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Early Assessment of Carbon Dioxide Storage on Seafloor Using Jet-Cooling Technology

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Abstract

It is generally recognized that using depleted oil and gas reservoirs as carbon dioxide (CO_2) storage is subject to the risk of CO_2 leak through wellbores due to the high corrosivity and mobility of CO_2 in gaseous and supercritical states. This work proposes an innovative process called jet-cooling to efficiently deposit and safely store CO_2 on the sea floor. It was found that, in typical seawater environments, if the water depth is greater than 4000 m, CO_2 can be deposited directly on the sea floor in liquid form. The liquid CO_2 will then form hydrates and stay on the sea floor. In the sea water depth ranging from 700 m to 4000 m, CO_2 can be injected through jetting-nozzles to reduce its temperature and generate hydrates. The generated hydrates will then settle down on the sea floor due to gravity. The jetting nozzles should be sized based on the required pressure differential and CO_2 flow rate. Sonic (critical) flow condition is preferred to maximize the cooling efficiency.

Keywords

CO_2 Storage, Gas Hydrate, Seafloor, Potential, Jet-Cooling

1. Introduction

Geological storage of carbon dioxide (CO_2) is considered a major measure for reducing carbon content in the earth's atmosphere. Depleted oil and gas reservoirs are widely used as geological structures for CO_2 storage [1]. A major advantage of using depleted oil and gas reservoirs as CO_2 storage is the elimination of the cost for drilling new wells. Another advantage of oil and gas reservoirs is their high injectivity and high-volumetric efficiency due to their high permeability and porosity. A disadvantage of using these wells is the risk of CO_2 leak through wellbores due to the high corrosivity and mobility of CO_2 in gaseous and supercritical state

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[2] [3]. Studies have shown that the risk of CO_2 leak is lower from subsea geological structures than from onshore oil and gas reservoirs due to the low mobility of CO_2 in liquid state in the former environment [4]. The pioneer process of CO_2 storage in subsea water zone is the Sleipnir project in the Norwegian Sea [5]. Another example process is the Hokkaido project [6]-[8]. However, storing CO_2 in liquid form in subsea structures is not an optimum solution because the stored CO_2 is still at risk of leaking.

CO_2 can exist in the form of gas hydrates which have no mobility and corrosivity. Gas hydrates are solids where small molecule gases are trapped inside the cages of water molecules at high-pressure and low-temperature conditions [9]. Numerous researchers have studied the formation of gas hydrates in static conditions in the past both experimentally and theoretically. The kinetics of formation of gas hydrates was formulated by Englezos *et al.* [10]. The gas hydrate formation rate was found directly proportional to the hydrate crystal particle area, and the difference in gas fugacity was found at the in-situ condition and the gas fugacity at the three-phase equilibrium conditions. Englezos *et al.*'s formula was modified by Skovborg and Rasmussen [11] by replacing the particle area with crystal film during hydrate formation. Chemical potential difference was used as the driving force for hydrate formation by Mohebbi [12] and Kashchiev [13]. The driving force was defined as the temperature difference between the equilibrium and testing conditions by Vysniauskas and Bishnoi [14], while it was defined as the pressure difference between the equilibrium and testing conditions by Natarajian [15]. Based on the knowledge accumulated in the past decades, the conditions (pressure and temperature) for CO_2 hydrate formation are accurately predictable. The pressure and temperature of CO_2 hydrate formation under static conditions are given by Sloan and Koh [9].

Previous studies proposed storing CO_2 in hydrate (solid) forms in subsea formations, including natural gas hydrate reservoirs, where the geological temperatures are below the hydrate-forming temperature of CO_2 [16] [17]. This can greatly reduce the leak probability because CO_2 is locked inside its hydrates, eliminating the problems of its high corrosivity and high mobility. However, efficient placement of CO_2 in the formation is a big issue. The low injectivity of CO_2 into the formations is due to the low permeability of the formations (normal silt-sand) [18]. The use of radial-lateral wells for placing CO2 in marine gas hydrate reservoirs may partially solve the injectivity problem, but it will induce additional costs for well drilling [19]. In this case, CO_2 can form hydrates in the formations during injection, which reduces injectivity [20]. Although using geothermal energy to enhance natural gas production from gas hydrate reservoirs by CO_2 swapping can be a long-term solution to the injectivity problem [21], a significant amount of initial investment is required.

