



A mass transfer model of absorption of carbon dioxide in a bubble column reactor by using magnesium hydroxide slurry



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ABSTRACT

A bubble column reactor was tested and evaluated for carbon dioxide removal from flue gases by using magnesium hydroxide slurry. The study showed that a high CO₂ removal efficiency could be achieved. An absorption model was developed for system analysis and CO₂ removal data analysis. The overall mass transfer coefficients for CO₂-H₂O and CO₂-Mg(OH)₂-H₂O systems have been calculated. The chemistry of the system, temperature effects, and changes of equilibrium constants also have been discussed.

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1. Introduction

The impact of CO₂ emissions on climate change has been widely discussed and increasing public awareness. According to the U.S. Energy Information Administration (EIA)'s annual report, fossil sources still account for 80% of today's world energy demand, which contributes to a large quantity of CO₂ emissions (EIA, 2011). Carbon capture and sequestration (CCS) is regarded as one of the approaches to manage the levels of anthropogenic CO₂ emitted into the atmosphere. Post combustion absorption technologies represent one of the most commercially ready technologies (Wang et al., 2011).

Solvent selection is the critical consideration in post-combustion absorption, and many research efforts have been made in the study areas of solvents improvement (Gonzalez-Salazar et al., 2012), process simulation (Mathias et al., 2010), modeling and optimization (Lawal et al., 2012), reaction kinetics in the process of absorption and desorption (Mores et al., 2012). Monoethanolamine (MEA), ammonia, potassium carbonate (K₂CO₃) and ionic liquid are the most widely studied scrubbing solvents. MEA is one of the earliest solvents that have been studied for CO₂ removal originally in the field of purification of natural gas, the major concern of which includes corrosivity and toxicity of solution itself and intensive energy consumption in the regeneration process. It has

been reported that >90% of CO₂ removal efficiency can be achieved at MEA to CO₂ ratio of 2:1 or even 1:1 when other amine such as DEA is used (Al-Baghli et al., 2001; Galindo et al., 2012). Hammond et al. estimated that MEA process will consume at least 20% and 30% of the total electricity production in the 500 MW natural gas fired plant and coal fired plant, respectively (Hammond and Ondo Akwe, 2007). Zahra et al. found that increasing the lean solution concentration can lower the energy demand for the desorption step, but will consequently increase the corrosivity of the solution and the solvent depletion rate (Abu-Zahra et al., 2007). Rochelle calculated out that the minimum work requirement for separate 90% of CO₂ via MEA process is 0.11 megawatt-h per metric ton of CO₂ (Rochelle, 2009).

Ammonia solution has been considered as a substitute solvent for amine solutions due to its lower cost and corrosivity. However, in addition to the high volatile nature of ammonia, the lack of a method to separate ammonia from CO₂ after thermal decomposition of ammonium bicarbonate also hinders the ammonia scrubbing technique from being applied in practice (Huang and Chang, 2002). There are two major ammonia processes based on the operation temperature: the chilled process has operation temperature ranged from 2 to 10 °C which can prevent the ammonia escape, but brings the scale and plugging problem because of the precipitation of ammonia carbonate salts (Kozak et al., 2009; Darde et al., 2010; Valenti et al., 2012); another ammonia process has the temperature ranged from 25 to 40 °C, this could not only prevent the precipitation of carbonate salts but also increase the absorption reaction activity (Bai and Yeh, 1997). Powerspan together

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Notation

a	surface area per unit volume, cm^2/cm^3
A_b	surface area of bubble, cm^2
A'	surface area of particle, cm^2
C_{Ai}	interfacial con. of CO_2 in the liquid, mol/cm^3
C_{AT}	conc. of CO_2 in the bulk liquid, mol/cm^3
C_{eq}	equilibrium concentration of total dissolved carbon, mol/cm^3
C_s	equilibrium concentration of $\text{Mg}(\text{OH})_2$, mol/cm^3
C_B	concentration of $\text{Mg}(\text{OH})_2$ in liquid, mol/cm^3
D_{AI}	liquid diffusivity of the dissolved CO_2 , cm^2/s
D_{BI}	liquid diffusivity of the dissolved magnesium ions, cm^2/s
$D_{\text{CO}_2 \text{ in air}}$	diffusivity of CO_2 in the air, cm^2/s
$D_{\text{CO}_2 \text{ in water}}$	diffusivity of CO_2 in the water cm^2/s
H_{CO_2}	Henry's law constant, $\text{atm}\cdot\text{cm}^3/\text{mole}$
He_{CO_2}	Effective Henry's law constant
K_w	dissociation constant of water, mol/cm^3
K_1, K_2	dissociation constants for reactions (12) and (13), mol/cm^3
K_G	overall mass transfer coefficient, $\text{mol}/\text{cm}^2\cdot\text{s}\cdot\text{atm}$
$k_G = k_g$	gas-side mass transfer coefficient, $\text{mol}/\text{cm}^2\cdot\text{s}\cdot\text{atm}$
k_{Ag}	gas-side mass transfer coefficient for CO_2 , $\text{mol}/\text{cm}^2\cdot\text{s}\cdot\text{atm}$
k_{Al}	liquid-side mass transfer coefficient for CO_2 , $\text{mol}/\text{cm}^2\cdot\text{s}\cdot\text{atm}$
$k_L = k_l$	liquid-side mass transfer coefficient, $\text{mol}/\text{cm}^2\cdot\text{s}\cdot\text{atm}$
k_s	dissolution mass transfer coefficient for $\text{Mg}(\text{OH})_2$, cm/s
k_{Bl}	liquid side mass transfer coefficient for Mg^{2+} , cm/s
L	height of liquid, cm
n_p	number of particles of $\text{Mg}(\text{OH})_2$ in container
p_A	concentration of CO_2 in the bulk gas, atm
p_{Ai}	interfacial concentration of CO_2 in the gas, atm
p_{avg}	average CO_2 partial pressure in bubble, atm
p_o	inlet CO_2 partial pressure in the gas stream (bubble), atm
p_{eq}	CO_2 partial pressure at equilibrium, atm
p_{in}, p_{out}	inlet and outlet gas pressure
Q_g	volumetric flow rate of the gas, cm^3/s
Q_{in}, Q_{out}	inlet and outlet gas flow rate, cm^3/s
R	gas constant, $\text{atm}\cdot\text{cm}^3/\text{mol}\cdot\text{K}$
$r = r_A$	absorption rate, $\text{mol}/\text{cm}^3\cdot\text{s}$
S	column cross-sectional area, cm^2
T	temperature, K
t	time, s
Δt_i	time interval
u	superficial gas velocity, cm/s
V_L	volume of liquid (without bubbles), cm^3
x_0	liquid film thickness, cm
x	distance of dissolved CO_2 travel from gas–liquid interface to the reaction zone, cm
$x_{\text{direction}}$	x direction of the column
$Y_{in,i}, Y_{out,i}$	inlet and outlet CO_2 volume fraction at during time interval i
z	normalized column height, from 0 to 1
φ	chemical enhancement factor
δ	film thickness around the solid particle, cm
$\delta_{\text{gas film}}$	gas film thickness, cm
$\delta_{\text{liquid film}}$	liquid film thickness, cm

