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A New Recovery Process of Carbon Dioxide from Alkaline Carbonate Solution via Electrodialysis

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DOI 10.1002/aic.11907

Bipolar membrane electrodialysis is applied to CO_2 recovery from alkaline carbonate solution. CO_2 in flue gas is captured by an alkaline hydroxide absorbing solution to form an alkaline carbonate solution. The captured CO_2 is recovered from the alkaline carbonate solution via bipolar membrane electrodialysis, and the alkaline solution is regenerated simultaneously. To reduce the power requirement for CO_2 recovery, this study considers optimal design and operation. Three membrane arrangements were compared, and the results indicate the membrane arrangement comprising a bipolar membrane and cation exchange membrane is the most energy saving. With further optimization of operation conditions, the minimum power requirement for CO_2 recovery was reduced to 2.1 MJ/kg-CO_2 (or 2.1 GJ/t-CO_2). © 2009 American Institute of Chemical Engineers *AIChE J,* 55: 3286–3293, 2009
Keywords: CO_2 capture, bipolar membrane electrodialysis, CCS

Introduction

Carbon dioxide capture and storage (CCS) has been recognized as an effective option to reduce atmospheric emissions of carbon dioxide (CO_2) and mitigate global warming. The CCS scenario is composed of capturing CO_2 from flue gas, liquefaction and transport of the captured CO_2, and injection of the liquid CO_2 into appropriate sites either underground or in the ocean.[1] Capture of CO_2 can be applied to large point sources.[1] The purpose of CO_2 capture is to produce a concentrated stream that can be readily transported to a CO_2 storage site.[1] The CO_2 capturing process is the most cost-intensive among the steps in the CCS scenario.[2] Therefore, cost reduction for CO_2 capture is essential for the effective implementation of CCS. Several kinds of CO_2 capturing methods have been proposed, such as chemical absorption, membrane separation, and pressure swing adsorption.[3–5] Chemical absorption methods using liquid absorbents are considered as the most promising option for CO_2 capture from the viewpoint of cost and capacity.[6–8] The liquid absorption method is composed of two steps, absorption and stripping. In the absorption step, liquid absorbents with a strong affinity to CO_2 such as alkanolamines or potassium carbonate are used to capture CO_2 from the flue gas.[9] In the

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stripping process, the captured CO_2 is recovered by a thermal treatment at about 200°C, and the absorbents are regenerated simultaneously. The thermal treatment requires a large amount of thermal energy, in the range of 3–4 MJ/kg-CO_2 for alkanolamines[9–12] and 3–4 MJ/kg-CO_2 for potassium carbonate.[9] This thermal energy required for recovering CO_2 accounts for a large portion of the total energy requirement for the CO_2 capture methods by chemical absorption, and these energy requirements would be directly reflected and a become major component in the cost for CO_2 capture.[12,13] Therefore, a reduction in the energy requirement for CO_2 recovery from the absorbents is necessary to reduce the cost of CCS.

An alternative method of CO_2 recovery is acidification of the absorbent solution. Since carbonate acid is a relatively weak acid, the reduction of pH in the CO_2 captured solution leads to the release of gaseous CO_2. When protons are provided into the solution of alkaline carbonate, gaseous CO_2 is generated according to the following reactions.

$$CO_3^{2-} + H^+ \rightarrow HCO_3^- \tag{1a}$$

$$HCO_3^- + H^+ \rightarrow H_2CO_3 \tag{1b}$$

$$H_2CO_3 \rightarrow H_2O + CO_2 \tag{1c}$$

To make these reactions proceed, it is necessary to provide protons in the solution and at the same time the alkaline metal ions should also be removed from the solution. Electrodialysis can be used for such an exchange of the alkaline metal ions with protons. Several authors have attempted to apply electrodialysis methods to recover CO_2 from the absorbent solutions.[14–16] However, the power requirement for the CO_2 recovery is reported to be as high as 7 MJ/kg-CO_2.[14–16] This power requirement is equivalent to a primary energy requirement of about 14 MJ/kg-CO_2 (assuming 50% power generation efficiency), which is in the range of or even higher than the total energy available from typical carbonaceous fuels (9–18 MJ/kg-CO_2).[17] Therefore, it is considered that electrodialysis is not competitive with the thermal method in recovering CO_2.

