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COMMUNICATION

Sustainable Circulating Energy System for Carbon Capture Usage and Storage (CCUS)

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ABSTRACT: Recently, we developed an innovative CO_2 capture and storage method based on simple chemical reactions using NaOH and $CaCl_2$. In this technology, it was newly found that the addition of CO_2 gas produced $CaCO_3$ (limestone) in the solution of NaOH and $CaCl_2$ at less than 0.2 N NaOH, while at more than 0.2 N NaOH, $Ca(OH)_2$ formation occurred merely without CO_2. The present study has been designed to develop an integrated system in which the electrolysis unit is combined with the CO_2 fixation unit. As the electrolysis of NaCl produces simultaneously not only electricity but also H_2 and Cl_2, the produced H_2 could be supplied to the hydrogen generator to produce further electricity, which could be used for the initial NaCl electrolysis for NaOH production. Contrarily, the combination of incinerators with electrolytic generators has already been established to supply electricity, as thermal power plants use coals or wastes. This electricity-providing unit could be replaced with a solar panel plant or with a storage buttery. The present integrated system, consisting of various electricity-providing methods and CO_2 fixation units, is a sustainable circulating energy system and carbon capture, usage, and storage (CCUS) system without environmental concerns. In addition, an unexpected-tremendous amount of the burned wood, which was produced by the big mountain or forest fires, could be disposed of by our integrated CO_2 fixing system with the incinerator without environmental concerns along with both H_2 and $CaCO_3$ productions. Thus, our simple technology must contribute immediately and economically to disaster recovery.

KEYWORDS: CO_2 fixation; CO_2 storage; circular energy system; climate change; limestone ($CaCO_3$); electrolysis; CCUS; SDGs; sustainability

1 Introduction

In our fresh memory, huge hurricanes Helene and Carolina hit the southwest area of the USA, one after another, in 2024, and a tremendously wide area was damaged seriously. In addition, many wildfires broke out in Los Angeles in January 2025. Regarding the weather that induced these wildfires, the World Weather Attribution (WWA) reported that the weather conditions might cause a 35% increase in the probability of wildfire occurrence based on high temperature, dry air, and light rain, compared with that before the Industrial Revolution. In Japan, there were several mountain fires in Iwate, Okayama, and Ehime Prefectures, and their burned areas were 2900, 565, and 442 hectares, respectively, within February 2025. Fortunately, these fires were extinguished by the rain. Contrarily, a tremendous amount of burned wood, as well as a large amount of CO_2 emission during the fire, was produced, although they had grown, followed by capturing a huge amount of CO_2 before the fires.

The Intergovernmental Panel on Climate Change (IPCC) concluded on 9 August 2021, that climate change has been caused by human activities that have produced carbon dioxide (CO_2) since the Industrial Revolution [1]. To reduce atmospheric CO_2 concentrations as a means of mitigating such effects, the so-called Paris Agreement was reached at the United Nations Climate Change Conference (COP20) in 2015. This agreement was based on the requirement to keep the increase in the mean global temperature below 2°C related to the temperature before the Industrial Revolution, preferably less than 1.5°C. China is the world's largest emitter of CO_2, while the President of the People's Republic of China, Xi Jinping, has declared that China will be carbon neutral by 2060. The former president of the USA, Joe Biden, rejoined the Paris Agreement on 20 January 2021, whereas the present US president, Donald Trump, quit the Paris Agreement on 21 January 2025. Incidentally, the USA is the world's second-largest emitter of CO_2. Scientists conclude that this fact will likely lead to dangerous irreversible levels of climate change [2]. People should notice that irreversible climate change cannot be repaired again with money and that we have induced the present climate change since the Industrial Revolution. Even if a carbon-neutral society could be immediately achieved, the accumulated atmospheric CO_2 would not be reduced at present.

CO_2 can be captured from the ambient air or gas via several technologies [3], including absorption [4], adsorption [5–9], and membrane gas separation [5,10]. Absorption with amines is currently the dominant technology. However, amines are organic solvents that chemical reactions must synthesize, and heat treatment is necessary to release CO_2 from the CO_2-amine complexes, resulting in heat decomposition of amines. Eventually, the amine technology is not used worldwide. Contrarily, Membrane and adsorption processes are still in the developmental stages, with the construction of primary pilot plants anticipated in the near future. However, to the best of our knowledge, these methods alone cannot achieve the necessary worldwide reductions in atmospheric CO_2.

