

# Entropy-Driven Carbon Dioxide Capture: The Role of High Salinity and Hydrophobic Monoethanolamine

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Addressing atmospheric CO<sub>2</sub> levels during the transition to carbon neutrality requires efficient CO<sub>2</sub> capture methods. Aqueous amine scrubbing dominates large-scale flue gas capture but is hampered by the energy-intensive regeneration step, sorbent loss, and consequent environmental concerns with volatile amines. Herein, hydrophobic non-volatile alkylated monoethanolamine (MEA) is introduced as a water-lean CO<sub>2</sub> absorbent in brine. The effects of alkylation of MEA, salinity, and aggregation of absorbents on the improved CO<sub>2</sub> capture process are systematically investigated. The CO<sub>2</sub> absorption facilitates spontaneous self-aggregation of hydrophobic absorbents, which increases the entropy of water in high-ion strength solutions. This effect is controlled by the salinity of aqueous solutions, affording comparative gravimetric CO<sub>2</sub> uptake performance to benchmark MEA. It is experimentally verified that the hydrophobicity of alkylated MEAs in saline water is responsible for facile absorption, and also for mild regeneration conditions. Therefore, the entropy-driven approach minimizes absorbent evaporation, corrosion, and decomposition, thus paving the way to realize energy-efficient carbon capture.


## 1. Introduction

Carbon capture and sequestration (CCS) is essential for mitigating global carbon emissions during the transition to cleaner energy sources.<sup>[1]</sup> The latest IPCC report underscores the urgency of limiting Earth's surface warming to 1.5–2 °C from pre-industrial levels to prevent irreversible damage.<sup>[2]</sup>

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Post-combustion carbon capture (PCC)<sup>[3]</sup> from point sources is a pivotal technology for achieving net-zero emissions. Aqueous amine scrubbing<sup>[4a-c]</sup> is an industrial standard PCC method<sup>[5]</sup> due to its maturity and suitability for deployment at a plant-scale level.<sup>[6,7]</sup> Amine absorbents have high chemoselectivity toward CO<sub>2</sub><sup>[8a,b]</sup> and a rapid absorption profile.<sup>[9]</sup> Moreover, they are cheap and easy to manufacture from platform chemicals<sup>[10]</sup> but suffer from high energy requirements for solvent regeneration causing a substantial parasitic burden to the operating power plant.<sup>[3]</sup> They corrode plant equipment<sup>[11]</sup> and form byproducts that reduce the capture performance,<sup>[12a,b]</sup> creating environmental concerns due to the evaporation and decomposition of amines.<sup>[13]</sup>

To address these challenges, water-lean systems for carbon capture have been explored. Substituting<sup>[11]</sup> high-heat capacity

water with organic diluents is to enhance the performance of absorbents while preserving their chemical characteristics. Various non-aqueous organic amine blends,<sup>[14a-c]</sup> amino silicates,<sup>[15a,b]</sup> superbases,<sup>[16]</sup> amino acids,<sup>[17a-c]</sup> alkyl carbonates,<sup>[14a,18a,b]</sup> and N-heterocyclic systems<sup>[19a,b]</sup> were developed to enhance physical absorption<sup>[20]</sup> (equimolar absorption capacity) with regeneration temperatures as low as 80–100 °C.<sup>[3]</sup> Jessop and coworkers developed switchable organic amines responsive to CO<sub>2</sub>,<sup>[21a-c]</sup> which can modulate the solution's physical properties – polarity, solubility, and hygroscopicity – under mild switchable conditions without significant heat energy input for regeneration. These water-lean absorbents are straightforward to synthesize, despite requiring multiple synthetic steps.<sup>[13]</sup>

Considering that PCC should be accompanied by large-scale operations and taking into account the global production volumes of amine-based absorbents,<sup>[13]</sup> it is evident that the use of the most common absorbent, MEA (alongside ammonia), is already maximized across all power plants.<sup>[4c]</sup> Introducing additional complexity into the chemical structures and thus the synthesis of amine absorbents may not be feasible.<sup>[22a,b]</sup> Although this view should not be taken as an absolute, it serves as a guide for designing novel sorbent systems.