The ideal energy requirement for the recovery of CO_2 from alkaline carbonate solution is about 0.8 MJ/kg-CO_2, which is one order of magnitude lower than the power requirement for the CO_2 recovery with electrodialysis reported in the literatures.[14–16] The difference is due to the electrical resistances across the electrodialysis cells and the reaction in the electrodes. The effects of these irreversible processes could be reduced by optimizing the cell configuration, which includes membrane arrangement and distance between the membranes, or by optimizing the operation conditions such as the current density, number of electrodialysis units between electrodes, and flow patterns of the solution in the cells. In previous studies, such optimization has not been thoroughly investigated even though it might greatly reduce the power requirement. In this study, effects of the cell configurations and operation conditions on the CO_2 recovery process from alkaline metal carbonate solution were investigated experimentally to examine the feasibility of electrodialysis.

Comparison of Three Membrane Arrangements

Membrane arrangements

In previous studies, the unit used for electrodialysis consists of three compartments divided by a bipolar membrane (BP), a cation exchange membrane (C), and an anion exchange membrane (A)[15] as shown in Figure 1a. An electrodialysis system is composed of repeated BP-C-A units with electrodes located at both ends. This membrane arrangement (BP-C-A type) is based on the conventional bipolar membrane electrodialysis system for converting a salt to its acid and base. The feed solution of the alkaline metal carbonate solution is introduced to the feed compartment sandwiched between the cation exchange membrane and the anion exchange membrane. The alkaline regeneration compartment is separated by the cation exchange membrane from the feed compartment while the bipolar membrane is placed on the other side. Lastly, the CO_2 recovery compartment is between the anion exchange membrane and the bipolar membrane placed on the other side. When the electric potential is applied, bicarbonate ions are transported through the anion exchange membrane to the CO_2 recovery compartment, and alkaline metal ions are transported through the cation exchange membrane to the alkaline regeneration compartment. In the CO_2 recovery compartment, protons are provided from the bipolar membrane, and gaseous CO_2 is generated under the acidic condition. In the alkaline regeneration compartment, alkaline solution is regenerated from alkaline metal ions and hydroxyl ions are provided from the bipolar membrane.

However, for the recovery of CO_2 from alkaline carbonate solution with the reaction in Eqs. 1a–1c, it is not necessary to remove both alkaline metal ions and bicarbonate ions from the feed solution because CO_2 can be automatically separated from the solution in the form of gas by reducing pH. In other words, either the CO_2 recovery compartment or alkaline regeneration compartment in the BP-C-A arrangement can be merged with the feed compartment. Thus, two simpler arrangements can be considered, namely the BP-C arrangement and BP-A arrangement. The BP-C arrangement is a configuration in which the anion exchange membrane has been removed from the BP-C-A arrangement to merge the CO_2 recovery compartment and feed compartment as shown in Figure 1b. The alkaline regeneration compartment is separated by the cation exchange membrane from the merged feed compartment. Protons are provided from the bipolar membrane to the feed compartment, and gaseous CO_2 is generated. The alkaline metal ions are removed from the feed compartment through the cation exchange membrane to the alkaline regeneration compartment, where alkaline metal hydroxide solution is regenerated with the hydroxyl ions provided from the bipolar membrane. In the BP-A arrangement shown in Figure 1c, the feed compartment and alkaline regeneration compartment are merged by removing the cation exchange membrane. In this case, bicarbonate ions are transported to the acid compartment from the feed compartment, where gaseous CO_2 is generated with the protons provided from the bipolar membrane.

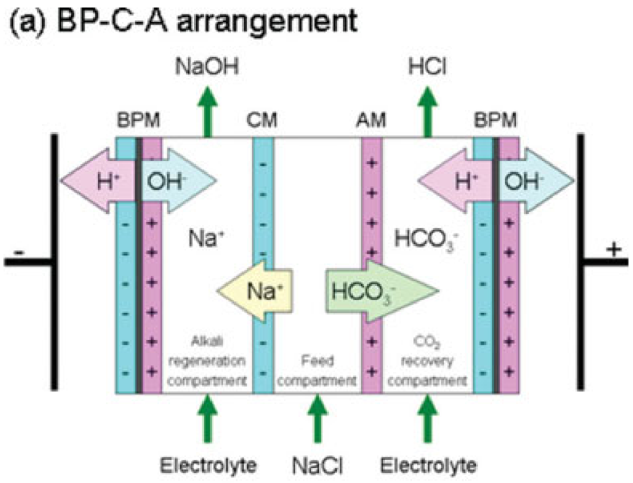
The CO_2 recovery performances for these three membrane arrangements are compared in terms of the CO_2 recovery rate and the power requirement per unit CO_2 recovered. It is expected that the simpler systems will be more advantageous in terms of the power requirement.

