



# Article A Review of CO<sub>2</sub> Marine Geological Sequestration

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**Abstract:** Carbon dioxide (CO<sub>2</sub>) sequestration plays a crucial role in reducing the levels of atmospheric CO<sub>2</sub> and mitigating the harmful effects of global warming. Among the various CO<sub>2</sub> sequestration technologies, CO<sub>2</sub> marine geological sequestration emerges as a safer and more efficient alternative compared with traditional terrestrial geological sequestration. This is highly attributed to its expansive potential, safe distance from aquifers, and stable temperature and pressure conditions. This paper reviews and evaluates the main CO<sub>2</sub> marine geological sequestration technologies, including CO<sub>2</sub> sequestrations in shallow marine sediments, CO<sub>2</sub>, sub-seabed aquifers, and CO<sub>2</sub>-CH<sub>4</sub> replacement. The goal of this paper is to shed light on the mechanism, potential, and challenges of each technology. Given the importance of safety in CO<sub>2</sub> sequestration, this review also explores the potential adverse effects of CO<sub>2</sub> leakage from reservoirs, particularly its impact on marine environments. Finally, we discuss potential development trends in CO<sub>2</sub> marine geological technology.

Keywords: CO<sub>2</sub> marine geological sequestration; sequestration mechanism; Marine environment

# 1. Introduction

With the continuous increase of carbon emissions, the global environment is gradually warming, and extreme weather frequently occurs, causing large-scale disasters such as drought, flood, and haze [1].  $CO_2$  is the main greenhouse gas, and it is important to control its atmospheric content so as to prohibit climate warming. CCS (Carbon Capture and Sequestration) technology is used to capture  $CO_2$  generated in industrial processes and store it in reservoirs for a long time to reduce  $CO_2$  emissions into the atmosphere [2–4].  $CO_2$  sequestration, as a core part of CCS technology, determines the path of  $CO_2$  from being released into the atmosphere, thereby efficiently mitigating the greenhouse effect [5]. Figure 1 shows China's annual  $CO_2$  emission is as high as 27% of the global total amount. In order to achieve the carbon emissions peak in 2030 and carbon neutrality in 2060, it is crucial to further develop  $CO_2$  sequestration technology [6].

Traditional territorial geological sequestration involves a variety of methods, including  $CO_2$ -enhanced oil and natural gas fields [7–9],  $CO_2$  sequestration in coal beds [10–12], saline aquifers [13], abandoned oil or gas fields [14], frozen soil [15], and enhanced geothermal systems [16]. Each method has its own limitations. For instance,  $CO_2$ -enhanced oil and natural gas fields, as well as  $CO_2$  sequestration in abandoned oil and gas fields, have limited sequestration capacity due to the finite resource reserves of the exploitation area. The availability of these methods is low, and their application is restricted to specific exploitation areas, resulting in a constrained sequestration constituency [17]. In the case of saline aquifers, frozen soil, and enhanced geothermal systems, challenges arise from the need for deep underground well drilling, high costs, and the complex geological conditions in the deep strata. Consequently, the evaluation of geological characteristics and sequestration capacity remains insufficient [18]. Furthermore, supercritical  $CO_2$  exhibits



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**Copyright:** © 2023 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/). high buoyancy and tends to accumulate at the interface between saline aquifers and cap layers. Over time, this accumulation may induce seismic activity, which adversely affects sequestration stability [19].



**Figure 1.** CO<sub>2</sub> emissions of major global carbon emitters, accumulated amout per year (left) and the amout per capita per year (right) [20].

