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The CO_2 capture performance of a high-intensity vortex spray scrubber

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ABSTRACT

The present study focuses on the enhancement of CO_2 capture efficiency using a high-intensity vortex spray scrubber by imparting swirl to the gas flow, which has the ability to augment the rates of heat and mass transfer. Experimental investigations into the reactive absorption of CO_2 from a mixture of air–CO_2 into an aqueous solution of NaOH in a laboratory-scale counter-current spray scrubber have been carried out. The mass transfer characteristics, in terms of the overall gas phase mass transfer coefficient (K_ga) were investigated for both the swirling and the non-swirling (axial) gas flows through the scrubber in order to quantify the effect of swirl. The effects of the gas/liquid flow rates, flow arrangements, scrubber height and spray nozzle type on the CO_2 capture performance were examined. For both the axial and the swirling flows, the K_ga increases initially with increasing gas flow rate up to a certain limit, beyond which it becomes essentially constant, whereas the K_ga increases continuously with the liquid flow rate within the measured range. The counter-current gas–droplets flow provides higher mass transfer rates compared with those in co-current flow. The K_ga deceases with the increase in the tower height. The spray nozzle producing finer droplets provides enhanced mass transfer rates. It is found that imparting swirl in the gas flow enhances the K_ga up to around 49% compared with that in axial flows.

1. Introduction

In recent years, there has been increasing demand for a significant abatement of carbon dioxide (CO_2) emissions from industrial sources in order to reduce global temperature rise. The main industrial sources of CO_2 are the thermal power plants fired with fossil fuels, incinerators and refineries. Amongst these sources, coal-fired power plants generate over 33% of the CO_2 emissions [1]. As cited in Ref. [2], according to the International Energy Agency *World Energy Outlook* (2002), the predicted increase in combustion generated CO_2 emissions is around 1.8% per year and by 2030 it will be 70% above 2000 levels. The global CO_2 emission is increasing by more than 3.3% per year [3]. This problem will be exacerbated because of the expected increase in burning of fossil fuels, in particular coal, for power generation in order to sustain the economic growth of developing countries around the world. There is therefore an urgent need for research into the development of viable and cost-effective technologies for the capture of CO_2 from flue gases emitting from power plants for sequestration in geological features (e.g., natural gas wells and deep sea).

In principle, the gas separation processes used in the chemical industries, such as absorption (scrubbing) in a chemical solvent with solvent regeneration via thermal stripping (desorption), adsorption in a solid adsorbent with thermal/pressure-swing regeneration, membrane and cryogenic separation, can be adapted for the post-combustion CO_2 capture from thermal power plants. New technologies, including photocatalytic and biological processes and chemical synthesis, are also under development. Pre-combustion CO_2 capture as in the integrated gasification and combined-cycle (IGCC) plants and combustion using pure oxygen instead of air (known as the oxyfuel combustion) for the production of sequestration-ready CO_2 are also being considered for this purpose. A collection of papers published in a special issue of *Industrial and Engineering Chemistry Research* [45(8), 2006] provides a good insight into the current status and future trends of post-combustion CO_2 capture technologies. However, most gas separation technologies mentioned above are not yet fully developed for the deployment for CO_2 removal from large quantities of flue gases emanating from thermal power plants and they may not be economically competitive. A viable option, at least for the short- and medium-term, appears to be the scrubbing/stripping process carried out in a suitable gas–liquid contactor using aqueous alkanolamine solutions (e.g., monoethanolamine and methyldiethanolamine or mixed amines) [2,4]. This process carried out in packed towers is a well-establish technology being used widely in the chemical industries and refineries for more than 60 years. Although this is a mature technology, there remains

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considerable scope for improving the process performance via the development of innovative configurations of scrubbing and stripping equipment and new chemical solvents.

In order to treat large volumes of flue gas (e.g., a typical 400 MW coal-fired power plant produces approximately 1.1×10^6 N m^3/h of gas [5]) with CO_2 concentration ranging from 3 to 15% (typically 3–5% for natural gas and 13–15% for coal combustion), huge size packed towers with a large diameter will be required involving high capital and maintenance costs. Packed towers also suffer from various operational problems including high gas phase pressure drop, liquid channelling and flooding, disintegration of packing materials caused by high temperature gases, and deposition on packings and clog up of void spaces by solid-laden gases. These problems are likely to occur in the flue gas scrubbing operations, which involve low liquid flow rates and high superficial gas velocities because of the large gas volumes with relatively low CO_2 concentrations and require low pressure drop as the flue gas fan typically operates slightly above the atmospheric pressure. In the flue gas scrubbing applications, a spray tower, which is a gas–liquid contacting device where a liquid solvent is sprayed as fine droplets in a continuous gas stream through an empty column, can offer a number of advantages over packed towers. These include low gas phase pressure drop, a wide range of gas to liquid loading ratios, simpler mechanical construction, and low equipment and maintenance costs [6,7]. Spray towers are already in use for the removal of SO_x via flue gas desulphurisation (FGD) in coal-fired power plants [8] and cleaning of acid gases emitted from furnaces and incinerators [7]. However, there are some disadvantages associated with this device such as low mass transfer rates resulting from the rapid reduction of interfacial area due to the droplets coalescence and impingement on the tower wall forming a liquid film, and high pumping cost for forcing the solvent through the spray nozzles for fine atomisation.

In the past, many experimental studies have been carried out to examine the mass transfer characteristics of spray towers for various gas–liquid systems, which include air–CO_2/NaOH [6,7,9,10], air–CO_2/H_2O [7,10], air–CO_2/Na_2CO_3/$NaHCO_3$ [11], air–SO_2/NaOH [6,9,11,12], and air–NH_3/H_2O [13–18]. The mass transfer performance of a spray tower depends not only on the operating conditions, gas/liquid flow directions and the physical properties of the fluids, but also strongly on the solvent spray characteristics, such as the liquid distribution and the droplets size and its distribution, which are in turn influenced by the spray nozzle specifications [19]. However, the main focus of most previous studies has been the understanding the influence of gas and liquid flow rates and tower height on the mass transfer rate.