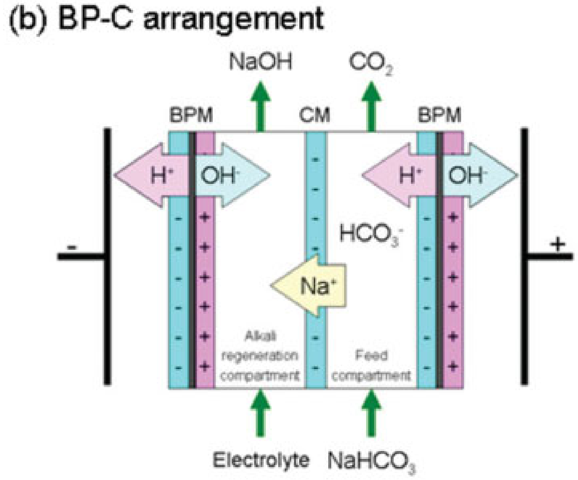
Experimental setup

A schematic diagram of the experimental apparatus is shown in Figure 2. The apparatus is composed of a laboratory-scale electrodialysis system (Asahi Glass, Japan) and a solution circulation system. The electrodialysis system consists of a given number of layered units for electrodialysis, in this case 10 layered units. In this study, only commercially available membranes were used. Neosepta BP-1 (Tokuyama, Japan) was used as a bipolar membrane. Selemion CMV and Selemion AMV (Asahi Glass, Japan) were used as a cation exchange membrane and an anion exchange membrane, respectively. The distance between the membranes was set at 0.75 mm. The effective area of each membrane was 117.5 cm^2.

Sodium bicarbonate solution (0.8 M) was used as a feed solution introduced to the feed compartment. Sodium chloride solution (0.1 M) was employed as an electrolyte solution to be introduced to the CO_2 recovery compartment or alkaline regeneration compartment. Each solution was circulated using magnetic pumps and the flows for each compartment were introduced in parallel. The flow rates of the feed, CO_2 recovery and alkaline regeneration solution were all set at 2.0 L/min. Thus, the experimental system can be recognized as a batch operation system circulating a fixed amount of solutions. Sodium sulfate solution (0.2 M) was used as electrode rinse solution.

The electrodialysis experiments were conducted under a constant current density condition of 17.0 mA/cm^2, and the potential differences across the experimental stack including and excluding electrode cells were measured. The CO_2 recovery rate from the CO_2 recovery compartment was measured with a gas flow meter at the location of the gas-liquid separator, which was installed on the downstream side of the CO_2 recovery compartment. The pH values of the solutions were monitored at the feed tanks. The temperatures of the solutions were kept between 25 and 30°C.


(a) BP-C-A arrangement


(b) BP-C arrangement

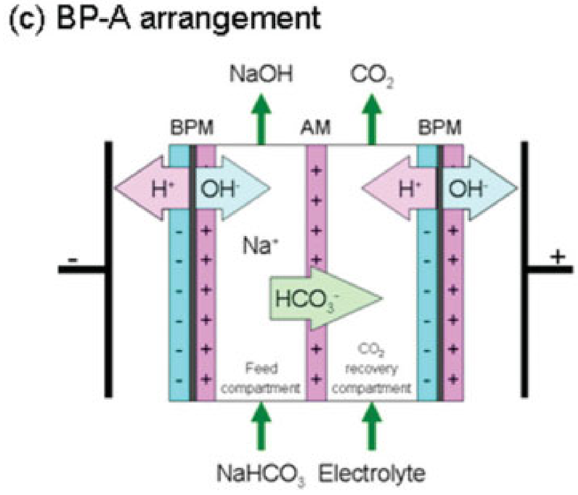

(c) BP-A arrangement

Figure 1. Process description of the CO_2 recovery system from an alkaline carbonate solution.

(a) BP-C-A arrangement, (b) BP-C arrangement, (c) BP-A arrangement. BPM, bipolar membrane; CM, cation exchange membrane; AM, anion exchange membrane. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

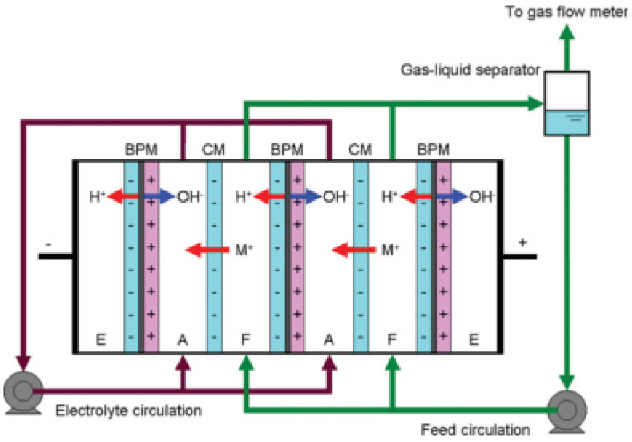


Figure 2. Experimental apparatus of the BP-C arrangement electrodialysis.