Considering these challenges, Marchetti [21] first proposed the innovative concept of  $CO_2$  marine sequestration in 1997. He suggested that  $CO_2$  should be directly injected into deep and dense seawater under low temperature and high-pressure conditions, estimating that the ocean could seal  $CO_2$  for thousands of years. This idea attracted widespread attention among scholars worldwide. However, direct dismission of  $CO_2$  into the seawater would seriously impact the ecological environment of the ocean. Therefore, three creative marine geological sequestration methods are proposed to sequestrate  $CO_2$  in the ocean area, including: sequestrations in shallow sediment, sub-seabed aquifers, and  $CO_2$ -CH<sub>4</sub> replacement. Zhao and Ikamura [3] published an article on carbon capture utilization and storage (CCUS) technologies, which are regarded as an economically feasible way to minimize greenhouse gas emissions. They also discussed the various aspects of CCUS. Each of these methods offers unique advantages and overcomes some of the limitations associated with traditional territorial geological sequestration technologies.

This paper investigates the storage mechanisms, capabilities, and main status related to  $CO_2$  marine geological storage technologies. Existing problems in these technologies are summarized and their technical complexities are clarified. This paper also puts forward the related solutions to solve these problems in the future. The overall goal of this paper is to provide a summary of the pros and cons of  $CO_2$  marine geological storage technologies, which will guide the development of the technologies, ensuring that they are both effective and environmentally friendly.

#### 2. Overview of CO<sub>2</sub> Marine Geological Sequestration

 $CO_2$  marine geological sequestration is a sophisticated technology that addresses the issue of anthropogenic climate change by capturing  $CO_2$ , a byproduct of industrial processes, and subsequently storing it in marine geological environments [17]. The advancement of this technology has been continuing, positioning it as a potential method for achieving carbon neutrality goals, first proposed by Marchetti [21]. Oceans, covering over 70% of the Earth's surface, provide a substantial theoretical sequestration capacity ranging from 2 to 13 trillion tons, making them a highly advantageous environment for large-scale  $CO_2$  storage.

Direct sequestration of  $CO_2$  into specific marine sedimentary strata has been demonstrated to effectively counteract the buoyancy of the gas, minimizing the risk of escape [22]. Researchers such as Fujioka et al. [23] thought that the deep seafloor offers an ideal geological space for  $CO_2$  sequestration in both liquid and hydrate states. Haszeldine et al. [24] proposed that, in comparison to alternative methods, seabed sequestration has advantages in terms of safety, reliability, and ease of monitoring, further emphasizing the potential of this technology.

Despite the numerous advantages associated with  $CO_2$  marine geological sequestration, it is crucial to acknowledge its limitations. One primary concern is that the method is not a permanent solution, as leakage can still occur, potentially leading to detrimental effects on marine organisms and the environment. This challenge highlights the primary importance of robust  $CO_2$  monitoring technology in reducing risks. Additionally, the feasibility of implementing  $CO_2$  marine geological sequestration technology varies across regions, as some areas present more favorable conditions for its application than others [25].

China has recognized the potential of marine geological sequestration technology in addressing its carbon emissions and has actively pursued studies on its feasibility within the country [25]. Researchers such as Chun et al. [26] have identified offshore basins, including the Pearl River Mouth and Beibu Gulf, as promising locations for  $CO_2$ sequestration. In August 2021, China took a significant step forward by launching an offshore  $CO_2$  demonstration project, which marked a major milestone in the advancement of the country's offshore  $CO_2$  marine geological sequestration technology [27]. This effort demonstrates the commitment of China to leveraging the technology as a vital support in achieving its ambitious carbon neutralization goals by 2060.

### 3. Advantages and Disadvantages of CO<sub>2</sub> Marine Geological Sequestration Methods

Compared with  $CO_2$  direct seawater sequestration,  $CO_2$  marine geological sequestration is a more feasible and economical method. Moreover, it maintains geomechanical stability and reduces the risk of seabed landslides and geological disasters, thereby protecting marine ecosystems and their biodiversity.  $CO_2$  marine geological sequestration can be roughly classified into three principal categories, including:  $CO_2$  sequestrations in shallow marine sediments, a sub-seabed aquifer, and  $CO_2$ -CH<sub>4</sub> replacement. Figure 2 shows the physical model of  $CO_2$  marine geological sequestration. In the shallow part of the  $CO_2$  reservoir, hydrates are formed to prevent the leakage of  $CO_2$ . In the middle part of the reservoir,  $CO_2$  can be trapped by gravity and flow downward to the deeper formations. In a deep reservoir,  $CO_2$  may exist in a super-critical state and form a plume flow in the pore network of the reservoir. It requires a proper permeability of the reservoir formations to store and migrate the flow.



