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Sustainable System for CO₂ Capturing with Multiple Products

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Abstract

Currently, we are facing several serious issues, including diseases, climate change, economics, and wars. Among them, which needs to be resolved imminently to protect the future generations. Recently, we developed an innovative CO2 fixation and storage method based on the use of chemical compounds such as NaOH and CaCl2, to prevent the climate crisis. Herein, the electrolysis of a NaCl solution or seawater was performed to make the resultant NaOH react with CO2. Amines that react with CO2 were also examined. Moreover, seawater was used to produce CaCO3 rather than the CaCl2 solution. Although amines are currently used to capture CO2 from exhaust gases, the thermal treatment of the amine—CO2 complex solution is necessary to release CO2. Thermal treatment requires energy that eventually produces CO2 and induces the degradation of organic amines. However, the CO2 captured in the amine or NaOH solution was released easily by acidifying the CO2-containing solutions. Moreover, HCl could release CO2 from hydro carbonates, Ca(OH)2, Mg(OH)2, and mMgCO3·Mg(OH)2·nH2O, as well as from mineral carbonates, CaCO3 and MgCO3. This simple method of releasing the captured CO2 from the amine—CO2 complexes, hydro carbonates, and mineral carbonates is crucial to obtain pure CO2 for additional usage as an original material.

Keywords: CO2, Climate Crisis, SDGs, DAC, CCS, Limestone, CaCO3, Seawater

1. Introduction

Post the Industrial Revolution, the increase in the CO2 levels in the atmosphere has led to a climate crisis and an increase in the global temperature [1]. On July 4, 2023, the British Broadcasting Corporation reported that at least seven people had been killed after being caught in an avalanche initiated by the collapse of the Marmolada Glacier in the Italian Alps. Similarly, on July 23, 2023, NHK (Japan Broadcasting Corporation) reported extremely high temperatures in over 40 European countries such as the UK, France, Spain, and Portugal. Over 1,300 people died in Spain and Portugal, and thunderbolts caused by unexpectedly high temperatures led to severe mountain fires in Spain. These incidents indicate that the accumulated CO2 in the atmosphere should be reduced immediately through global cooperation to prevent the Earth from irreversible ruin [2]. CO2 can be captured from ambient air or flue gas using several methods, such as absorption, adsorption [3-9], and membrane separation [10,11]. However, achieving a carbon-neutral society by 2050 is unlikely. Practically, based on their nucleophilic characteristics, amine solutions have been used to capture CO2 [12-14]. Therefore, we evaluated practical methods for reducing atmospheric CO2 using amines. We recently developed an innovative method for CO2 fixation and storage using low concentrations of NaOH and CaCl₂ [16-19]. Certain improvements in our CO₂ fixation

method would contribute to the prevention of the climate crisis.

Although renewable energy sources, including solar radiation and wind, can reduce CO₂ emissions, these alternative systems require energy expenditure and may involve CO₂ production. Furthermore, they do not remove the accumulated CO₂ in the atmosphere or address the ongoing problem of CO₂ generation from exhaust gases and industrial sources. Thus, even if a carbonneutral society could be immediately realized, the accumulated CO₂ in the atmosphere remains to be a problem. Therefore, it is important to develop immediate methods to reduce the current CO₂ level in the Earth's atmosphere. Geosequestration by injecting CO₂ into underground geological formations, such as oil fields, gas fields, and saline formations, has potential for CO₂ storage [20,21].

However, these methods are projects for the future.

2. Materials and Methods

2.1. Chemicals

Reagent grade NaOH, CaCl₂, ethanolamine, and MgCl₂ were purchased from Fujifilm Wako Pure Chemical Corp. (Osaka, Japan). Gaseous CO₂ was purchased from Keni's (Osaka, Japan).

2.2 Electrolysis

The electrolysis of the NaCl solution or seawater was performed using an electric current converter (SKY Top power) from Shines Tenakee Degen Co. Ltd. (Shines, China).

2.3. CO₂ Concentration Measurements

CO2 was captured and released in polyethylene terephthalate (PET) bottles. Small volumes (5–10 mL) of solutions such as NaOH, seawater, and electrolyzed NaCl and seawater were poured into the PET bottles (300–600 mL). Subsequently, the CO2 concentration within the bottles was measured under various conditions using CO2 analysers, CX-6000 ((for high concentration; Riken Keiki, Co. Ltd. (Tokyo, Japan)), and XP-3140 ((for low concentration; Cosmo, Co., Ltd. (Tokyo, Japan)).