BPM, bipolar membrane; CM, cation exchange membrane; E, electrode compartment; A, alkali regeneration compartment; F, Feed compartment. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

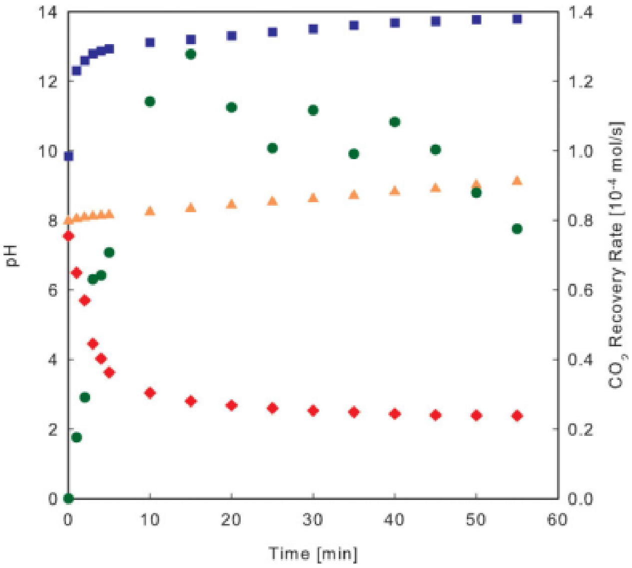


Figure 3. Time variation of pH in each compartment and CO_2 recovery rate of the BP-C-A arrangement.

Alkali regeneration compartment (■), feed compartment (▲), CO_2 recovery compartment (◆) and CO_2 recovery rate (●). Number of electrodialysis units: 10, alkali regeneration solution: 0.2 M NaCl, feed solution: 0.8 M $NaHCO_3$, CO_2 recovery solution: 0.2 M NaCl, current density: 17.0 mA/cm^2. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

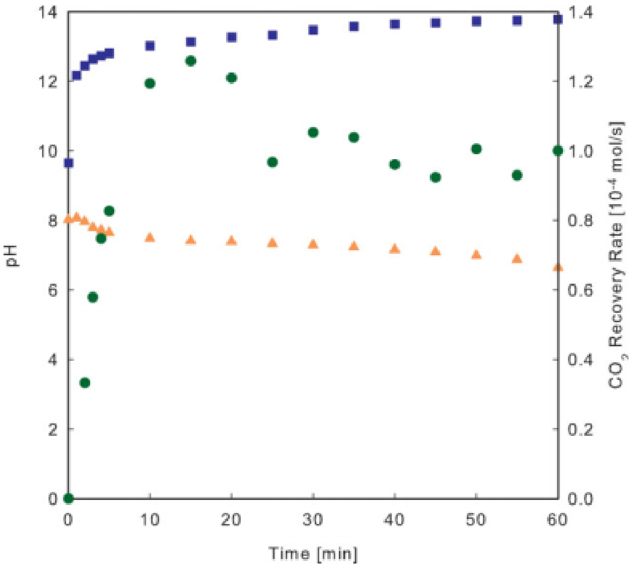


Figure 4. Time variation of pH in each compartment and CO_2 recovery rate of the BP-C arrangement.

Alkali regeneration compartment (■), feed compartment (▲) and CO_2 recovery rate (●). Number of electrodialysis units: 10, alkali regeneration solution: 0.2 M NaCl, feed solution: 0.8 M $NaHCO_3$, current density: 17.0 mA/cm^2. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

Results and discussion

The time variation of pH in each compartment for the BP-C-A arrangement is shown in Figure 3. After applying the electric potential, the pH value in the CO_2 recovery compartment decreased sharply for up to 10 min and then gradually decreased to reach about pH 2.0 at 60 min. The pH value in the alkaline regeneration compartment increased initially for 5 min and then more gradually to reach about pH 14.0 at 60 min. The pH value in the feed compartment increased with time to reach about pH 9.0. In Figure 3, the time variation of the CO_2 recovery rate is also shown. The CO_2 recovery rate was almost constant during the experiment. Thus, CO_2 recovery and alkaline regeneration can be realized for the basic system with the BP-C-A arrangement.