This study proposes an innovative process to permanently store CO_2 in solid form on the sea floor to solve the problems of injectivity and leak risk. Site selection criteria and required technologies are discussed. It should be considered the ultimate process for safely and economically storing CO_2 for carbon reduction in the atmosphere of the Earth.

2. Application Potential

This section describes required pressures and temperatures for stable CO_2 hydrates, seawater temperature, and densities of seawater, CO_2 liquid and CO_2 hydrates. The favorable seawater depths for disposals of CO_2 will be identified accordingly.

2.1. CO_2 Hydrate Forming Condition

Figure 1 presents a contour map of pressure and temperature for stable CO_2 hydrates as compared to that for methane hydrates [9]. It shows that the pressure required for forming CO_2 hydrates is much lower than that for methane at the same temperature. At a sea surface temperature of 20˚C (293.15 K), CO_2 can form hydrates at a pressure of 2000 psi, as indicated by the intersection point with the straight line going through point A. However, at a sea water temperature of 0˚C (273.15 K), CO_2 can form hydrates at a pressure of 160 psi, which is equivalent to a water depth of 109 m (360 feet). These are conditions for CO_2 in fresh water of extreme pressure and temperature. Considering the effect of seawater salinity on the CO_2 hydrate stability, the line between points B and C should be employed to determine the required pressure for stable CO_2 hydrates in seawater.

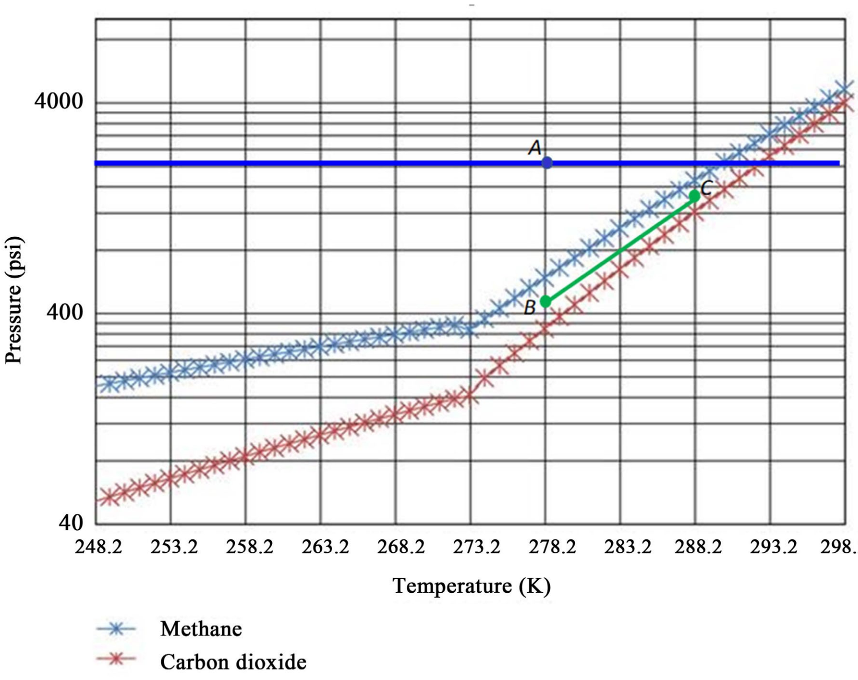

Figure 1. Static conditions for forming CO_2-hydrate and methane-hydrate [9].

2.2. Temperature of Sea Water

Figure 2 shows variations in the ocean's surface temperature and salinity with latitude [22]. The temperatures of sea surfaces around these oceans are similar. Figure 2 presents variations in the ocean's surface temperature and salinity with latitude [22]. The temperatures of sea surfaces around these oceans are similar. It shows that sea surface temperature drops with latitude. **Figure 3** illustrates variations in ocean water temperature with depths at low and high latitudes [23]. The temperature profiles of seawater around the corresponding oceans are similar. In low altitude regions, the surface layer water is an epipelagic zone, also called the sunlight zone, extending from the surface to about 200 meters (660 feet). Most of the sunlight is visible in this zone where the sea surface temperature is as high as 97˚F (36˚C) in the Persian Gulf. In high altitude regions, the sea surface temperature is as low as 28˚F (−2˚C) near the North Pole. Below the epipelagic zone is the mesopelagic zone (twilight zone or the midwater zone) which extends from about 200 meters (660 feet) to about 1000 meters (3300 feet). Water temperature decreases rapidly with increasing depth. The depth of the thermocline varies seasonally. It is found in this work that the temperature in the mesopelagic zone can be generally expressed as