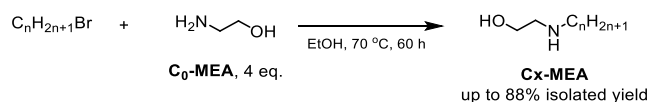
In this context, we envisioned how to access ideal solvent systems for amine-based CO<sub>2</sub> capture based on the following criteria: 1) a water-lean system with high processability (e.g., low foaming); 2) low volatility of the absorbents to minimize evaporation loss and environmental concerns; 3) altering the absorbent to offset  $\Delta H_{\text{abs}}$  and high-temperature requirement; and 4) high

absorption, regeneration rate, and recycling performance. Based on our previous work on CO<sub>2</sub>-mediated desalination using amphiphilic diamines, which are prone to dynamically self-aggregate by CO<sub>2</sub>,<sup>[23]</sup> alkylated amino alcohols have emerged as potential candidates for achieving these criteria (Figure 1A). These amphiphilic amines exhibit switchable properties<sup>[24a-c]</sup> triggered by CO<sub>2</sub>, leading to self-aggregation in aqueous solutions.<sup>[25]</sup> CO<sub>2</sub>-Induced formation of micellar structures can facilitate water release, impacting the spontaneity of the capture process by increasing the entropy. CO<sub>2</sub> has already been identified as an ideal trigger for reversible self-assembly in aqueous media with polymeric materials applied in for example forward osmosis desalination and membrane separation.<sup>[26]</sup>

The ion strength or salinity of aqueous solutions can affect CO<sub>2</sub> capture in aqueous systems.<sup>[27]</sup> In this context, seawater is an abundant source of ions that have previously been utilized in large-scale geological carbon storage and capture. For these processes, low salinity of brine solutions are preferred for both CO<sub>2</sub> reservoirs and CO<sub>2</sub> mineralization.<sup>[28a,b]</sup>

Herein, we showcase how altering the hydrophobicity of MEA and the high salinity of the solution can lead to high-performing CO<sub>2</sub> absorbent systems in terms of CO<sub>2</sub> capture capacity, kinetics, and regeneration temperatures. Various alkylated-MEA derivatives were prepared to study positive hydrophobic effects in CO<sub>2</sub> capture in water-lean brine solutions. The ionic strength of the solution is found to effectively enhance the CO<sub>2</sub> capture capacity, owing to the hydrophobicity-driven self-aggregation of the amphiphile and the increased entropy of water (Figure 1B,C).

We commenced our investigation with alkylated MEAs and unmodified MEA to compare their performances directly. To test our absorbent system, we used 35 g L<sup>-1</sup> NaCl (0.6 M) solution to mimic the average seawater salt concentration, which is abundant and more accessible than pure water. A comparative study on CO<sub>2</sub> capture (Figure 1D) revealed that C12-MEA showed higher capture capacity in saline water than in deionized water (0.70 compared to 0.47 mol<sub>CO<sub>2</sub></sub>:mol<sub>absorbent</sub> loading, respectively). Unmodified MEA showed negligible differences in terms of total capacity in pure and saline water.



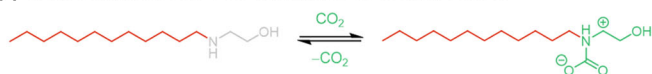
**Scheme 1.** A general synthesis scheme for C<sub>n</sub>-MEA absorbents.

Based on this preliminary result, we hypothesized that the increased performance of alkylated MEA in a brine solution is directly related to the hydrophobicity of the absorbent. To test this hypothesis, we varied the chain length of alkylated-MEA molecules via a one-step substitution reaction from the MEA molecule and a corresponding alkyl bromide (Scheme 1).