Figure 4 shows the time variations of the pH values and the CO_2 recovery rate for the system with the BP-C arrangement. The time variation of the pH value in the alkaline regeneration compartment is almost equivalent to that observed for the case with the BP-C-A arrangement. The CO_2 recovery rate was also equal to that observed for the BP-C-A arrangement. On the other hand, pH in the feed compartment, which is merged with the CO_2 recovery compartment, decreased with time. Therefore, the BP-C arrangement is almost equivalent to the BP-C-A arrangement in terms of the CO_2 recovery rate and the alkaline regeneration.

Figure 5 shows the time variation of the pH value for each compartment and the CO_2 recovery rate for the BP-A arrangement. The pH value in the CO_2 recovery compartment decreased for the initial 20 min, but became almost constant afterward, and an abrupt increase in the pH value was observed at about 40 min. The pH value in the feed compartment, which is merged with the alkali regeneration compartment, slightly increased for the initial 40 min, and a rapid increase was observed at about 40 min. However, the

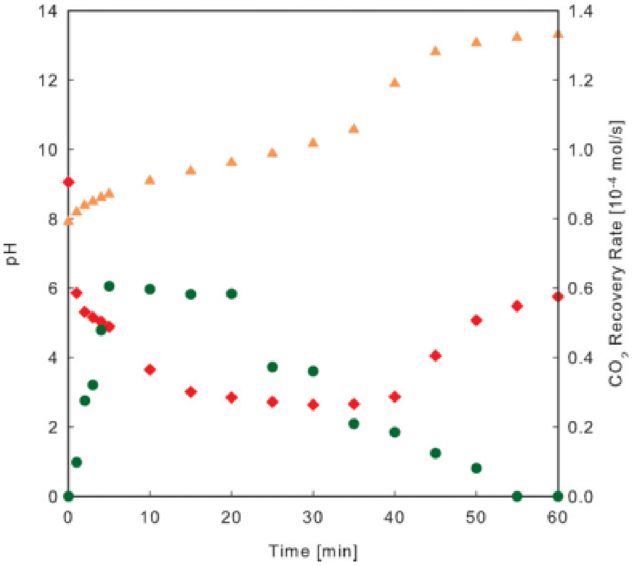


Figure 5. Time variation of pH in each compartment and CO_2 recovery rate of the BP-A arrangement.

Feed compartment (▲), CO_2 recovery compartment (◆) and CO_2 recovery rate (●). Number of electrodialysis units: 10, feed solution: 0.8 M $NaHCO_3$, CO_2 recovery solution: 0.2 M NaCl, current density: 17.0 mA/cm^2. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

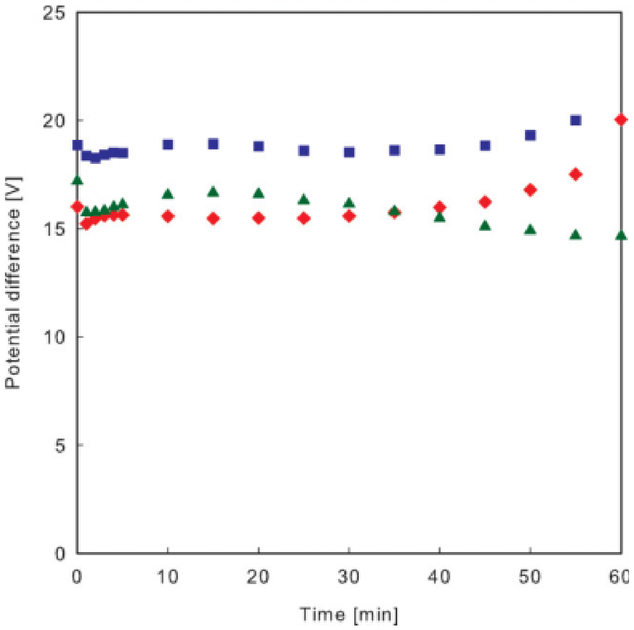


Figure 6. Potential difference across the electrodialysis cell.

BP-C-A arrangement (■), BP-C arrangement (◆), and BP-A arrangement (▲). [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

pH value after 60 min is about pH 13. The CO_2 recovery rate was much lower than in the case of the BP-C-A arrangement, and the CO_2 recovery rate significantly decreased after 40 min. Thus, the CO_2 recovery and alkaline regeneration performances for the BP-A arrangement is lower than the base case of the BP-C-A arrangement or the BP-C arrangement.

