



Carbon dioxide utilization using a pretreated brine solution at normal temperature and pressure



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HIGHLIGHTS

- This research deals with carbonate salts formation using pretreated brine solutions.
- Precipitated calcium carbonate salts was formed in solutions.
- XRD measurements were performed and SEM images were obtained to verify products.
- Precipitated calcium carbonate was proven to be in three different crystal structures.
- With this research, economic feasibility and wastewater treatment can be achieved at the same time.

ARTICLE INFO

Article history:

Received 15 June 2015

Received in revised form 15 August 2015

Accepted 8 September 2015

Available online 25 September 2015

Keywords:

Carbon capture and utilization (CCU)

Industrial wastewater

Metal cations

Precipitated calcium carbonate (PCC)

Brine solution

ABSTRACT

Herein, we discuss carbon capture and utilization processes using industrial wastewater. 5, 10, and 30 wt % aqueous monoethanolamine (MEA) solutions were used as the absorbent solution. A pretreated brine solution, in which contained calcium ion with concentration of 21972.64 ppm, was used to supply calcium cations. All of the experiments were done under ambient pressure and in a temperature range between 298.15 K and 343.15 K. 0.79, 0.34 and 0.19 mol of CO₂ was captured by 5, 10 and 30 wt% of aqueous MEA solutions, respectively. When 100 ml of pretreated brine solutions were added to each saturated MEA solution, 24.65%, 36.96%, and 41.65% (mol basis) of captured CO₂ was converted to calcium carbonate salts by carbon fixation process. To investigate possibility of absorbent recycle after precipitation step, second absorption experiments were performed. As a result 0.55, 0.27, and 0.16 mol of CO₂ were captured by 5, 10, and 30 wt% MEA solution, respectively. Through X-ray diffraction (XRD) analysis and scanning electron microscope (SEM) analysis, it was proven that the calcium carbonate formation was possible and the majority of calcium carbonate was in the form of calcite.

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1. Introduction

Many of the nations in the world are concerned about climate change caused by massive emissions of greenhouse gases, especially for carbon dioxide. In order to achieve the reduction goals for carbon dioxide, many developed nations have conducted related research and have applied carbon reduction technologies known as carbon capture and storage (CCS). In this processes, as shown in Fig. 1, carbon dioxide gas emitted from processes are captured by liquid absorbents in an absorber. Those absorbents become saturated by carbon dioxide gas and are transported to a desorber where they are separated at a relatively high temperature. This separated or desorbed carbon dioxide gas is compressed at high pressure and the CO₂-lean liquid absorbent is returned to

the absorber to form a continuous process. Then, the compressed carbon dioxide gas is transported and disposed of deep in the ocean or underground. However, after carbon dioxide gas is disposed of deep in the ocean, it is dissolved and produces proton (H⁺ ion), carbonate, and bicarbonate ions by reacting with water. The production of H⁺ ions can acidify peripheral ocean ecosystems. Additionally, when captured carbon dioxide is stored underground, there are problems related to CO₂ leaking or sudden explosions. In order to prevent these types of problems caused by conventional storage, a new method should be utilized in the carbon capture technologies. One such method is to convert captured carbon dioxide into a solid state, such as a metal carbonate salt. There are two representative ways in the carbon fixation process: direct and aqueous carbon fixation methods. In direct carbon fixation processes, gaseous carbon dioxide reacts directly with calcium oxide in the solid state, as shown in Eq. (1)



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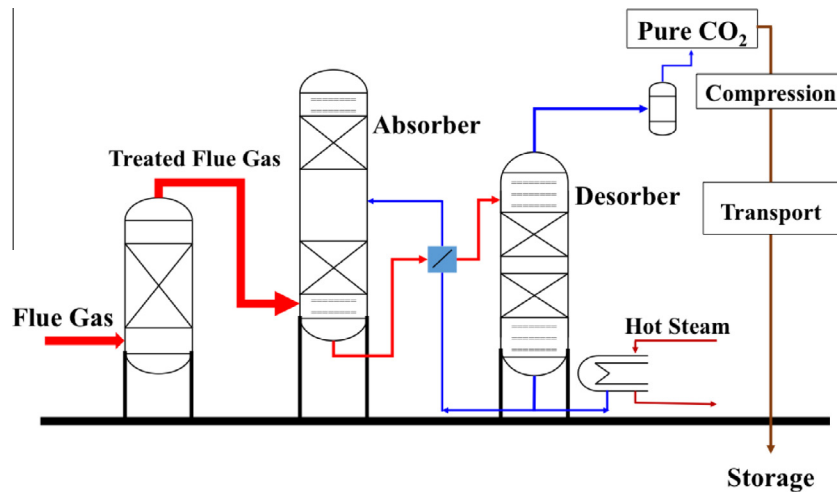
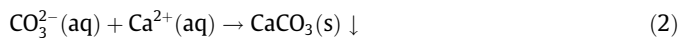


