



Mass transfer and kinetics of CO₂ absorption into aqueous monoethanolamine/1-hydroxyethyl-3-methyl imidazolium glycinate solution



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HIGHLIGHTS

- Kinetics of the reaction of CO₂ and MEA/[C₂OHmim][Gly] was investigated.
- $k_{2,mix}$ and E of this new system were all higher than those of MEA solution.
- $k_{2,mix}$ into MEA/[C₂OHmim][Gly] solution was 6506.4 m³ kmol⁻¹ s⁻¹ at 303.15 K.

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ABSTRACT

The aqueous blend of monoethanolamine (MEA) and 1-hydroxyethyl-3-methyl imidazolium glycinate ([C₂OHmim][Gly]) was considered as a promising CO₂-capturing solvent because of the advantages of fast absorption rate, high absorption capacity, great thermal stability and good resistance to O₂. In the present work, kinetics of the reaction of CO₂ and MEA/[C₂OHmim][Gly] in aqueous solutions had been investigated using a double stirred-cell absorber with a defined gas/liquid interface. The zwitterion mechanism was used as the kinetic model to describe the absorption of CO₂ in the mixed solution. The kinetic and mass transfer parameters, e.g., the reaction rate constant ($k_{2,mix}$), the overall reaction rate constant (k_{ov}) and the enhancement factor (E), were evaluated at different absorbent concentrations and temperature. At 303.15 K, the values of k_2 into MEA/[C₂OHmim][Gly] and [C₂OHmim][Gly] solution were respectively calculated as 6506.4 and 8980.6 m³ kmol⁻¹ s⁻¹. The relationship between k_2 and the reaction temperature was determined by the Arrhenius Equation, and was presented as:

$$k_{2,mix} = 7.12 \times 10^6 \exp\left(\frac{-2126.3}{T}\right)$$

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1. Introduction

The concept of carbon dioxide (CO₂) capture and sequestration (CCS) has attracted considerable interest over the past two decades, as a result of the emerging problem of climate change [1]. One of the most potent techniques widely used in capturing CO₂ from low pressure flue gas streams in power plants is chemical absorption using aqueous amine-based absorbents [2].

Alkanolamines such as monoethanolamine (MEA), diethanolamine (DEA), N-methyldiethanolamine (MDEA) and 2-amino-2-methyl-1-propanol (AMP) are the most commonly used absorbents in the past few years. The most advantage of MEA is high reactivity with CO₂, but this process still suffers some disadvantages [3–6], e.g., low CO₂ capacity (0.5 mol CO₂/mol absorbent), amine degradation and oxidation and high energy consumption. Thus the mixed solvents of amines with various additives are developed to overcome these existing drawbacks.

To date, ionic liquids (ILs) have received an advantage in various applications including in CO₂ capture [7,8]. More interestingly, introducing some special groups to the anion or the cation of ILs

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Nomenclature

$C_{\text{Absorbent}}$	concentration of the absorbent (kmol m^{-3})	$k_{2, i}$	second-order reaction rate constant in i solution ($\text{m}^3 \text{kmol}^{-1} \text{s}^{-1}$)
C_B	concentration of base B in bulk liquid (kmol m^{-3})	k_{ov}	overall reaction rate constant (s^{-1})
C_{CO_2}	concentration of CO_2 (kmol m^{-3})	L	liquid-phase
$D_{i, j}$	diffusivity of i in the solution of j ($\text{m}^2 \text{s}^{-1}$)	n	the stirring speed (rpm)
E	enhancement factor (–)	N	absorption rate of CO_2 ($\text{kmol m}^{-2} \text{s}^{-1}$)
E_{∞}	the instantaneous enhancement factor (–)	P_{CO_2}	partial pressure of CO_2 (kPa)
G	gas-phase	Q_{in}	the inlet gas-flow rate ($\text{m}^3 \text{s}^{-1}$)
$H_{i, j}$	solubility of i in the solution of j ($\text{kPa m}^3 \text{kmol}^{-1}$)	Q_{out}	the outlet gas-flow rate ($\text{m}^3 \text{s}^{-1}$)
Ha	Hatta number (–)	r	the reaction rate ($\text{kmol m}^{-2} \text{s}^{-1}$)
K_G	overall gas phase mass transfer coefficient ($\text{kmol m}^{-2} \text{s}^{-1} \text{kPa}^{-1}$)	R	perfect gas constant ($8.314 \text{ J mol}^{-1} \text{ K}^{-1}$)
k_G	gas-phase mass transfer coefficient ($\text{kmol m}^{-2} \text{s}^{-1} \text{kPa}^{-1}$)	T	temperature (K)
k_L	liquid-phase mass transfer coefficient (m s^{-1})	μ	viscosity of solution (mPa s)
		z	stoichiometric coefficient (–)

to obtain functionalized ILs can achieve excellent performance in CO_2 capture due to the following characteristics [9–14]: low volatility, low heat required for reaction, low thermal and oxidative degradation, high absorption capacity and easy to be regenerated.