with NETL successfully demonstrated 90% CO_2 removal efficiency at experiment conditions of 54°C , 5 s gas residence time, and liquid to gas ratio of 65 gpm/1000 acf (McLarnon and Duncan, 2009). Yeh et al. found that in the rich solution, product NH_4HCO_3 requires the least energy to be regenerated, overall, the energy requirement in the regeneration step is 62% less than the MEA desorption step (Yeh et al., 2005; Versteeg and Rubin, 2011).

K_2CO_3 solution has wide applications in the natural gas or synthetic gas sweetening processes due to the high reaction activity under high CO_2 partial pressures. However, when CO_2 partial pressure decreases to the power plant flue gas level, the absorption rate is significantly decreased. Piperazine (PZ) is one of the widely studied chemicals added into the solution in order to increase the absorption reactivity. It is reported that PZ has the advantage of faster absorption rate and larger CO_2 capacity than MEA (Oexmann et al., 2008). It is also more resistant to oxidative and thermal degradation than MEA and ammonia (Cullinane and Rochelle, 2004; Oexmann and Kather, 2009; Gouedard et al., 2012). The energy consumption of the process mainly depends on the overall concentration of PZ and K_2CO_3 , while the absorption kinetic mainly depends on the formation and concentration of PZ (Mudhasakul et al., 2013). Cullinane et al. showed that the solution consists of 20% K_2CO_3 and 5% PZ has comparable absorption rate as 23% MEA solution; while the solution consists of 36% K_2CO_3 and 11% PZ has up to 5 times faster rate than 30% MEA solution (Tim Cullinane et al., 2005). In addition, the PZ- K_2CO_3 process has an average energy saving of 35% compared with MEA process (Tim Cullinane et al., 2005). However, the price of PZ is 3–5 times higher than MEA, and only bench scale and small pilot scale tests have been performed.

Besides the selection of solvent, the configuration mass transfer device is also an important factor that influences the performance of the capture of CO_2 . Bubble contactors in which the gas is dispersed in the liquid and/or solid phases in the shape of bubbles are frequently used in the chemical industry as absorbers (Álvarez et al., 2008). They provide several advantages such as high heat and mass transfer rates, compactness, low maintenance and operating cost. Mass transfer is one of the main topics of recent research with bubble column (Behkish et al., 2002, 2007; Maalej et al., 2003). Table 1 summarizes the literature findings of bubble column mass transfer studies.

Scientifically, CO_2 can be separated from a gas mixture by all the methods that are being studied, while the challenge lies in practical feasibility and ultimately the cost. In this study, we proposed to use magnesium hydroxide ($\text{Mg}(\text{OH})_2$) as the absorbent, which is environmentally friendly and regenerable. And the bubble column reactor was selected as the mass transfer device in which the mass transfer coefficient was calculated for both water and $\text{Mg}(\text{OH})_2$ slurry.

The purpose of this study is to investigate the chemical reaction mechanisms and the mass transfer phenomena in the absorption process, and to develop a bubble column absorber model to assist in the understanding of the mass transportation of CO_2 removal within a gas–liquid–solid system.

2. Model development

The primary goal of this modeling work is to help understanding the mass transfer phenomenon of CO_2 - $\text{Mg}(\text{OH})_2$ - H_2O system in the bubble column reactor, and to calculate the overall mass transfer coefficient K_G , which is a key design parameter for FGDC absorption unit.

CO_2 - $\text{Mg}(\text{OH})_2$ - H_2O system is a gas–liquid–solid reaction system. The CO_2 in the gas phase need to be dissolved in the water in order to react with magnesium ions, which are dissolved from magnesium hydroxide particles. The whole scrubbing process may be

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