 4.6×10^6 t and 9.2×10^6 t, respectively [14]. Notably, China's CO_2 demand has grown rapidly in recent years, with its market share already increasing from 21% in 2018 to 28% in 2020, overtaking the US as the world's largest CO2-consuming market. According to estimation China's CO_2 consumption in the future will reach 6.2×10^8 t~8.7×10^8 t in 2060 [12].

3. Geological CCUS

Undoubtedly, the amount of sequestration that can be achieved by geological storage of CO_2 is enormous compared to fixed products, such as building materials, food, beverage, etc. Estimations have shown that China's geological storage potential of CO_2 is roughly 1.21×10^{12}~4.13×10^{12} t [12]. Obviously, CO_2 can be widely used in underground energy development and storage. Among them, using carbon dioxide to flood oil or gas is one of the methods of CO_2 utilization (Figure 1a,b), which has been practiced worldwide in the oil and gas industry [15]. Injection of CO_2 into hydrocarbon formations is able to provide significant underground storage for CO_2 while enhancing hydrocarbon recovery (CO_2-EOR, CO_2-EGR) which cuts down the expenses. Moreover, as emerging technologies with significant potential and application interest, CO_2 fracturing and CO_2 application in geothermal energy development have also attracted increasing attention from both academics and industries (Figure 1c,d). Generally, considering a portion of carbon dioxide is sequestrated permanently below the surface in geological utilization, CCUS technologies can accomplish negative carbon emissions.

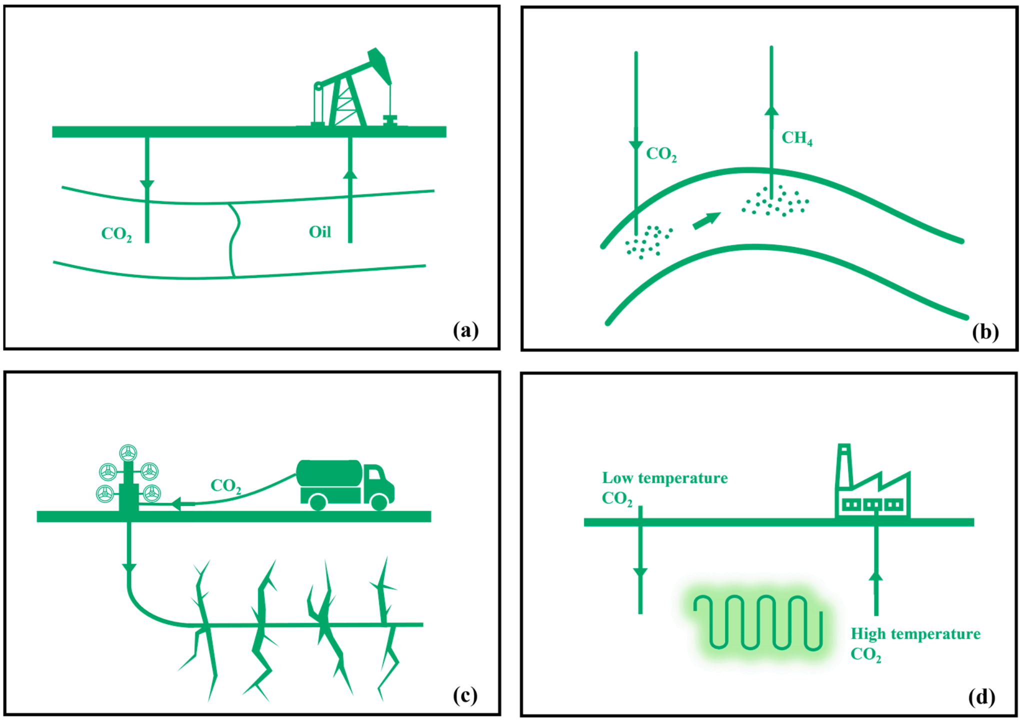


Figure 1. The diagrammatic sketch of geological CCUS technologies. (**a**) CO_2-enhanced oil recovery; (**b**) CO_2-enhanced gas recovery; (**c**) CO_2 fracturing; (**d**) CO_2-enhanced geothermal system (modified from [16–19]).