2.4. Precipitate Measurement

A small portion (1.5 mL) of the resulting solutions was poured into microcentrifuge tubes and centrifuged at 6200 rpm for 1 min to separate the precipitates. The weight of the wet precipitates was obtained by measuring the difference in the centrifuge tube

weight before and after centrifugation [16].

3. Results

3.1. Electrolysis of NaCl

Electrolysis converts H₂O into hydrogen (H₂) and oxygen (O₂) [16,17,22]. Herein, the NaCl solution was electrolyzed, and NaOH was eventually produced. The produced NaOH easily reacted with CO₂ in the solution and formed Na₂CO₃ and NaHCO₃. The carbonate ions, CO₃²⁺ and HCO₃⁺, react with Ca²⁺ and form CaCO³ precipitates, existing as limestone in nature. Interestingly, CaCO³ is a component of corals.

When the electrolyzed NaCl solution was treated with air bubbles in the vinyl chloride pipe, CO_2 (~600 ppm) in the atmosphere was completely captured in the solution (Fig. 1a). The air containing 12%–18% CO_2 in the same system reduced the CO2 concentration to 4% when passed through the electrolyzed solution (Fig. 1b). These results demonstrate that the electrolyzed NaCl solution is extremely effective in capturing CO_2 in the air.

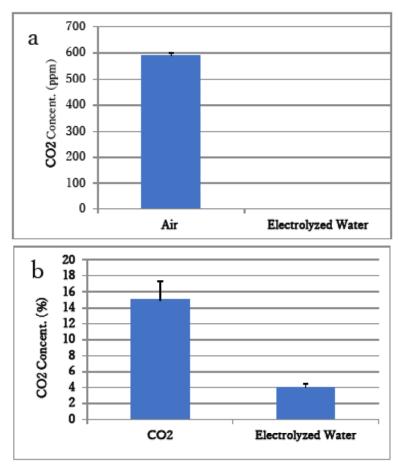


Figure1: CO2 absorption by electrolyzed NaCl solution. A 200 mL solution of the electrolyzed NaCl solution was poured into the vinyl chloride pipe (40-mm diameter), and air (a) or CO2-added air (b) was bubbled from the bottom of the pipe through the bubble former. The presented value is the mean of three to five experiments.

3.2. Electrolysis of Seawater

Seawater that contains many minerals, such as Na, Mg, and Ca, is preferred for electrolysis rather than a NaCl solution. Electrolyzing seawater yielded white precipitates because a large amount of Mg2+ readily formed magnesium hydrate, Mg(OH)2, under alkaline conditions. In fact, 1.5 g of wet precipitate was obtained in 40 mL of electrolyzed solution. Bubbling CO2 in the supernatants after centrifuging the solution yielded no white precipitates. However, the addition of CaCl2 before bubbling CO2 yielded white precipitates of CaCO3, suggesting that CaCO3 precipitation might have occurred during the electrolysis of seawater.

3.3. Mg(OH)2 and Ca(OH)2 Precipitations

A MgCl₂ solution was mixed with the NaOH solution to vary the pH levels. A small amount of Mg(OH)₂ precipitates, which are semi-transparent white colloids, appeared at low NaOH concentrations of 0.01–0.05 M, while clear precipitates were observed at concentrations of >0.1 M NaOH (Fig. 2). Mg(OH)₂

precipitate formation plateaued at 0.1 M NaOH.

When CaCl₂ and NaOH solutions were mixed to form varying pH levels, a clear white precipitate, Ca(OH)₂, was observed at a concentration of >0.2 M NaOH (Fig. 2b). Apparently, the solution of concentration below 0.1 M NaOH did not show any precipitate because of Ca(OH)₂. This result is consistent with our previous results [16]. The critical concentration at which hydrate precipitation occurs differs for Mg₂+ and Ca₂+ (Fig. 2).

