

## Carbon dioxide absorption using ammonia solution in a microchannel



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### ABSTRACT

This work dealt with the application of microchannel for CO<sub>2</sub> absorption by using ammonia solution. Statistical analysis was used to investigate the main and interaction effects of pressure, temperature, concentration and flow rate of ammonia solution. The feed gas was 10 vol.% CO<sub>2</sub> in nitrogen and a T-type microchannel (0.5 × 0.5 × 60 mm<sup>3</sup>) was used. Increasing temperature, pressure, and concentration enhanced the CO<sub>2</sub> capture. At 30 °C, ammonia concentration of 10% with the flow rate of 0.0003 m<sup>3</sup> h<sup>-1</sup> and 300 kPa, the absorption efficiency was 96.45%. We also studied the effect of operating parameters on the overall volumetric mass transfer coefficient.

### 1. Introduction

Carbon dioxide (CO<sub>2</sub>) is the most important gas that causes the greenhouse effect (Molina and Bouallou, 2015) due to the heat capacity, the accumulation, and the increasing trend of CO<sub>2</sub> global emission. The sources for CO<sub>2</sub> come from agricultures and industries such as manufacturing, transportation, combustion and oil rigs (Davison, 2007). CO<sub>2</sub> is also a cause for a decrease in heating value of a fuel such as natural gas (Ayandotun et al., 2012) and biogas (Tan and Ai, 2016). Due to the low heating value, such fuel is used in greater quantities and it requires larger storage space compared to that of standard fuel. The CO<sub>2</sub> capture is very useful to upgrade the properties of fuel. For instance, the properties biogas can be upgraded close to the properties of natural gas (Nock et al., 2014). Thus, CO<sub>2</sub> removal is very important for both greenhouse effect and fuel upgrading. Moreover, the separated CO<sub>2</sub> can be further purified and used in many industries such as dry ice, beverage, extraction process (Raventos et al., 2002), enhanced oil recovery (Dai et al., 2014), and chemicals (methane (Aziz et al., 2015), methanol (Wang et al., 2011) and salicylic acid (Zou and Liu, 2010)).

There are several methods for CO<sub>2</sub> separation such as physical absorption (Haszeldine, 2009), chemical absorption (Giuffrida et al., 2013), adsorption (Zhao et al., 2010), cryogenic distillation (Hart and Gnanendran, 2009) and membrane separation (Kovvali and Kamalesh, 2002). In early years, water scrubbing was the most widely used technique for CO<sub>2</sub> absorption from fuels because its simplicity and low operating cost compared to the use of amine solution. However, water scrubbing has low CO<sub>2</sub> specificity and can also absorb methane (CH<sub>4</sub>) which leads to the loss of CH<sub>4</sub> in a process and low purity of CH<sub>4</sub> (Bauer

et al., 2013). At present, chemical absorption is the most suitable and widely used method (Davison, 2007; Puxty et al., 2010; Zeng et al., 2011) because of high removal efficiency, selectivity (Ma et al., 2013), and cost effectiveness for large scale plant (Tobiesen et al., 2008). There are many absorbents that can be used for CO<sub>2</sub> absorption such as monoethanolamine (MEA), sodium hydroxide (NaOH), ionic liquid, ammonia solution, etc. MEA is the most widely used absorbent for CO<sub>2</sub> absorption because its high reactivity and thermal stability (Lin and Kuo, 2016). However, some drawbacks for using MEA include corrosion of equipment, low CO<sub>2</sub> capacity, degradation by sulfur dioxide (SO<sub>2</sub>), nitrogen dioxide (NO<sub>2</sub>) and hydrochloric acid (HCL) (Rivera-Tinoco and Bouallou, 2010) and the energy requirement of MEA regeneration (Hanak et al., 2015). NaOH is another absorbent for CO<sub>2</sub> absorption but the regeneration of NaOH is difficult due to the fact that NaHCO<sub>3</sub>, a product from CO<sub>2</sub> absorption process, is easily dissolved in water (Yoo et al., 2013). Ionic liquid is not widely used in industry because it is expensive and highly viscous (Camper et al., 2008). In order to overcome these problems, many researchers apply ammonia solution for CO<sub>2</sub> absorption. Not only that ammonia solution is cheaper, it also has high CO<sub>2</sub> capacity, high absorption efficiency, compatibility with SO<sub>x</sub> and NO<sub>x</sub> (Han et al., 2013). It requires much less energy for the regeneration compared to that of amine solution (Puxty et al., 2010; Diao et al., 2004; Yeh and Bai, 1999). Moreover, the product from chemical absorption using ammonia solution can be used for fertilizer (Ma et al., 2013; Bak et al., 2015) such as urea (Barzagli et al., 2016) and ammonium sulphate (Bonalumi and Giuffrida, 2016).