3.1. CO_2-EOR

Using CO_2 as an injectant to enhance oil recovery has gained increasing attention in the oil and gas industry. It is not only an environmentally friendly method of greenhouse gas treatment, but also an important technology for the development of conventional and unconventional oil reservoirs [20,21]. The primary displacement mechanisms of CO_2-EOR are: (a) reduced oil viscosity and density; (b) reduced oil swelling due to CO_2 solution; (c) vaporization and extraction of light hydrocarbon component; (d) dissolved gas drive; (e) reduced interfacial tensions; and (f) improved permeability due to acidification effect [22–24]. In terms of reservoir fluid properties and reservoir pressure and temperature conditions, the injection scheme of a typical CO_2-EOR process can be classified as miscible or immiscible based on CO_2 and oil miscibility. Basically, the main mechanism depends on the CO_2 solubility in oil in the rock matrix and fractures, which can reduce oil density and viscosity, increase oil mobility, and result in improved oil recovery [25]. Moreover, diffusion is a crucial influencing factor for CO2-enhanced oil recovery in tight or shale oil reservoirs with well-developed natural and artificial fractures generated by hydraulic fracturing at the initiation of development. Hence, the characterization of the flow behavior of CO_2 and oil in low permeability and pressure conditions, as well as the diffusion mechanism are encouraged to be emphasized in future studies. Since the 1960s, USA and China have been pioneers in CO_2-EOR [26]. Recently, it has been gaining more attention worldwide, including in Brazil, Norway, and Trinidad, which have recognized CO_2-EOR storage as a vital step toward reducing carbon emissions from industrial and energy sources via large-scale carbon capture projects [27]. Wei et al. [28] found that the technical potential for CO_2-EOR in China is approximately 1.0 Gt, with the attendant capability of storing 2.2 Gt of CO_2 in the process. Furthermore, according to a recent report by the Ministry of Environmental Protection of China, CO_2-EOR can sequester approximately 5.1 Gt of CO_2 in the subsurface [12]. China has focused heavily on developing CO_2-EOR projects to utilize and eventually store CO_2 to meet the carbon neutralization target.

However, when compared to reservoir characteristics and properties in North America, the geological conditions of oil reservoirs in China are relatively poor, particularly for tight reservoirs, which are characterized by ultra-low permeability, low brittleness of rock, and poor mobility. Furthermore, gas displacement requires a large-scale and stable supply of CO_2, so the match between carbon sources and carbon demand is also crucial. Usually, CO_2 is separated from nearby coal-fired power plants and piped to the fields. Thereby, this also places a demand on the corrosion resistance of equipment such as pipes and pumping equipment. It should be noted that economic issues are vital concerns in the practice process of CO_2-EOR. As current carbon capture and storage technologies for enhanced oil recovery are not cost-competitive with low oil prices, thus many planned carbon capture, utilization, and storage projects have been canceled over the past few years. Technically, CO_2-EOR also has some requirements, including reservoir characteristics (reservoir thickness, porosity, permeability, heterogeneity, geochemical and geomechanical parameters, etc.), well pattern and spacing, and CO_2 leakage control, etc. Generally, the main challenges of the CO_2-EOR process including:

(1) Control the cost of the overall industrial chain covering carbon capture and carbon transportation to improve economics.
(2) Reservoir screening and evaluation (seepage and storage conditions, mechanical and chemical properties).
(3) Stable carbon source and large-scale CO_2 transport technology.
(4) Underground carbon dioxide monitoring technologies and anti-corrosion, anti-blocking pipes, operating strategies, and service life extension.
(5) Infrastructure investments, such as pipelines and surface separation and recycling facilities for CO_2, should be improved.