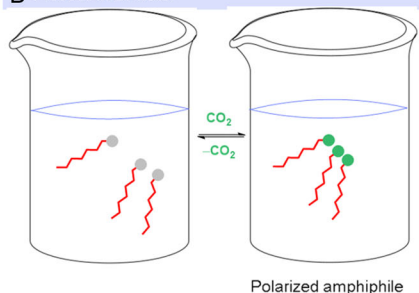
All absorbents were tested under identical conditions (20 wt% absorbent in 80 wt% DI water or brine 0.6 M, with pure CO<sub>2</sub> used at a low flow rate (2 mL min<sup>-1</sup>, room temperature to minimize evaporative water loss). Results show that up to C9-MEA the performance between experiments in DI water (blue bars) and brine (orange bars) were close to identical and C10-MEA was identified as the first molecule that exhibited higher performance in brine (Figure 2A). Interestingly, the longer the chain length the greater the difference between experiments in brine and DI water (C10, 12, and C16-MEA). To avoid a high energy penalty at the regeneration step, we reduced the water content of the solvent. We tested various absorbent loadings (5–90 wt%) of C10-MEA in 35 g L<sup>-1</sup> brine to maximize the capacity of CO<sub>2</sub> absorbed. The results show that 80 wt% C10-MEA solution reaches up to CO<sub>2</sub> loading of 8.6 wt%, approaching the gravimetric capacity of the standard MEA solution (9.5 wt%, Figure 2B).

Realizing that the ion strength of the solution plays a crucial role in water-rich systems for CO<sub>2</sub> capture, we anticipated a similar effect holds for water-lean systems. We subjected the 80 wt% C10-MEA to a salinity screening by preparing model aqueous NaCl(aq) solutions of 5, 35, 100, 175, 250, 300, and 360 (saturation point of NaCl in water) g L<sup>-1</sup> NaCl concentrations as illustrated in Figure 2C. With C10-MEA, high ion strength solutions were superior for CO<sub>2</sub> capture to low-salinity solutions; 250 g L<sup>-1</sup> salinity was optimal in terms of total CO<sub>2</sub> capture capacity (9.65 wt%) reaching the 20 wt% MEA benchmark (9.71 mmol

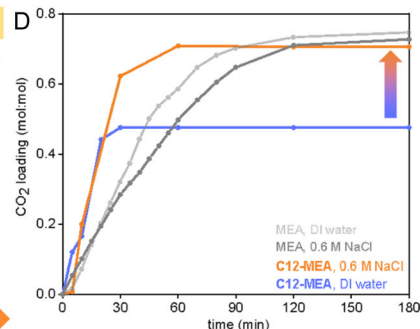
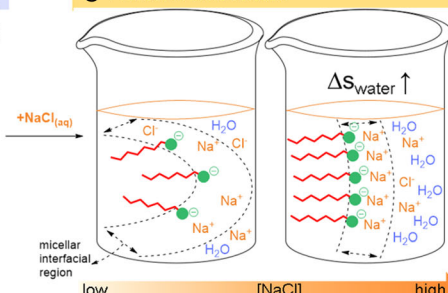
**A** Reversible polarity switch with alkylated MEA (C12-MEA) and CO<sub>2</sub>