On the other hand, we recently developed an innovative method for CO_2 fixation and storage [11]. This method is based on simple chemical reactions involving NaOH and $CaCl_2$. Using low concentrations of these chemicals prevented the formation of $Ca(OH)_2$ in the absence of CO_2 but resulted in $CaCO_3$ formation in the presence of CO_2 bubbling. Additionally, a polyethylene tunnel-based improvement method for CO_2 fixation with NaOH mists was proposed as an "artificial forest" model [12]. In this model, CO_2 penetrates a large polyethylene tunnel and is then converted into $CaCO_3$, which is stable and harmless. Namely, this CO_2-fixing process is likened to photosynthesis, in which CO_2 is converted to carbohydrates in the plant. The present study has been designed to develop a sustainable energy circulating system based on the integration of an energy circulating unit and CO_2 capturing unit, consisting of low-cost based on simple chemical reactions and simple facilities to spread worldwide.

2 Methods

2.1 Chemicals

Reagent grade NaOH and $CaCl_2$ were purchased from Wako Junyaku Kogyo (Tokyo, Japan).

2.2 CO_2 Fixation

The chimney model was prepared by combining two 1-L paper milk boxes, after which air (at approximately 100 cm^3/s) and CO_2 (approximately 10 cm^3/s) were supplied into the lower box. A layer of gauze was placed between the two boxes, and approximately 4 mL of the solution, consisting of 0.05 N NaOH and 0.05 M $CaCl_2$, was sprayed into the middle part of the lower box. The CO_2 concentration (in %) was subsequently determined at the central point of the upper box using an XP-3140 instrument (COSMOS). All experiments were carried out at room temperature of around 25°C.

3 Results

Our previous studies [11] showed that the NaOH mist can efficiently capture CO_2 in the plastic pet bottle, representing the closed system. In experiments using a chimney model (Fig. 1a), when the chimney contained high CO_2 concentrations, the amounts of NaOH and $CaCl_2$ in the solution were insufficient to react with all the CO_2 at a gas flow rate of approximately 110 cm^3/s (Fig. 1b). Thus, the solution could only capture a relatively small amount of the CO_2 in the chimney model.

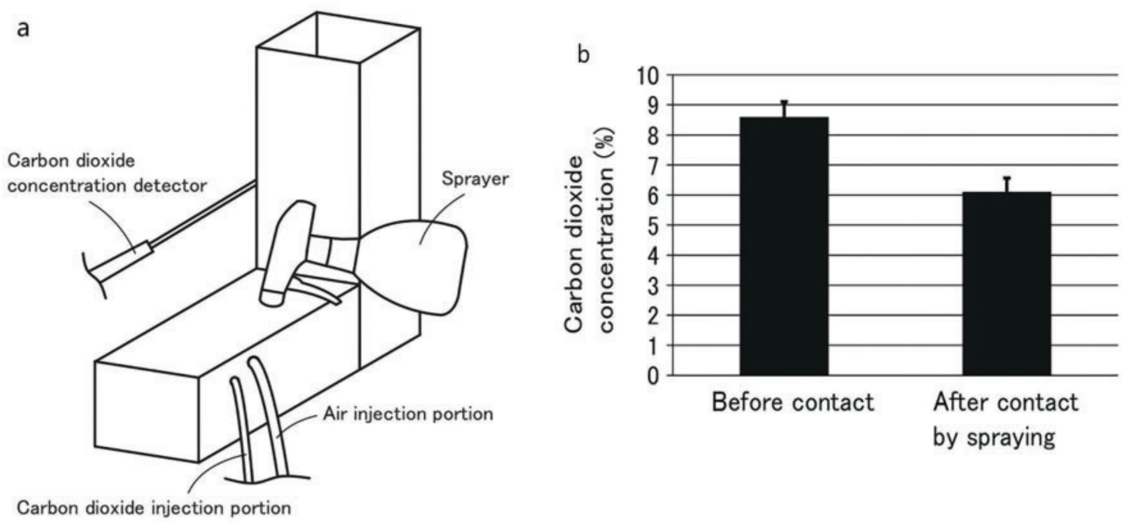

Figure 1: Schematic diagram of chimney model and its data. The data was published in Scientific Reports [12]. Two values are the means plus or minus one standard deviation based on either six or ten replicates

