



Mass transfer and energy consumption for CO₂ absorption by ammonia solution in bubble column



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HIGHLIGHTS

- Both the CO₂ capture and ammonia escape in the BCR are investigated.
- Influences of the BCR geometries and operating conditions are obtained.
- Increased orifice size can enhance CO₂ mass transfer and restrain ammonia escape.
- Consumed energy increases with increasing operating pressure, orifice number, size.
- Recommended height-to-diameter ratio is 5.76 for energy consumption and CO₂ removal.

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ABSTRACT

The CO₂ absorption in the bubble column using ammonia solution has been proved a viable approach for the post-combustion CO₂ capture, so it is of benefit to the energy saving of CO₂ absorption and the bubble column design to clarify the impacts of the geometry and running parameters on the mass transfer and energy consumption of CO₂ capture. Based on the representative elementary volume method, a computational model of both the CO₂ capture and ammonia slip was developed and validated by the experimental data, by which the hydrodynamic characteristics, mass transfer performances and energy consumption in CO₂ capture process were investigated. The results show that at the same inlet velocity of CO₂, the higher height-to-diameter ratio of the column can improve the mass transfer performances of both the CO₂ capture and ammonia escape. What's more, the energy consumption decreases with the reduced orifice number on the bottom of column, and increases as the orifice size increases. An optimal height-to-diameter ratio of 5.76 is recommended when taking both the energy consumption and CO₂ removal efficiency into consideration. This work can provide the viable guidance and suggestions of the better mass transfer performance and energy saving for the industrial application of CO₂ capture, as well as the inhibition of ammonia escape.

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1. Introduction

The climate change has become a serious environmental issue all over the world, which is mainly caused by the emission of greenhouse gases (GHGs). Carbon dioxide (CO₂) is regarded as one of the most important GHGs [1]. As the excessive consumption of coal, the annual emission from the coal-fired power plants occupies nearly 35% of the CO₂ emission in the world [2]. The anthropogenic CO₂ emission accounts for 80% of the total GHGs and makes a significant contribution to the global climate change [3,4]. The carbon capture and storage (CCS) is an essential measure

against the CO₂ discharge from the coal-fired power plants and other related industries, whereas with the tremendous energy consumption.

Various approaches have been proposed to deal with the CO₂ emission, including physical adsorption, chemical absorption, membrane separation, cryogenic methods, biological fixation, photocatalytic reduction and O₂/CO₂ combustion [5–11]. Among these methods, the chemical absorption is regarded as an alternative and potential choice with the high efficiency and mature technology [12]. Due to the higher absorption capacity than the monoethanolamine (MEA) at the same operating temperature and pressure, the aqueous ammonia is basically applied to the chemical absorption [13–17]. Farid et al. [17] studied the aqueous ammonia absorption in comparison with aqueous amine, finding that the aqueous