3.2. CO_2-EGR

CO_2-EGR is another CCUS technology that improves gas recovery by injecting CO_2. The gas displacement effect and re-pressurization in depleting or depleted gas reservoirs are the primary mechanisms. On the one hand, CO_2 injection can lower the dew point pressure of reservoir fluids in wet gas reservoirs. The separated CO_2 from the produced gas, on the other hand, can be injected back into the reservoir to improve gas recovery [29]. After natural gas production and pressure reduction, gas reservoirs have sufficient pore space to store injected CO_2 molecules. Moreover, they enable the long-term sequestration of CO_2 within a reservoir sealed by impermeable cap rocks. Notably, depleted gas reservoirs showed a greater potential to store CO_2 compared to oil reservoirs due to the high primary recovery in gas reservoirs (>60%), which is nearly double that of oil recovery [30]. However, the process is complicated because of the gas adsorption on the surface of reservoir rocks, the mixture of CO_2 and natural gas, and CO_2 breakthrough production wells. As is described in many studies, an incremental recovery of up to 11% can be achieved by CO_2 injection [31]. Furthermore, several experimental and numerical simulation studies have been conducted to assess the feasibility of CO_2-EGR [32–35]. According to the observations, the CO_2 breakthrough is the most significant concern in EGR. Early CO_2 breakthroughs actually resulted in unfavorable gas production. As a result, geological formations, particularly microstructures, are thought to have a remark impact on CO_2 flow behavior and sweep the region. Furthermore, irreducible water saturation influences the mixing of CO_2 and CH_4. In addition, engineering parameters also play an important role in determining CO_2-EGR performance. The findings showed that injecting CO_2 with a horizontal well into the lower reservoir while extracting CH_4 from the upper reservoir would reduce CO_2 breakthrough into the production well. Likewise, CO_2 injection during the early decline phase of natural gas production is advantageous for achieving a high CH_4 recovery because it ensures supercritical displacement and reduces CO_2 and CH_4 mixing. To ensure the supercritical phase in the displacement process, it is suggested that CO_2 be injected at relatively high pressure. In terms of injection rate, numerous studies have shown that a high injection rate is beneficial for gas recovery. Moreover, it is proposed that the CO_2 injection rate be lower than the CH_4 production rate to prevent early CO_2 breakthrough. Furthermore, parameters such as diffusion coefficient, viscosity, and permeability are important in gas displacement as well.

Several CO_2-EGR projects have been implemented in North America, Europe, and Asia. China's gas reservoirs are primarily distributed in the Ordos Basin, Sichuan Basin, Bohai Bay Basin, and Tarim Basin, and CO2-enhanced natural gas extraction technology can sequester approximately 9 billion tons of CO_2 [12].

As it is still a developing technology, the main technological challenges of CO_2-EGR cover the following four parts:

(1) Key technologies (e.g., CO_2 breakthrough) and engineering parameters should be overcome and optimized to improve technical maturity.
(2) Reservoir storage capacity assessment and evaluation of site selection criteria.
(3) Similarly, the amount of CO_2 consumed by EGR poses specific demands on the carbon source, as well as sufficient and cost-effective CO_2 is the basis for the commercialization of CO_2-EGR.
(4) Monitoring technology of downhole CO_2 and anti-corrosion and anti-blocking technologies of equipment and pipelines.

3.3. CO_2 Fracturing

Carbon dioxide fracturing refers to a new type of fracturing technology that uses carbon dioxide as the fracturing fluid. According to the phase state of CO_2 after reaching the reservoir, carbon dioxide fracturing can be divided into liquid CO_2 fracturing (carbon dioxide dry fracturing) and supercritical CO_2 fracturing, and the latter generally has a wellhead heating device, as shown in Figure 1c. During the fracturing process, after carbon dioxide is filtered into the matrix, part of the carbon dioxide reacts with water to form H_2CO_3, which is further dissociated to form HCO_3^- and CO_3^{2-}, so carbon dioxide will be dissolved and stored in reservoir solution in the form of CO_2(aq), H_2CO_3, HCO_3^-, CO_3^{2-} [36,37]. On the other hand, the injection of CO_2 will form a large number of acidic zones, which will lead to the dissolution of feldspar, clay minerals, and carbonate minerals, resulting in a large number of metal cations such as Ca^{2+} and Fe^{2+}, which are combined with CO_3^{2-} and other anions to produce carbon-containing minerals such as calcite, dolomite, etc., and realize the mineral sequestration of carbon dioxide [38]. When CO_2 fracturing technology is applied to unconventional oil and gas reservoirs, carbon dioxide can compete with methane adsorbed on the rock surface, thereby realizing the adsorption and storage of CO_2 [39]. In addition, compared with hydraulic fracturing, carbon dioxide fracturing also has the advantages of reducing rock breakdown pressure, forming complex fracture networks, and reducing reservoir damage and water resources dependence [40,41]. Therefore, the use of carbon dioxide for fracturing can also realize the geological sequestration of CO_2 during the efficient development of oil and gas reservoirs.