**Figure 2.** Physical model of  $CO_2$  marine geological sequestration: in the shallow sediments, hydrates are formed to prevent the leakage of  $CO_2$ , which can be used as a caprock for the  $CO_2$  reservoir. Beneath the hydrate caprock,  $CO_2$  can be trapped by gravity and flow downward to the deeper formations. In a deep reservoir,  $CO_2$  exists in a super-critical state and forms a plume flow in the reservoir.

#### 3.1. Sequestration as Hydrate in Shallow Sediment

 $CO_2$  sequestration in shallow marine sediments represents a distinctive CCS strategy, which notably diverges from conventional geological sequestration. This innovative approach entails the extraction of  $CO_2$  from emission sources, followed by its injection beneath the mud line of seabed sediment within a depth of approximately 300 m from the seabed surface. When the stored seawater surpasses a depth of 2800 m, liquid  $CO_2$  exhibits greater density than seawater, thereby being stored within sediment pores [28]. Meanwhile, under specific temperature and pressure conditions,  $CO_2$  and water molecules interact to form hydrates within the sediment pores, which is called hydrate-based  $CO_2$  sequestration (HCS), and Equation 1 shows the equation describing the chemical reaction of hydrate formation:

$$n_h H_2 O + C O_2 = C O_2 \cdot n_h H_2 O \tag{1}$$

In the equation,  $n_h$  is the hydration number, which is ideally 5.7. Under standard conditions, 1 m<sup>3</sup> of hydrate can store 160–180 m<sup>3</sup> of CO<sub>2</sub> [29]. These hydrates play a crucial role in effectively capturing CO<sub>2</sub>, minimizing sedimentary porosity, and inhibiting CO<sub>2</sub> escape, ultimately generating a relatively stable cap layer [22].

 $CO_2$  sequestration in shallow marine sediments offers several distinct advantages over traditional geological sequestration. In geological sequestration,  $CO_2$  exhibits buoyancy, which causes it to accumulate at the interface between saltwater and cap layers, potentially inducing seismic activity and undermining the stability of the sequestration process [19]. Additionally, geological sequestration requires extensive drilling, which increases the risk of  $CO_2$  leakage following the injection process [30]. In contrast, sequestration in seabed sediments leverages the abundant pore space and unconsolidated skeleton structure of sediments, negating the need for complex drilling technology and reducing both costs and the potential for  $CO_2$  leakage [31]. Furthermore, the high permeability of  $CO_2$  facilitates its dispersion to remote areas from the injection site, resulting in enhanced sequestration efficiency [32]. By combining the negative buoyancy zone (NBZ) and hydrate forming zone (HFZ) within the sediment, an effective sequestration trap can be established, ensuring the safety, commercial viability, and scalability of this novel CCS approach [33,34]. To predict the leakage, monitoring technology is applied for safe sequestration, and seabed node seismic monitoring is used to detect  $CO_2$  migration [35,36].