3.4. CO₂ Treatment of Authentic Seawater

The characterized mineral concentrations in seawater for Na+, Mg₂+, and C₂+ are known to be 0.3, 0.05, and 0.01 M, respectively. Herein, their concentrations were 0.2, 0.05 and 0.01 M, respectively. At 0.2 M NaOH solution, Mg₂+ was precipitated by its hydrate formation, Mg(OH)₂, as shown in Fig. 3. The precipitate was isolated by centrifuging at 3,000 rpm for 10 min. It was found that the precipitate was dominantly Mg(OH)₂ with a small amount of Ca(OH)₂ (Fig. 2b).

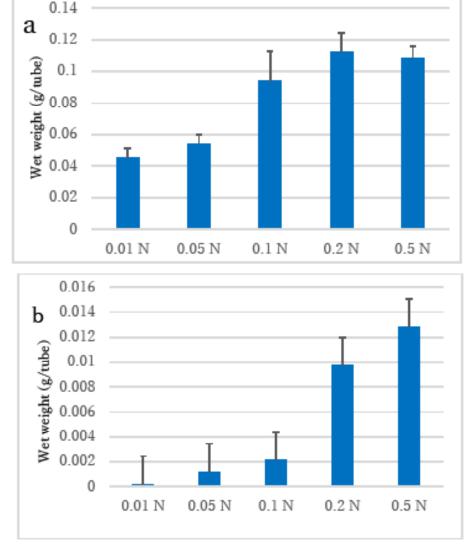


Figure 2: Mg(OH)₂ and Ca(OH)₂ precipitations at different NaOH concentrations. 0.5 M MgCl₂ (a) or 0.5 M CaCl₂ (b) was mixed with NaOH at different concentrations. The precipitates were separated via centrifugation at 6,200 rpm for 1 min. The weight of the wet precipitates is the mean of four to five experiments.

When the supernatant was treated with bubbling CO₂, Ca₂+ was precipitated as CaCO₃ in the solution (Fig. 3). The CaCO₃ precipitate could be observed clearly and was separated via centrifugation. However, the CaCO₃ precipitate amount was considerably lesser than that of MgCO₃ (Fig. 3). This result is reasonable based on their initial concentrations.

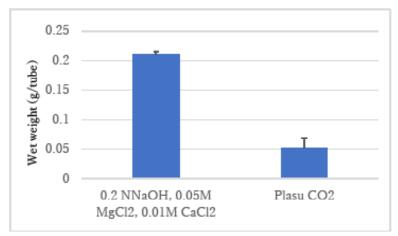


Figure 3: Mg(OH)₂ precipitation and further CaCO₃ precipitation by CO₂ bubbling. Authentic seawater comprising 0.2 M NaOH, 0.05 M MgCl₂, and 0.01 M CaCl₂ was prepared, and the obtained Mg(OH)₂ precipitate was first separated via centrifugation at 6,200 rpm for 1 min. After separating Mg(OH)₂, CO₂ gas was bubbled into the supernatant and the weight of the wet CaCO₃ precipitate was measured. The presented value is the mean for five experiments.

3.5. CO2 Treatment of Seawater

Seawater was alkalized by adding solid NaOH to prepare a 0.2 M NaOH solution. Air was bubbled using a microbubble former stone at the bottom of the vinyl chloride pipe. The CO₂ concentration in the air at the upper part of the pipe was measured at different times (Fig. 4a). The CO₂ concentration decreased considerably from 630 to 120 ppm within 1 h. This reduction occurred in 5 min. The effect of the alkaline solution on CO₂ capture continued until 6 h, and a slight effect persisted

even after 24 h (Fig. 4a).

The seawater pH value was changed at different CO₂ bubbling times (Fig. 4b). The pH value was almost constant for 6 h of bubbling and then gradually decreased along with decreasing bubbling time. This pH change period correlated with the CO₂ capture ability of the alkaline seawater (Fig. 4). Thus, the result indicates that CO₂ capture depends on the solution ph.

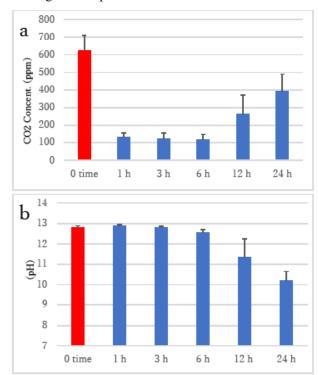


Figure 4: CO₂ absorption by alkalized seawater. Solid NaOH (1.6 g) was dissolved into 200 mL seawater in a 300 mL PET bottle. Air was bubbled from the PET bottle bottom, and CO₂ concentration in the upper space of the bottle at different times (a) and the pH value (b) in the solution were measured. The presented value is the mean of four to fiveexperiments.

