Carbon dioxide capture by sodium hydroxide-glycerol aqueous solution in a rotating packed bed

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ABSTRACT

Carbon dioxide capture by chemical absorption has been one of the most popular approaches. However, low solubility of CO_2 in water and high energy demand for absorbent regeneration hinder its feasibility. In this study, glycerol, a biodiesel waste by-product, was added to sodium hydroxide (NaOH) aqueous solution to relax these drawbacks, and to serve as an efficient absorbent in a rotating packed bed (RPB) for CO_2 capture. The experimental results suggested that the addition of glycerol in the NaOH aqueous solution boosted the overall CO_2 absorption efficiency to more than 90% easily. Under the high G (rotating speed) condition, the (chemical) enhancement factor could go up to more than 50. Furthermore, at same NaOH concentration, the mass transfer coefficients (K_Ga) could be doubled even when the viscous glycerol was added because of intensified centrifugal field. It is clearly indicated that, in an RPB, glycerol addition in NaOH solution showed very positive effect on CO_2 capture not only for better mass transfer performance but also for the potential utilization of biodiesel by-product.

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1. Introduction

Carbon dioxide capture is one of the most important approaches to reduce the global climate change. Among several CO_2 capture methods, absorption perhaps is considered to be one of the most attractive and efficient unit operations. In general, CO_2 absorption processes can be grouped into physical absorption and chemical absorption. For physical absorption, the solubility of CO_2, or the maximal amount can be absorbed, in water is very much limited by its nature. In contrast, the chemical absorption, enhanced by the acid-base neutralization or other reactions after CO_2 diffusing into the liquid phase, is widely used in the industry to overcome the limitation. Additives for chemical reactions frequently involved are sodium hydroxide, monoethanol-amine (MEA), 2-methyl-1-propanol (AMP), diethylenetetramine (DETA), diethanolamine (DEA), and piperamine (PZ) [1–6]. These CO_2 capture processes with the aid of chemical reaction are much more feasible and deserve more attention.

For both physical absorption and chemical absorption, the regeneration of the absorbent after the absorption is often necessary. However, during the absorbent regeneration, mainly a heating process, the majority of the energy (∼60%) is used in heating up water (∼4.2 J/g °C), which has little contribution in capturing CO_2 [7]. This energy waste might be reduced by increasing the concentrations of "functional" absorbents (NaOH, MEA, and AMP) but it could increase the viscosity and corrosivity inevitably. As a result, using solvents soluble in water with a good affinity toward CO_2 and a lower heat capacity such as diethylene glycol (DEG, 2.470 J/g °C) and glycerol (2.301 J/g °C) were proposed [5,8]. However, the viscosity for pure DEG and glycerol at 20 °C are 35.7 cp and 1410 cp, respectively. Hence, the merit of low heat capacity might be hindered by the poor mass transfer due to their viscosities if the CO_2 capture were carried out in a conventional CO_2 absorption bed. Therefore, a design that could handle viscous fluids and still possess superior mass transfer performance, i.e. rotating packed bed (RPB), becomes a good candidate. Comprehensive list concerning CO_2 capture by chemical absorption using high gravity can be found in a recent review article [9].

A rotating packed bed also known as a Higee system, which replaces gravity with centrifugal force up to hundreds of gravitational value ($g = 9.8$ m/s^2), was first introduced in 1981 as a high mass transfer gas/liquid contactor [10]. Under a high centrifugal field, tiny liquid droplets as well as thin liquid films are generated in the bed leading to a much higher gas/liquid interfacial area and also resulting in a reduction of mass transfer resistance. In literatures, the mass transfer coefficients in an RPB sometimes can be enhanced up to 1–2 orders of magnitude as compared with a

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Nomenclature

A	absorption percentage (%)
a	effective gas–liquid interfacial area per unit volume (m^2/m^3)
a_t	total particle surface area per unit volume of the packed bed (m^2/m^3)
a'_p	surface area per unit volume of beads (2 mm in diameter) packing (m^2/m^3)
D_G	diffusion coefficient in gas (m^2/s)
D_L	diffusion coefficient in liquid (m^2/s)
d_p	spherical equivalent diameter of the packing $= \frac{6(1-\varepsilon)}{a_t \psi}$ (m)
E	enhancement factor (–)
G'	gas mass flux (kg/m^2 s)
H_c	Henry's constant [(mole/m^3)/(mole/m^3)]
k_G	gas-phase mass transfer coefficient (m/s)
k_L	liquid-phase mass transfer coefficient (m/s)
$K_G a$	overall volumetric gas-film mass-transfer coefficient (1/s)
L'	liquid mass flux (kg/m^2 s)
Q_G''	liquid volumetric flow rate (m_3/s)
r	radius of the packed bed (m)
r_i	inner radius of the packed bed (m)
r_o	outer radius of the packed bed (m)
V_i	volume between the inner radius (m^3)
V_o	volume between the outer radius and stationary housing (m^3)
V_t	total volume of the bed (m^3)
Y	molar ratio of solute in the gas stream (mole/mole)
Y_i	molar ratio of solute in the inlet gas stream (mole/mole)
Y_o	molar ratio of solute in the outlet gas stream (mole/mole)
Y^*	equilibrium molar ratio associate with the liquid molar ratio (mole/mole)
z_b	axial height of the packing (m^3)