To continuously capture a large amount of CO_2 from the ambient air or exhaust gases, a new CO_2 capture model was developed, as shown in Fig. 2. Using a large chamber equipped with many spray nozzles, CO_2 can be efficiently captured by droplets or mists of the NaOH solution. It is possible to expand the chamber in both directions, such as vertical and horizontal directions, increasing the chamber volume. It is easy to increase the number of nozzles that are equipped with pipelines, expanding three dimensions. Thus, it seems possible to use natural caves, mine galleries, used tunnels, and used buildings, which have a large space for the CO_2 reaction with OH^-, instead of new constructions, when the floor, wall, and ceiling are covered with polymer sheets. This unit could also be combined with the NaOH generating unit, which produces electricity.

This system is applicable to thermal power plants, chemical plants, large ships, combustion operations, incinerators, and automobiles. For example, the incinerator could produce electricity, integrating with the generator, as shown in Fig. 3. The thermal power plant is already established, so it is easy to integrate the thermal power unit with the present CO_2 capturing unit. In addition, as the electrolysis unit can produce HCl, the addition of HCl can convert Na_2CO_3, which is formed from CO_2 and NaOH to pure concentrated CO_2 and NaCl. Similarly, $CaCO_3$ can be converted into CO_2 and $CaCl_2$ by the addition of HCl. The focused CO_2 could be used for further chemical reaction materials. The synthesis of methanol from CO_2 is practically important because methanol is a primary raw material for the production of numerous other chemicals [13]. Methane is produced from CO_2 [14], and hydrocarbons are made from the ambient air by using a photo catalyzer in the presence of H_2O [15–17]. These facts indicate that the presently proposed CO_2-capturing technology is applicable to produce energy from CO_2.

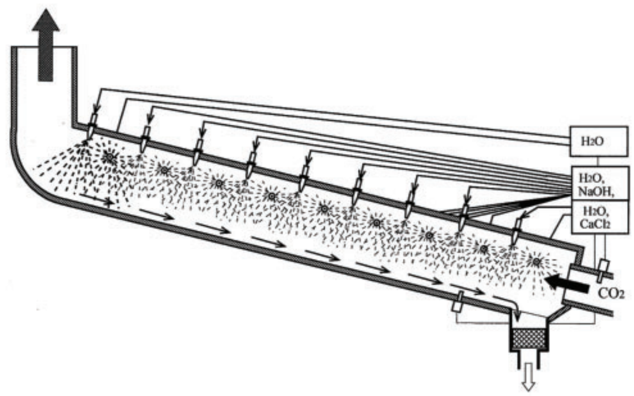

Figure 2: The proposed CO_2 fixation chamber. The original diagram was drawn by the author, and it was formally traced by the Matsushima Patent Office, using the software "Hanako" added in "Ichitaro". The figure was already published in Scientific Reports [12]

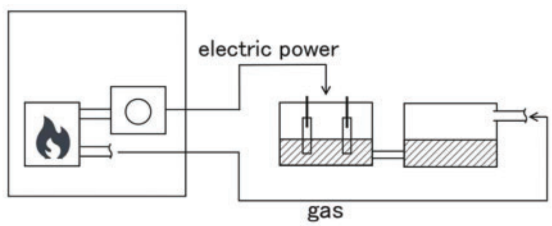

Figure 3: Flow sheet of CO_2 capturing system

Electrolysis of NaCl solution produces H_2 and Cl_2 from the anode and cathode, respectively, as follows:

$2Na^+ + 2Cl^- + 2H_2O \rightarrow H_2\uparrow + Cl_2\uparrow + 2OH^- + 2Na^+$

CO_2 easily reacts with OH^- to produce CO_3^{2-}.