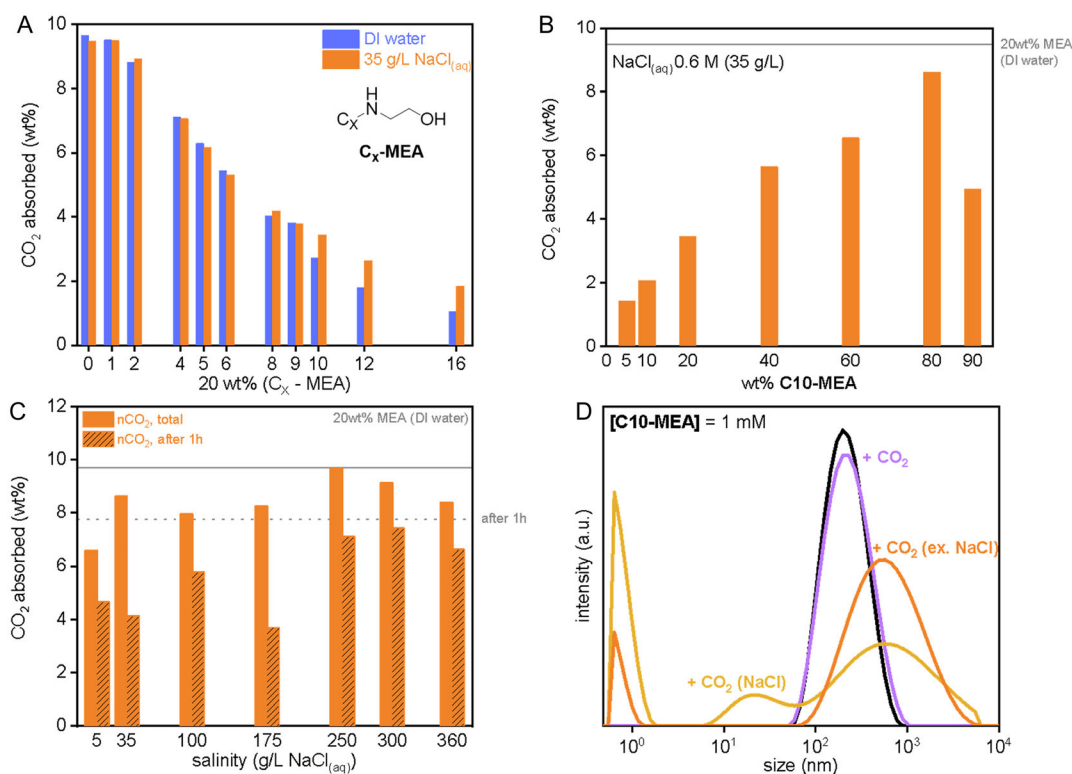
**B** Process in DI water



**C** Process in brine solution



**Figure 1.** A) CO<sub>2</sub>-induced polarity switch of C12-MEA as a model substrate. B) Hydrophobic MEA in DI water. C) Salinity effect on CO<sub>2</sub> responsive self-aggregation of amphiphiles in water-lean system. D) Preliminary experiments with enhanced CO<sub>2</sub> capture performance of C12-MEA in brine (orange) compared to DI water (blue).



**Figure 2.** A) The chain length screening. B) Screening of the absorbent loading. C) The effect of increasing salt concentration on CO<sub>2</sub> capturing performance of 80 wt% C10-MEA solution. D) Salt effect on C10-MEA aggregation in 1 mM DMSO solution measured in dynamic light scattering (DLS).

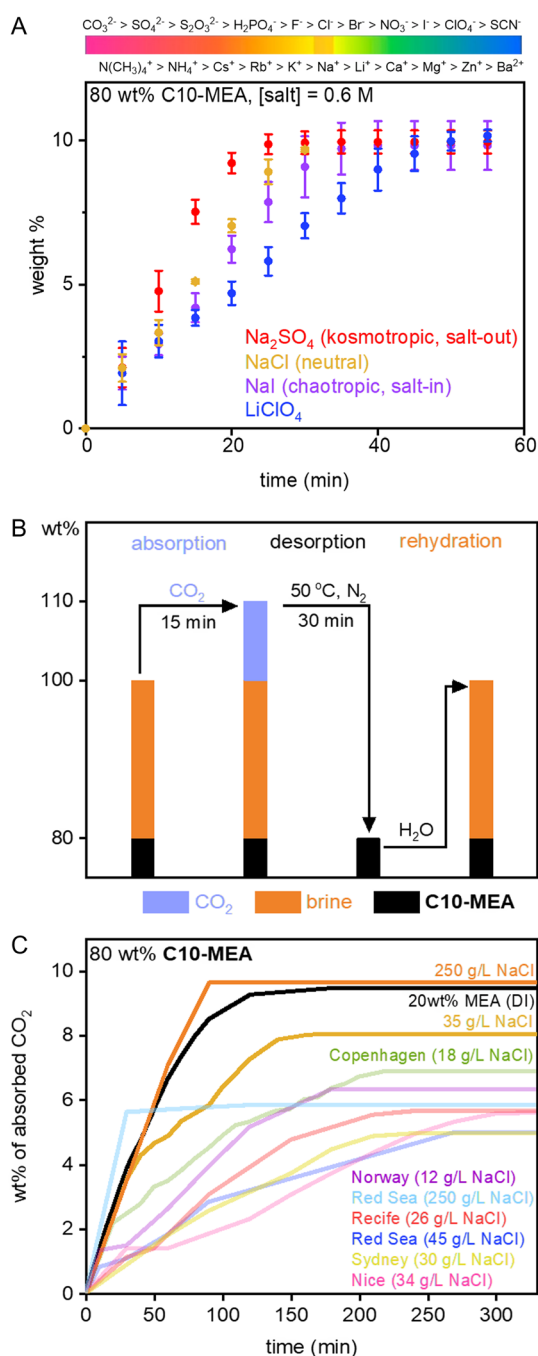
CO<sub>2</sub> per g of absorbent). The high salt concentration provides not only a high CO<sub>2</sub> loading capacity but also in terms of absorption rate. The three solutions (250, 300, and 360 g L<sup>-1</sup>) that are highest in capacity also exhibited the highest absorption rates at 1 h of CO<sub>2</sub> absorption experiments (Figure 2C, orange bars, shaded). An optimal salt concentration with high CO<sub>2</sub> loading and absorption rate was 250 g L<sup>-1</sup>, which might be ascribed to effective self-aggregation, and the critical micelle concentration (CMC) of surfactants like amphiphile C10-MEA. This prompted us to further investigate the effect of salts on the aggregation and micelle formation of C10-MEA (see Figure S21, Supporting Information, for an overview of salinity and absorbent loading screening).