The current efficiency for the CO_2 recovery represents the molar ratio of the recovered CO_2 to the number of electric charges passing through the cell per unit time. In an ideal case, all the electric charges should be used for the recovery of CO_2 ($\eta = 1$). The current efficiency for the CO_2 recovery, η, is given by

$$\eta = \frac{r_{CO_2} \times F}{n \times i \times S} \tag{2}$$

where r_{CO_2} (mol/s) is the observed CO_2 recovery rate, F (C/mol) is the Faraday constant, n is the number of unit cell-pairs, i (A/cm^2) is the current density, and S (cm^2) is the effective membrane area. The observed current efficiency for the BP-C arrangement is 40–50%, which is almost to the same as that for the BP-C-A arrangement. However, the current efficiency for the BP-A arrangement was 30%. The most significant factor in reducing the current efficiency is the leak transport of protons through the cation exchange membrane and the leak transport of hydroxyl ions through the anion exchange membrane. As protons and hydroxyl ions have much higher mobility than alkali metal ions and carbonate ions, protons would be preferentially transported even at lower concentration. The leakages of protons into the alkaline compartment would decrease the pH value and the leakages of hydroxyl ions into the CO_2 recovery compartment would increase the pH value; both effects would reduce the CO_2 recovery efficiency. The lower current efficiency of the case with the BP-A arrangement suggests that the leakage of hydroxyl ions from the feed compartment to the CO_2 recovery compartment through the anion exchange membrane is more significant for the efficiency reduction than is the leakage of protons from the feed compartment to the alkaline regeneration compartment through the cation exchange membrane in the present system.

Figure 6 shows the time variation of electric potential differences across the electrodialysis stack including the electrode compartments. The potential immediately increased after applying the constant current, and reached an almost steady value during the experiment. The potential increased with time for the cases with the BP-C-A arrangement and the BP-C arrangement. These increases in potential would be due to the increase in the electrical resistance in the feed compartments caused by the decrease in the ion concentration with time. The smallest potential was required for the case with the BP-C arrangement, and the highest potential was needed for the case with the BP-C-A arrangement. The difference in potentials including and excluding the potentials of the electrode cells was about 2.0–2.5 V in all cases. This indicates that about 20% of the electrical potential is required for the electrolysis of water in both electrode compartments in the present systems.

The power requirement for the CO_2 recovery can be estimated based on the current density and potential difference. All the experiments were carried out under the constant current density condition. The voltage necessary to realize a constant value for the current density would depend on the cell configuration. More specifically, a higher resistance would result in a higher necessary potential across the cells. The difference in the potential would reflect the power requirement for the CO_2 recovery. The power requirement per unit amount of CO_2 recovery, E (J/kg-CO_2), is

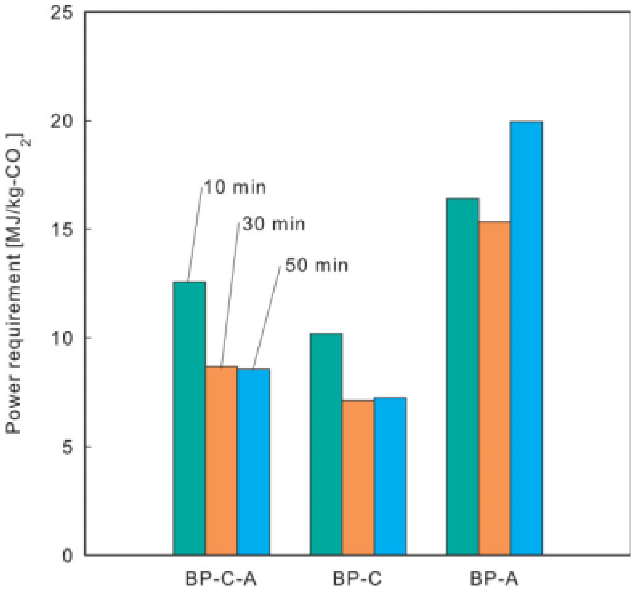


Figure 7. Power requirement per unit amount of CO_2 recovery.

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$$E = \frac{V \times i \times S}{m_{CO_2} \times r_{CO_2}} \tag{3}$$

where V (V) is the potential difference across the stack, i (A/cm^2) is the current density, S (cm^2) is the membrane effective area, m_{CO_2} (kg/mol) is the molecular weight of CO_2, and r_{CO_2} (mol/s) is the amount of CO_2 recovered per unit time. Figure 7 shows the power requirement per unit amount of CO_2 recovery. It is clear that the case with the BP-C arrangement is the most effective for CO_2 recovery in terms of the power requirement per unit amount of CO_2 recovery. The case with the BP-C-A arrangement requires higher power, which is almost equivalent to the value reported in the literature. The difference in the power requirements for the BP-C-A and BP-C arrangements might be due to the lower electric resistance for the BP-C arrangement because of fewer membranes. However, the case with the BP-C-A arrangement would require a higher capital cost because the number of compartments is 1.5 times that of the BP-C arrangement, which may increase the system size as well as the number of membranes. From the aforementioned results, the BP-C arrangement is the most appropriate configuration for CO_2 recovery in terms of the power requirement among the three types of arrangements studied.