As for potential, current field data show that one fracturing well consumes around 1000 t of liquid carbon dioxide [42]. Furthermore, according to the US Energy Agency (EIA), the number of horizontal wells fractured in North America in 2020 exceeded 15.3×10^4 [43]. With China's continued investment in unconventional oil and gas development, it is roughly estimated that the number of fractured wells in China will reach between 1×10^5 and 1.5×10^5. Consequently, the potential of CO_2 utilized in fracturing could be 1×10^8 t~1.5×10^8 t.

However, the low viscosity and high filtration of carbon dioxide fracturing fluid lead to narrow fracture width, poor sand carrying effect, and easy sand plugging [36], which limits the large-scale popularization and application in the field and causes several challenges:

(1) The fracturing mechanisms still need to be further investigated, including supercritical carbon dioxide fracturing initiation and fracture expansion mechanism.
(2) The improvement of sand-carrying ability and the development of a thickening agent, resistance-reducing agent, and new lightweight proppant.
(3) Precision phase control and strict requirements of equipment sealing and corrosion resistance.

(4) Cost control and economics for large-scale industrial applications.

3.4. CO_2-EGS

Earth is regarded as a tremendous thermal energy resource due to its hot core and the decay of radioactive minerals [44,45]. Furthermore, geothermal energy has a higher utilization rate than solar and wind energy (5.2 times and 3.5 times, respectively) [46]. Consequently, the exploitation of geothermal energy sources has attracted significant attention because of their exceptional properties of being stable, sustainable, environmentally friendly, and weather-independent. Despite sufficient geothermal energy resources, commercial geothermal energy extraction is constrained to shallow geothermal energy and hydrothermal resources that enable fluid circulation through natural high-permeable formations currently. To exploit geothermal energy in deep and low permeability formations, a number of studies and pilot projects are being carried out, namely the development of an enhanced geothermal system (EGS) based on hot dry rock (HDR) through hydraulic fracturing. Conventionally, water has been used as fracturing and heat extraction fluid due to its high density and specific heat levels in EGS. Generally, a large amount of water will be consumed for continuous operations during fracturing and heat extraction, with more than 20% water loss [47,48]. Employing supercritical CO_2 to replace water for EGS development has been proposed due to its significant advantages, such as reduced pumping power requirements, lower fluid density, less risk of scaling, and improved thermodynamic efficiency [49]. Evidence also suggests that supercritical CO_2 has more compressibility and expansivity in HDR, making it a mobile fluid of interest. According to estimates, heat flow rates utilizing CO_2 as the working fluid might be up to five times higher than those using formation brine [50].

China's geothermal resources are significant, accounting for 7.9% of the world's reserves, with an annual availability of 3.06×10^{18} kWh [51]. Specifically, the annual recoverable resources of shallow geothermal energy and hydrothermal resources are equivalent to 700 million tons and 1.865 billion tons of standard coal, respectively. In addition, the resources of HDR within 5500 m are approximately 106 trillion tons of standard coal, which is also the emphasis for future geothermal energy exploration and development [52]. Only considering the EGS development of HDR geothermal energy, referring to the Soultz project in France and the Hijiori project in Japan, the water consumption of a single EGS well is between 30,000 and 60,000 m^3. Therefore, it can be calculated that the potential for applying CO_2 in developing geothermal energy from hot dry rock in China will be 3.3×10^7 t~6.6×10^7 t (density of liquid CO_2: 1101 kg/m^3).