In recent years, there have been renewed interests in studying mass transfer characteristics of the CO_2 absorption processes in various solvents with potential applications for the post-combustion capture of CO_2 from flue gases. These studies have been carried out mainly in packed towers with different types of packing using aqueous solutions of NaOH and monoethanolamine (MEA) [e.g., 20–23] and, to a much lesser extent, in spray towers [24,25] (also see earlier studies cited above). Recently, Kuntz and Aroonwilas [24] have examined the performance of a small-scale spray tower for the removal of CO_2 from air–CO_2 mixture using aqueous solutions of MEA for a range of process conditions. The comparison between the performances of the spray tower and a packed tower of the same height reveals that the former is capable of removing CO_2 at a higher rate. Stolaroff et al. [25] have studied removal of CO_2 from atmospheric air using NaOH sprays in a large-scale spray contactor.

In the past, a few attempts have been made to enhance the mass transfer efficiency using rotating (or swirling) gas flow in high-intensity cyclone spray towers. These include early studies in the 1930s and 1940s on the removal of ash and SO_2 from boiler flue gases using water spray [26,27] and the absorption of SO_2 in sprays of NaOH solutions [28] in co-current towers, and a later study on the selective absorption of H_2S and CO_2 from a mixture of N_2–H_2S–CO_2 into alkanol amine solutions in a co-current spray tower [29]. These studies, however, did not compare between the mass transfer rates in cyclone spray towers and the conventional spray towers using axial (i.e., non-swirling) gas flow. In our previous study [18], we have demonstrated that using swirling gas flow in a laboratory-scale spray tower, referred to as the *vortex spray scrubber*, the rate of mass transfer for the air–NH_3/H_2O system can be enhanced by 20% compared with that using axial gas flow.

This paper reports an experimental investigation into the CO_2 capture performance of the high-intensity vortex spray scrubber [18], with potential applications for CO_2 removal from flue gases as well as synthesis gases used, for example, for the production of H_2 and NH_3. The mass transfer study is carried out by absorbing CO_2 from air–CO_2 mixtures into the spray of aqueous NaOH solution produced by a single pressure-swirl atomiser. The performance of the vortex spray scrubber is characterised in terms of the overall gas phase mass transfer coefficient. The effects of the gas and liquid flow rates, flow arrangements, tower height and initial droplet size on the mass transfer rate are examined. The CO_2 capture performance of the vortex spray scrubber is also compared with the performance of the scrubber when operating as a conventional spray tower with axial gas flow.

2. Review of CO_2 absorption mechanism into alkaline solutions

The reactive absorption of CO_2 into alkaline solutions of hydroxide (NaOH or KOH) has been studied extensively both theoretically and experimentally in packed and wetted-wall columns. Astarita [30] and Danckwerts [31] have dealt with this topic theoretically in detail and also reviewed the earlier experimental work. An overview of the mechanism of chemical absorption of CO_2 in NaOH solutions is presented below.

2.1. Process chemistry

The absorption of CO_2 into an alkaline solution is accompanied by chemical reactions, which can be represented by the following scheme [30,31]:

$$CO_2 + H_2O \rightarrow HCO_3{}^- + H^+ \tag{R.1}$$

$$CO_2 + OH^- \rightarrow HCO_3{}^- \tag{R.2}$$

$$HCO_3{}^- + OH^- \rightarrow CO_3{}^{--} + H_2O \tag{R.3}$$

Reaction (R.1) is the first-order with a rate constant of 0.02 s^{-1} at 20 °C whilst Reaction (R.2) is the second-order and has a finite rate with the rate constant at 20 °C and infinite dilution being 6000 m^3/kmol s [31]. Reaction (R.1) has a negligible effect on the rate of CO_2 absorption in alkaline solutions with $pH > 10$ (see for details, Ref. [31]). Reaction (R.2) is followed by an instantaneous reaction (R.3). The overall reaction between CO_2 and NaOH solution can be expressed as:

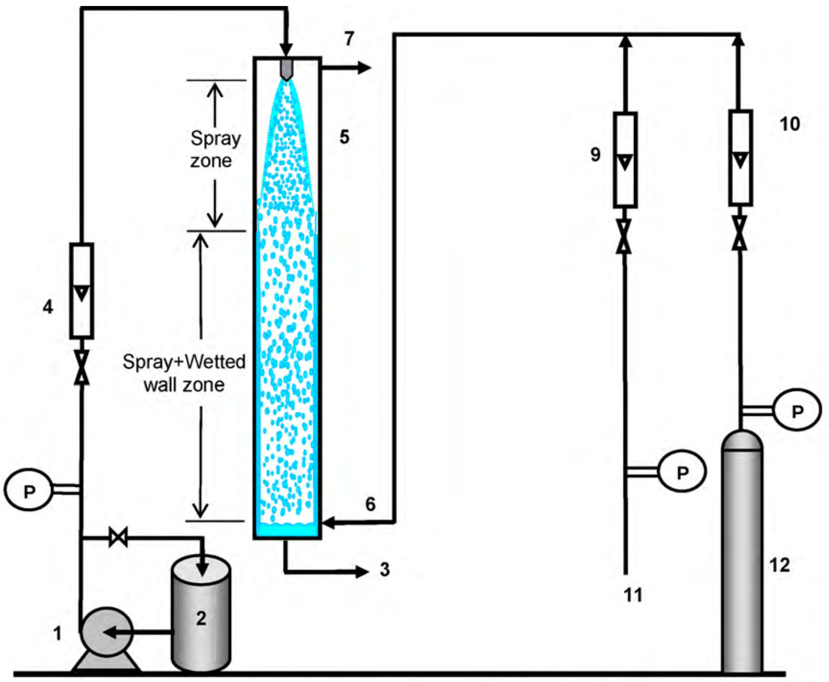
$$CO_2 + 2OH^- \rightarrow CO_3{}^{--} + H_2O \tag{R.4}$$

Reaction (R.4) is the second-order and maybe considered to be irreversible [31]. The rate of reaction is given by:

$$r_{CO_2} = -k_2 C_{CO_2} C_{OH^-} \tag{1}$$

The second-order rate constant (k_2) depends on the temperature and the ionic strength (I) of the solution. Pohorecki and Moniuk [32] have provided a correlation for k_2 as a function of these parameters for values of practical interests:

$$\log k_2 = 11.895 - 2382/T + 0.221I - 0.016I^2 \tag{2}$$



1. Pump, 2. Solution tank, 3. Liquid outlet, 4. Liquid rotameter, 5. Vortex spray scrubber, 6. Gas inlet, 7. Gas outlet, 9. Air rotameter, 10. CO_2 rotameter, 11. Compressed air supply, 12. CO_2 cylinder.