Optimization of the BP-C Arrangement

Experimental setting

For the BP-C arrangement, the power requirement for the CO_2 recovery is lower than that reported in the literature using a system based on the BP-C-A arrangement. There would be greater potential for reduction in the power requirement by optimizing the configuration of the electrodialysis stack and the operation conditions. Several factors affecting the CO_2 recovery efficiency can be considered. In this section, effects of the configuration of the electrodialysis stack, specifically the distance between membranes and number of electrodialysis units, and effects of the operation conditions, specifically the current density and concentration of the feed solution, were experimentally examined.

The experiment for the optimization of the BP-C arrangement was carried out using a laboratory-scale electrodialysis system (Asahi Glass, Japan) with a slight modification. The number of electrodialysis units between electrodes was varied between 5 and 10, the effective membrane area was 210 cm^2, and the distance between membranes was varied between 0.75 mm and 1.5 mm. The concentration of the feed solution (sodium bicarbonate) was varied between 0.6 and 1.0 M. The concentration of the electrolyte solution (sodium carbonate) was varied between 0.1 and 0.5 M. Sodium sulfate solution (0.5 M) was used as electrode rinse solution. A set amount (1000 mL) of each solution was circulated in the system driven by magnetic pumps. The current density was varied between 2.7 and 9.7 mA/cm^2.

Results and discussion

Effects of the Configuration of the Electrodialysis Stack. The effect of the number of electrodialysis units was examined for 5–10 units. As shown in Figure 8, the power requirement per unit amount of CO_2 recovery decreased with an increase in the number of units. This is because the contribution of the power requirement for the electrode compartments to the total power requirement is reduced by increasing the number of electrodialysis units.

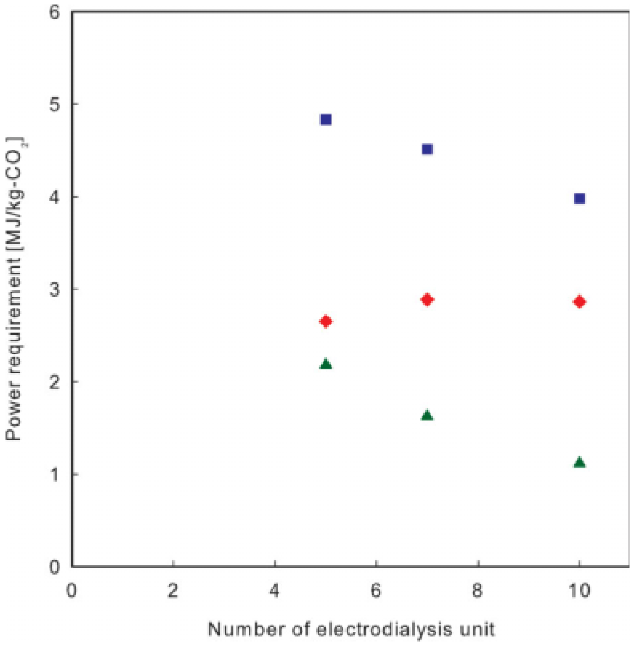

Figure 8. Effect of the number of electrodialysis units on the total power requirement (■), the power requirement for one electrodialysis unit (◆) and power requirement for electrode compartments (▲).

[Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

Also, the effect of distance between membranes was examined for 0.75–1.5 mm. The potential difference slightly increased as the distance increased, which resulted in a higher power requirement. This is because the electric resistance due to the solution increased with an increase in the membrane distance.

Effect of the Operation Conditions. The current density was varied between 2.4 and 9.5 mA/cm^2 (a current between 0.5 and 2.0 A) and other conditions are fixed; feed cell: 1.0 M $NaHCO_3$, base cell: 0.35 M Na_2CO_3. The CO_2 recovery rate increased proportionally to the current density. The current efficiency was in the range of 70–75% in the present case. The factor limiting the current efficiency is the leaching of protons through the cation exchange membrane.

The potential differences, both including and excluding the potentials of the electrode compartments, increased linearly with an increase in the current density. The effect of the current density on the power requirement is shown in Figure 9. The power requirement increased with an increase in the current density. This is due to the increase in the ohmic energy dissipated by electric resistance. The power requirement per unit amount of CO_2 recovery was in the range of 2.1–2.8 MJ/kg-CO_2 excluding the power requirement of the electrode compartments, and 3.0–4.0 MJ/kg-CO_2 including the power requirement of the electrode compartments.

