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Post-combustion CO_2 capture with ammonia by vortex flow-based multistage spraying: Process intensification and performance characteristics

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ABSTRACT

To improve the process and performance of CO_2 capture with ammonia by chemical absorption, a vortex flow-based multistage spray reactor was designed to evaluate the enhancement effect for post-combustion CO_2 capture with ammonia. The process intensification analysis based on flow patterns from a CFD (computational fluid dynamics) simulation indicated that the vortex flow presented multi-dimensional velocities including a V-shaped tangential velocity profile and non-uniform axial velocity profile, which resulted in enhancement of gas–liquid contact, mixing, mass transfer, and reaction compared to non-vortex flow. Furthermore, the CO_2 capture characteristics were examined at varied operating parameters. It was found that the capture efficiency E increased with increasing ammonia concentration and liquid flow rate but decreased with increasing CO_2 inlet concentration and gas flow rate. Meanwhile, the overall gas phase mass transfer coefficient K_ga increased with increasing ammonia concentration, liquid flow rate, and gas flow rates but decreased with increasing CO_2 inlet concentration. Within the measured range, the E and K_ga varied from 72.05 to 86.72% and $0.31–0.49 \times 10^{-3}$ kmol/m^3 kPa s, respectively. Importantly, vortex flow presents relative enhancements of 7–15% in E and 18–33% in K_ga compared with non-vortex flow depending on the operating parameters.

1. Introduction

Anthropogenic CO_2 emissions have been demonstrated to be responsible for greenhouse effect and global warming, and have become an important issue in relation to energy and the environment. Statistics indicated that CO_2 concentrations have increased by 40% since pre-industrial times, primarily due to fossil fuel combustion [1,2]. Considering the current large-scale utilization of fossil fuels, it is necessary to develop stable, safe, and environmentally acceptable approaches and technologies for CCS (CO_2 capture and storage). Compared with pre-combustion and oxy-fuel combustion technology, large decreases in post-combustion CO_2 emissions via capture and separation are urgently needed to maintain CO_2 at controllable levels [3–5]. Among the many post-combustion CO_2 capture approaches, e.g., physicochemical adsorption, membrane separation, cryogenics, chemical looping combustion and biological sequestration [6], chemical absorption via gas–liquid contact scrubbing is considered to be a viable and mature option for gas separation and purification, including CO_2 capture [7].

Over the last few decades, CO_2 capture approaches via gas–liquid contact and chemical absorption have been successfully developed in both bench-scale investigations and pilot-scale applications. These approaches include: bubble columns [8,9], packed towers [10–14] and spray columns with the use of: air–CO_2/NaOH [15–19], air–CO_2/H_2O [18,19], and air–CO_2/Na_2CO_3/$NaHCO_3$ [20–22]. Capture efficiencies of these systems have been found to be in the range of 55–95% [23]. However, process intensification and performance improvement of these conventional gas–liquid mass transfer processes still face great challenges, particularly due to mass transfer limitations that hinder improvement of CO_2 capture performance and do not allow for device size reduction.

To enhance performance in the CO_2 capture process, a series of methods based on extreme physical environment improvement have been proposed in recent years, including: hollow fiber membrane-based [24,25], high gravity-based [26–28], and impact stream-based approaches [29,30]. Experiments indicated that these approaches were able to achieve high CO_2 capture

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performances with 50–99% CO_2 capture efficiencies found at the specific operating conditions. However, the main focus of most these studies has not been comprehensively understood, namely the influence of operating parameters on both CO_2 capture efficiency and mass transfer rate. Moreover, these approaches usually include high-performance media which requires relatively complicated fabrication and high cost, or often have rotors and coupled structures that require high energy consumption, or strongly depend on harsh operating environments such as high pressure that partially cause limitations of development and application.

Gas-liquid contact and reaction via vortex gas flow is an effective and simple way to achieve both process intensification and energy consumption reduction. This technique was originally used via co-current flow to enhance the capture of low-concentration pollutants, including successfully capturing SO_2 with water-based ash [31,32], SO_2 with NaOH, and also H_2S–CO_2 with alkanolamine [33,34]. The mass transfer rate for an air–NH_3–H_2O physical absorption system was demonstrated to be increased by 20% for vortex gas flow in a laboratory-scale spray tower compared with that using axial gas flow [35]. Similar attempts were made for CO_2 capture more recently. The enhancing effect based on vortex flow increased the overall gas volumetric mass transfer coefficients by 31–49% for a NaOH-based CO_2 (2.5%, v/v) system in a laboratory-scale spray tower [36] and, increased the capture efficiency by 20% (from 50% to 70%) for a NaOH-based CO_2 (1.5%, v/v) system in a reverse-flow cyclone contactor [37], respectively.