Despite its huge potential,  $CO_2$  sequestration in shallow marine sediments faces several challenges, including the relatively rapid formation of  $CO_2$  hydrate in the sediment layer. This process reduces the porosity and permeability of the sediment, inhibiting the injection and transport of  $CO_2$ , and subsequently reducing sequestration efficiency. To address this limitation, researchers have proposed the addition of nitrogen  $(N_2)$  to the CO<sub>2</sub> stream, which can decelerate hydrate formation and minimize blockage. Hassanpouryouzband et al. [37] proved that injecting a certain proportion of  $CO_2-N_2$  into the sedimentary layer can promote the formation of  $CO_2$  hydrate. When the temperature of the porous medium rises,  $N_2$  hydrate will decompose ahead of the sequestration of CO<sub>2</sub> hydrate resulting in a longer and more stable sequestration. Yu et al. [38] carried out a numerical simulation of the injection of single CO<sub>2</sub> and CO<sub>2</sub>-N<sub>2</sub> mixture into a single wellbore installed in the seabed sediment. Their results showed that the injection of CO<sub>2</sub>-N<sub>2</sub> mixture improved the  $CO_2$  sequestration efficiency while maintaining a relatively small deformation. This study emphasized the importance of injection capacity and the hydrate formation rate for the presence of  $CO_2$  as a solid hydrate in seafloor sediments. Therefore, the addition of  $N_2$  can accelerate the efficiency of  $CO_2$  sequestration in shallow sediments, extending the period of the sequestration and keeping the mechanical properties of the seabed relatively stable.

The stability and security of  $CO_2$  reservoirs in shallow sediments remain a concern. The process of  $CO_2$  injection and migration can alter the skeleton of sediment structure and shear strength, leading to a dynamic disturbance of the  $CO_2$  reservoir [39]. Changes in pore structure can negatively impact  $CO_2$  transport, modifying the porosity and permeability of the reservoir, and generating uncertainty in the  $CO_2$  sealing volume and sequestration efficiency [40]. In addition, over time, the  $CO_2$  hydrate may partially dissolve or decompose, triggering sediment creep, reducing strength and stiffness, ultimately affecting the integrity and stability of the cap [41]. It is, therefore, necessary to develop a geotechnical theory and a coupled multiphysical modeling framework suitable for  $CO_2$  marine geological sequestration to ensure the successful execution of efficient and safe CCS projects in an increasingly ecologically conscious global landscape.

#### 3.2. Sequestration in Sub-Seabed Aquifers

The geological structure of the seafloor, which extends from the terrestrial landscape, offers an opportunity to harness the potential of sub-seabed aquifers for  $CO_2$  sequestration. This method involves injecting captured  $CO_2$  into rock strata containing groundwater with a closed structure beneath the seabed using a wellbore [22]. The sequestration mechanism within these aquifers relies on several key factors.  $CO_2$  is introduced at high pressure, occupying space within the aquifer. Due to density differences, the lighter gas floats on the cap rock, while some  $CO_2$  is trapped in a dissolved state within a brine solution. Another portion of the  $CO_2$  mineralizes with water and hypertonic rocks, eventually forming carbonate trapped in the rock layers [42]. However, this mineralization process takes thousands of years, making the first two mechanisms the primary methods of sequestration [43].

Sub-seabed aquifers are considered the most promising option among marine geological sequestration technologies due to their potential and feasibility. One of the main reasons is the presence of sediment layers between the aquifers and the seabed surface, which are conducive to the formation of  $CO_2$  hydrates [44]. These hydrates provide a secondary barrier to  $CO_2$  leakage in the event of cap rock cracks or faults. Furthermore, when the density of  $CO_2$  reaches 90% of seawater under specific temperature and pressure conditions, the gas becomes immobile due to gravity and capillary action, further ensuring the stability of sequestration in sub-seabed aquifer [28].

