



Mass transfer performance of CO₂ absorption by alkanolamine aqueous solution for biogas purification



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ABSTRACT

The CO₂ absorption of an aqueous alkanolamine solution is an important research topic in the field of biogas purification. In this paper, an experimental packed tower was set up to investigate the high-concentration CO₂ absorption of an ethanolamine (MEA) solution. A mathematical model of the enhancement factor based on a second-order chemical reaction was proposed. This model was used to calculate and analyse the enhancement factor and the mass transfer performance of high-concentration CO₂ absorption by an MEA aqueous solution, respectively. The results show that the assumption of a pseudo-first-order reaction is not suitable for the high-concentration CO₂ absorption process and the enhancement factor obtained via mathematical prediction by this model is in agreement with the experimental value. Moreover, the high-concentration CO₂ absorption of the MEA aqueous solution is a liquid-controlled process. Furthermore, parameters such as gas phase overall mass transfer coefficient K_G , liquid phase mass transfer coefficient k_L and enhancement factor E increase with increasing MEA concentration but decrease with increasing CO₂ concentration. When the gas flow rate is increased, the above parameters initially increase, then decrease, and finally level-off gradually. The results of this study may provide meaningful insights for the research and application of biogas purification technology.

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1. Introduction

Renewable energy derived from biomass sources has great potential for development to meet our future energy demands. Biogas, which is produced via the anaerobic decomposition of organics, is a mixture composed of approximately 55–70% CH₄, 30–45% CO₂, and trace amounts of NH₃ (80–100 ppm), H₂S (1000–3000 ppm) and hydrocarbons (<100 ppm) [1–4]. Biogas is a very important source of renewable methane after removal of CO₂ and other impurities. Various technologies have been previously developed for CO₂ separation from gas streams, including absorption by chemical solvents, physical absorption, cryogenic separation, membrane separation, and CO₂ fixation by biological or chemical methods [5,6]. The removal of CO₂ by an alkanolamine solution has received increasing attention because of its suitable scale, high concentration of the product gas and low energy consumption [7,8].

Numerous investigations have been reported on the absorption of both low-concentration CO₂ (1–15%) and high-concentration CO₂ (30–45%) by alkanolamine. Aroonwilas and Ton-tiwachwuthikul [9] studied the feasibility of the use of high-

efficiency structured packing in the CO₂ separation processes with 2-amino-2-methyl-1-propanol (AMP). Tang et al. [10] experimentally studied the volumetric overall mass transfer coefficients of CO₂ absorption (<10%) into aqua ammonia in a structured packed column. Cheng and Tan [11] studied the removal of CO₂ (<1%) from indoor air by using alkanolamine in a rotating packed bed. Hasibur-rahman and Faical [12] investigated the kinetic behaviour of CO₂ (<10%) absorption in amine-room-temperature ionic liquids blends and simultaneously discussed the influence of experimental parameters on CO₂ uptake behaviour, such as amine concentration, CO₂ partial pressure, agitation speed, and temperature variation. Liu et al. [13] experimentally studied the performance of aqueous sodium glycinate for biogas purification. Kim et al. [14] took into account the species distributions of the polyamine absorbent after CO₂ absorption, thereby establishing CO₂ (30%) absorption mechanisms, which indicated that multiple amino groups promote CO₂ absorption capacity and decrease the heat of absorption. Lu et al. [15] studied the performances of piperazine and phosphates that were served as additives and injected into an aqueous N-methyl monoethanolamine (NMEA) to form complex absorbents for CO₂ capture in a bubble column reactor. The impact of various factors, such as additive types, concentrations, and gas flow rate, on the overall mass transfer has yet to be determined. Meanwhile, the