Fig. 1. Experimental set-up for the vortex spray scrubber.

It should be noted that the diffusivity of CO_2 in solution and Henry's law constant are also affected by ionic strength.

2.2. *Regime of chemical absorption*

The reaction regime can vary between the fast and the instantaneous reaction depending on the CO_2 partial pressure and OH^- concentration according to Eq. (1). Astarita [30] has argued that since the ranges of possible values of the groups, $(k_2 C_{OH}\, t_d)^{1/2}$ and $C_{OH}/bC_{CO_2,i}$ (where b is the stoichiometric factor of reaction (R.4), which is 2; $C_{CO_2,i}$ is the interfacial concentration of CO_2; and t_d is the diffusion time), widely overlap, both the instantaneous and the fast reaction regimes can be encounter for the chemical absorption of CO_2 into alkaline solutions, but the conditions for the former regime may not completely be fulfilled. The fast reaction condition, i.e., $(k_2 C_{OH}\, t_d)^{1/2} \ll C_{OH}/bC_{CO_2,i}$, is likely to be satisfied at low CO_2 partial pressures and high concentrations of OH^-, whilst the instantaneous reaction condition, $(k_2 C_{OH}\, t_d)^{1/2} \gg C_{OH}/bC_{CO_2,i}$, is satisfied at high CO_2 partial pressures and low concentrations of OH^-. In earlier studies on the CO_2–NaOH system reviewed by Astarita [30], the fast-reaction regime was encountered in packed towers whereas the instantaneous reaction conditions were satisfied in wetted-wall columns. Such information is not available from previous studies in spray towers.

2.3. *Rate of mass transfer*

The overall rate of chemical absorption of CO_2, accounting for both the gas- and the liquid-film resistances, can be expressed based on the two-film theory as [33]:

$$N_{CO_2} = K_g a P (y_{CO_2} - y^*_{CO_2}) \tag{3}$$

where a is the gas–liquid interfacial area, P is the total pressure, y_{CO_2} is the mole fraction of CO_2 in the bulk gas and $y^*_{CO_2}$ is the mole fraction in the gas phase in equilibrium with bulk liquid CO_2 concentration, and K_g is the overall gas phase mass transfer coefficient, which is defined as:

$$\frac{1}{K_g a} = \frac{1}{k_g a} + \frac{H}{k_l a E} \tag{4}$$

where E is the enhancement factor, H is the Henry's law constant, k_g is the gas-film coefficient, and k_l is the liquid-film coefficient for physical absorption. The enhancement factor is defined as the ratio of the liquid-film coefficient for chemical absorption to the liquid-film coefficient for physical absorption, and can be determined from the models developed for fast second-order reactions [e.g., 34–36].

3. Experimental set-up and procedure

3.1. *Vortex spray scrubber and ancillary equipment*

Experiments were carried out in a laboratory-scale vortex spray scrubber, which was used in our previous study and described in detail by Javed et al. [18]. The scrubber, shown schematically in Fig. 1, consisted of a cylindrical perspex tower of 1.25 m in height and 0.1 m in diameter with modular construction. The gas, a mixture of air and CO_2, was introduced at the bottom of the tower while the liquid, an aqueous NaOH solution, was sprayed from the top using an atomiser to provide a counter-current flow. A pressure-swirl atomiser with 30° nominal cone angle and 2 mm orifice diameter (B1/2-B1590, Delavan Spray Technology), referred to as 30°-NAC nozzle, was used to produce solid cone sprays. Some experiments were also carried out using sprays produced by the same type of atomiser but with 15° nominal cone angle and 5 mm orifice diameter (B1/2GG-SS1590, Delavan Spray Technology), referred to as 15°-NAC nozzle. The NaOH solution was supplied to the nozzle by a centrifugal pump and the flow rates were measured by a calibrated rotameter. The gas was introduced through the inlet pipes, as shown in detail in Fig. 2. Swirling flow in the tower was generated by introducing the gas through the tangential inlet pipe, while axial flow was produced by introducing the gas through the side inlet pipe. A flow distributor was placed

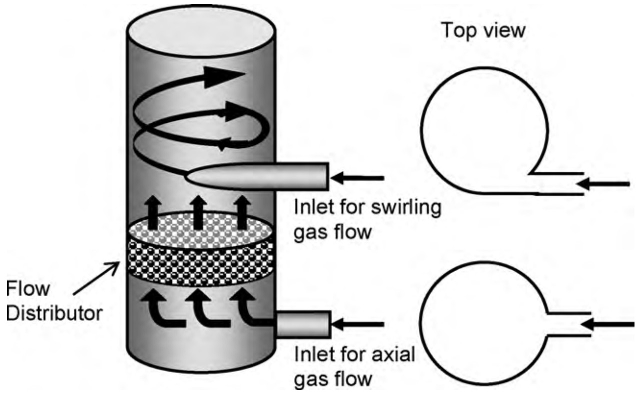


Fig. 2. Schematic diagram of the gas injection systems for the spray scrubber.

above the side inlet to ensure a uniform gas flow through the cross-section of the tower. CO_2 was supplied from a cylinder and was mixed with the air before entering the tower (see Fig. 1). To ensure complete mixing between CO_2 and air, sufficient pipe length was provided. The flow rates of gases were also measured using calibrated rotameters.