However, not all sub-seabed aquifers are suitable for  $CO_2$  sequestration. To be considered suitable, an aquifer must have high porosity and permeability, as well as a stable hydrate or low permeability clay layer and a soft mud cap layer to prevent leakage [45]. Additionally, the reservoir should contain high permeability sandstone that can directly store  $CO_2$  and react with it to rapidly form stable carbonate. Among various sub-seabed aquifers, basalt is an excellent candidate for sequestration [46]. First, basalt is highly porous and permeable because it erupts from volcanic ridges in all of the world's oceans, forming pillow lava and lava flows on the seafloor, which are buried over time, creating highly permeable aquifers within the oceanic crust. Secondly, basalt aquifer for geological CO<sub>2</sub> sequestration arises from providing multiple physical/chemical trapping mechanisms magnesium, which makes deep-sea basalt acts as a natural, in situ weathering reactor. Moreover, basalt aquifer has a high iron content, reaction rate, and large reservoir capacities, which makes it an excellent reservoir for  $CO_2$  sequestration [45,47,48]. Its dense cap layer on the ocean crust further enhances its suitability [49]. Luhmann et al. [47] found that the flow rate had a significant effect on the permeability and the outlet fluid chemistry of the basalt cores. At a higher flow rate, permeability increased and Fe, Mg, and Si concentrations were relatively stable. At a lower flow rate, permeability decreased and Fe, Mg, and Si concentrations showed complex trends. The authors also observed secondary mineralization of Al- and Si-rich phases and an Fe2O3-rich phase on the post-experimental cores. They suggested that siderite formation was thermodynamically favorable at low pH, and that alkali metals were highly mobile during fluid-basalt interaction. Goldberg et al. [45] studied the fluid-basalt interaction in CO<sub>2</sub>-rich systems and its implications for carbon sequestration and reservoir properties in basaltic formations. They highlighted the importance of flow rate, pH, and CO<sub>2</sub> concentration on the alteration processes and the reservoir properties. They also suggested that the composition of secondary carbonates could be used to infer the environmental conditions during their formation. Goldberg et. al. [48] discussed the potential of deep-sea basalt sites for carbon sequestration and proposed a global site assessment strategy to identify the most secure oceanic basalt sites that provided all trapping

mechanisms. They suggested that deep-sea basalt sites have several advantages over other carbon sequestration options, such as terrestrial storage and oceanic injection. They also highlighted the importance of understanding the geological, hydrological, and biological factors that affect the carbon sequestration capacity of deep-sea basalt sites.

Despite the potential benefits of sequestration in sub-seabed aquifers, many factors such as temperature distribution, geothermal gradient, sediment thickness, and reservoir porosity and permeability can limit its effectiveness. One successful example of the sequestration in sub-seabed aquifer is Norway's Sleipner field CCS project in the North Sea, which has sequestrated over 8 million tons of  $CO_2$  into the subsea brackish water layer since its inception in August 1996 [44]. This sequestration project used an alkamine solvent to absorb  $CO_2$  from natural gas and stored it in the Utsira formation, which consists of a 200–300 m thick sandstone reservoir with high porosity and permeability and a relatively thin shale interval (1–2 m thick) under the seabed at a depth of 1000 m, and it is covered by a 250 m thick shale cap, which can effectively avoid the leakage of  $CO_2$ from the storage layer. The Sleipner project as a whole has two distinctive features: a very regular and stable injection history, and continuous geophysical monitoring. These factors have made Norway's Sleipner field one of the most successful CO<sub>2</sub> marine geological sequestration projects in the world [50,51]. To maximize the potential of this promising  $CO_2$  sequestration method, it is essential to address these limiting factors and develop a comprehensive understanding of the geological and environmental conditions, such as geothermal gradient, reservoir porosity, and permeability, that contribute to successful sequestration. Temperature distribution played a crucial role in the Sleipner field CCS project. The injection of  $CO_2$  into the Utsira formation required an understanding of the temperature distribution within the reservoir. The temperature distribution affected the behavior of  $CO_2$  and its interaction with the surrounding rock formations [52]. This study compared the geomechanical deformation induced by CO<sub>2</sub> storage at Sleipner, Weyburn, and In Salah, highlighting the importance of temperature distribution in the success of the CCS project [52]. Sediment thickness was another important factor in the success of the Sleipner field CCS project. The thickness of the sediment layer affected the storage capacity and containment of  $CO_2$  within the reservoir. A thicker sediment layer provided a larger storage volume for CO<sub>2</sub> [53]. The study by Pegler et al. [54] estimated the parameters for the ongoing CCS operation at Sleipner, including sediment thickness, to understand the fluid migration between confined aquifers. Reservoir porosity and permeability were critical factors in the Sleipner field CCS project. Porosity referred to the volume of empty spaces within the reservoir rock, while permeability referred to the ability of fluids to flow through the rock. High porosity and permeability were desirable for efficient  $CO_2$  storage and injection [55]. The study by Shahkarami et al. [56] modeled the pressure and saturation distribution in a  $CO_2$  storage project, highlighting the importance of reservoir porosity and permeability.