$$T = T_\infty + \frac{T_0 - T_\infty}{2}\left[2 - \left[1 + erf\left(2\left(\frac{D}{D_m} - 1\right)\right)\right]\right] \tag{1}$$

where T is water temperature in ˚C, T_∞ is the stable water temperature below the mesopelagic zone, T_0 is the seasonal-averaged water temperature at sea surface, D is water depth of interest in meters, D_m is the water depth at the mid-point of the mesopelagic zone. The water temperature below the mesopelagic zone tends to remain at a low level T_∞.

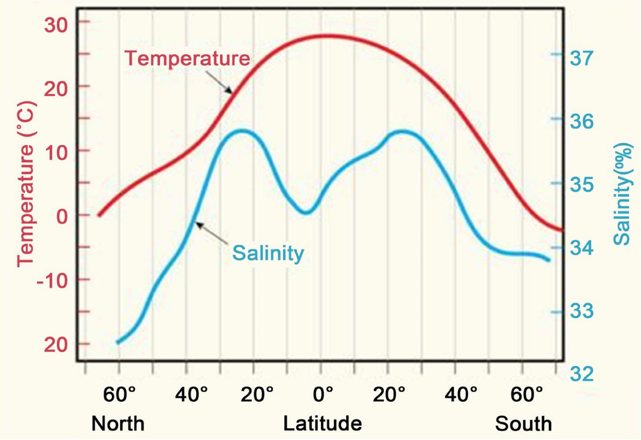

Figure 2. Variations in the ocean's surface temperature and salinity with latitude [22].

2.3. Densities of Sea Water, CO_2 Liquid and CO_2 Hydrates

The data presented by Klemm *et al.* [23] shows that seawater density depends on seawater temperature at depth. Seawater density is determined by the temperature-dependent salinity [24]. It varies in a narrow range between 1.025 g/cc at sea surface and 1.028 g/cc at depth greater than 1000 m. **Figure 4** shows data on the density of CO_2 at various pressures and temperatures [24]. It indicates that, at the

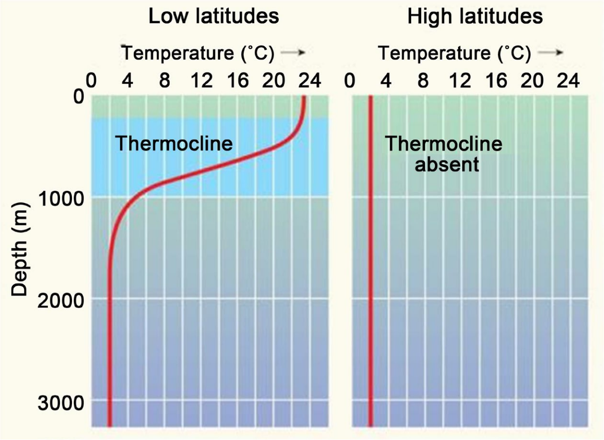

Figure 3. Variations in ocean water temperature with depth [23].

temperature of 0˚C, the density of CO_2 is greater than that of seawater (1.028 g/cc) if the pressure is greater than 170 bars (2465 psi), or deeper than 5540 feet. At the temperature of 30˚C, the density of CO_2 is greater than that of seawater if the pressure is greater than 470 bars (6815 psi), or deeper than 15,315 feet of water. Figure 5 presents a chart for the minimum required pressure and the corresponding water depth for CO_2 liquid to reach a density of 1.028 g/cc based on the interpolation of data given by Calsep [25]. This chart indicates that significant water depth is required to deposit CO_2 in liquid form on the seafloor. For example, 4000 ft of water depth is required even if the in-situ temperature is 2˚C. The density of CO_2 hydrates varies from 1.09 g/cc to 1.11 g/cc, depending on the fraction of CO_2 in hydrate form [9], which is greater than the maximum density of seawater. Therefore, if CO_2 forms hydrates in seawater, the hydrate should settle down to the seafloor due to gravity.