To achieve excellent performance, the complexities of amine and ILs or functionalized ILs are proposed as new solvents for CO_2 capture [15–18]. It was found that CO_2 solubilities in binary and ternary systems of guanidinium trifluoromethanesulfonate ([gua][OTf]) in MDEA and/or water were higher compared to other ILs, and decreased as the composition of [gua][OTf] in the systems increased [19]. Regeneration performance of $[\text{N}_{1111}][\text{Gly}]$ and MDEA aqueous solutions was performed, and it was found that the regeneration efficiency of 30 wt% MDEA + 15 wt% IL solution was a little higher than the other solution with different compositions [20]. In the other mixed solution, such as $[\text{N}_{1111}][\text{Gly}]/\text{AMP}$ system, the addition of $[\text{N}_{1111}][\text{Gly}]$ had greatly promoted the absorption of CO_2 in AMP aqueous solution [21].

Besides, many works focus on kinetics analysis in order to investigate the mass transfer process. Our previous work found that $[\text{Bmim}][\text{BF}_4]$ had an active effect on the CO_2 hydration and the values of enhancement factor (E) and the second-order reaction rate constant ($k_{2, \text{mix}}$) for CO_2 absorption into $\text{MEA}/[\text{Bmim}][\text{BF}_4]$ solution were higher than that of MEA solution [22]. Kinetics of DEA dispersion in $[\text{Hmim}][\text{Tf}_2\text{N}]$ for efficient CO_2 capture was investigated by Iliuta et al., and the kinetic model used to describe the chemical reaction was based on the zwitterion mechanism followed by the removal of a proton by the amine and formation of immiscible solid carbamate crystals in the ionic liquid phase [23]. The kinetics region of absorption CO_2 into aqueous $[\text{N}_{1111}][\text{Gly}] + \text{AMP}$ solution was found to be the fast pseudo-first order reaction regime and the activation energy of CO_2 capture into $[\text{N}_{1111}][\text{Gly}] + \text{AMP}$ aqueous solution found to be 40.7 kJ mol^{-1} [21]. A summary of aqueous blend of amine and ILs or functionalized ILs for CO_2 capture at low partial pressure were presented in Table 1, which indicated that the mixture seemed to be a suitable candidate for CO_2 capture.

In our previous work, a dual functionalized ILs ($[\text{C}_2\text{OHmim}][\text{Gly}]$) was designed and prepared based on the imidazolium ionic liquid with amino acid and hydroxyl group. Then the new hydrophilic amino acid ionic liquid was utilized to enhance the process of MEA for CO_2 capture from low-pressure flue gas. The new blend of MEA and $[\text{C}_2\text{OHmim}][\text{Gly}]$ was proved to have an enhanced absorption capacity, a higher regeneration efficiency and a better resistance to O_2 for the CO_2 capture [26]. As a promising CO_2 -capturing solvent, although the absorption and desorption performance of CO_2 capture into $\text{MEA}/[\text{C}_2\text{OHmim}][\text{Gly}]$ solution had been extensively studied, there is uncertainty about the mass

transfer and the reaction kinetics of this new system. Herein, kinetics of the reaction of CO_2 and $\text{MEA}/[\text{C}_2\text{OHmim}][\text{Gly}]$ in aqueous solutions was investigated using a double stirred-cell absorber with a defined gas/liquid interface. The kinetics model and parameters were measured in the presented work.

2. Materials and methods

2.1. Chemicals

Monoethanolamine (MEA) with 99.0% purity was supplied by Shanghai Ling Feng Chemical Reagent Co. Ltd, China. The gas of CO_2 (>99.99%), O_2 (>99.99%) and N_2 (>99.99%) supplied from the steel cylinder was purchased from Zhejiang Jin-gong Gas Co, China. The dual functionalized ionic liquid $[\text{C}_2\text{OHmim}][\text{Gly}]$ was prepared in our laboratory. No further purification was performed on the materials used.

2.2. Kinetic experiments

The experiments were carried out in a double stirred-cell absorber [22], which had a defined gas/liquid interface as the mass transfer area and was convenient for kinetic investigation. The experimental setup is shown in Fig. 1. CO_2 was mixed with N_2 to simulate the flue gas. The total gas-flow rate was controlled at 1 L min^{-1} by using a mass flow controller. The stirring speeds of the gas phase and liquid phase were kept at 250 rpm and 100 rpm, respectively. The absorption rate of CO_2 into aqueous blend of $\text{MEA}/[\text{C}_2\text{OHmim}][\text{Gly}]$ were measured under different concentrations of initial mixed absorbent ($1\text{--}3 \text{ kmol m}^{-3}$) and different temperatures ($303.15\text{--}333.15 \text{ K}$), respectively. To establish pseudo-first order reaction conditions, the simulated gas and fresh aqueous solution were both continuously supplied to the absorber. The CO_2 partial pressure of the gas phase remained unchanged under each condition during the experiments because of steady conditions. The inlet and outlet concentrations of CO_2 were measured by using gas chromatography (GC-7890, Agilent, USA). All data shown in this paper were the mean values of duplicate or triplicate experiments.

3. Theoretical analysis

3.1. Reaction mechanisms

The total reaction of CO_2 absorption into aqueous blend of $\text{MEA}/[\text{C}_2\text{OHmim}][\text{Gly}]$ can be simplified as the sum of the reaction

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