Moreover, continued research, development, and collaboration among stakeholders are crucial for optimizing this technology and ensuring its sustainable integration into global CCS strategies.

#### 3.3. CO<sub>2</sub>-CH<sub>4</sub> Replacement Sequestration

Methane hydrate reserves on Earth are extensive, with a vast amount present in the world's oceans, sufficient to supply human consumption for approximately 1000 years. Traditionally, the extraction of natural gas hydrate (NGH) has been dependent on the method of direct exploitation. However, this approach has been associated with serious marine geological disasters, such as seabed settlement, landslides, borehole breakage, and gas leakage, which pose significant risks to the surrounding environment [57–59]. In 1996, Ohgaki et al. [60] introduced a concept of replacing CH<sub>4</sub> in hydrate-bearing sediments (HBS) with CO<sub>2</sub>. Subsequently, Wilder [61] and Kim [62] provided evidence supporting the theoretical feasibility of this innovative process by conducting an in-depth analysis and comparison of the phase equilibrium curves of methane and CO<sub>2</sub> hydrates, as shown in

Figure 3. The figure shows that the yellow zone is suitable for hydrate formation under 1200 m depth in the ocean, and the  $CO_2$ -CH<sub>4</sub> replacement sequestration occurs here. The replacement reaction is also influenced by the geothermal gradient. The replacement rate is very low when the replacement pressure is higher than the equilibrium pressure of the CH<sub>4</sub> hydrate. It is difficult to change the temperature of sediment but easy to reduce its pressure using a depressurizing well. Their findings revealed that the pressure required for CO<sub>2</sub> hydrate formation is lower than that for NGH at the same temperature. Furthermore,  $CO_2$  demonstrates a stronger affinity for water than  $CH_4$ . As a result, the replacement process involving CH<sub>4</sub> and CO<sub>2</sub> has been identified as spontaneous and exothermic in nature, which promotes the decomposition of NGH and accelerates the overall replacement process in turn [63]. Espinoza et al. [64] utilized seismic wave monitoring technology to study the process of replacing  $CO_2$  with NGH in HBS. Their research indicated that the replacement process of CO<sub>2</sub> and CH<sub>4</sub> hydrate did not induce any changes in sedimentary strength. Moreover,  $CO_2$  injection was found to maintain the stability of the formation structure. Despite these encouraging findings, both the rate of the  $CO_2$  replacement reaction and the rate of gas hydrate sequestration are relatively low, with a maximum theoretical value of only 75%.



**Figure 3.** Phase equilibrium curve of methane and  $CO_2$  [65]: The replacement reaction is influenced by the geothermal gradient. The blue and yellow parts enveloped by the curves are the hydrate stability zone, where hydrate can form in the sediments. However, due to the low concentration of gases in seawater, most of the hydrates are formed in the sediments, shown in the yellow part.

There are several factors that limit the practical application of the replacement and sequestration processes. Firstly, the replacement method is not universally applicable to all seabed reservoirs, as  $CO_2$  replacement of  $CH_4$  necessitates specific temperature and pressure conditions. If these conditions are not met, the replacement process cannot be effectively carried out. Additionally, during the extraction of other forms of combustible ice, the stability of  $CO_2$  hydrate must be taken into account. Although  $CO_2$  hydrate exhibits greater stability than combustible ice, it may decompose beyond particular temperature and pressure ranges [66]. Nevertheless, the replacement method possesses significant potential for delivering substantial environmental benefits, such as  $CO_2$  sequestration, as well as economic advantages stemming from methane gas extraction. Importantly, the process does not interfere with the hydrate formation structure, ensuring seabed geomechanical stability throughout the procedure [65].