Salts and ions play a crucial role in influencing the behavior of surfactant molecules in water, particularly in terms of aggregation and micellization of amphiphiles.<sup>[29]</sup> Molecular dynamic simulations on sodium dodecyl sulfate reveal that increased salt concentration enhances the packing of the surfactants, fostering aggregation at lower CMCs.<sup>[30]</sup> Therefore, higher aggregation of surfactants results in the expulsion of water molecules, which can be explained by a more pronounced hydrophobic effect and increased entropy of the system. This finding is in agreement with the study on the cation effect on the micelle-to-vesicle transition in the micellar interfacial region.<sup>[31]</sup> Based on our qualitative studies by <sup>1</sup>H and <sup>13</sup>C NMR spectroscopy (see Figures S1–S3, Supporting Information), we presumed that NaCl concentration correlates with the rise of the carbamate (RR'N-CO<sub>2</sub>)-sodium ion pairs in micellar interfacial regions,

significantly reducing interfacial water volume (Figure 1C, at a higher [NaCl]). This phenomenon arises because NaCl ions, being more hydrated, outcompeting hydrophobic ammonium carbamate ion pairs. The hydrophobic effect of aliphatic chains elevates the entropy of the solution, as water at the micellar interfacial region has lower entropy than bulk water.

To shed light on this hypothesis, dynamic light scattering (DLS) experiments were conducted with C10-MEA (Figure 2D, black). We observed that CO<sub>2</sub> introduction to C10-MEA has a minimal impact (purple) on particle size distribution without any salts. In contrast, the addition of NaCl significantly increased the particle sizes to higher micellar structures (~500 nm, Figure 2D, orange). These results suggest a unique entropy-driven CO<sub>2</sub> capture mechanism where the presence of salts contributes to the spontaneity of self-aggregation of amphiphiles. This finding is consistent with our observation that absorbents with longer alkyl chains (longer than C9) display a larger difference in terms of CO<sub>2</sub> capture capacity in deionized water and brine solutions (Figure 2A).

Next, we performed a Hofmeister series analysis with NaCl as a neutral salt solution (orange, Figure 3A). Under our standard CO<sub>2</sub> absorption conditions (1 atm CO<sub>2</sub>, 2 mL min<sup>-1</sup>, room temperature), a chaotropic salt, NaI (purple), showed a poorer performance due to micelle dissolution (salting-in) while kosmotropic NaSO<sub>4</sub> (red) showed superior performance due to micelle forming (salting-out).<sup>[32]</sup> These results further confirm the CO<sub>2</sub>-induced micelle formation and its effect on the capture rate and the capacity.



**Figure 3.** A) Hofmeister anion series effect on  $\text{CO}_2$  capture rate and performance (experiments performed in triplicates).  $\text{LiClO}_4$  was added as a common micelle-breaking salt. B) Absorption/desorption cycle of the 80 wt% C10-MEA solution and 20 wt% 0.6 M brine. C)  $\text{CO}_2$  absorption curves of model seawater (orange shades) and real sea water solutions with 80 wt% C10-MEA.