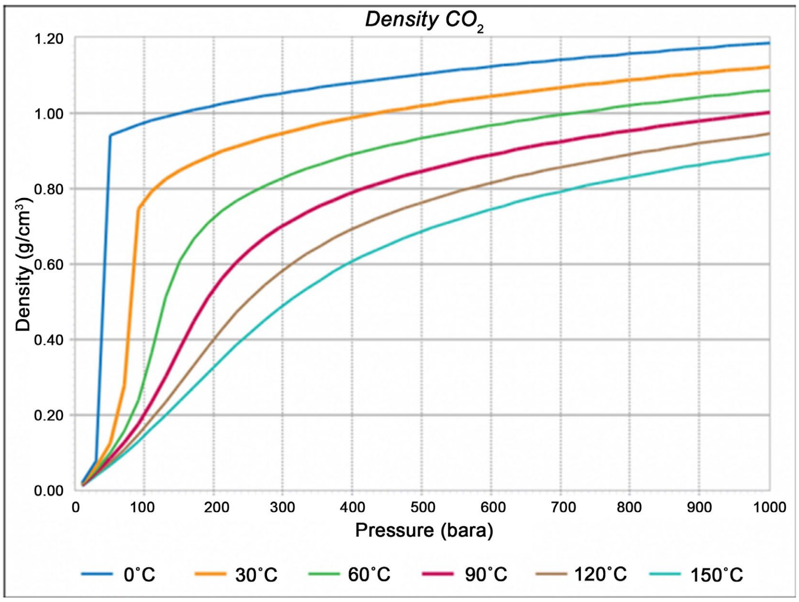

Figure 4. Density of CO_2 at various pressures and temperatures [24].

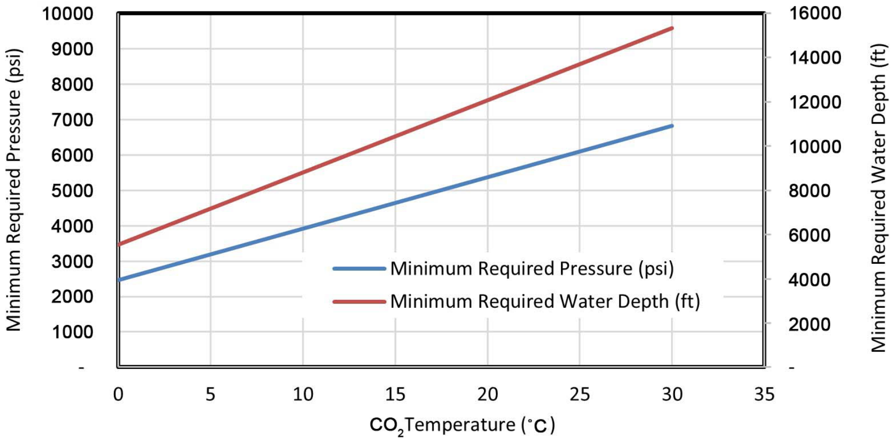

Figure 5. The minimum required pressure and the corresponding water depth for CO_2 liquid to reach a density of 1.028 g/cc based on the interpolation of data given by Calsep [25].

2.4. Favorable Seawater Depths for CO_2 Disposal

Although the water depths given in **Figure 5** allow for CO_2 disposal in liquid form, such deep seafloors are not always accessible. It is desirable to deposit CO_2 on a shallow seafloor. **Figure 6** plots the minimum required pressures for CO_2 hydrate formation in an example seawater with temperature profile given by Equation (1) for $T_0 = 24˚C$, $T_\infty = 2˚C$, and $D_m = 700$ m. The data for the minimum required pressure was read out from **Figure 1**. Also plotted is the seawater pressure with a gradient 0.444 psi/ft based on seawater density of 1.025 g/cc for comparison. This graph indicates that the seawater pressure is greater than the minimum required pressure for CO_2 hydrate formation below about 700 m, suggesting that CO_2 hydrates should form and remain stable below a water depth of about 700 m.