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Nomenclature

c	concentration, mol L ⁻¹	p	pressure, kPa
D_G	diffusivity of CO ₂ in mixed gas, m ² s ⁻¹	Δp	pressure drop, kPa
D_{MEA}	diffusivity of MEA in MEA solution, m ² s ⁻¹	r	chemical reaction rate, kmol m ⁻³ s ⁻¹
E	enhancement factor	R	gas–liquid ratio
E_{cal}	theoretical value of the enhancement factor	T	temperature, K
E_{exp}	experimental value of the enhancement factor	y	molar ratio of the gas phase
E_∞	enhancement factor of the instantaneous reaction	z	packing height, m
G	gas flow rate, kmol m ⁻² s ⁻¹		
G_B	gas flow rate of N ₂ , kmol m ⁻² s ⁻¹		
H	Henry's constant, m ³ kPa kmol ⁻¹		
Ha	Hatta number		
K_G	gas phase overall mass transfer coefficient, kmol m ⁻² s ⁻¹ kPa ⁻¹		
k_G	gas phase mass transfer coefficient, kmol m ⁻² s ⁻¹ kPa ⁻¹		
k_L^0	liquid phase mass transfer coefficient with no chemical reaction, m s ⁻¹		
k_L	liquid phase mass transfer coefficient, m s ⁻¹		
k_2, k_H^*	reaction rate constant, kmol ⁻¹ m ³ s ⁻¹		
k^{-1}	reaction rate constant, s ⁻¹		
L	liquid flow rate, m ³ m ⁻² s ⁻¹		
N_A	mass transfer rate, kmol m ⁻² s ⁻¹		
		Greek letter	
		α	Loading of CO ₂ in MEA, mol mol MEA ⁻¹
		a_e	gas–liquid interfacial area, m ² m ⁻³
		μ	dynamic viscosity, Pa s
		δ	thickness of liquid film, m
		Subscript	
		A_i, B_i	component A, component B in the interface
		A_L, B_L	component A, component B in the main body of liquid
		G	gas phase
		L	liquid phase

effect of coefficient orifice parameter and stirring rate on the average absorption velocity was also presented. Palmeri et al. [16] carried out experimental research on CO₂ separation for biogas purification. A first-order semi-empirical model was proposed for monoethanolamine concentration, which described the absorption of CO₂ in MEA as a function of temperature, CO₂ flow, and time. However, research on mass transfer performance, especially the enhancement factor, of high-concentration CO₂ absorption by an ethanolamine (MEA) aqueous solution in a packed tower has not yet been reported.

Furthermore, with respect to the kinetics of CO₂ absorption in MEA, previous studies have almost always been based on the pseudo-first-order reaction [17,18]; nevertheless, for the high-concentration CO₂ absorption by MEA in a packed tower, the variation in absorbent concentration along the height of the tower is significant enough that the assumption of pseudo-first-order reaction is no longer reasonable and needs to be discussed further.

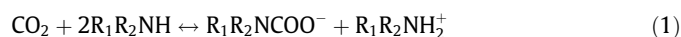
Therefore, an enhancement factor mathematical model, which was based on the second-order reaction and could be used to calculate the enhancement factor of high-concentration CO₂ absorption by MEA, was built. A series of experiments were conducted in an experimental packed tower in this work. The enhancement factor of the mathematical model was verified by the experimental results, and the mass transfer performance of high-concentration CO₂ absorption by MEA aqueous solution was analysed. The results of this study could provide important insights for the research and practical application of biogas purification technology.

2. Theoretical section

2.1. Reaction mechanism

In aqueous solutions of primary alkanolamines, the following reactions with CO₂ occur:

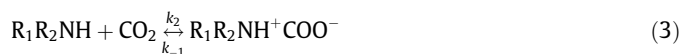
carbamate formation:



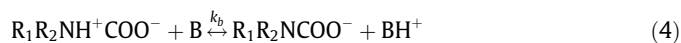
bicarbonate formation:



The carbamate formation above by reaction (1) takes place in accordance with the approach given by Danckwerts [19], which is derived from Caplow's work [20]. The reaction steps successively involve the formation of a “zwitterion”:



The subsequent removal of the proton by a base B (base catalysis) [21]:



For the reaction rate of reaction (3), Danckwerts derived the forward reaction rate equation at the quasi-steady state [19]:

$$r_{(1)} = \frac{k_2[\text{CO}_2][\text{R}_1\text{R}_2\text{NH}]}{1 + k_{-1}/\sum k_b[\text{B}]} \quad (5)$$

where $\sum k_b[\text{B}]$ indicates the contribution to the proton removal reaction (4) by all bases present in the solution.

Similar to the case of MEA, Danckwerts [19] indicated $k_{-1}/\sum k_b[\text{B}] \ll 1$. Thus, a simple second-order kinetics that ignores the contribution of reaction (2) could be approximated as:

$$r = k_2[\text{CO}_2][\text{R}_1\text{R}_2\text{NH}] \quad (6)$$

The kinetic constant k_2 in reaction (3) was proposed by Blauwhoff et al. [22]:

$$\lg k_2 = 10.99 - 2152/T \quad (7)$$

2.2. Mathematical model of the enhancement factor

When the second-order irreversible reaction takes place between absorbate A and absorbent B:



The liquid film diffusion differential equations for the reaction are written as:

$$\frac{d^2 c_A}{dx^2} = \frac{k_2 c_A c_B}{D_{AL}} \quad (9)$$

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