$CO_2 + 2OH^- \rightarrow CO_3^{2-} + H_2O$

CO_3^{2-} reacts with Ca^{2+} to produce $CaCO_3$.

$CO_3^{2-} + Ca^{2+} \rightarrow CaCO_3\downarrow$

The total reaction is:

$H_2O + CO_2 + Ca^{2+}\ 2Cl^- \rightarrow H_2\uparrow + Cl_2\uparrow + CaCO_3\downarrow$

Using the electrolysis of NaCl solution in a CO_2 fixing unit, CO_2 is converted to $CaCO_3$ along with H_2 and Cl_2 productions followed by $CaCl_2$ addition. Namely, the electrolysis of 2 mol NaCl produces 1 mol H_2, 1 mol Cl_2, and 1 mol $CaCO_3$. When low-cost electricity could be provided for the electrolysis, the cost reduction of CO_2 fixation could be obtained along with H_2 production which has high potential as fuels and chemical materials.

The H_2 produced from the electrolysis based on the thermal power plant could be supplied not only to the hydrogen generator, which produces electricity but also to the chemical industry as fuel or starting materials of methane and ammonia [18,19]. Neglecting the energy loss due to the electrolysis and hydrogen generator, the integrated system is a complete energy circulation system. At the same time, the total energy might exceed the energy loss by the thermal power (Fig. 4A). Solar power panels are also capable of CO_2 capturing unit, as shown in Fig. 4A. In this case, the integrated system consists of a CO_2 capturing unit and solar power panel. The system is very simple without a thermal power unit (Fig. 4A). Similarly, not only renewable energy such as hydro power, wind power, geothermal power, and biomass plants but also even nuclear plants, could be applicable. The CO_2 capturing from a thermal power unit is not necessary. In addition, by incorporating the storage battery into the integrated system, the system would be independent not only of weather conditions but also of time (Fig. 4B). This system must be very beneficial for the mass reduction of CO_2 from the atmosphere. In addition, the system could be applied to the brown H_2 production industry from brown coal and H_2O, exhausting a large amount of CO_2 which could be captured, and to the methane fermentation which produces simultaneously methane and CO_2 to remove CO_2.

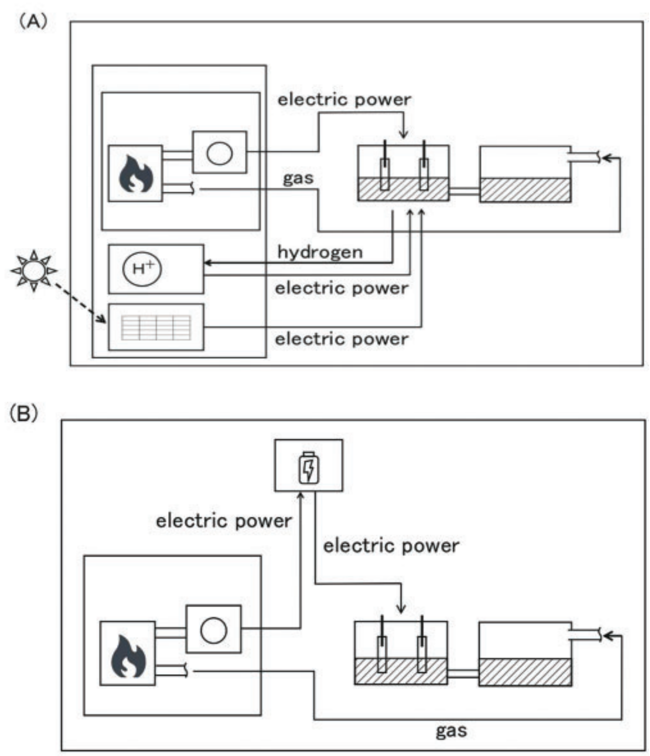

Figure 4: Integrated flow sheets of energy and CO_2 supplying systems. **(A)** The solar panel and hydrogen generator unit are incorporated into the integrated system. **(B)** The battery unit is incorporated into the integrated system

In our proposed CO_2 fixation flow sheets (Figs. 3 and 4), the relationships between the energy-generating unit and the CO_2 fixation unit are shown. Namely, only human beings found that burning fossil fuels, coal, oil, and natural gas, produces energy as well as wood, resulting in the accumulation of CO_2 in the atmosphere. On the other hand, the accumulated CO_2 could be removed by our technology without environmental concerns, as shown in Fig. 5.

