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Carbon dioxide recovery from carbonate solutions using bipolar membrane electrodialysis

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ABSTRACT

Process design and cost estimation were conducted for CO₂ recovery by liquid absorption with alkaline solution, coupled with bipolar membrane electrodialysis for CO₂ gas separation and alkaline solution regeneration. The electrodialysis performances and power consumption for CO₂ recovery and alkaline solution regeneration were examined using laboratory-scale electrodialysis equipment under various conditions; degree of CO₂ absorption, CO₂ recovery ratio, alkaline concentration, and type of cation exchange membrane. The total cost of CO₂ recovery was estimated for the treatment of CO₂ emitted from a 400 MW coal-fired thermal power plant. The minimum cost for the optimum condition was about US\$180 per ton-CO₂ removed, which is higher than that for the conventional process using amine absorption and thermal desorption. However, a sensitivity analysis indicates that the overall cost could be significantly reduced if the cost of membranes was reduced.

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1. Introduction

Carbon capture and storage (CCS) has been widely recognized as an effective countermeasure to greenhouse gas emissions that mitigates global warming [1,2]. In the CCS process, carbon dioxide emitted from point sources would be captured and transported to storage sites, where liquid CO₂ would be injected into either underground or the ocean waters. The main concern regarding CCS implementation is the need to reduce cost and power consumption for capturing CO₂ from flue gas. More than 80% of the total power consumption and cost for CCS can be attributed to those for CO₂ capture process. There are several methods for capturing and recovering CO2 from point sources, e.g., chemical absorption [3-8], physical adsorption including pressure swing adsorption (PSA) [9,10] and temperature swing adsorption (TSA) [11,12], membrane separation [13–17]. Among these methods, chemical absorption method using amines has been considered to be the most promising method from the viewpoints of both cost and energy penalty [1]. Aqueous solutions of mono ethanol amine (MEA) and its similar substances are used for capturing CO_2 in the flue gas emitted from point sources. These amines form CO_2 -amine compounds at the lower temperature conditions. It is necessary to increase the temperature of the solution to recover CO_2 . The heating process would require a lot of heat; almost all the power consumption for the CO_2 capture can be attributed to this regeneration process, which in turn increases the cost for CCS [1].

Electrochemical methods have been proposed for recovery of CO_2 from chemically absorbed solutions [18–20]. Electrodialysis using bipolar membranes is utilized for these recovering methods of CO_2 from solutions. In a previous paper [18], we proposed a method for recovering CO_2 from carbonate solutions and demonstrated its recovery performances from sodium carbonate with a laboratory-scale experimental apparatus.

The concept behind the method is as follows. First, CO₂ in the flue gas is absorbed in an alkaline solution such as NaOH.

$$NaOH_{(aq)} + CO_{2(g)} \rightarrow HCO_{3(qq)}^{-} + Na_{(qq)}^{+}$$
 (1)

The absorbed CO_2 exists mainly in the form of bicarbonate ions, HCO_3^- , in the solution, depending on the amount absorbed.

The solution is then fed into electrodialysis cells, where the carbonate ions react with protons and are converted back to CO₂ gas. Fig. 1 shows a simple cell configuration for the electro-

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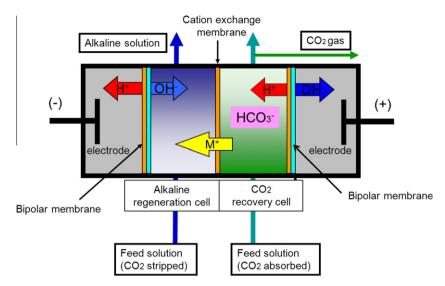


Fig. 1. Configuration of electrodialysis cells for CO₂ recovery from carbonate solution.