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Nomenclature

$C_{CO_2,G}$	CO ₂ mass concentration in gas phase, dimensionless	S_{NH_3}	source term of the NH ₃ mass fraction equation in liquid phase, kg/m ³ /s
$C_{HCO_3,L}$	NH ₄ HCO ₃ mass concentration in liquid phase, dimensionless	$S_{NH_3_Esc}$	source term of the NH ₃ mass fraction equation in gas phase, kg/m ³ /s
$C_{NH_3,G}$	NH ₃ mass concentration in gas phase, dimensionless	t	time, s
$C_{NH_3,L}$	NH ₃ mass concentration in liquid phase, dimensionless	u_G	averaged velocity vector of gas phase, m/s
D_{G,CO_2}	molecular diffusivity of CO ₂ in gas phase, m ² /s	u_L	averaged velocity vector of liquid phase, m/s
D_{G,NH_3}	molecular diffusivity of NH ₃ in gas phase, m ² /s	V_L	volume of the solvent in BCR, L
D_{L,HCO_3}	molecular diffusivity of NH ₄ HCO ₃ in liquid phase, m ² /s	W	total energy consumption, dimensionless
D_{L,NH_3}	molecular diffusivity of NH ₃ in liquid phase, m ² /s	W_C	energy consumption of compressor, kW
E	enhancement factor, dimensionless	W_f	energy consumption of fan, kW
F_G	momentum source term of gas phase, N/m ³	$X_{CO_2,i}$	CO ₂ concentration at the interface, kmol/m ³
F_L	momentum source term of liquid phase, N/m ³	$X_{CO_2,L}$	CO ₂ concentration in liquid phase, kmol/m ³
g	gravitational acceleration, m/s ²	X_{NH_3}	NH ₃ concentration in liquid phase, kmol/m ³
G_F	gas flow rate, kmol/s	$X_{NH_3,i}$	NH ₃ concentration at the interface, kmol/m ³
$G_{CO_2,in}$	CO ₂ gas molar flow rate, kmol/s	Y^{CO_2}	molar ratio of CO ₂ to N ₂ , dimensionless
G_{N_2}	N ₂ molar flow rate, kmol/s	y_1, y_2	CO ₂ mole ratio in gas phase entering and leaving BCR, dimensionless
H_{CO_2}	Henry's constant of CO ₂ , kPa m ³ /kmol	$y_{NH_3,out}$	NH ₃ mole ratio in gas phase at outlet of BCR, dimensionless
$K_G^{CO_2,A}$	volumetric overall mass transfer coefficient of CO ₂ absorption, kmol/(L s kPa)	$y_{N_2,out}$	N ₂ mole ratio in gas phase at outlet of BCR, dimensionless
$K_G^{NH_3,A}$	volumetric overall mass transfer coefficient of ammonia escape, kmol/(L s kPa)	Y_{out}	NH ₃ to N ₂ mole ratio in the gas phase at the outlet of the BCR, dimensionless
k_{G,NH_3}	gas phase mass transfer coefficient of NH ₃ , m/s	z	number of compression stage, $z = 1$
k_{L,CO_2}	liquid phase mass transfer coefficient, m/s	<i>Greek symbols</i>	
k_{L,NH_3}	liquid phase mass transfer coefficient of NH ₃ , m/s	α_G	volume fraction of gas phase, dimensionless
$p_{CO_2,i}$	partial pressure of CO ₂ in main gas, kPa	α_L	volume fraction of liquid phase, dimensionless
p_{in}	inlet pressure of compressor, kPa	δ_G	thicknesses of gas film, m
p_{NH_3}	partial pressure of NH ₃ in main gas, kPa	δ_L	thicknesses of liquid film, m
$p_{NH_3,i}$	partial pressure of NH ₃ at the interface, kPa	ε	gas hold up, dimensionless
p_{out}	outlet pressure of compressor, kPa	η	CO ₂ removal efficiency, dimensionless
Q_L	volumetric flow rate of gas phase, m ³ /s	η_c	efficiency of compressor, $\eta_c = 0.85$
R_{CO_2}	mean chemical reaction rate of CO ₂ absorption in BCR, kmol/s/L	η_p	efficiency of fan, $\eta_p = 0.8$
$S_{CO_2,G}$	source term of CO ₂ mass fraction equation, kg/m ² /s	μ_G	viscosity of gas phase, Pa s
S_{HCO_3}	source term of NH ₄ HCO ₃ mass fraction equation in liquid phase, kg/m ² /s	μ_L	viscosity of liquid phase, Pa s
S_G	source term of the continuity equation for gas phase, kg/m ³ /s	ρ_G	density of gas phase, kg/m ³
S_L	source term of the continuity equation for liquid phase, kg/m ³ /s	ρ_L	density of liquid phase, kg/m ³

ammonia requires a much lower energy for regeneration. Moreover, SO₂ can be simultaneously absorbed by the aqueous ammonia [18]. Yu et al. [19] and Li et al. [20] confirmed the technical feasibility of the CO₂ capture by aqueous ammonia solution based on the pilot plant trials. Han et al. [21] concluded that the waste heat recovery system can supply the energy for the regeneration of absorbent solution. Bonalumi and Giuffrida [22] proposed the integration of CO₂ capture at three different levels, showing that the specific primary energy consumption for the 90% of CO₂ avoided was as low as 2.3 MJ/(kgCO₂). Though the CO₂ removal by ammonia is treated as the most viable technology in practical engineering thanks to its high removal efficiency and adequate adaptability [23], it suffers the ammonia escape disadvantage, which leads to the ammonia loss and secondary atmosphere pollution [24]. Moreover, the absorbing and ammonia escape processes are affected by the reaction conditions and reactor configuration, so should be further investigated.

The conventional gas and liquid reactors have been investigated for several decades. Especially, the packed tower and bubble column have been successfully applied to the post-combustion CO₂ capture in pilot and industrial scales [25]. The packed column is good at the lower pressure drop [26], but inferior to the bubbling

reactor in mass transfer performance [27]. Thanks to its high heat and mass transfer rates and low operation and maintenance costs, the bubble column reactor (BCR) has been widely used in chemical and power engineering [28]. However, the fundamental studies of CO₂ capture in bubble column reactors mainly focused on the CO₂ removal efficiency or the ammonia escape inhibition, and the mass transfer of ammonia escape process as well as CO₂ absorbing process was rarely involved. Although the geometry of BCR is relatively simple, it is difficult to predict the mass transfer and energy consumption inside the BCR due to the complex multiphase hydrodynamics and reaction mechanism. The mass transfer performance and energy consumption are influenced significantly by the operating conditions, gas and liquid properties and the configuration of BCR. Bai and Yeh [13,29] studied the CO₂ capture by ammonia in BCR, finding that the removal efficiency can reach 95%. Zhao et al. [4] experimentally investigated the influence of BCR geometry on the CO₂ capture efficiency, pointing out that the height-to-diameter ratio plays an important role in CO₂ capture. Zhang et al. [30] investigated the ammonia escape in a super-gravity bed with disks, finding that the outlet molar fraction of ammonia arrived at 1.3–3.6%. Ma et al. [27] studied the impacts of mass transfer of ammonia escaping, and obtained the volumetric mass

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