

Experimental and numerical study on CO₂ absorption mass transfer enhancement for a diameter-varying spray tower

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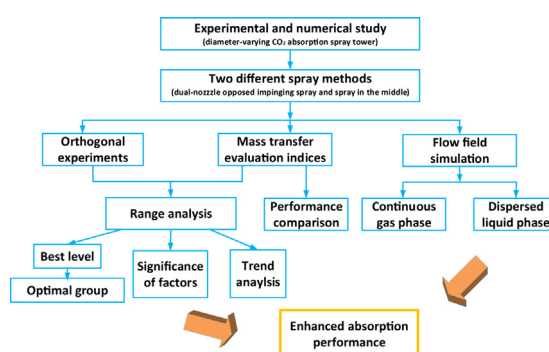
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HIGHLIGHTS

- Orthogonal tests were used to evaluate importance of operating parameters in CO₂ capture.
- Two different spray methods were discussed for performance comparison.
- Optimal conditions for different evaluation indices were obtained by range analysis.
- MEA and CO₂ concentrations were major factors affecting absorption performance.
- CFD simulation was performed to illustrate the enhancement of gas-liquid flow field.

GRAPHICAL ABSTRACT



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ABSTRACT

In this research article, orthogonal experiments were performed in a proposed diameter-varying spray tower system to evaluate the importance of operating parameters and analyse the relationship between factors and the mass transfer evaluation indices (the CO₂ removal rate and overall absorption rate). Two different spray methods such as dual-nozzle opposed impinging and spray in the middle method were discussed for performance comparison. Optimal operating conditions for different evaluation indices were obtained by using range analysis. After analysis it was found that the absorption performance was mainly affected by two parameters, monoethanolamine concentration and CO₂ concentration. The effects of different operating parameters on the CO₂ removal rate and overall absorption rate were also discussed by using trend analysis. Furthermore, gas phase mass transfer coefficient and effective contacting area were determined by experimental data. A comparison with reported conventional reactor systems demonstrated a great application potential for proposed spray tower in CO₂ capture. Finally, simulation results were illustrated which shows that the gas phase and liquid phase distributions for the dual-nozzle opposed impinging spray method enhance CO₂ absorption performance.

1. Introduction

Carbon dioxide (CO₂) emission from fossil fuel combustion makes a major contribution to the global warming and greenhouse gas effect [1–4]. Global warming leads to negative impacts like increasing global

average temperature, elevating sea level, coastal region erosion, endangerment of arctic species, and increasing mortality associated with extreme weathers [5,6]. These side effects are proven to be catastrophic for both natural ecosystem and human civilization. Hence, CO₂ reduction become a vital international scientific and environmental issue

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Nomenclature

a_e	effective contacting area, $\text{m}^2 \text{qm}^{-3}$	k_1	reaction rate constant, s^{-1}
C	molar concentration, kmol m^{-3}	P	pressure, kPa
C_G	CO_2 concentration, vol%	Q_G	gas flow rate, $\text{m}^3 \text{h}^{-1}$
C_L	MEA concentration, wt%	Q_L	gas flow rate, L h^{-1}
D	diffusion coefficient, $\text{m}^2 \text{s}^{-1}$	q_G	molar gas flow rate, $\text{kmol m}^{-3} \text{h}^{-1}$
d	diameter, m	S	cross-sectional area, m^2
H	Henry's coefficient, $\text{kPa m}^3 \text{kmol}^{-1}$	T	temperature, $^\circ\text{C}$
K_G	overall mass transfer coefficient of gas phase, $\text{kmol m}^{-2} \text{h}^{-1}$	V_r	reactor volume, m^3
$K_G a_e$	volumetric overall mass transfer coefficient, $\text{kmol m}^{-3} \text{h}^{-1} \text{kPa}^{-1}$	Y_1, Y_2	inlet and outlet mole ratio in gas phase
k_G	gas phase mass transfer coefficient, $\text{kmol m}^{-2} \text{h}^{-1}$	y_1, y_2	inlet and outlet mole fraction in gas phase
k_L	liquid phase mass transfer coefficient, $\text{kmol m}^{-2} \text{h}^{-1}$	β	enhancement factor of chemical reaction
		η	CO_2 removal rate
		λ_1, λ_2	proportionality coefficient
		Φ	overall absorption rate, $\text{kmol m}^{-3} \text{h}^{-1}$

[7,8].

Carbon capture and storage (CCS) technology has been recognized as a promising method to significantly reduce CO_2 emissions from large-point sources such as power plants and heavy manufacturing industries [9,10]. Compared with other CCS methods, post-combustion carbon capture is a better route in near and middle term on account of its economic and feasible retrofit for existing plants without radical changes [11–14]. Meanwhile, chemical absorption with monoethanolamine (MEA) absorbent is the most well-established technology for post-combustion carbon capture [15,16].