Future research directions for CO<sub>2</sub> replacement sequestration may concentrate on investigating the underlying replacement mechanism and determining strategies for enhancing replacement efficiency. Yahaya et al. [67] discovered that replacement efficiency is positively correlated with both temperature and pressure, with the effect of tempera-

ture being more pronounced than that of pressure under specific experimental conditions. Therefore, it is recommended to take appropriate measures to increase the replacement time and temperature. Mohammadi et al. [68] and Wang et al. [69] suggested the incorporation of catalysts to improve the replacement rate and the utilization of  $CO_2$  emulsions in the  $CO_2$ -CH<sub>4</sub> replacement process to improve replacement efficiency. In addition, studies conducted by Pivnyak et al. [70] and Shin et al. [71] have demonstrated that the inclusion of an appropriate concentration of N<sub>2</sub> during the replacement process can accelerate the replacement rate. Through experimentation, Niu et al. [72] established that injecting a  $CO_2$ -N<sub>2</sub> mixture in the sequestration zone is a viable method for enhancing the efficiency of  $CO_2$  sequestration. However, it is crucial to exercise control over the components of the injected mixture. Geng et al. [73] identified that electrostatic interaction plays a pivotal role in the replacement process and that optimizing the electrostatic interaction with H2O is an effective way to improve the replacement efficiency.

At present, the application of NGH replacement for  $CO_2$  sequestration remains in the experimental stage, and the relevant technologies and sequestration methods are predominantly in the realm of theoretical exploration. Table 1 shows the methods to improve the efficiency of replacement. There is an urgent need for continued investigation and development in this area of research. Future research should focus on a more comprehensive understanding of the alternative mechanisms and structures of various seafloor sediments. This approach will help establish a sequestration mechanism suitable for different sediment types, thus laying a solid foundation for potential large-scale commercial applications in the future.

Measures	Reference	Conclusion
temperature and pressure	Yahaya et al. [67]	The influence on the temperature is more significant than the replacement on pressure
catalysts	Mohammadi et al. [68] and Wang et al. [69]	Catalysts or using CO <sub>2</sub> emulsion can improve the replacement rate.
adding N <sub>2</sub>	Pivnyak et al. [70] and Shin et al. [71] and Niu et al. [72]	Appropriate concentration of N <sub>2</sub> could accelerate the replacement rate.
electrostatic interaction	Geng et al. [73]	Improving the electrostatic interaction with H2O is an effective way to improve the

Table 1. The measures to improve the effectiveness of replacement.

#### 4. Effects of CO<sub>2</sub> Marine Geological Sequestration on the Marine Environment

Although  $CO_2$  marine geological sequestration offers promising potential, gas leakage during the process of  $CO_2$  sequestration is inevitable, leading to environmental concerns such as ocean acidification, geological hazards, and enhanced greenhouse effects.

replacement efficiency.

In marine geological sequestration, converting  $CO_2$  into seabed sediment can create issues with unstable pore pressure, increasing hydrate formation temperature, and  $CO_2$ leakage. These could lead to acidification of the sediment environment, causing chemical alterations that change the solubility or permeability of certain elements, releasing harmful metal ions that can damage marine ecosystems and organisms [74–76]. Additionally, during the process of substituting  $CO_2$  for NGH, CH<sub>4</sub> leakage from pipeline transportation poses an even greater risk than  $CO_2$ , as the greenhouse effect of methane is significantly more severe. It is, therefore, essential to reduce this as much as possible.

Marine geological sequestration is a potential future solution for mitigating the accumulation of anthropogenic  $CO_2$  in the atmosphere. However, during the implementation of marine geological sequestration, it is essential to detect and monitor the impact of stored  $CO_2$  on the marine environment. Thus, comprehensive marine geological sequestration technology for  $CO_2$  leakage detection and monitoring must be developed and established before commercial seabed sequestration can be implemented [77].