Despite the considerable advantages of CO_2-EGS and numerous numerical efforts, it is still at the conceptual stage around the world to the best of our knowledge, and the following challenges are still needed to be addressed:

(1) Reasonable and acceptable CO_2 capture cost is a necessary prerequisite for CO_2 application in geology.
(2) The potential threats of CO_2 leakage during storage in deep formations have been the major constraints in promoting CO_2 geological storage techniques.
(3) Complex phase transition mechanisms and migration processes of CO_2 in geothermal reservoirs.
(4) Generation of large-area hydraulic fracture network and heat exchange surface.

Nevertheless, combining geothermal heat extraction and CO_2 geological storage in deep EGS is a promising alternative to improve the economic feasibility of CCUS.

4. CCCUS

Aiming to promote carbon cycle economy, underground energy storage, and enhance the scale of CO_2 utilization, the authors propose a new concept of CO_2 capture, circular utilization, and storage, namely underground biomethanation (Figure 2). H_2 and CO_2 are mixed or injected sequentially into the depleted or depleting natural gas reservoirs in the CCCUS process. A very small percentage of injected carbon dioxide is sequestrated underground through mineralization or dissolution, and most of the injected gas is bio-converted to water and renewable methane under the catalysis of methanogens during the long-time well shut-in [53]. The reaction equation of methanation is as follows:

$$CO_2 + 4H_2 \rightarrow CH_4 + 2H_2O;\ \Delta G = -136\ \text{kJ/mol} \tag{1}$$

Natural gas composed of renewable methane and other impurities can be produced for industrial or domestic usage. CO_2 in the industrial exhaust gas or emitted into the atmosphere is captured and then used for the next stage of biomethanation, to realize the circular utilization and continuous underground storage of carbon dioxide. In the CCCUS process, H_2 can be produced by electrolysis of water from renewable energy (e.g., wind and solar), or by coal-based gasification, an industrial by-product, and so on. As a result, this technology can also realize the storage and partially circular utilization of CO_2. China has become the world's largest H_2 producer, and its annual output is about 33 million tons. Almost 80% and 20% of the output comes from H_2 produced by fossil energy and industrial by-product H_2, respectively [54].

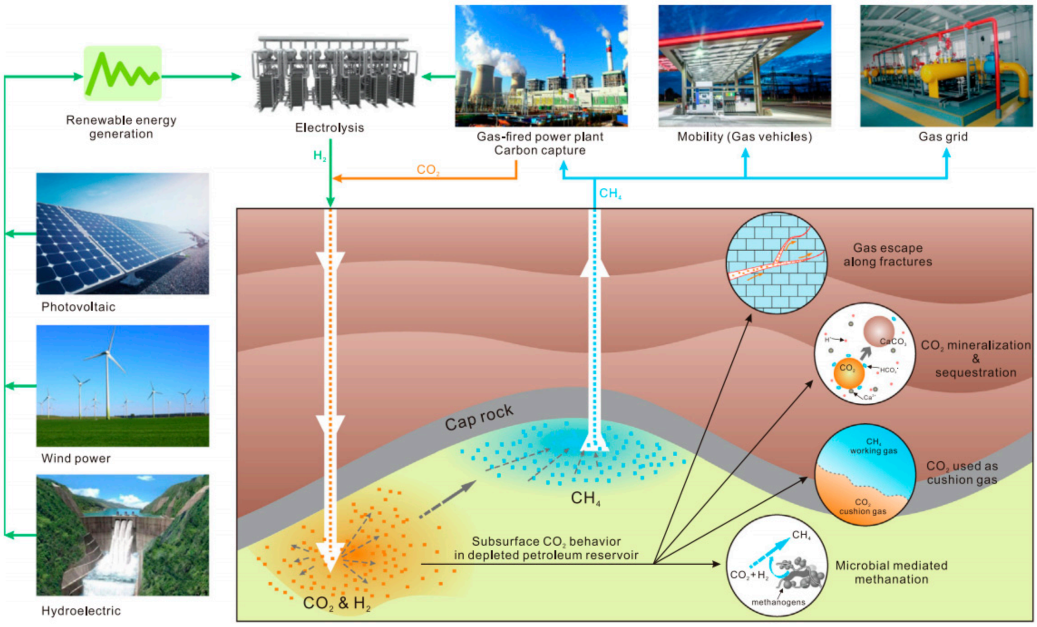


Figure 2. Simplified concept of biomethanation [55].