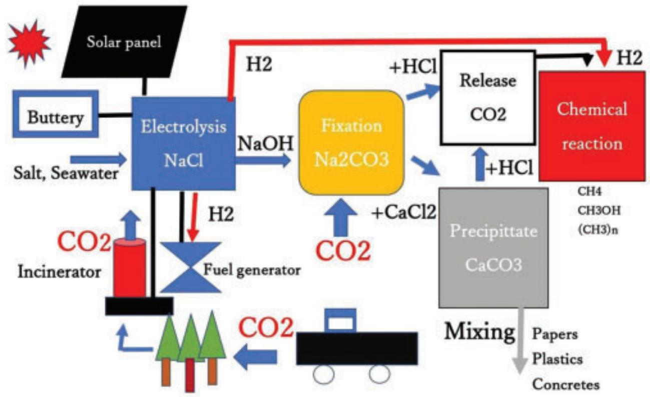

Figure 5: Flow sheet of energy and chemical components

4 Discussion

Plants capture large quantities of CO_2 based on photosynthesis and are distributed on the large parts of the earth, not only on the land surface but also in the shallow sea. However, the planet's large forest, the Amazon, which greatly contributes to the removal of atmospheric CO_2, is continuously shrinking because of commercial development and serious fires. The plant can spontaneously capture CO_2 from the extremely low concentrations of CO_2 in the ambient air and produce wood, leaves, fruits, and crops under sunlight. Eventually, as these natural products consist of organic compounds such as cellulose, lignin, and carbohydrates, they are basically flammable materials that potentially have high energy which was obtained from the concentrated thin sunlight energy.

Recently, we discovered that the high penetrability of CO_2 in the gas phase caused "Pseudo-osmosis" against polymer elasticity not only cellulose membranes but also plastic membranes based on the CO_2 concentration gradient [20]. In this phenomenon, CO_2 can penetrate through the polymer membranes, such as latex and cellulose, while O_2 and N_2 whose penetrability is extremely lower compared with that of CO_2, can't penetrate the membrane. Therefore, when a latex glove partially filled with air is left in a glass bottle filled with high-concentration CO_2 gas, the latex glove expands spontaneously. The CO_2, air, and polymer membranes resemble water, impermeable compounds, and semipermeable membranes, respectively, in the osmosis. We named this discovery "pseudo-osmosis". This characteristic of CO_2 contributes to CO_2 absorption from the air or water by plants as well as their pores.

The dead plants were converted to coals by geothermal heat and pressure deep underground in the Carboniferous period, while the remains of plants and animals were converted to oil in liquid condition and natural gas in the gas condition, respectively. Namely, these fossil fuels are high-energy materials, although they produce a large amount of CO_2 at the time of combustion. Consequently, we have used valuable fossil fuels since the Industrial Revolution because of their easy handiness, whether people living on the earth are wealthy or not. We make a certain amount of waste based on papers and foods in our daily lives, while our integrated CO_2 capturing system is very beneficial for us to dispose of waste without environmental concerns.

Cyanobacteria have been thought that large quantities of CO_2 were produced about 2.7 billion years ago by the photosynthesis of chlorophylls with CO_2 and sunlight in the presence of H_2O, in the shallow ocean, resulting in stromatolite, $CaCO_3$, formation. Even at present, this organism still lives in Shark Bay, Australia. In the food chain, a large amount of phytoplankton breeds under sunlight, and CO_2 exists in the ocean followed by the zooplankton prosperity which leads to animal foods. These processes contribute obviously to CO_2 fixation in the ocean, although the organisms produce CO_2 via their respiration [20]. Of course, organisms including humans release CO_2 via the metabolism of carbohydrates, the TCA cycle, which are produced by plants. Interestingly, we found that not only sodium carbonates, $NaHCO_3$, and Na_2CO_3 [21], but also amines [22] accelerated glucose consumption in cultured cells. These results indicate that the CO_2 balance is reserved in nature but also organisms if it were not for CO_2 production based on using the fossil fuel.