3.6. Mg(OH)₂ Reacts with CO₂

The reaction of Mg(OH)₂ with CO₂ is well characterized [22]. When Mg(OH)₂ is treated with CO₂ in an aqueous solution, mMgCO₃ Mg(OH)₂ nH₂O is formed. Mg(OH)₂ was formed at 0.2 M NaOH in 0.05 M MgCl₂ solution, followed by CO₂

bubbling for 30 s. The solution was centrifuged to separate the precipitates at different times. After 5 min, the precipitate amount considerably reduced and a slight reduction continued until 30 min (Fig. 5).

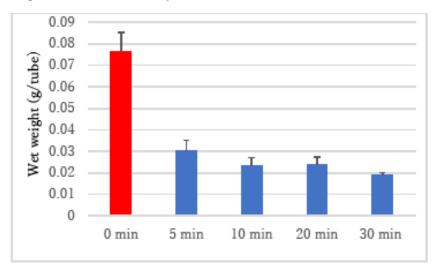


Figure 5: Reaction of Mg(OH)₂ with CO₂. CO₂ gas was bubbled into 0.05 M Mg(OH)₂ solution for 30 s, and then the weight of the wet precipitates was measured. The presented value is the mean of four experiments.

3.7. Release of the Captured CO2

When 0.05 M NaOH solution bubbled with air was acidified by HCl addition, its pH reduced from 10.0 to 1.5 with increasing HCl addition (Fig. 6a). However, the CO₂ release from the bubbled

solution increased from 1,500 to 8,000 ppm with increasing HCl addition (Fig. 6b). Drastic CO₂ release was observed below pH 3.0. It plateaued at 0.05 M NaOH, indicating the pH and CO₂ release were inversely related.

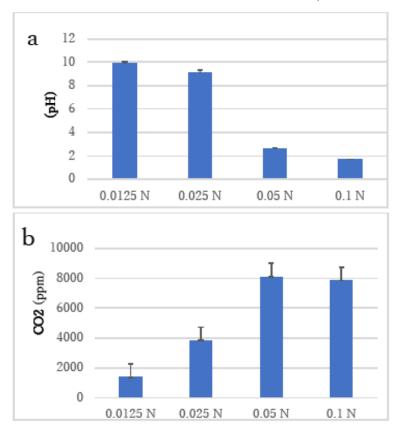


Figure 6: Change of pH and CO₂ release by HCl addition. Air was bubbled into 200 mL of 0.05 M NaOH solution for 6 h, followed by adding different volumes of HCl to the bubbled solution. CO₂ concentration was measured after 5 min of HCl addition. The presented value is the mean of five experiments.

Mixing 0.1 N NaOH solution with CO₂ vigorously in a PET bottle reduced the CO₂ concentration to 75 ppm (Fig. 7a). This result was consistent with other studies [16]. Acidifying the CO₂-treated solution by adding HCl released CO₂ and increased its concentration to 780 ppm in the bottle. Further, alkalizing the solution by NaOH addition reduced the CO₂ concentration to 75 ppm. In the first step, OH– and CO₂ formed CO₃^{2–}, which was

subsequently converted to CO₂ and H₂O by adding HCl in the second step. Finally, alkalizing with NaOH led to CO₂ capture by the solution. Adding CaCl₂ to the solution in the first step caused Ca₂+ and CO₃²⁻ to react and precipitate CaCO₃. Although CaCl₂ addition occurred at higher NaOH concentration of >0.2 N, Ca(OH)₂ precipitates were formed [17]. The pH values were 13.06, 3.38, and 12.76 at the three steps, respectively (Fig. 7b).