During experiments, the CO_2 concentration in the gas at the exit of the tower was measured once steady state was reached using an Infra-Red CO_2-analyser (ADC RF CO_2, Series No. 1201-3076). The measurement error associated with the Infra-Red CO_2-analyser was ±2%. The concentrations of hydroxyl ion (OH^-) in the NaOH solution at the inlet and outlet of the tower were determined by titration with 0.5 M HCl solution. However, the changes in the OH^- concentration in all runs were too small to estimate the amount of CO_2 absorbed into the solution accurately. The measurement accuracy was also checked by repeating experimental runs and by checking the overall mass balance for CO_2. For each experimental condition, measurements were repeated at least 3 times and data with more than 5% deviation were discarded. From the mass balance discrepancies between the calculated and the measured amount of solute leaving the tower with the gas stream were found to be less than 5%.

It should be noted that despite the important influence of spray properties on the mass transfer between gas and droplets, liquid sprays were characterised in a very few previous studies [e.g., 7,9,14]. The performance of the pressure-swirl nozzles used in this study was characterised by measuring the droplet size distributions at different axial locations using Malvern 2600 Particle Size Analyser (Malvern Instruments Ltd., Malvern, Worchestershire, England) for different liquid flow rates [18]. For 30°-NAC nozzle, the increase in liquid flow rate from 2 to 5 L/min produced finer droplets with the initial *Sauter Mean Diameter* (SMD), D_{32}, reducing from 155.5 to 57.3 μm. Whereas for 15°-NAC nozzle, D_{32} reduced from 290 to 210 μm for flow rates between 10 and 20 L/min. Measurements also revealed that the mean droplet diameter increased with axial distance from the atomiser for all the liquid flow rates. The distributions of sprayed liquid along the diameter of the tower were also measured using a patternator at different locations downstream of the nozzle. A solid cone spray pattern was formed and the radial liquid distributions at all locations revealed good symmetry about the spray axis. The axial gas flow distributions along the diameter of the tower and variation of pressure drop over the tower height as a function of the gas flow rate were measured and reported in [18].

3.2. Experimental conditions

The experimental conditions are given in Table 1. The concentration of CO_2 in the gas stream was 2.5% by volume at the inlet of the scrubber. The concentration of NaOH solution at the inlet of the scrubber was 1.25 M (5% by mass), with an alkalinity of pH = 14, for all the experimental runs. These inlet CO_2 and NaOH solution concentrations were selected from within the range of values used in previous studies cited before. The gas flow rate (G) was varied from 50 to 800 L/min, equivalent to 382–6112 m^3/m^2 h. These give average gas velocities (V) in the tower in the range of 0.11–1.7 m/s with corresponding Reynolds number (Re) of 700–11,323. The maximum gas velocity was set to 1.7 m/s in order to avoid entrainment of fine droplets by the gas flowing in the opposite direction [18]. The liquid flow rate (L) was varied between 2 and 5 L/min (3.3×10^{-5}–8.3×10^{-5} m^3/s), equivalent to 15.1–38.0 m^3/m^2 h. All experiments were carried out using swirling gas flow through the spray tower. For the purpose of comparison, experiments were also performed with axial gas flow.

Table 1
Experimental conditions.

Gas flow rate	50–800 L/min
Gas velocity	0.11–1.7 m/s
Liquid flow rate	2–5 L/min
Inlet CO_2 concentration	2.5 vol.%
NaOH solution concentration	1.25 M

4. Results and discussion

The experimental data for the absorption of CO_2 in NaOH solutions in both packed and spray towers were commonly expressed in terms of the overall gas phase mass transfer coefficient (K_ga) in previous studies. This is because the changes between the inlet and the outlet solution concentrations were usually very small for an accurate estimation of the rate of CO_2 absorption. This, however, does not imply that the gas-film resistance is the main controlling mechanism for the mass transfer in the air–CO_2/NaOH system [37]. It should be noted that in determining K_ga in the present study, the spray tower was treated as a single unit consisting of a spray zone near the nozzle and a spray plus wetted-wall zone downstream of the spray impingement point on the wall, as shown in Fig. 1. The overall gas phase mass transfer coefficient, in common with Tepe and Dodge [37], is calculated from the measured inlet and outlet CO_2 concentrations in the gas stream using the formula:

$$K_ga = \frac{G(y_{CO_2,\text{in}} - y_{CO_2,\text{out}})}{Z\ P(y_{CO_2} - y^*_{CO_2})_{\text{lm}}} \tag{5}$$

where G is the gas flow rate (kmol/m^2 s), P is the pressure (atm), and Z is the tower height (m), $y_{CO_2,\text{in}}$ and $y_{CO_2,\text{out}}$ are the inlet and outlet mol fractions of CO_2 in the gas, respectively, and $(y_{CO_2} - y^*_{CO_2})_{\text{lm}}$ is the logarithmic-mean concentration driving force. The K_ga (kmol/m^3 s atm) can be expressed in terms of the individual film mass transfer coefficients according to Eq. (4).

For the liquid flow rates (2–5 L/min) through the 30°-NAC nozzle used in the experiments, the droplets injection velocities were estimated [18] to vary between 3.1 and 6.2 m/s, with the flight time ranging from 0.2 to 0.4 s. During these short flight times of the droplets, the increase in liquid temperature due to the exothermic nature of absorption and reaction (R.4) is considered to be very small [10,31]. Therefore, the effect of droplets temperature variation on the CO_2 absorption rate has been neglected.

