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Promoter characteristic study on the K_2CO_3 absorbents for CO_2 capture: Mass transfer according to functional group and chain length of promoter

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Abstract

The studies on the characteristics of absorbents are important to develop process and evaluate performance. Among the absorbent studies, the overall mass transfer coefficient was used as a key data for design packing type and absorber height to achieve cost effective process.

In this study, the overall mass transfer rates of the K_2CO_3 based absorbents were measured by a wetted-wall column (WWC) apparatus. Experimental conditions of temperature and pressure were maintained at 313.15 K ~ 333.15 K and 1.15 bar, respectively. The injected CO_2 concentrations were 3 vol.% ~ 12 vol.% (N_2 balanced). As a result of experiment, the mass transfer rates increased following the order: MEA > K_2CO_3+CL-4 > K_2CO_3+CL-2 > K_2CO_3+CL-3 > K_2CO_3+CL-1 > K_2CO_3+FG-1 > K_2CO_3+FG-2 > K_2CO_3+FG-3 at 333.15 K. In case of promoters containing hydroxyl functional groups, the equilibrium partial pressure of CO_2 increased and the slope of the flux decreased. The effect was greater than those of promoters containing increased chain length. The mass transport rate of K_2CO_3+CL-4 solution was similar to that of the commercial MEA solution.

Keywords: mass transfer; carbon dioxide; potassium carbonate; wetted wall column; post combustion capture

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1. Introduction

The post-combustion capture has been widely applied for CO_2 reduction in the coal fired power plant, steel, oil refining, etc [1,2]. The CO_2 capture process using K_2CO_3 absorbent has many advantages such as low cost of raw material, low toxicity, and easy regeneration. However, there are some limitations for applying the process to other industries required operation conditions with lower temperature and low CO_2 partial pressure: slow reaction rate, precipitation of the absorbent [3,4]. The hot potassium carbonate (Benfield) process was developed for the synthesis gas purification. The process was designed for the synthesis gas conditions (high CO_2 partial pressure and high temperature) with the objective of reducing cost. The further developed process using an amine activator (diethanolamine) had the results of lower capital and operating costs and higher gas purity. The commercial activated hot potassium carbonate (AHPC) processes, Exxon's Flexsorb hot potassium carbonate (HP) process and Giammarco-Vetrocoke process were used hindered amines and organic activators, respectively [5]. Recently, Benfield process is being reviewed for application of carbon capture and storage (CCS) besides synthesis gas purification. Generally, the Benfield processes are commercially offered for applications at a minimum CO_2 partial pressure of 210 to 345 kPa and an optimum operating pressure of 700 kPa [6]. AHPC processes are still required to operate at high temperature and pressure resulting high

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operating cost [7]. Devries (2014) studied for various amine promoters under conditions of typical HP process. These amine promoters increased the absorption rates into the concentrated $K_2CO_3/KHCO_3$ solution by 3.5 to 50 times at 70 °C [4]. Cullinane and Rochelle (2005) studied thermodynamics between the promoted K_2CO_3 aqueous solution and CO_2 at 313 K~ 383 K [8]. Furthermore, Cullinane and Rochelle (2006) showed the absorption rate of concentrated K^+/piperazine (PZ) mixtures was 3 times faster than that of 30 wt% MEA [9]. Kim et al. (2012) presented the CO_2 equilibrium partial pressure and the absorption rate using a vapor-liquid equilibrium equipment [10]. As a result of the experiment, absorption rate of K_2CO_3 aqueous solution was improved by a promoter (ethylenediamine). Recently, an economical process, KIERSOL™, has been developed by Korea Institute of Energy Research (KIER) using high efficient K_2CO_3 based absorbents for application in the large point source of CO_2 such as cement and steel industries, coal fired power plant, and bio-methane production. Most of the previous studies focused on the carbon dioxide absorption rate of promoted K_2CO_3 aqueous solution. However, studies have not been sufficiently conducted to design process and optimize operating conditions. Further kinetic studies are necessary to understand characteristics of the absorbent and process. The mass transfer coefficient is generally used as a key data for design of packing and absorber height to achieve cost effective process [11].

In this study, mass transfer of the promoted K_2CO_3 aqueous solutions were investigated. The functional group and chain length were considered selecting promoters to K_2CO_3 aqueous solutions. The most conventional absorbent, monoethanolamine (MEA) aqueous solution, was used for comparison.

Nomenclature

P_{CO2}^*	equilibrium partial pressure of CO_2, kPa
T	absolute temperature (K)
R	universal gas constant (8.314 J/mol•K)
K_G	overall mass transfer coefficient (mol/cm^2•sec•Pa)
g	gravitational acceleration (9.80665 m/s^2)
d	diameter of the tube (cm)
π	pi
ρ	density of liquid (g/cm^3)
μ	viscosity of liquid (cP)
ν	volumetric liquid flow-rate
h	column height (cm)
us	speed of the column surface
n	mole (g/mol)
Flux	rate of movement of molecules across a unit area (mol/cm^2•sec)

2. Reaction mechanism

2.1. K_2CO_3

The overall chemical reaction between the K_2CO_3 aqueous solution and CO_2 is as follows:

$$K_2CO_3 + H_2O + CO_2 \longleftrightarrow 2KHCO_3 \quad (1)$$

The chemical species formed by eq. (1) exists as bicarbonate/carbonate (HCO_3^- /CO_3^{2-}) ions in the aqueous solution. The species distribution between bicarbonate and carbonate is changed with pH of the solution. The reactions can be explained by the eqs. (2) ~ (5). The principal mechanism is a direct reaction of dissolved CO_2 with OH^- at pH > 10. At pH < 8, hydration of dissolved CO_2 is predominant [12]:

At base condition of pH > 10;

$$CO_2 + OH^- \longleftrightarrow HCO_3^- \quad \text{(fast reaction rate)} \quad (2)$$

$$HCO_3^- + OH^- + \longleftrightarrow CO_3^{2-} + H_2O \quad \text{(instantaneous)} \quad (3)$$

At acidic and neutral condition of pH < 8;

$$CO_2 + H_2O \longleftrightarrow H_2CO_3 \quad \text{(slow reaction rate)} \quad (4)$$