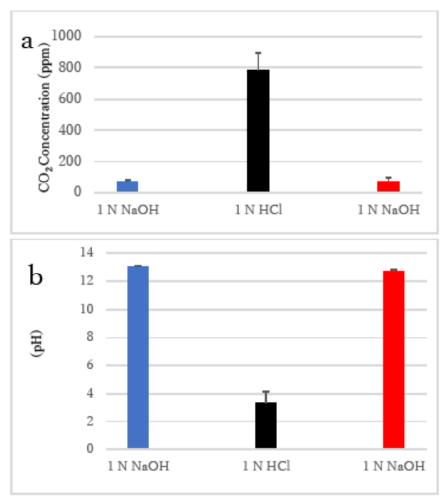


Figure 7: CO₂ capture, release, and recapture. 0.1 M (10 mL) NaOH solution was placed in the PET bottle. First, 0.5 mL of 1 N HCl followed by 0.5 mL of 1 N NaOH were added to the solution. CO₂ concentration in the PET bottle (a) and pH values (b).

3.8. CO₂ Capture by Ethanolamine and Release of Captured CO₂ by HCl.

Ethanolamine is a liquid compound that can capture CO₂ because of its fundamental characteristics [12-15]. The atmospheric CO₂ in the PET bottle was captured with different aqueous ethanolamine solution concentrations (0.25%, 1.0%, and 10% v/v). The initial CO₂ concentration was 504 ppm, which increased with increases in ethanolamine concentrations to 205, 140, and 0 ppm, respectively (Fig. 8a). The pH values of these solutions were 7.58, 10.43, 11.56, and 12.20, respectively (Fig. 8b). Once the ethanolamine solution was treated with CO₂, CaCO₃ precipitation was observed with CaCl2 addition (data not shown).

Atmospheric CO₂ was captured by the ethanolamine—water solution in a PET bottle, and the solution was acidified by adding HCl. The first ethanolamine treatment reduced the CO₂ concentration from 480 to 110–117 ppm (Fig. 8c). Acidifying the CO₂-containing ethanolamine solution with HCl increased the CO₂ concentration to 453.3 ppm in the PET bottle. However, alkalizing the solution with NaOH reduced the CO₂ concentration to 120 ppm (Fig. 8c). This indicates that the interaction between CO₂ and ethanolamine is reversible and pH-based. The result regarding CO₂ capture and CO₂ release by HCl addition was consistent with results obtained from other amines, such as 2-amino-1-phenylethanol and polyethyleneimine (data not shown).

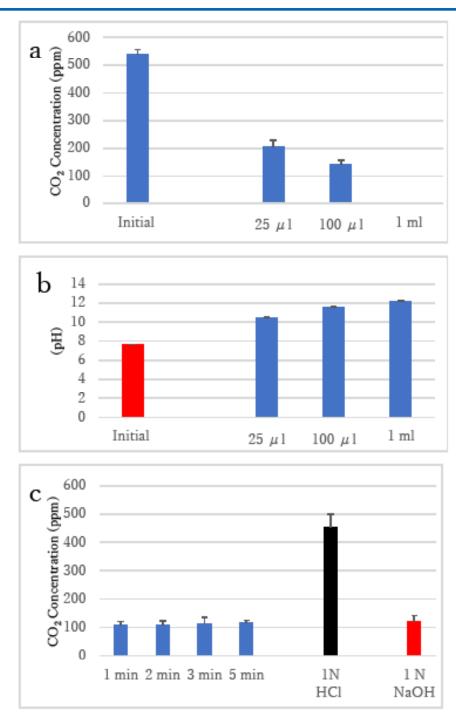
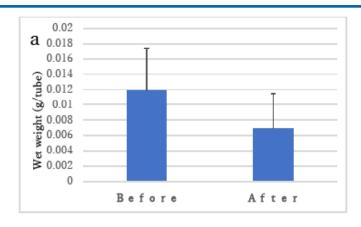


Figure 8: Atmospheric CO₂ capture by aqueous ethanolamine solution in the PET bottle. The ethanolamine concentrations were 25, 100, and 1,000 μ L/10 mL H₂O (a) the pH values at different ethanolamine concentrations (b) CO₂ captured by ethanolamine and then released by adding 4 mL of 1 N HCl, followed by the addition of 4 mL of 1 N NaOH (c).