4.1. CO_2 absorption in axial gas flow

The CO_2 capture performance of the spray tower was first examined using axial (i.e., non-swirling) gas flow similar to that in conventional spray towers used in previous studies cited before. The experiments were carried out using sprays produced by the

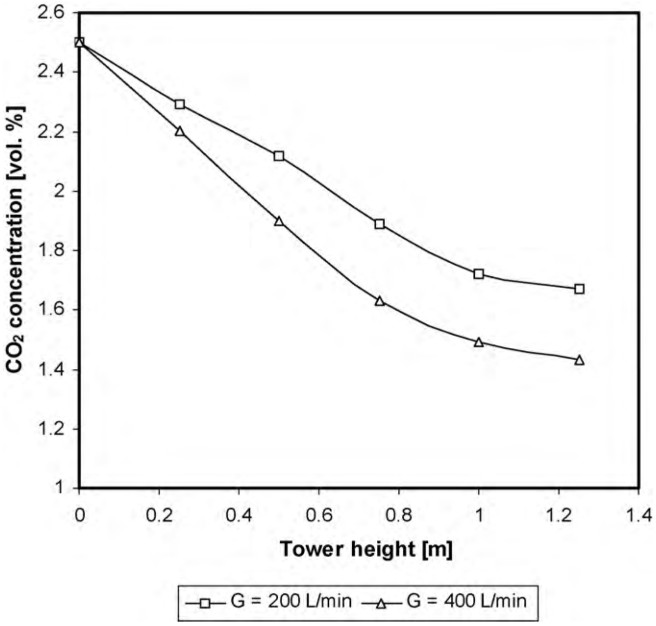


Fig. 3. Gas phase CO_2 concentration distributions along the tower height in axial gas flow for a liquid flow rate of 3 L/min (measurement errors ± 2%).

30°-NCA nozzle at various gas and liquid flow rates (see Table 1) with the counter-current flow arrangement. Some experiments were also conducted in the co-current gas–droplets flow.

4.1.1. Effect of gas flow rate in counter-current flow

Typical gas phase CO_2 concentration profiles along the length of the tower for gas flow rates of 200 and 400 L/min are shown in Fig. 3. The CO_2 concentration decreases rapidly with the tower height up to 0.75 m and more gradually beyond this point. At the higher gas flow rate, the CO_2 concentration decreases more rapidly. Fig. 4 shows the variation of the overall gas phase mass transfer coefficient as a function of the gas flow rate for four different liquid flow rates. As can be seen, the K_ga increases initially with increasing gas flow rate up to a value of 400 L/min (0.85 m^3/m^2 s) beyond which the K_ga becomes essentially independent of the flow rate. The same trend can be observed for all liquid flow rates. This suggests that at low gas flow rates (<400 L/min) the rate of mass transfer is largely controlled by the gas-film resistance, whereas at higher flow rates it is liquid-film controlled. A similar trend has been report by Kuntz and Aroonwilas [24] for the chemical absorption of CO_2 into MEA solutions in a spray tower with axial gas flow, but the transition from the gas-film controlled regime to the liquid-film controlled regime occurs at a much lower gas flow rate of 0.083 m^3/m^2 s. Tepe and Dodge [37] have observed no effect of the gas flow rate on K_ga for the air–CO_2/NaOH system over the experimental range from 0.114 to 0.503 m^3/m^2 s in a packed tower.

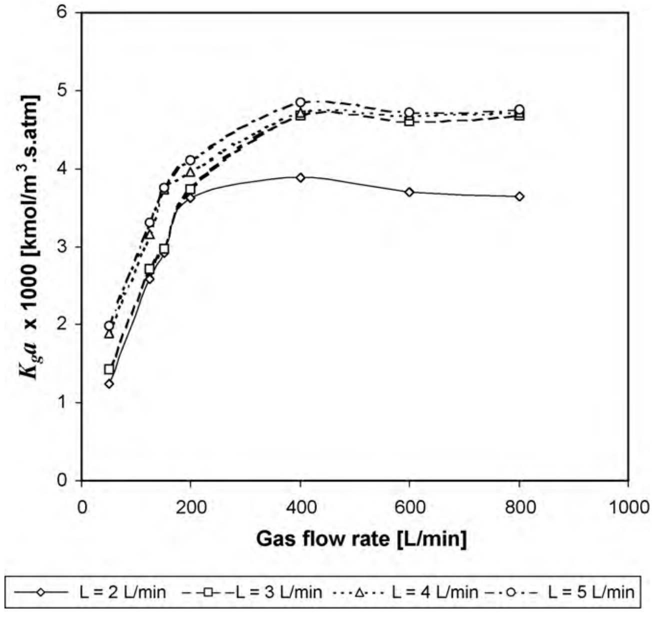


Fig. 4. Variation of K_ga as a function of axial gas flow rate (uncertainty in $K_ga < 5\%$).

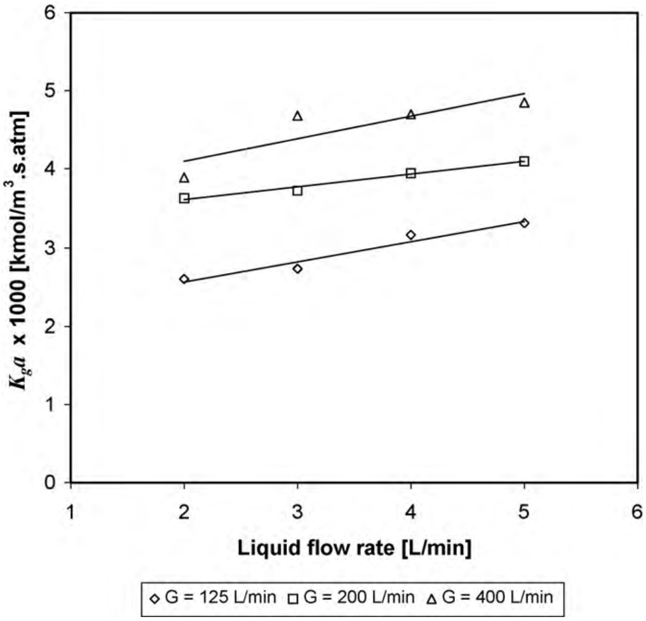


Fig. 5. Variation of K_ga as a function of liquid flow rate in axial gas flow (uncertainty in $K_ga < 5\%$).