Greek letters

ε	porosity of the packing (–)
μ_G	viscosity of the gas
μ_L	viscosity of the liquid
ρ_G	density of the gas (kg/m^3)
ρ_L	density of the liquid (kg/m^3)
σ_c	critical surface tension of packing (kg/s^2)
σ_w	surface tension of water (kg/s^2)
ψ	sphericity of packing (–)

Dimensionless quantities

Gr_G	Grashof number of gas $= d_p^3 \rho_G^2 a_c / \mu_G^2$
Gr_L	Grashof number of liquid $= d_p^3 \rho_L^2 a_c / \mu_L^2$
Re_G	Reynolds number of gas $= G'/a_t \mu_G$
Re_L	Reynolds number of liquid $= L'/a_t \mu_L$
Sc_L	Schmidt number of liquid $= \mu_L/\rho_L D_L$
We_L	Webber number of liquid $= L'^2/\rho_L a_t \sigma_L$

traditional packed bed in some cases. One additional and important merit of an RPB comparing to traditional packed beds is that an RPB is capable of dealing with highly viscous fluids, such as glycerol [11,12], silicone oil [13], CMC solution [14], diethylene glycol [5] while maintaining its superior mass transfer performance. In other words, RPBs open a window for a wider range of the solvent choices that is very unlikely feasible in the traditional bed operations and still possess outstanding mass transfer efficiency. More importantly, with higher mass transfer efficiency,

much smaller absorption unit is required and thus lower the capital cost which is usually accounted more than half of total cost in a conventional absorption system [15].

Hence, in this study, glycerol, a co-solvent of lower heat capacity and higher CO_2 solubility, was added into an aqueous sodium hydroxide solution in an RPB for CO_2 capture. Among several possible co-solvents with similar properties, glycerol was chosen simply because glycerol is overproduced as a by-product in the biodiesel industry [16]. Each year, roughly 2.4 million tons of glycerol was produced in Europe only, much more than what is needed in industry (*ca.* 1 million ton) in the world [16]. Furthermore, this by-product stream, glycerol, in a biodiesel plant actually may contain the transesterification catalyst, NaOH. As a result, this study aims to demonstrate the possibility that using the waste from biodiesel industry to further decrease the carbon footprint by CO_2 absorption. This CO_2 rich glycerol–NaOH aqueous solution might be later used in an electrolysis system to produce useful chemicals [17,18].

2. Material and method

2.1. Experimental setup

The schematic experimental setup is shown in Fig. 1. The sodium hydroxide aqueous solution was added with glycerol at various ratios to serve as the CO_2 absorbent. The absorbent was pumped into the center pipe (marked as liquid in) and sprayed outward by the centrifugal force with the rotor speed from 300 to 1500 rpm, roughly corresponding to 4–101 g (using arithmetic average radius of the bed). A 10% CO_2 gas stream, by mixing CO_2 with air, at rates of 4.3 L/min and 8.8 L/min were used. Gas stream entered at the bottom of the housing and then contacted with the liquid absorbent within the rotating bed counter-currently. The inner and outer radii of the bed were 2 and 6 cm while the height was 2 cm and the stationary housing was 9 cm in radius. The packing used was stainless steel wire mesh with 0.22 mm in diameter and its porosity and interfacial area were 0.93 and 1514 m^2/m^3, respectively.

The concentration of CO_2 was measured by a gas chromatography (China Chromatography GC9800) equipped with a TCD detector and a Porapak Q column (6 ft in length, 2 mm ID). The temperatures of injector, oven and detector were all 40 °C. The flow rates of nitrogen (carrier gas) was set to be 20 mL/min.

2.2. Definition of the efficiency indicators (A, $K_G a$, E)

2.2.1. Absorption percentage (A)

Absorption percentage is used as a direct index of the efficiency for the absorption process. The definition is given as the following:

$$A = \frac{Y_i - Y_o}{Y_i} \times 100\% \tag{1}$$

where Y_i (mole/mole) and Y_o (mole/mole) are the molar ratio of CO_2 at inlet and outlet, respectively.

2.2.2. Mass transfer coefficient ($K_G a$)

The overall volumetric gas-phase mass transfer coefficient ($K_G a$) in an RPB is derived from the concept of mass balance and transfer unit. Considering a differential volume with cross-section area of $2\pi r z_b$ and thickness dr, where r is the arithmetic average radius and z_b is the height of the packed bed. The mass balance of CO_2 in this differential volume for a dilute system is

$$Q_G'' dY = K_G a(Y - Y^*) 2\pi r z_b dr \tag{2}$$

where Q''_G and Y are the gas flow rate (m^3/s) and the CO_2 molar ratio in gas phase (mole/mole), and Y^* represents the equilibrium concentration associated with the liquid concentration. Then,