3.9. Release of CO2 from Electrolyzed Seawater

When seawater was alkalized via electrolysis, Mg(OH)₂ precipitates were formed by the newly produced NaOH. This study demonstrated that the weight of the wet Mg(OH)₂ precipitates was reduced by treating the solution with CO₂ based on the formation of mMgCO₃ Mg(OH)₂ nH₂O (Fig. 5). The same phenomenon was observed in case of electrolyzed seawater (Fig. 9a). The phenomenon of CO₂ bubbling of the electrolyzed seawater reduced the weight of the wet precipitates. This result is

also observed in case of authentic (Fig. 5) and natural seawaters (Fig. 9a). After the CO₂ bubbling of the electrolyzed seawater, the solution was acidified via HCl addition. The CO₂ captured in the electrolyzed seawater rapidly released into the air (Fig. 9b), indicating that CO₂ was released from CaCO₃ as well as from mMgCO₃ Mg(OH)₂ nH₂O. The other precipitates, such as Ca(OH)₂ and MgCO₃, might have converted into CO₂ and metal ions.



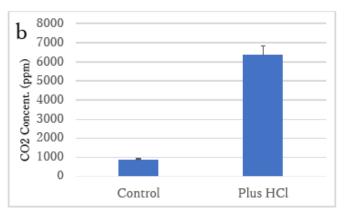


Figure 9: Mg(OH)₂ precipitate and its CO₂ complex in seawater and CO₂ release by HCl addition. Solid NaOH (1.6 g) was dissolved into 200 mL of seawater, and then the weight of wet Mg(OH)₂ precipitate was measured (a). Furthermore, the air was bubbled in the seawater for 3 h, and the wet weight of mMgCO₃· Mg(OH)₂· nH₂O was measured. After bubbling air into the solution, HCl was added to release CO₂.

4. Applications of CO₂ Capturing in Waste Treatment Facility

The proposed CO₂ capturing technology will have potential deployment strategies in industrial and transportation applications and community infrastructures to support reduction of CO₂ hence climate change. The proposed CO₂ capturing technology is shown in Fig. 10, where input and output materials

are shown with benefits of multiple output products. One potential application is the integration within waste treatment facility. The proposed technology could be implemented as a standalone CO₂ capturing unit with multiple products or as integrated with waste-to-energy gasification process where it will clean the syngas by removing CO₂ for improved clean fuel and energy generation, as shown in Fig. 11.

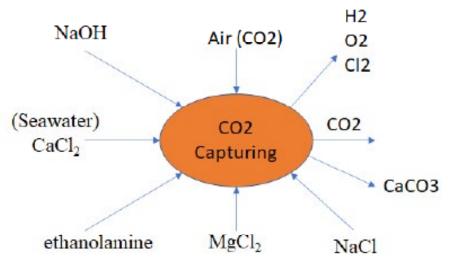


Figure 10: CO2 Capturing Technology Conceptual Design

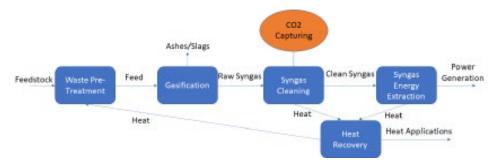


Figure 11: Waste-to-Energy Process Integration with CO₂ Capturing.

5. Discussion

Primitive life is considered to have appeared 3.6 billion years ago on the Earth. After a long biological evolution period, human beings became the most evolved organism based on genomic evolution and the five-fingernail functional cooperation [23]. Organisms developed a respiratory system, including the lung organ, in which CO₂ easily penetrates through tissue membranes, based on CO₂ characteristics. The diffusion force of CO₂ gas through plastic polymer membranes and bio membranes, including cellulose membranes, was considerably greater than that of O₂ or N₂ because of concentration differences [17,24]. Interestingly, the extensive use of fossil fuels, such as coal, oil, and natural gas, by the developing human civilization has induced the present climate change.

Charles Darwin made a statement based on his natural selection theory that "It is not the strongest of the species that survives, nor the most intelligent that survives. Rather, the one that is most adaptable to changes." However, some humans believe in "the law of the jungle" and that humans are ranked at the top among all the organisms on the Earth. Humans are majorly responsible for the climate crisis. Therefore, our moral duty is to address the situation through global cooperation to protect our beautiful blue planet for future generations and other organisms that are not responsible for this climate crisis.