The initial increase in K_ga with the gas flow rate (<400 L/min) is mainly due to the increase in the gas-film coefficient (k_g) which can be attributed to the fact that increasing gas velocity reduces the film thickness and increases the level of interfacial turbulence, both of which can lead to an enhancement of the transport of solute from the gas to liquid phase. For higher gas flow rates (>400 L/min) where the liquid-film resistance is controlling, a further increase in the k_g has no significant effect on the overall mass transfer coefficient. In this regime, the mass transfer rate predominantly depends on the liquid-film coefficient, k_l, and the chemical reaction rate. It is worth noting that as the gas velocity increases so does the applied shear stress on the droplet surface, which can enhance internal circulation of the liquid in droplets larger than a certain size, usually 50 μm in diameter [6], and thus increases the mass transfer rate across the liquid film. The droplets may also oscillate at high gas velocities, which increases the mass transfer rate compared with non-oscillating droplets [38]. Both these phenomena are expected to increase the k_l. However, it appears that these effects are not significant over the range of the droplet size and gas flow rate used in the present experiments. Taniguchi et al. [7] have suggested, based on good agreement between the measured absorption rates and those obtained from a solid sphere penetration model [39], that the effects of internal circulation and oscillation could be ignored for 120–250 μm droplets.

4.1.2. Effect of liquid flow rate in counter-current flow

Fig. 5 shows the effect of liquid flow rate on the K_ga for different gas flow rates. The results are presented for the gas flow rates up to 400 L/min because the K_ga is essentially constant at higher flow rates as shown in Fig. 4. As can be seen in the figure, the K_ga increases, but rather slowly, with increasing liquid flow rate within the measured range. The K_ga varies between the 0.16 and 0.29 power of the liquid flow rate. The spray characterisation study [18] has shown that the initial mean diameter of the droplets reduces from 155.5 to 57.3 μm with the increase in the liquid flow rate from 2 to 5 L/min, which generates larger interfacial surface area per unit volume of the sprayed liquid and hence enhances the mass transfer rate. On the other hand, the injection velocity of the droplets also increases from 3.1 to 6.2 m/s with the liquid flow rate within the same range [18], which leads to a reduction of the droplets residence time in the tower. These two opposing phenomena might have resulted in a slow increase in the K_ga with increasing liquid flow rate. Previous studies on the absorption of CO_2 into NaOH [6] and MEA [24] solutions in spray towers and into NaOH solutions in a packed tower [37] have also revealed a similar trend of increasing K_ga with increasing liquid flow rate, but generally more rapidly.

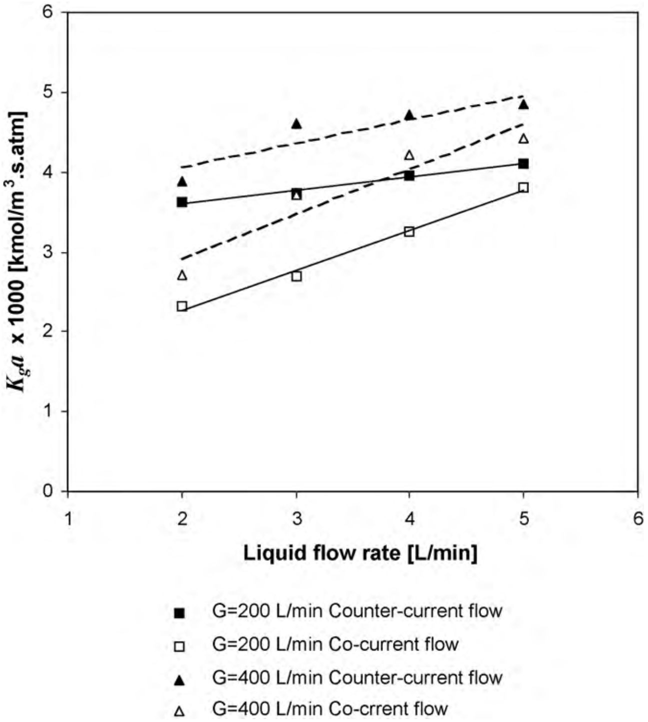


Fig. 6. Variation of K_ga as a function of liquid flow rate in counter-current and co-current gas–droplets flow (uncertainty in K_ga < 5%).

4.1.3. Effect of gas–droplets flow direction

Fig. 6 shows the effect of co-current and counter-current gas and droplets flow arrangements on the K_ga as a function of the liquid flow rate for G = 200 and 400 L/min. The K_ga increases with increasing liquid flow rate in the co-current flow, as also found in previous studies [7,9], but higher mass transfer rates are obtained for the counter-current flow arrangement. In a spray scrubber, the most effective zone for the mass transfer is considered to be in the vicinity of the nozzle, as the mass transfer rate is high during the first few seconds of the droplet formation, release and acceleration [40,41]. Therefore, supplying the solute directly to this zone by means of the co-current gas and liquid flow arrangement is expected to result