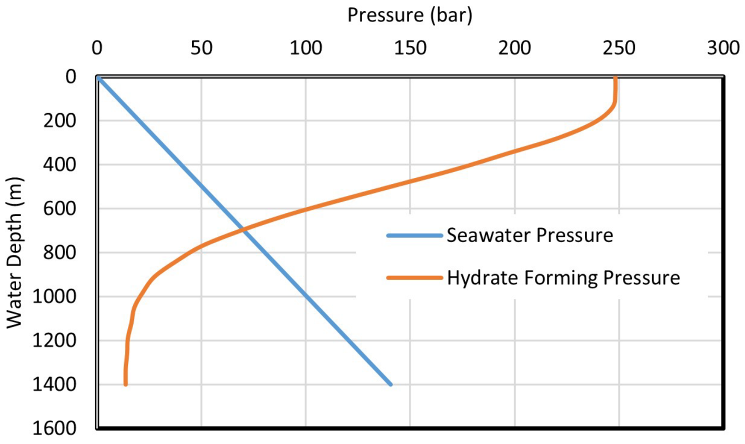

Figure 6. The minimum required pressure for CO_2 hydrate formation compared to seawater pressure.

3. Technology Requirement

Deposition of CO_2 in hydrae status on seafloor below the critical depth requires that CO_2 hydrates are generated instantly in seawater unless the water depth is greater than the depth for stable CO_2 liquid where hydrates can form slowly without concern of hydrate-floating up. To generate CO_2 hydrates instantly, a super-cooling technique is proposed using the Joule-Thomson cooling effect. **Figure 7** presents a schematic diagram of a CO_2 hydrate generation process using a jet-cooling method. Ship 1 is loaded with liquid CO_2 from carrier connector 2 or pipeline hose to CO_2 tank 3. The liquid CO_2 is cooled by cooler 4 to increase density. The cooled CO_2 is pumped by pump 5 into hose 6, leading to the injection head 7 where the CO_2 is expanded through the jetting nozzles 8. Due to the Joule-Thomson cooling effect the jetted CO_2 stream should be further cooled below the seawater temperature, causing the formation of CO_2 hydrates immediately. The remaining CO_2, if there is any, will form hydrates in buffer chamber 9. Depth control room 10 receives seawater depth data from the sonar depth surveyor 11 and adjusts the location of the injection head 7 to keep it close to the seafloor. CO_2 monitoring room 12 receives data from CO_2 detector 13 and sends a signal to the CO_2 pump control to reduce the CO_2 injection rate if free CO_2 is detected. The CO_2 hydrates generated should remain on the seafloor due to their density being higher than seawater. The ship moves forward slowly as needed.

The total cross-sectional area of the nozzles should be designed to cause a significant pressure drop at the nozzles to generate adequate Joule-Thomson cooling effect. The temperature at the downstream of the nozzles can be estimated based on choke flow theory [26]:

$$T_{\mathrm{dn}} = T_{\mathrm{up}} \frac{z_{\mathrm{up}}}{z_{\mathrm{outlet}}} \left(\frac{p_{\mathrm{outlet}}}{p_{\mathrm{up}}} \right)^{\frac{k-1}{k}} \tag{2}$$

where p_{outlet} is the pressure at nozzles' outlet, p_{up} is the upstream pressure, and k is the specific heat ratio of CO_2. The outlet pressure is equal to the sea water pressure in subsonic (subcritical) flow conditions. Sonic (critical) flow conditions can be used to achieve the maximum temperature drop. The critical pressure ratio through nozzles is expressed as [26]:

$$\left(\frac{p_{\mathrm{outlet}}}{p_{\mathrm{up}}} \right)_c = \left(\frac{2}{k+1} \right)^{\frac{k}{k-1}}, \tag{3}$$

where the value of k for CO_2 is about 1.28, the critical pressure ratio is about 0.55 for CO_2. This suggests that the upstream pressure should be more than double the downstream pressure which is the in-situ seawater pressure. For example, if the in-situ seawater temperature is 276˚ K (3˚C), the in-situ seawater pressure should be selected to be at least 16.6 bar for hydrate formation. The nozzle size should be designed to create upstream pressure of at least 13.2 bars for instant hydrate formation. The relation between the upstream pressure, CO_2 flow rate, and total nozzle area can be found in literature [26].