## 5. Conclusions and Future Perspectives

In summary, the advantages and constrains of the three marine geological sequestration technologies are shown in Table 2.

Table 2. Comparison of advantages and constrains of each sequestration technology.

Marine Geological Sequestration Technology	Advantages	Constrains	Gaps and Technical Barriers
Sequestration in shallow sediment	With abundant pore space and unconsolidated skeleton structure, the sequestration process does not need drilling	Permeability is reduced, causing the diffusion difficult to the far field	Complexity of early exploration work and the massive pressure of pipeline
Sequestration in sub-seabed aquifers	High sequestration security and reliability	Sequestration cost is high, pipeline pressure is too high	Complexity of early exploration work and the massive pressure of pipeline
CO <sub>2</sub> -CH <sub>4</sub> replacement	It can not only store $CO_2$ , but also extract $CH_4$ without disturbing formation stability.	Low replacement efficiency	Unclear replacement mechanisms and low replacement efficiency

Compared with terrestrial sequestration, marine geological sequestration has the characteristics of being far away from aquifers, stable high-temperature pressure, sealing, and perfect security, so it has greater potential. However, in terms of strengthening offshore oil,  $CO_2$  marine geological sequestration technology still has many problems to solve:

- Implementing CO<sub>2</sub> sequestration in shallow marine sediments is a challenge due to the complexity of early exploration work and the enormous pressure of the pipeline. At present, there are still shortcomings, such as unclear CH<sub>4</sub>- CO<sub>2</sub> replacement mechanism and low replacement efficiency.
- 2. A significant number of existing studies have primarily focused on theoretical aspects, with a noticeable lack of concrete case studies and accurate data feedback. This limitation has emerged as a significant obstacle to advancing the research. Furthermore, even some well-versed individuals have a limited understanding of CO<sub>2</sub> sequestration. National policies, laws, and regulations regarding CO<sub>2</sub> marine geological sequestration are notably absent, stifling motivation for further study among both enterprises and individuals. As a result, the development of CO<sub>2</sub> marine geological sequestration technology still requires substantial advancement.
- 3. From an engineering perspective, the aggregate cost of CO<sub>2</sub> capture, transportation, and sequestration in CO<sub>2</sub> marine geological sequestration is markedly higher than that of geological sequestration. In the specific case of the transportation phase, marine pipelines demand significantly higher pressure and lower temperature conditions. This requires the use of more flexible pipe materials and, therefore, costs more. Challenges such as geological exploration in the marine environment, unique aspects of marine construction, trenching and backfilling in deep ocean high-pressure environments, and pipeline installation all lead to considerable cost implications. In addition, preliminary tasks such as deep drilling and seismic exploration, which are essential for offshore operations, are much more complex and costly than those performed on land.

While CO<sub>2</sub> marine geological sequestration still confronts numerous challenges, this technology undeniably represents the future direction of CO<sub>2</sub> sequestration. Bearing the mission of achieving carbon neutrality and peak carbon emissions in the world, CO<sub>2</sub> marine geological sequestration undoubtedly has a considerable journey ahead. To reach these goals, robust support and encouragement from the state and government are essential. The improvement of related laws and regulations can serve to motivate enterprises and individuals to study CO<sub>2</sub> marine geological sequestration, boost public awareness, and establish new experimental sites that provide tangible data feedback. In addition, research should prioritize strengthening CO<sub>2</sub> leakage monitoring mechanisms and continuously

improve the accuracy and authenticity of various monitoring technologies. Meanwhile, a commitment to ensuring quality is essential to minimize the investment costs associated with these technologies. Only through the implementation of such comprehensive measures can we encourage continued collaboration between enterprises and individuals toward achieving the development goals of CO<sub>2</sub> marine geological sequestration.

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## Abbreviations

- NGH natural gas hydrate
- HBS hydrate-bearing sediment
- HCS Hydrate-based CO<sub>2</sub> sequestration
- NBZ negative buoyancy zone
- HFZ hydrate forming zone

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