As mentioned above, amine solutions capture CO2 from exhaust fumes [12-15]. Except for their pharmacological effects in vivo, the behaviour of amines is relatively obscure, although our body comprises amino derivatives, including polymers and monomers. The reason that CO2 produced through the glucose metabolism is not captured in vitro and in vivo remains unknown. Industrial high-temperature treatments release the CO₂ captured by amine solutions. This study demonstrated that the neutralization of the amine-CO2 solution could easily release CO2 without thermal treatment. As our body does not accumulate CO2, our body fluids maintain a neutral ph. The CO2 fixation and storage method converted CO2 into CaCO3, limestone, and coral using low concentrations of NaOH and CaCl₂ [16]. This method converts NaHCO3 and Na2CO3, the first compounds formed after the CO2 and NaOH reaction, secondarily to CaCO3 by CaCl2 addition [16-19]. Therefore, this suggests that neutralizing or acidifying the NaHCO3 and Na2CO3 solutions can release pure CO2 without thermal addition.

Space and artificial technology, including computer science,

have considerably developed during the present half-century. These technologies visualized natural phenomena, such as ozone-hole formation, glacier and polar ice reduction, and artificial reduction in rain forests by commercial tree cutting and agricultural land. Moreover, computer simulations can predict future climate changes based on various abovementioned phenomena. However, these modern technologies cannot capture the accumulated CO₂ in air; therefore, climate change remains unresolved. Thus, the most reliable method to practically improve climate change is capturing the accumulated CO₂ in the air. Furthermore, this method must be simple and cost-effective to develop practical plants worldwide.

NaCl solution or seawater electrolysis requires electric power to produce NaOH while producing H₂ and Cl₂. Therefore, the generated H₂ can be used in the fuel battery to supply electric power and as a clean energy source [25,26]. Cl₂ can be used as reaction materials for other chemical compounds. Moreover, the final products are CaCO₃, a component of limestone, and NaCl via Na₂CO₃ and NaHCO₃ solutions. NaCl can be used as a starting material for electrolysis. This fact shows that sequential reactions form a whole cycle. It confirms that the system based on NaCl or seawater electrolysis capturing the accumulated CO₂ in the air can help mitigate climate change. Therefore, this system can achieve the sustainable development goals (SDGs).

Only the method that uses amines has been established to capture CO₂ from the exhaust gases [12-15]. Toxic organic solvents are a major reason why this amine method is not used worldwide. Furthermore, thermal treatment is necessary to release the captured CO2 from the amine-CO2 complexes. This process eventually induces amine degradation, which requires regeneration for further use. However, our developed method is based on the use of NaOH and CaCl2, which are inorganic, water soluble, and heat stable [16]. Both compounds are cost-effective, although a high concentration of NaOH is extremely toxic. In the developed method, however, low NaOH concentrations of <0.2 M should be used to prevent CaCO₃ formation from Ca(OH)₂ formation [16]. Under this condition, only CaCO₃, which is harmless, is produced. Eventually, this method can convert CO2 to naturally stable and harmless CaCO3 limestone without requiring complex and expensive constructions. The fact that a large amount of pure CO2 can be concentrated in the air in which CO₂ concentration is extremely low by our developed CO2 fixation and storage method is crucial for future CO2 usage. Further, CO₂ is a source of other chemical compounds

[27,28]. Conversely, direct air capture (DAC) technologies are challenging to improve in three areas, contactor, sorbent, and regeneration, to drive down the cost [29].

6. Conclusions

The electrolysis of NaCl or seawater produces NaOH, which reacts with CO₂ and yields carbonate compounds with metal ions such as Ca₂+ and Mg₂+. CaCO₃, a limestone component, was produced by adding CaCl₂ to the electrolyzed NaCl solution via CO₂ bubbling. Furthermore, using seawater rather than NaCl solution, CaCO₃ could be produced without adding CaCl₂. Thus, the system for CO₂ fixation based on NaCl or seawater electrolysis fits the SDGs.

Acknowledgments

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Author Contributions

K.S. conceived, designed, and conducted the study and wrote the manuscript.

H.G. analysed, defined energy and process models, validated, and edited manuscript.

Competing Financial Interests

The author declares that the present data have been used to support an application to the Japan

Patent Office (JP2022-199049, JP2023-7321 and PCT/JP2023/021554)

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