dialysis process, namely the two-cell configuration [18]. In this configuration, the two types of cells, the CO2 recovery cell and the alkali regeneration cell are alternately stacked to form the electrodialysis system. Both cells are sandwiched by a bipolar membrane and a cation exchange membrane. Cation exchange membranes (CEMs) are polymeric membranes containing chemical groups with fixed negative charge and mobile cations as a counter ion. Cations can permeate through the CEM, but anions are repelled by the fixed negative charges in the CEM, and cannot permeate through the CEM. The bipolar membrane (BPM) is a membrane composed of two types of ion-exchange membranes, a cation exchange membrane and an anion exchange membrane stacked together. Anion exchange membranes (AEMs) are the membranes with fixed positive charges in the membrane by the chemical groups connected to the main polymer chains of the membrane, and mobile anions are included in the membrane as a counter ion. When applied a voltage across the BPM that is higher than the voltage necessary for water splitting, the water contained in the BPM would dissociate into a proton and a hydroxyl ion. The proton can permeate through the CEM constituting the BPM, while the hydroxyl ion can permeate through the AEM constituting the BPM. As a result, protons can be obtained at the lower potential side of the BPM and hydroxyl ions can be obtained at the higher potential side of the BPM. In the two-cell type configuration of BPM and CEM, the BPM is located at the higher potential side of the CO₂ recovery cell and the CEM is located at the lower potential side. The membranes in the alkali regeneration cell are configured conversely. Other kinds of the membrane configuration can be considered. It was demonstrated that the two cell type configuration with BPM and CEM is most efficient for the CO2 recovery from sodium carbonate solutions.

Alkaline carbonate solution is supplied to the CO_2 recovery cell, where protons are supplied from the bipolar membrane and react with bicarbonate ions to form CO_2 gas.

$$HCO_{3(aq)}^{-} + H_{(aq)}^{+} \rightarrow H_{2}O + CO_{2(g)} \uparrow \eqno(2)$$

The alkaline metal ions in the CO_2 recovery cell are transported to the alkali regeneration cell through the cation exchange membrane, driven by the potential slope. Hydroxide ions are transferred from the bipolar membrane to the alkali regeneration cells and the alkaline solution is thus regenerated by the alkaline ions transferred from the CO_2 recovery cell.

$$Na^+ + OH^- \rightarrow NaOH$$
 (3)

The protons that reduce the pH in the absorption solution can be supplied by the water splitting reaction in the bipolar membrane. At the same time, hydroxyl ions formed by water splitting regenerate the alkaline solution for CO₂ absorption. Because CO₂ gas recovery and alkaline regeneration should be performed separately, it is necessary to design the cell configuration for efficient electrodialysis.

In our previous study, it was demonstrated that gaseous CO_2 can be recovered immediately after the CO_2 recovery cell but this was examined under limited conditions, using laboratory scale experimental apparatus. To examine process feasibility, it is necessary to examine the CO_2 recovery performance and power consumption in the process under a range of conditions. It is also necessary to consider the process of CO_2 absorption from flue gas by alkaline solution. This study aimed to evaluate the total cost of the process, based on the experimental results, and to identify the key parameters involved in reducing CO_2 recovery costs.

2. Outline of the CO₂ absorption and recovery process

A flow diagram for the overall process is shown in Fig. 2. The process comprises the following three steps:

- (1) CO₂ absorption in an absorption tower: CO₂ from the flue gas stream is passed through an alkaline solution such as sodium hydroxide in an absorption tower to form carbonate solution.
- (2) CO₂ recovery by electrodialysis: the carbonate solution is fed to the CO₂ recovery cells of the electrodialysis system, where gaseous CO₂ is recovered.
- (3) Alkaline regeneration by electrodialysis: the carbonate solution, after releasing CO₂, is fed into the alkali regeneration cells in the electrodialysis system, where alkaline solution is regenerated. The regenerated alkaline solution is then reused for CO₂ capture in the absorption tower.

Overall, the net inlet flow to the system is the flue gas, and the net outlet flow from the system is CO₂ and the off-gas. The performance of the above process would depend on various operating parameters. Fig. 3 shows a diagram of the process operation for the present system, as a function of the concentrations of sodium ions and total carbonate. Point A in Fig. 3 represents the initial con-

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