The effect of the initial concentration of carbonate ($NaHCO_3$) in the feed solution on the CO_2 recovery was examined in the range of 0.6–1.0 M. It was found that the CO_2 recovery rate was almost unaffected by the concentration in the range studied. The potential difference slightly decreased as the concentration increased. This is because the electric resistance of carbonate solution decreases with an increase in concentration. As a result, the power requirement per unit CO_2 recovery decreased with an increase in the carbonate concentration.

The effect of the concentration of the electrolyte solution (Na_2CO_3) in the alkaline regeneration compartment on the CO_2 recovery rate was examined in the range of 0.1–0.4 M. No remarkable effect on the CO_2 recovery rate was observed. However, the potential difference slightly decreased as the concentration increased. This is also due to the decrease in the electric resistance with an increase of the concentration. The power requirement per unit CO_2 recovery decreased with an increase in the concentration of the electrolyte solution as well.

The effects of the initial concentrations of the feed solution and the electrolyte solution might be more significant when the electrodialysis operations are conducted under more severe conditions, such as high current densities for which the electric resistances of the solutions affect the total performance.

Process Evaluation. The power requirement for the CO_2 recovery could be reduced to 2.1 MJ/kg-CO_2 (or 2.1 GJ/t-CO_2) for the case of the BP-C arrangement with the operating condition presented in Table 1. This power should be supplied in the form of electricity. Assuming the power generation efficiency of a thermal power plant at 50%, the thermal energy required for generating this amount of electricity is 4.1 MJ/kg-CO_2. This value is highly competitive with the thermal energy required for the thermal recovery methods of CO_2.

Table 1. The Operating Condition for Minimum Power Requirement

CO_2 recovery compartment	1.0 M $NaHCO_3$
Alkaline regeneration compartment	0.35 M Na_2CO_3
Electrode rinse solution	0.5 M Na_2SO_4
Current density	2.4 mA/cm^2

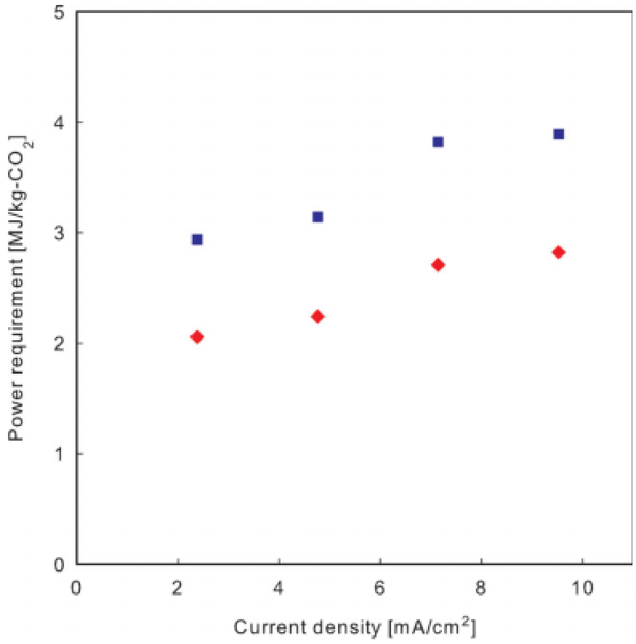

Figure 9. Effect of the current density on the power requirement including electrode compartments (■) and excluding electrode compartments (◆).

[Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

The power requirement could be further reduced by reducing the electricity required by the system by decreasing the permeation resistance of alkaline metal ions through the cation exchange membrane. In the present study, the minimum power requirement is achieved for a current density of 2.4 mA/cm^2, which was the lowest current density but the CO_2 recovery rate was also the lowest among the conditions studied. Although the lower current density is favorable to reduce the power requirement, the CO_2 recovery rate is low. The recovery rate would increase with an increase in the size of the electrodialysis cell, which may lead to a subsequent increase in equipment costs. Such a trade-off relationship between the current density and efficiency should be carefully considered in the design of the recovery process. A more detailed study on the optimization will be reported in the future.

Conclusions

In this study, three membrane arrangements for CO_2 recovery process from alkaline metal carbonate solution by using bipolar membrane electrodialysis were compared. The results indicate that the BP-C arrangement is the most appropriate configuration for CO_2 recovery in terms of the power requirement among the three arrangements studied. Further experimentation was carried out for the case of the BP-C arrangement to optimize operation conditions and reduce the power requirement for CO_2 recovery. Subsequent results showed that the minimum power requirement for CO_2 recovery was reduced to 2.1 MJ/kg-CO_2 (or 2.1 GJ/t-CO_2).

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Manuscript received July 4, 2008, and revision received Feb. 21, 2009.