The chemical separation process for CO<sub>2</sub> absorption relies heavily on the interfacial mass transfer. There are many contacting devices that

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Nomenclature		$V_r$	Volume of reactor, $m^3$
$a_v$	Interfacial area, $m^2 m^{-3}$	$y$	Mole fraction of carbon dioxide
$C$	Concentration of ammonia solution, wt. %	$\beta$	Enhancement factor
$C_{CO_2}$	Concentration of $CO_2$ at the bulk liquid, wt. %	$\phi$	Overall absorption rate, $kmol h^{-1} m^{-3}$
$F$	Flow rate of ammonia, $m^3 h^{-1}$	<i>Superscripts</i>	
$H$	Henry's law constant, $kPa m^3 kmol^{-1}$	*	Gas-liquid equilibrium
$K$	Overall mass transfer coefficient, $kmol h^{-1} m^{-2} kPa^{-1}$	Sol	Aqueous ammonia solution
$K_{Ga_v}$	Overall volumetric mass transfer coefficient, $kmol h^{-1} m^{-3} kPa^{-1}$	<i>Subscripts</i>	
$k_L$	Liquid mass transfer coefficient, $m h^{-1}$	G	Gas phase
$D_{CO_2}$	Diffusion coefficient of $CO_2$ in an ammonia solution, $m^2 s^{-1}$	L	Liquid phase
$N$	The mass flux, $kmol h^{-1} m^{-2}$	i	At the gas-liquid interface
$P$	Total pressure gauge, kPa	$CO_2$	Carbon dioxide
$P_{CO_2}$	Partial pressure of carbon dioxide, kPa	in	Inlet
$q$	Molar flow rate, $kmol h^{-1}$	out	Outlet
$T$	Temperature, $^{\circ}C$		

are used for  $CO_2$  absorption such as packed column (Lin and Kuo, 2016), spray column (Zhao et al., 2016a), and bubble column (Chu et al., 2017). These gas-liquid contactors have limited gas-liquid interface and rate of mass transfer (Lin et al., 2003). By and large,  $CO_2$  absorption process requires large gas-liquid interface and high rate of mass transfer (Ma et al., 2013); hence, high performance and high throughput reactors are necessary. Note that large reactors are normally associated with high operating cost, difficulty of maintenance and safety issues. Thus, in order to enhance the  $CO_2$  absorption efficiency, a microchannel reactor is proposed. The microchannel reactor can improve the absorption efficiency for gas-liquid absorption due to high surface-to-volume ratio, short transport distances and high driving force gradients (Lam et al., 2013), resulting in rapid rates of reaction, heat transfer and mass transfer compared to the conventional devices. Instead of scaling up, the numbering-up principle (or scaling out) is used to adjust the production capacity of microchannels. Zafir et al. (2005) applied a microstructured reactor with dimensions of  $300 \mu m \times 100 \mu m \times 66.4 mm$  for carbon dioxide absorption by using sodium hydroxide solution. Results suggested that  $CO_2$  was rapidly used at the gas-liquid interface and the microstructured reactor helped enhancing the  $CO_2$  absorption efficiency by increasing the ratio of gas-liquid interface per liquid volume and reducing waste. The mass

transfer coefficient and specific interfacial area of gas-liquid absorption have been studied in small-scale device such as microchannel reactor using MEA (Li et al., 2014), minichannel reactor using DEA (Ganapathy et al., 2014). Both values were higher than those obtained from spray column, packed column, bubble column and venture reactor.

By using ammonia solution as an absorbent, both physical and the chemical absorption will take place simultaneously (Zhao et al., 2016b). First,  $CO_2$  from the gas stream diffuses to the gas-liquid interface prior to dissolving into the liquid film, which is known as physical absorption (Walozi et al., 2016). Then the dissolved  $CO_2$  reacts with ammonium hydroxide in the liquid phase, which is considered as chemical absorption. The reaction product includes  $CO_2$ -containing ammonium salts such as ammonium bicarbonate ( $NH_4HCO_3$ ), ammonium carbonate ( $(NH_4)_2CO_3 \cdot H_2O$ ), ammonium carbamate ( $NH_2COONH_4$ ) and other products (Zeng et al., 2011) depending on the operating conditions applied such as temperature, pressure, pH of solution, concentration of  $CO_2$  and concentration of ammonia solution (Yeh et al., 2005). A study by Darde et al. (2010) and Sutter et al. (2015) analyzed the phase diagram suggesting the main product formed for different operating conditions of  $CO_2$  absorption using ammonia solution. The reaction to form ammonium carbamate is fast and exothermic as shown below (Zeng et al., 2011).

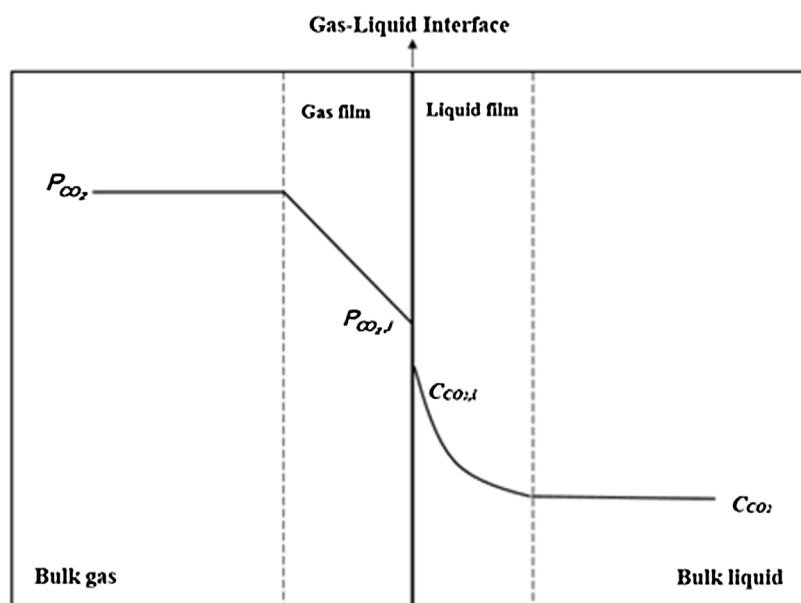


Fig. 1. The schematic diagram of the model of two-film theory.

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