To realize the ideal biomethanation of injected H_2 and CO_2 underground, two conditions need to be met: (i) the injected gas will not leak underground; (ii) the methanogens have high biological activity. As for the first aspect, trapping (e.g., anticline and fault) and impermeable cap rock are necessary to prevent the migration and leakage of the injected gas. As a large amount of oil and gas was stored in depleted reservoirs before production, the sealing performance of depleted reservoirs is more reliable than that of aquifers. In terms of the methanogens' activity, it is affected by temperature, porosity, water saturation, pH, salinity, and so on (Figure 3): (1) Methanogen growth also requires pore space and water. Strobel et al. [56] suggested that the minimum reservoir porosity and permeability for biomethanation are 10% and 10mD, respectively, and pores should be connected for easier injection. Moreover, the minimum water saturation is 10%; (2) Higher and lower temperatures affect the activity of enzymes in methanogens, and even lead to the death of methanogens. Most methanogens can survive in the temperature range of 15 to 90 °C, and the favorable temperature range of 30 to 70 °C [57]; (3) The brine pH affects the methanogen's growth by directly affecting metabolism or indirectly through redox reactions. Most methanogens will die if the pH range is outside the range of 4 to 9.5, and the favorable pH range is 6.5 to 7.5; (4) The salinity affects the osmotic pressure of methanogen cells, and higher salinity causes methanogen cells to lose too much water and die. The upper limit of salt content for the growth of methanogen is 150 g/L. The environmental requirements for underground biomethanantion are summarized in Table 1.

According to the requirements of formation tightness and a suitable environment for the biomethanation process (Table 1) mentioned above, the depleted reservoirs or aquifers suitable for biomethanation can be screened out. If there are no natural methanogens in the formation water, the methanogens cultivated on the ground can be artificially injected into the underground bio-reactors. Moreover, when the temperature is so high that the activity of methanogens is low, pre-injection or circulating injection of cold fluid can be used to reduce the temperature.

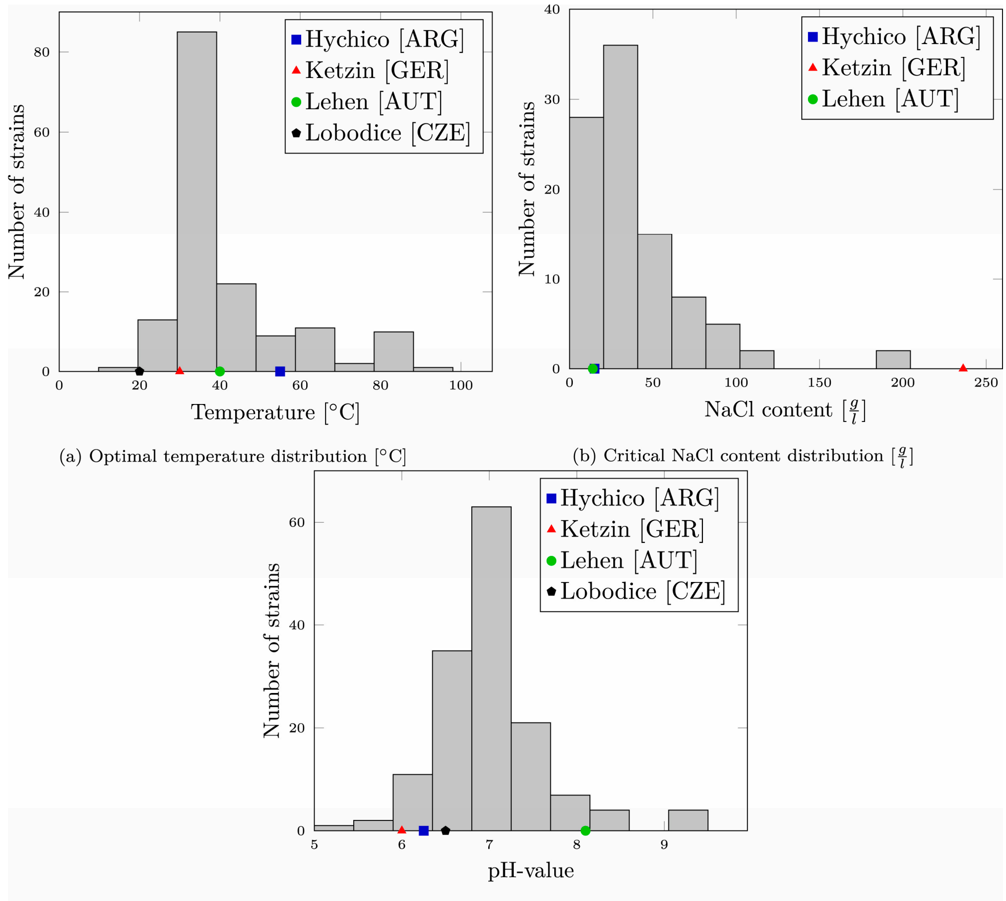
Table 1. Environmental requirements for underground biomethanation.