Recently, plastic waste has been shown to be a significant environmental pollutant, and microplastics have been found to affect marine organisms [23]. A small portion of the plastics used daily in human activities is recycled, while the remainder is simply treated as waste. Many of these materials could be incinerated but are typically sent to landfills. However, if the present CO_2 fixing system becomes available, this waste could be readily disposed of by burning without environmental concerns and with the potential to generate energy.

CO_2 also dissolves in the oceans to form H_2CO_3, HCO^{3-} and $CO_3{}^{2-}$, there is approximately 50 times as much carbon dissolved in the oceans in the atmosphere [24]. However, even though a certain amount of Ca^{2+} (0.4%) is dissolved in the ocean, $CaCO_3$ formation does not take place in nature. This fact indicates that this reaction does not take place under natural inorganic conditions without organisms. Indeed, limestone, $CaCO_3$, consists of the fossils of *Fuslinids*, belonging to protozoa in the Paleozoic and Mesozoic eras, several hundred million years ago, and at present, coral reefs and shells are formed by coral and shellfishes, respectively. In our experiments, the bubbling of the atmosphere into the seawater did not produce obvious $CaCO_3$, whereas white precipitates of $CaCO_3$ were formed by the addition of a small amount of NaOH (unpublished data). This experimental fact indicates that limestone and coral reef formations were carried out by living organisms based on biological reactions, not chemical reactions. These living organisms are indeed capturing spontaneously CO_2 from the atmosphere or oceans on large parts of the earth. Therefore, environmental conditions where coal and plants can live continuously must be reserved to prevent climate change.

In our previous study [11], we found that CO_2 is converted into $CaCO_3$ as a result of NaOH and $CaCl_2$. $CaCO_3$ is a main component in limestone or coral and is almost insoluble in water and harmless to organisms. Contrarily, for CO_2 storage, geo-sequestration by injecting CO_2 into underground geological formations, such as oil fields, gas fields, and saline formations, has been suggested [25,26], although these systems are still projects for the future. Considering CO_2 condensation, transportation, and storage technologies, the total CO_2 storage cost might be extremely high, compared with our integrated CO_2 technology.

5 Conclusions

It is impossible not to use fossil fuels, such as coal, oil, and natural gas, in our daily lives because the present acquired civilization is based on the consumption of fossil fuels since the Industrial Revolution. However, it seems possible to preserve the present daily lives using continuously fossil fuels if our proposed sustainable circulating energy system, which does not emit CO_2 into the atmosphere, could be incorporated into our society immediately. Adopting the present system could improve climate change without social confusion.

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Author Contributions: Kenji Sorimachi conceived, designed, and conducted the study and also wrote the manuscript. Toshinori Tsukad discussed the study and supported a partial financial support. Hossam A. Gabbar discussed the present study and revised the manuscript. All authors revied the results and approved the final revision of the manuscript.

Availability of Data and Materials: The authors confirm that the data supporting the findings of this study are available within the article.

Ethics Approval: Not applicable.

Conflicts of Interest: The authors declare no conflicts of interest to report regarding the present study.

Appendix A

• Patents

[US 11,305,228 b2] Inventor Kenji Sorimachi; Assignee Kenji Sorimachi, Assignee Shinko Inc. Ind. Title of the patent; Method for fixing carbon dioxide, method for producing fixed carbon dioxide, and fixed carbon dioxide production apparatus. 2021, 8, 26/2022, 4, 19.

⟦JP 7408125⟧ Inventor Kenji Sorimachi; Assignee Kenji Sorimachi. Title of the patent; The carbon dioxide fixing device. 2022, 3, 28/2022, 4, 5.

The other related patents.

[JP 6739680], [JP 6830564], [JP 6878666], [JP 7433694], [JP 6817485], [JP 6788170], [JP 6788169], [JP 6788162], [JP 7048125], [JP 6864143], [JP 6783436], [JP 6906112], [JP 7221553], [JP 6906111].

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