4. Discussions

The study proposes a jet-cooling process that exploits Joule-Thomson expansion

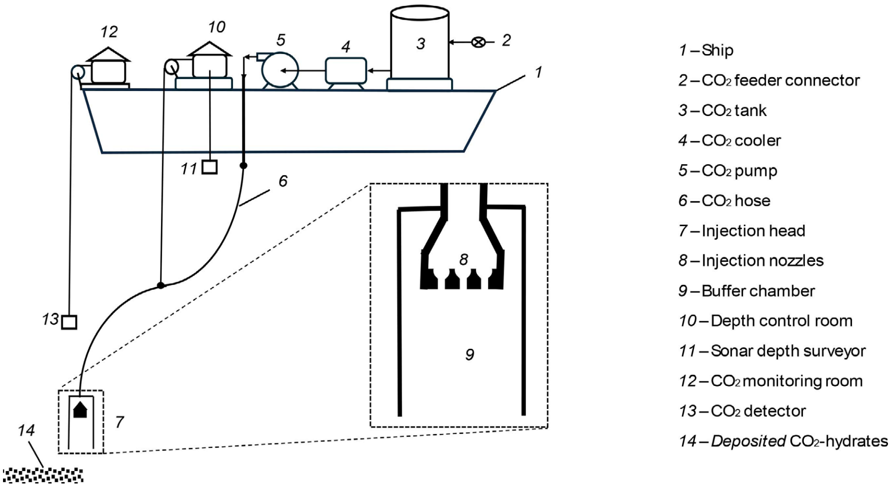

Figure 7. A schematic diagram of a CO_2 hydrate generation process using a jet-cooling method.

to form CO_2 hydrates immediately above the seabed, thereby immobilizing the gas and lowering leakage risk. Pressure-temperature envelopes, seawater density data and hydrate stability charts are combined to define two operational depth windows (>4000 m for liquid CO_2; 700 - 4000 m for hydrate formation). A schematic system layout with CO_2 cooler, pump, hoses, sonic nozzles, buffer chamber, floor survey and depth control, and CO_2 monitoring is outlined. A guideline for designing sonic nozzles is provided. The technique could offer a virtually unlimited, leakage-free offshore storage capacity for captured CO_2. This document provides an early technical assessment of the process only. More research work needs to be conducted to support the engineering design of the system and assess the potential impact of the process on the environment of seafloors. The following work is specially recommended for future focus:

1) Conducting experimental investigations of the proposed jet-cooling process to validate the concept. This is being designed and prepared using a high-pressure-low-temperature windowed cell to observe the settling of CO_2 hydrates and CO_2 liquid due to gravitational segregation.

2) Performing a quantitative hydrate formation kinetics analysis to determine the minimum required residence time of the jet plume for full solidification under the seawater condition of the proposed process. This information will be used to design the geometry (diameter and length) of the buffer chamber to ensure 100% conversion of CO_2 to hydrates before the CO_2 plume exits the buffer chamber.

3) Carrying out research work to analyze the long-term stability of the deposited hydrate layer, including potential resuspension by sea bottom currents, sediment interaction, warming scenarios, and aviation of artificial objects.

4) It is understood that Equation (1) was established based on the data in Figure 3. Under other seawater conditions, either the actual temperature data from local survey or Equation (1) validated using the actual data should be used to determine the critical water depth for CO_2 hydrate formation.

5. Conclusions

An efficient process called jetting-cooling and a technology suite to realize the process for safely depositing CO_2 on the sea floor are presented in this study. The advantages of CO_2 storage on the sea floor in hydrate form include no limitation to storage capacity and stability. There is no concern about the size of geological traps and CO_2 leakage to the atmosphere. The following conclusions are drawn regarding the favorable water depth and necessary equipment design.

CO_2 can be injected and stored on the sea floor in its liquid-then-hydrate form if the sea water is deep enough. In typical seawater environments, if the water depth is greater than 4000 m, CO_2 can be deposited directly to the sea floor in liquid form. The liquid CO_2 will then form hydrates and stay on the sea floor.

CO_2 can be stored on the sea floor in its hydrate form if the sea water is deep enough. In typical sea water environments, if the water depth is greater than 700 m, CO_2 can be injected through nozzles to reduce its temperature and generate hydrates. The generated hydrates will then be deposited on the sea floor due to gravity.

The nozzles for reducing CO_2 temperature to generate hydrate are the key component of the proposed technology suite. The nozzles should be sized based on the required pressure differential and CO_2 flow rate. Sonic (critical) flow condition is preferred to maximize the cooling efficiency.

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Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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