Items	Description
Underground structure	Trapping (e.g., anticline and fault) and impermeable cap rock
Porosity	Minimum: 10%
Permeability	Minimum: 10 mD
Water saturation	Minimum: 10%
Temperature	Survival range: 15–98 °C; favorable range: 30–70 °C
pH	Survival range: 4–9.5; favorable range: 6.5–7.5
Salinity	Maximum: 150 g/L

The CCCUS technology stores and utilizes among others green H_2, which means that this method enables large-scale underground storage of renewable hydrogen while consuming a large amount of green power. Moreover, this method effectively couples zero (negative) emission economic utilization of impure H_2, CO_2 recycling, and sequestration, underground natural gas synthesis and storage, and geothermal parallel development, thus boosting the development of a low-carbon circular economy. In terms of operation management, safety requirements, etc., it is also necessary to develop unified norms and standards. Underground biochemical mechanization technology is currently at the stage of mechanism research and small-scale field trials, even in Europe and USA. Before large-scale industrial applications, the following challenges are still needed to be addressed:

(1) Screening of specific strains and large-scale efficient and low-cost nurture.
(2) Dynamic tracking and monitoring system for biochemical catalytic reaction processes.
(3) Unknown biochemical processes and influencing factors in real reservoirs.
(4) Activation and control of microbial populations in the reservoir environment and inhibition of competing organisms.
(5) Energy conversion efficiency and site selection and evaluation criteria.
(6) Uncertainty of economic benefits, operating mechanisms, and business models. It is needed for quantitative financial model assessment on field tests (the Sichuan Province of China is suitable for such tests) to find the appropriate business mechanism.
(7) The full supply chain of hydrogen, including production (cost-effective, method, etc.), transportation (related materials, mode, etc.), and storage (corrosion, leakage, etc.) are also key issues of CCCUS.

Regarding H_2 being extremely flammable and easy to form an explosive gas mixture with air, safety management measures need to be implemented during transportation and injection, mainly from the following four aspects: (1) Hydrogenation facilities should be placed, installed, and running in designated dedicated locations. Lightning protection facilities, safety fences, anti-collision columns, and grounding devices should be set up in the operation area; (2) Implement uninterrupted monitoring and periodic maintenance system to improve equipment reliability; (3) Vehicles transporting hydrogen should comply with relevant regulations, use special motor vehicles, and the bottom of the vehicle should be equipped with static-conducting mopping tapes that meet the regulations; (4) Personnel engaged in the safety management of hydrogenation facilities should undergo professional training in pressure vessel safety management to understand the characteristics of pressurized hydrogenation equipment, the use characteristics of packaging containers, and emergency measures.



(a) Optimal temperature distribution [°C] (b) Critical NaCl content distribution [$\frac{g}{l}$] (c) Optimal pH-value distribution

Figure 3. Effects of (**a**) temperature, (**b**) salinity, and (**c**) pH on the activity of methanogens [56].