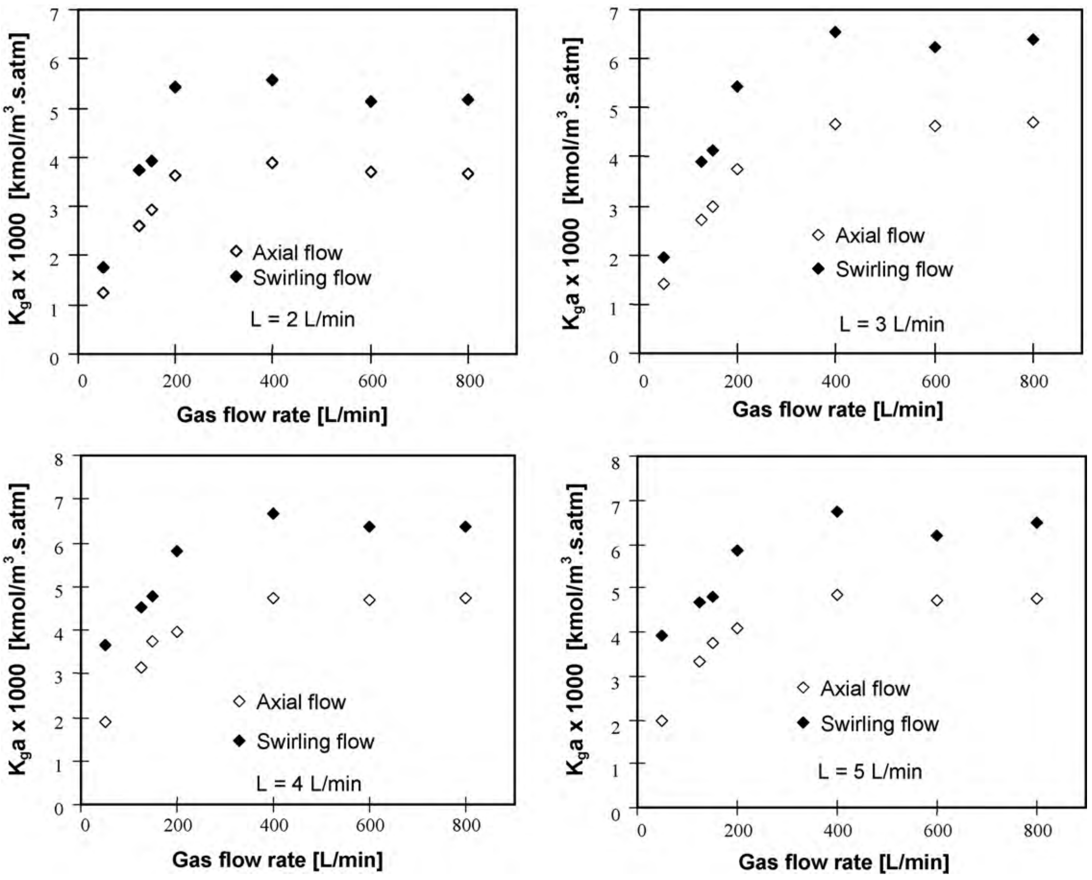


Fig. 7. Comparison between K_ga in the axial and swirling gas flows at various liquid flow rates (uncertainty in K_ga < 5%).

in a higher overall mass transfer rate. However, the K_ga data presented in Fig. 6 reveal that the overall concentration driving force, which is higher in the counter-current flow compared with that in co-current, plays a dominant role in determining the total mass transfer in the scrubber. Previous CO_2 absorption studies in spray towers have been conducted either in counter-current [6,10,11] or co-current [7,9] flows. No comparison between the two flow arrangements has been reported. Fig. 6 also shows that the K_ga increases more rapidly with the liquid flow rate in the co-current flow (proportional to the 0.5 and 0.56 power of L) compared with that in the counter-current flow (proportional to the 0.16 and 0.29 power of L) and hence the difference between the K_ga for these two flow arrangements diminishes with increasing liquid flow rate.

4.2. CO_2 absorption in swirling gas flow

The mass transfer characteristics of the counter-current vortex spray scrubber with swirling gas flow using sprays produced by the 30°-NCA nozzle were examined for the same ranges of gas and liquid flow rates as used for the axial gas flow. In this section, the CO_2 capture performance of the vortex spray scrubber is compared with that of the scrubber when operating in the conventional mode with axial gas flow.

4.2.1. Effect of gas flow rate

Fig. 7 shows the comparison between the overall mass transfer coefficients obtained in the swirling gas flow through the scrubber with those in the axial flow at various gas and liquid flow rates. As can be seen, the variation of K_ga with gas flow rate in swirling flows shows a trend similar to that in the axial flow, which has been explained in Section 4.1.1. However, it is important to note that for the same range of gas flow rate, the mass transfer rates in swirling flows are higher than those obtained in axial flows with an enhancement of K_ga from 31 to 49%.

In the present study, swirling flow was generated by introducing the gas into the tower through a single tangential inlet, as shown in Fig. 2, which creates a helical motion superimposed on the axial motion. In this method of imparting swirl, the initial intensity of swirl, referred to as the *swirl number* (see for details, Ref. [42]), depends upon the ratio of the tangential inlet diameter to the tower diameter, which is constant for the scrubber used in this study, and the average gas velocity through the inlet pipe. Therefore, the swirl intensity increases with increasing gas flow rate and it can be seen in the panels of Fig. 7 that, in general, the level of enhancement of K_ga increases with the gas flow rate up to 400 L/min.

The enhancement of K_ga due to the swirl in the gas flow maybe attributed to the high levels of gas phase turbulence and the reduction of the boundary layer thickness. Previous studies on swirling flows in pipes (for example, [43,44]) have revealed that the turbulence intensity and Reynolds stresses increase with increasing swirl intensity and are significantly larger compared with those in corresponding axial flows. Unfortunately, very little is known about the fluid dynamic characteristics around a solid spherical particle or a liquid drop in swirling flow compared with that in non-swirling axial flow. A recent experimental and theoretical study [45] on flow with and without swirl around a solid sphere has revealed that increasing swirl intensity causes significant changes in the characteristics of the boundary-layer, exhibiting unsteady spiral flow structure and thinning of the boundary-layer as well as enhancement of the turbulence level. Measurements in flow past a solid sphere have also revealed that the drag coefficient in swirling flows is at least an order of magnitude larger than that measured in axial flows [46]. This suggests that in the case of a liquid drop, the increased shear at the surface would enhance the internal liquid circulation and hence the mass transfer rate, but, as mentioned before, this effect may not be significant in the present study.

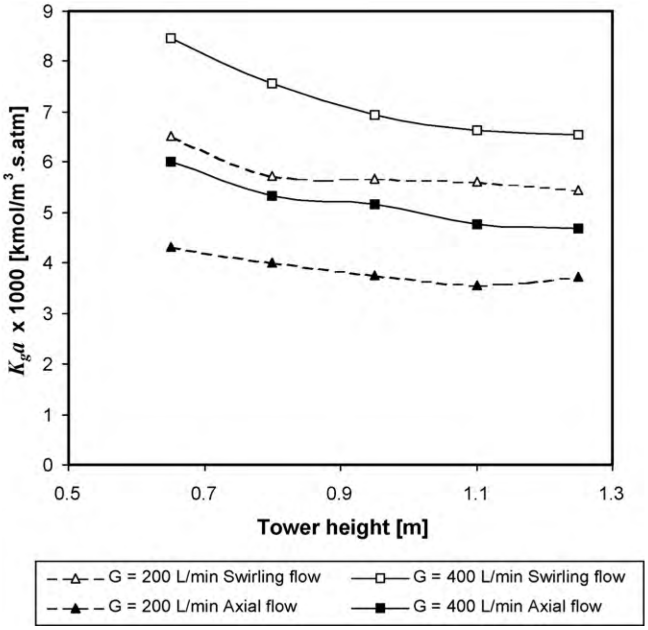


Fig. 8. Effect of scrubber height on K_ga in axial and swirling gas flows at a liquid flow rate of 3 L/min (uncertainty in $K_ga < 5\%$).

