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# Comparison of mass transfer coefficients and desorption rates of CO<sub>2</sub> absorption into aqueous MEA + ionic liquids solution

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## ABSTRACT

The mixed solvent of ionic liquids and ethanolamine (MEA) was used to absorb CO<sub>2</sub> from mixed gases using a packed column. The density and viscosity of aqueous MEA + ionic liquids solution were determined at temperatures ranging from 303.15 to 323.15 K and ionic liquids mass concentrations from 20% to 30%, respectively. A comparison among the mass transfer of CO<sub>2</sub> absorption into MEA + ionic liquids ([Bmim][Br], [Emim][Br]) and MEA was performed at 303.15–323.15 K. Meanwhile, the effects of the temperature, liquid flow rate and gas flow rate on the  $K_G a_v$  value were discussed in this paper, respectively. At last, the desorption rates of rich solution were calculated by determining the volume of CO<sub>2</sub> released from rich solution. The result showed that aqueous MEA + 20% [Emim][Br] solution had superior performance for CO<sub>2</sub> capture. Moreover, as CO<sub>2</sub> was absorbed by the mixture of MEA and ionic liquids, the catalytic action of ionic liquids was not obvious.

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## 1. Introduction

Carbon dioxide (CO<sub>2</sub>) is known as the main greenhouse gas which causes the global warming nowadays (Ma et al., 2013; Godini and Mowla, 2008). Therefore, the exploitation of economical and practical capture technology is becoming a hot issue in the field of greenhouse gas emission (Ober et al., 2012; Shiflett et al., 2010; Holst et al., 2009; Liu et al., 2013; Kavoshi et al., 2013). The commercial way on CO<sub>2</sub> capture depends on chemical absorption with the most widely used alkanolamines (Moioli and Pellegrini, 2015), such as monoethanolamine (MEA), diethanolamine (DEA) and methyldiethanolamine (MDEA). The alkanolamines are known as good absorption performance for the separation of CO<sub>2</sub> from flue gas and crude natural gas (Bidart et al., 2011).

Among these alkanolamines, MEA is the most common absorbent for CO<sub>2</sub> capture due to its advantages of low cost, high capacity and reasonable thermal stability. However, aqueous MEA solution has also many drawbacks, such as high solvent loss, corrosion, high energy demand for regeneration and instability to oxidant (Gutowski and Maginn, 2008; Wang et al., 2010; Privalova et al., 2013).

As a potential absorbent, room temperature ionic liquids (RTILs) have attracted more and more attention compared to alkanolamines solvents owing to their advantages, such as negligible volatility, low energy consumption for regeneration, superior thermal stability and good performance of CO<sub>2</sub> absorption (Wang et al., 2010; Privalova et al., 2013; Galán Sánchez et al., 2011; Bara et al., 2009; Kühne et al., 2008; Zhang et al., 2013). In general, the absorption of CO<sub>2</sub> into conven-

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### Nomenclature

$C_{CO_2}$	CO <sub>2</sub> concentration in liquid phase [mol/m <sup>3</sup> ]
$C_{MEA}$	MEA concentration in liquid phase [mol/m <sup>3</sup> ]
$C_{MEA H^+}$	MEA concentration in liquid phase [mol/m <sup>3</sup> ]
$k_{CO_2}$	Reaction rate constant [m <sup>3</sup> /(mol s)]
$G$	Inert gas flow rate [mol/(m <sup>2</sup> s)]
$K_G a_v$	Volumetric overall mass transfer coefficient [mol/(m <sup>3</sup> s Pa)]
$L$	Liquid flow rate [mol/(m <sup>2</sup> s)]
$P$	Operating pressure [Pa]
$r_{CO_2}$	Desorption rate [mol/s]
wt%	Weight percent
$y_A$	The CO <sub>2</sub> mole fraction in gas bulk
$y_A^*$	CO <sub>2</sub> equilibrium mole fraction at interface
$V_{tot}$	The total volume of CO <sub>2</sub> desorption [m <sup>3</sup> ]
Abb	The solution before absorbing CO <sub>2</sub>
Abs	The solution after absorbing CO <sub>2</sub>
$\rho$	Density [g/cm <sup>3</sup> ]
$\mu$	Viscosity [cP, mPa s]

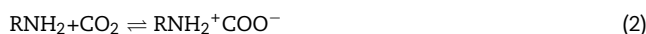
tional ionic liquids is a physical process. Different with alkanolamines, the physical absorption of CO<sub>2</sub> in ionic liquids largely depends on its solubility and operating conditions, and owns low value at ambient pressure and temperature (Zhang et al., 2008). Therefore, there is a growing trend for research amino-functionalized ionic liquids (NH<sub>2</sub>-RTIL), because both physical and chemical absorption takes place during the process of absorption that lead to 2:1 molar ratio of NH<sub>2</sub>-IL to CO<sub>2</sub> (Gutowski and Maginn, 2008; Wang et al., 2010; Galán Sánchez et al., 2011). However, it had been reported that the pure task-specific ionic liquids (TSILs) had high viscosities and their viscosities increased dramatically after contacting with CO<sub>2</sub>, which would lead to many drawbacks in industrial application (Gutowski and Maginn, 2008).