As a key carbon-negative technology, CCCUS will have an unprecedented development opportunity and significant potential. According to preliminary estimates, depleted gas reservoirs can sequester approximately 1.53×10^{10} tons of CO_2 in China, which shows that China has sufficient underground gas reservoir space for CCCUS. In addition, in terms of energy storage, the demand for reserved power in China would be 5×10^{11} kWh~1×10^{12} kWh in 2030, and 6×10^{12} kWh~7×10^{12} kWh in 2060 based on our studies. Among them, China's large-scale underground energy storage will reach 4.8×10^{12} kWh~5.6×10^{12} kWh (80% of the total power storage demand) in 2060. In the case of full capacity utilization to produce hydrogen and the industrial by-product hydrogen, underground biochemical synthesis of methane with CO_2, energy storage of 3.29×10^{12} kWh~4.42×10^{12} kWh, and consumption of 5.94×10^{8} t~7.98×10^{8} t of carbon dioxide can be achieved (energy efficiency: 68.6~79.2%).

5. Outlook

As a crucial artificial carbon sink and underground large-scale energy storage method in the future, CCCUS technology represented by underground biomethanation has a promising development prospect, particularly in China, where renewable energy and hydrogen are booming significantly. According to conservative estimations, underground biomethanation has significant potential, with a corresponding energy storage capacity of over 4×10^{12} kWh and a carbon dioxide uptake capacity close to 8×10^{8} t. Under the limited growth of natural carbon sinks, vigorously promoting CCUS development and proposing new CCCUS modes will play a key role, particularly in China, where the planned period from carbon peak to carbon neutrality is strictly restricted. Comparatively, underground biomethanation incorporates renewable energy consumption, reduces wind and solar abandonment, and simultaneously enables energy storage resulting in an efficient carbon circular economy, hence making the investigation highly valuable as a potential carbon neutrality technology and solution.

According to the analysis of the potentialities and challenges in each method (see Table 2), as well as the evaluation of its development trend, combined with the consideration of China's status quo, the following suggestions have been made: deepening field studies of depleted oil and gas reservoirs as natural biochemical reactors to synthesize methane is necessary. Moreover, the primary control parameters and evaluation indicators to prepare for field testing can be investigated. Combined with the author's investigation and research, it is recommended that the pilot work of CCCUS should be implemented in all localities appropriately. For instance, the Sichuan Basin, where gas reservoirs are widely distributed, also has a large number of depleted oil and gas reservoirs, the pilot study of depleted oil and gas reservoirs as a natural biochemical reactor to synthesize methane can be organized.

Table 2. Characteristics and potentialities of various technologies in China.

Technical Type		Main Characteristics	Technology Maturity	Potential
CU/CCU	Direct or conversion utilization	CO_2 fixation is simple and convenient	Mature	CO_2 consumption: 6.2×10^7 t~8.7×10^7 t
Geological CCUS	EOR	Large-scale, partially permanent sequestration of CO_2, more economical than pure CO_2 sequestration	Almost mature	CO_2 storage: 5.1×10^9 t
	EGR		Almost mature	CO_2 storage: 9.0×10^9 t
	Fracturing		Pilot stage	CO_2 consumption: 1×10^8 t~1.5×10^8 t
	Geothermal		Theoretical stage	CO_2 consumption: 3.3×10^7 t~6.6×10^7 t
CCCUS	Underground Biomethanation	Designed carbon sink, large scale, low cost of underground methanation, storage of renewable methane, carbon circular economy	Theoretical stage	CO_2 consumption: 5.94×10^8 t~7.98×10^8 t Energy storage: 3.29×10^{12} kWh~ 4.42×10^{12} kWh

Note: These summaries of characteristics and maturity is based on references [12,14,16–19,55,58,59].

Nevertheless, there are also various development opportunities for geological CCUS considering the significant CO_2 uptake capacity, particularly, CO_2 fracturing and CO_2-EGS, although they are not as widespread and commercialized as CO_2-EOR and CO_2-EGR. Establishing a complete set of site selection and evaluation criteria, and promoting demonstration projects of geological CCUS are necessary. Likewise in CO_2 fracturing and CO_2-EGS development, relevant research needs to be implemented urgently. Furthermore, investigations on key technologies related to carbon dioxide applications, such as CO_2 transportation, phase control, and related equipment can be performed.

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