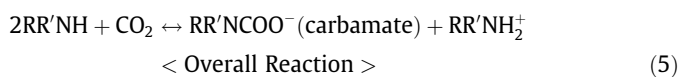
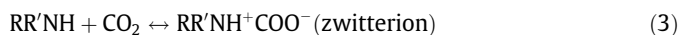
Fig. 1. Carbon capture and storage process for the post-combustion method.

However, large amounts of heat and pressure are required for this process. This means that carbon capture systems using direct carbonation consume large amounts of energy. A large amount of energy is required because that activation energy of this reaction is high due to the low reactivity between the solid and gas phases [1]. It has been reported that an activation energy of 78 kJ/mol is required to ignite the direct reaction between gaseous carbon dioxide and calcium oxide in the solid phase [2]. The other carbon fixation method utilizes an aqueous approach, which is observed in nature (especially in the ocean), as shown in Eq. (2)

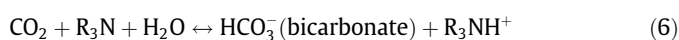


When carbon dioxide is in an ionic state, such as carbonate (CO_3^{2-}) or bicarbonate (HCO_3^-), it can react with calcium ions and precipitate to form calcium carbonate (PCC) at normal conditions, that is, near room temperature (303.15 K) and atmospheric pressure without the need for further energy. This reaction naturally occurs in the bodies of many animal species (e.g., crustaceans and coral). This aqueous approach seems to have great potential for application in the development of carbon capture systems. Also, unlike conventional CCS technologies, the final product can be reused or utilized. This type of technology is referred to as a carbon capture and utilization (CCU) technology. In this research, the aqueous carbonation process found in nature was modified and used to develop our proposed CCU process. Instead of using water as the carbon dioxide absorbent, alkanolamine solutions are used as the carbon dioxide absorbents because they react with carbon dioxide very quickly and convert it into ionic carbon dioxides such as carbonate, bicarbonate, and carbamate, as shown in Eqs. (3)–(6) [3–5].

1.1. For primary and secondary amines (R' for H in primary amine)



1.2. For ternary amines



Since monoethanolamine solutions are the most commonly used absorbents in commercial carbon capture and storage plants [6–7], aqueous monoethanolamine solutions of 5, 10, and 30 wt% were used in this research. There are many sources of calcium that could be utilized [8–9]. Since industrial solid waste such as waste cement or concrete. Since major component of them are calcium, calcium ions can be extracted using acid based solvents [10]. However, wastewater came after their extraction should be treated and the extraction yields are not that high. Also, when acids are used as extraction solvents, calcium ions which is extracted from waste will be supplied to the carbonation reactor with remaining acid solution [11]. This can make the performances of amine based absorbent solutions lower. Hence, pretreated brine solutions were used in this research due to their ready availability and the lack of concern that the brine might be depleted. Also, there are no concern for the pretreated brine solution to lower the absorbent performances. And pretreated brine solution produced after making salt shows high concentrations of metal ions, this can affect marine environment when discharged without treatment. So we can achieve both wastewater treatment and calcium supplying by using pretreated brine solution. To investigate the properties of the precipitated calcium carbonate (PCC) salt, X-ray diffraction (XRD), scanning electron microscope (SEM) were performed. The brine pretreatment method and details of these experiments will be dealt with in the next section.

2. Experimental section

2.1. Materials

Monoethanolamine (purity > 0.99 mass fraction, CAS 141-43-5) was purchased from Alfa-Aesar and was used as-received without further purification. A pretreated brine solution was obtained from Hanju Corporation (35 Sapyeong-ro, Nam-gu, Ulsan, Republic of Korea). Schematics of the brine pretreatment method and the concentrations of the dissolved metal ions are shown in Fig. 2 and Table 1, respectively. For the experiments, 30, 10 and 5 wt% aqueous MEA solutions were prepared using the purchased reagent and deionized water. The concentrations of the solutions were set as described above to demonstrate the difference between conventional CCS processes (where no precipitation occurs) and CCU process (where some/most of the carbon dioxide is precipitated). To make the precipitated calcium carbonate salts, the pretreated brine solution without further treatment was used. To filter the precipitated salts, glass fiber filter paper, manufactured by CHMLAB

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