Recently, it had been reported that the mixture of alkanolamine and ionic liquids had good performance for CO<sub>2</sub> capture even at very low pressure (Bara et al., 2009; Kühne et al., 2008). Alkanolamine can be dissolved and reacted with CO<sub>2</sub> in ionic liquids readily. Meanwhile, CO<sub>2</sub> can be released easily from these mixtures by heating solution. It overcomes the deficiency of high viscosities and volatile, heavy corrosion caused by other methods on CO<sub>2</sub> capture. The CO<sub>2</sub> absorption into aqueous MEA and ionic liquids solution has been reported at high pressure in many papers (Kühne et al., 2008; Shin et al., 2008; Shariati and Peters, 2004). For instance, Shiflett et al. (2010) studied the absorption process of CO<sub>2</sub> using 1-butyl-3-methylimidazolium acetate at atmospheric pressure. Ahmady and Zhao discussed the absorption performance of CO<sub>2</sub> into several blends combined by alkanolamine and ionic liquids at wide range of carbon dioxide partial pressures, and the activation energy of the mixture of 1-butyl-3-methylimidazolium tetrafluoroborate and MDEA solution has been proved to be reduced by compared to pure MDEA solution (Ahmady et al., 2012; Zhao et al., 2010). And it was reported that ionic liquids take a role of catalyst in the process of CO<sub>2</sub> absorption into the blend of MDEA and [Bmim][BF<sub>4</sub>] in their paper. Meanwhile, the study of absorbing CO<sub>2</sub> with the mixtures of MEA and imidazolium-based ionic liquids in packed column has not been reported at present. Since MEA is used widely in industry, also in order to check catalytic action of ionic liquids and design column for CO<sub>2</sub> absorption process, it is necessary to study the mass transfer coefficient of CO<sub>2</sub> absorption into aqueous MEA+ionic liquids solution. Therefore, 1-ethyl-3-methylimidazolium bromine ([Emim][Br]) and 1-butyl-3-methylimidazolium bromine ([Bmim][Br]) were added into aqueous MEA solution, respectively. And the volumetric overall mass transfer coefficients ( $K_G a_v$ ) of CO<sub>2</sub> absorption into these solution were calculated in this article. It will be helpful to learn the action of ionic liquids in the process of CO<sub>2</sub> absorption. The effect of temperature, liquid flow rate and CO<sub>2</sub> flow rate on the mass transfer of CO<sub>2</sub> absorption into aqueous MEA+ionic liquids([Emim][Br], [Bmim][Br])

solution was discussed, respectively. Meanwhile, in order to determine the capacity of CO<sub>2</sub> absorption and the effect of ionic liquids on CO<sub>2</sub> desorption, the performance of CO<sub>2</sub> desorption from different rich solution was studied at different temperatures.

## 2. Theory

### 2.1. CO<sub>2</sub> absorption in aqueous MEA solution



The mechanism of CO<sub>2</sub> absorption into aqueous MEA solution was described by Danckwerts (Danckwerts 1979; Sada et al., 1976). In this process, Eq. (1) represents that CO<sub>2</sub> is dissolved in aqueous MEA solution, it is a physical process accompanied with heat emission. As shown in Reactions (2) and (3), the reaction between CO<sub>2</sub> and MEA is consisted of two-step mechanism, which lead to the formation of carbamate. In general, the Reaction (4) can be neglected because the time of liquid–gas contact is very short in industrial absorbers (Liu et al., 2006). Since CO<sub>2</sub> is reacted with MEA promptly, the Reaction (6) can be also neglected in this process. As described in the literatures (Liu et al., 2006; Khan et al., 2011), the whole process of CO<sub>2</sub> absorption was an irreversible second-order reaction and reaction rate was conditional on MEA concentration and CO<sub>2</sub> partial pressure. As MEA concentration is constant, this process can be regarded as the first order reaction.

### 2.2. Mass transfer

It is well known that the gas film and the liquid film are formed on the gas side and liquid side in the process of absorption, respectively. The process of mass transfer is regarded to take place in the interface of gas side and liquid side. In our study, the process of CO<sub>2</sub> absorption is assumed to consist of diffusion and chemical reaction. According to two-film theory (Puxty et al., 2010), the mass transfer process can be described as shown in Fig. 1. At the side of gas film, CO<sub>2</sub> concentration profile is sharply reduced toward the interface of gas and liquid. As it reaches the interface, CO<sub>2</sub> is reacted with absorbent. Then CO<sub>2</sub> diffuses in equilibrium state through the zone toward the bulk part of the falling liquid film. At the side of the liquid film, carbon dioxide diffuses toward the bulk part until completed conversion. Since the experiment was performed at atmospheric pressure, the concentration of CO<sub>2</sub> in bulk liquid was assumed to be negligible compared to its concentration at the interface of gas–liquid due to its instantaneous reaction with MEA (Ahmady et al., 2012).

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