To assess the energy cost of entropy-driven carbon capture, we first conducted calorimetry to determine the heat of absorption of amphiphile C10-MEA in different concentrations of salt solutions (Figure S4, S5, Supporting Information). Theoretically, our water-lean system should exhibit a smaller heat of

absorption<sup>[33]</sup> than that of the benchmark water-rich solution (MEA in 70–80 wt%  $\text{H}_2\text{O}$ ). We repeatedly injected small amounts of gaseous  $\text{CO}_2$  (20  $\mu\text{L}$ , 0.16 equiv. to C10-MEA) and measured the energy input of the calorimeter to match the energy released in the exothermic capture process. At  $25^\circ\text{C}$  and ambient pressure, we obtained  $\Delta H_{\text{abs}} = -63.5 \text{ kJ mol}^{-1}$  in  $\text{NaCl}$   $35 \text{ g L}^{-1}$  and  $\Delta H_{\text{abs}} = -78.0 \text{ kJ mol}^{-1}$  for a  $250 \text{ g L}^{-1}$  brine solution system, which is in good agreement with their respective performances. These values fall within the range for water-lean systems and are significantly lower than  $\Delta H_{\text{abs}}$  of the MEA system (80–100  $\text{kJ mol}^{-1}$ ), implying a reduced energy penalty at the desorption step in our system.<sup>[34a,b]</sup> We then performed desorption experiments with C10-MEA in brine, which showed complete desorption at  $50^\circ\text{C}$  under the  $\text{N}_2$  stream within 30 min (Figure 3B). Thermal gravimetric analysis also confirmed the facile desorption of water and  $\text{CO}_2$  starting at  $\approx 70^\circ\text{C}$  (Figure S18, Supporting Information).

We also demonstrated  $\text{CO}_2$  capture using real seawater samples with salinities ranging approximately from 12 to  $45 \text{ g L}^{-1}$ , testing their  $\text{CO}_2$  capture capacities (Figure 3C). Our experimental data show comparable gravimetric capture capacities to other model saline solutions, achieving  $\text{CO}_2$  absorption of up to 8–10 wt%. Notably, regardless of the varying salinities and compositions of the real seawater samples, we detected 4–7 wt%  $\text{CO}_2$  absorption in our water-lean systems. This underscores the robustness of C10-MEA for entropy-driven  $\text{CO}_2$  capture. We observed a higher absorption rate in a high salinity real seawater solution (Red Sea), to which  $\text{NaCl}$  was added to adjust the salinity to  $250 \text{ g L}^{-1}$  (light blue, Figure 3C). For practical applications, it is noteworthy that C10-MEA exhibits insolubility in deionized (DI) water when exposed to  $\text{CO}_2$ . However, a homogeneous solution of C10-MEA was observed in brine, without significant foaming, during  $\text{CO}_2$  capture experiments (Figure S22, Supporting Information). This suggests that alkylated MEAs are feasible for carbon dioxide removal on a larger scale.

## 2. Conclusion

We have demonstrated a  $\text{CO}_2$  capturing method comparable to the industry's current best technology in terms of performance and regeneration temperature while achieving several advantages over conventional MEA-based aqueous solutions: a higher absorption rate, lower regeneration energy, minimal thermal, and oxidative degradation (see Figure S20, Supporting Information, for thermal stability of C10-MEA under air at high temperatures), and absorbent loss due to the minimized evaporation and corrosion. The amphiphilic MEAs exhibit high carbon capture performance in an entropy-driven water-lean brine system, controlling the aggregation of amphiphiles, thus increasing the entropy of the water. A large-scale test with an improved performance of MEA-based amine absorbents in water-lean systems is under investigation.

## Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.



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## Conflict of Interest

A.P., R.L., and J.W.L. filed a preliminary patent application.

## Author Contributions

**Aleksa Petrović:** Conceptualization (lead); Data curation (lead); Formal analysis (lead); Investigation (lead); Methodology (lead); Writing—original draft (lead); Writing—review & editing (lead). **Rodrigo Lima:** Methodology (equal); Resources (lead); Writing—original draft (supporting); Writing—review & editing (supporting). **Peter Westh:** Formal analysis (lead); Methodology (lead); Supervision (supporting); Writing—original draft (supporting); Writing—review & editing (supporting). **Ji-Woong Lee:** Conceptualization (lead); Data curation (lead); Formal analysis (lead); Funding acquisition (lead); Investigation (lead); Methodology (lead); Project administration (lead); Supervision (lead); Writing—original draft (lead); Writing—review & editing (lead).

## Data Availability Statement

The data that support the findings of this study are available in the supplementary material of this article.

## Keywords

carbon capture, CO<sub>2</sub>, entropy, hydrophobicity, sea water

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