The packed tower, one of the conventional gas-liquid reactor for CO_2 chemical absorption process, was commonly used absorber on both lab and industrial scale systems [17–19]. According to number of meaningful conclusions, it was concluded that this technology was not yet fully developed for large-scale industrial applications and may not be economically competitive because of its high capital and operating costs [20–22]. There are some drawbacks for packed tower like high gas-phase pressure drop, liquid channeling and flooding and deposition onto packing material [23]. Therefore, it is necessary to develop efficient gas-liquid reactors for CO_2 capture process to enhance the separation performance.

In the past decades, spray tower has been mainly used for sulfur dioxide removal. Theoretical and experimental studies were conducted to investigate the promising features like simple construction, lower pressure drop, large handling capacity, low investment and maintenance cost [24–26]. Despite being widely used in the desulfurization process, the application of spray tower for CO_2 scrubbing process is a relatively recent development trend. Some research has been published on spray absorption of CO_2 by NaOH and ammonia. However, less attention is paid on the utilization of spray tower in CO_2 scrubbing process with aqueous amine solvents so far. Kuntz and Aroonwilas [27,28] compared the mass transfer performance of spray tower with packed tower from lab-scale CO_2 absorption with MEA solvent. The spray tower was declared as a very promising reactor for CO_2 capture due to its higher overall mass transfer coefficient. Niu et al. [29] conducted experiments by using spray tower to study CO_2 absorption with MEA solution and investigated effects of different operating parameters. Experimental results showed that the mass transfer performance of scrubbing process was improved by ejecting absorbent through the nozzle. It was also found that the spray tower achieved more than 95% CO_2 removal rate. Seyboth et al. [30] proposed a bench-scale two-way experimental approach to investigate the absorption of CO_2 by single spray droplet and demonstrate the applicability of spray scrubbing process. The results showed that effective contacting area is crucial for CO_2 absorption. Tamhankar et al. [31,32] provided the experimental data on droplet size distribution for amine sprays and quantified the available surface area from droplet size measurements for the first time. Koller et al. [33] investigated a larger pilot spray scrubber plant having

gas flow rate up to $160 \text{ m}^3/\text{h}$. They found that the measured absorption performance achieved by using single nozzle was not sufficient. It was proposed that spray tower design was improved in future by using multi-nozzles. The available literature review has revealed the feasibility of spray tower used in CO_2 capture process with amine-based solvents because the existence of spray nozzle significantly increases interfacial surface area between absorbent and flue gas [34].

The mass transfer performance of traditional spray tower, however, is not high enough for industrial application. Most of the previous experiments were conducted in a cylindrical tower by using a single spray nozzle. The configuration of this cylindrical tower was different from that used in actual industry makes the results of these studies far away from application. Keeping in view the above discussion, the current study has been carried out for the enhancement of CO_2 absorption process by using an improved diameter-varying spray tower. As previously studied, CO_2 absorption in spray tower mainly occurs in the nozzle exit, hence increasing the space of nozzle exit is a feasible way to enhance the absorption performance [30,33]. The reaction sections of the proposed diameter-varying spray tower comprising of major two parts namely: the cylindrical section and the conical section. The existence of the conical section would increase the effective contacting area and gas-liquid contacting time, which will improve the absorption performance. Some interesting results have been obtained by using the proposed diameter-varying spray tower in our published work [35,36].

The effects of operating parameters such as MEA concentration, liquid flow rate, CO_2 concentration and gas flow rate on the CO_2 capture performance has been discussed by the single variable experimental method in many researches, but few focused on the degree of significance of the factors on absorption performance [27,37–39]. Liao et al. using orthogonal tests to study the mass transfer performance (in terms of volumetric overall mass transfer coefficient) of blended DEEA-MEA solution for capturing CO_2 in a lab-scale packed reactor and obtained the influence degree order of different operating parameters [40]. It verified the possibility of orthogonal experimental method using in CO_2 absorption system. However, little attention has been paid to the spray tower. In this research, orthogonal experiments were conducted to evaluate the importance of factors (operating parameters) and analyse the relationship between factors and the mass transfer evaluation indices (the CO_2 removal rate and overall absorption rate) for the proposed diameter-varying spray tower. Range analysis has been carried out to obtain the optimal conditions for different performance evaluation indices and indicate the significance of the factors on CO_2 absorption performance. The relationship between the mean values of each factor and the evaluation indices was obtained by trend analysis. Two different spray methods were discussed in this work. One is the dual-nozzle opposed impinging spray method proposed by our research group, the other is mostly literature discussed spray in the middle method. Orthogonal test were also performed in order to compare the

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