Nevertheless, these studies mainly focused on the capture of low-concentration CO_2 (<2.5%, v/v). The results and conclusions would not be applicable to the capture of CO_2 from post-combustion flue gas, which usually has higher concentrations between 10 and 15% (v/v). Also, most of the previous investigations used NaOH solution as an absorbent, which does not have a good regeneration potential. Hence, understanding the behavior of CO_2 capture with renewable, recyclable, and advanced absorbents is necessary. Aqueous ammonia, an alternative absorbent, has advantages including: high absorption capacity, low energy requirement for regeneration compared to NaOH, good inoxidability compared to alkanolamine, and a wide distribution of sources. Aqueous ammonia-based CO_2 capture has received more attention in recent years, but its availability when using gas vortex flow needs to be further evaluated. Moreover, in the view of multiphase flow, the co-current flow used in the most previous investigations is considered to be limited to improvement of mass transfer and reaction rate. Also, the mechanism of the vortex gas flow pattern has not yet been completely understood. Questions remain relating to the gas velocity distribution inside the reactor and its effects on enhancement of capture efficiency and mass transfer. Additionally, the single stage spraying used in most of the studies proved unfavorable to enhancement of mass transfer driven by concentration gradient of the liquid phase.

In this work, a novel vortex flow-based multistage spray gas–liquid reactor was designed to achieve an improved process enhancement configuration for CO_2 capture. The countercurrent vortex gas flow was generated by a rectangular cross-section inlet that helped enhance mixing and reduce axial decay of the vortex flow. The liquid phase was sprayed using a multistage arrangement to overcome problems caused by the single-stage spray such as reduction of the mass transfer driving force due to a reduction of the absorbent concentration gradient, and also reduction of the wall-film effect due to diffusion of the cone-jet atomization. This configuration has the advantages of simple structure, low cost, and easy scale-up. Using the vortex flow-based multistage spraying reactor, we employed aqueous ammonia solutions as the absorbents to examine the feasibility of capturing high concentration CO_2 from simulated flue gas. Firstly, we explored the process intensification effect of gas vortex flow on fluid flow and mass transfer using a CFD (computational fluid dynamics) approach. Consequently, we addressed the response characteristics between the influence factors and CO_2 capture performance. Also, the CO_2 capture performance was analyzed and compared with the performance of the reactor when operating as a spray reactor with non-vortex gas flow, a bubble column reactor, a two-stage spray reactor with axial flow, and a conventional packed bed reactor.

2. Process chemistry

CO_2 capture with ammonia is usually accompanied by a series of complex multi-step reactions [38–42], as described in Eqs. (1)–(5). The overall reaction is usually expressed by Eq. (6).

$$2NH_3 + CO_2 \rightleftharpoons NH_2COONH_4 \tag{1}$$

$$NH_2COONH_4 + H_2O \rightleftharpoons NH_4HCO_3 + NH_3 \tag{2}$$

$$NH_3 + H_2O \rightleftharpoons NH_4OH \tag{3}$$

$$NH_4HCO_3 + NH_4OH \rightleftharpoons (NH_4)_2CO_3 + H_2O \tag{4}$$

$$(NH_4)_2CO_3 + CO_2 + H_2O \rightleftharpoons 2NH_4HCO_3 \tag{5}$$

$$NH_3 + CO_2 + H_2O \rightarrow NH_4HCO_3 \tag{6}$$

The reaction (6) can be considered to be irreversible and, the kinetic parameter can be modeled using the Arrhenius equation based on a single equation reaction mechanism [43–46].

$$\ln k = \ln A - E_a/RT \tag{7}$$

where $\ln A = 9.4867\text{–}25.6278$, $E_a = 26.730\text{–}48.529$ kJ/mol ($T = 0\text{–}50$ °C).

3. Experiment and methods

3.1. Experimental setup

The experiments were conducted in a laboratory-scale system as shown in Fig. 1. Simulated flue gas containing varied concentration CO_2 (balanced with N_2 which is the primary fixed component in flue gas) was flowed into the bottom of the reactor while the absorbent solution (aqueous ammonia) was sprayed using atomizers. The absorbent solution was supplied to the nozzle by a centrifugal pump and had characteristics of: solid cone with a nominal cone angle of 30° and, initial mean droplet diameter of 125–75 μm which decreased with increasing working pressure (0.7–1.0 MPa) and liquid flow rate within the experimental range. The gas and liquid flow rates were controlled by a series of rotameters. Moreover, a gas dryer and a gas-washing bottle of sulfuric acid solution were used to prevent the volatilized absorbent and water vapor from entering into the simulated flue gas monitored. Finally, CO_2 concentration was measured using an online infrared flue gas analyzer (Gasboard-3000, ±1% measuring accuracy), which was calibrated with a relative CO_2 concentration uncertainty of less than 5%.

The vortex flow-based multistage spray reactor is schematically illustrated in Fig. 2. It consisted of a perspex cylinder of 50 mm in diameter. Gas vortex flow in the reactor was generated by guiding the gas via a tangential inlet with rectangular cross-section to enhance the gas–liquid turbulence and mixing. Also, it was