4.2.2. Effect of tower height

The effect of the spray tower height on the CO_2 absorption rate has been examined for both the axial and the swirling gas flows at a liquid flow rate of 3 L/min and gas flow rates of 200 and 400 L/min using the 30°-NCA nozzle. Fig. 8 shows that the K_ga decreases with the increase in the tower height from 0.65 to 1.25 m for both types of flow. However, higher K_ga values are obtained in swirling flows. Previous spray characterisation study [18] has revealed that the mean droplet diameter increases with distance from the nozzle exit due to the coalescence of droplets. It is also expected that the number of droplets will decrease along the length of the tower due to the impingement of droplets on the wall forming a liquid film (see Fig. 1). Both these factors result in a reduction of the interfacial area with the distance leading to a reduced mass transfer rate. Mehta and Sharma [6] have reported that the interfacial area deceases as the power −0.38 of the tower height. The results presented in Fig. 8 show that the K_ga is proportional to the −0.24 to −0.41 power of the tower height. A similar trend has been found in previous experimental studies [13,14,47]; however, the values of the exponent, ranging from −0.34 to −1.03, depend on the gas–liquid system and the operating conditions.

4.2.3. Effect of nozzle type

The effect of the spray nozzle type on the CO_2 capture performance was examined by producing sprays using the 30°-NCA and 15°-NCA nozzles for both the swirling and the axial gas flows. It should be noted that because of different orifice diameters, these two nozzles could not be operated at the same liquid flow rates. The 15°-NCA nozzle can only produce good quality sprays at much higher liquid flow rates between 10 and 22 L/min compared with the 30°-NCA nozzle using 2–5 L/min of liquid flow rates. Consequently, comparisons between the mass transfer performances of the sprays produced by these two nozzle types can only be made by operating the spray tower at the same gas to liquid flow rate ratios (*G*/*L*). The variations of the K_ga as a function of *G*/*L* for the two nozzles are shown in Fig. 9. As can be seen, the K_ga values for the 30°-NCA nozzle are larger than those for the 15°-NCA nozzle over the same range of *G*/*L*. There are two possible reasons for this trend. Firstly, the measurement of droplets size distributions

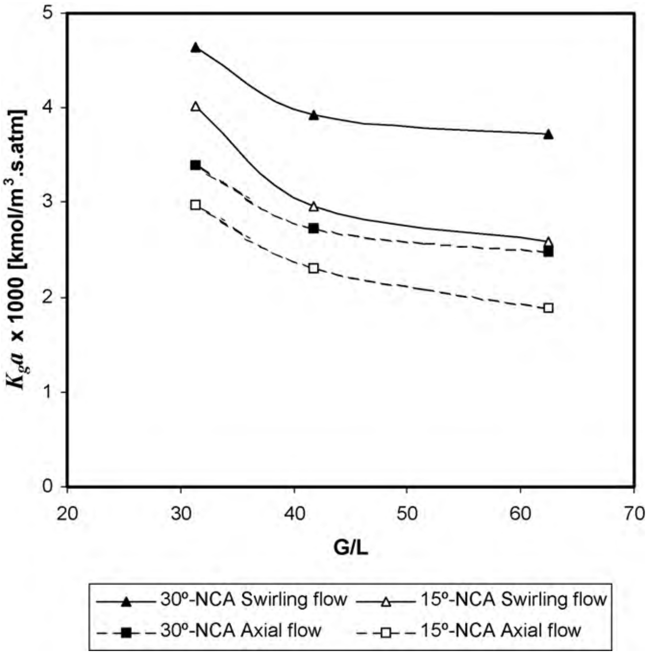


Fig. 9. Effect of spray nozzle type on $K_g a$ in axial and swirling gas flows for different gas to liquid ratios (uncertainty in $K_g a < 5\%$).

shows that the 30°-NCA nozzle produces smaller droplets with mean diameter ranging between 57.3 and 155.5 μm, while the 15°-NCA nozzle produces much larger droplets with diameter between 210 and 290 μm. Secondly, the 15°-NCA nozzle with a smaller cone angle produces a denser spray, which causes more droplets to coalesce resulting in a greater reduction of the interfacial area with the distance from the nozzle exit. Consequently, the 30°-NCA nozzle generates sprays with a larger interfacial area per unit volume of liquid than the 15°-NCA nozzle. On the other hand, the latter nozzle provides a longer length of the spray zone because of the narrower cone angle resulting in a lesser lose of droplets due to the impingement on the tower wall and hence more area available for mass transfer. However, this effect appears not to be significant.

5. Concluding remarks

An experimental investigation into the enhancement of CO_2 capture performance of a spray scrubber using swirling gas flow has been carried out and compared with the performance in axial gas flow. Experiments included measurements of the absorption of CO_2 from an air–CO_2 mixture in sprays of aqueous solution of NaOH for a range of gas and liquid flow rates, tower height and nozzle type in counter- and co-current flows.

The mass transfer performance of the spray scrubber is reported in terms of the overall gas phase mass transfer coefficient, $K_g a$. In both the axial and the swirling gas flow, the $K_g a$ initially increases with increasing gas flow rate within the range of 50–400 L/min, but at higher flow rates it is essentially constant. The $K_g a$ increases linearly with liquid flow rates between 2 and 5 L/min. In axial gas flows, the $K_g a$ varies from 1.24×10^{-3} to 4.75×10^{-3} kmol/m^3 s atm within these ranges of gas/liquid flow rates, whereas in swirling flows it varies between 1.77×10^{-3} and 6.49×10^{-3} kmol/m^3 s atm. It has been demonstrated that the CO_2 absorption rate can be augmented by applying swirl to the gas flow and, in the present study, around 31–49% enhancement of $K_g a$ is achieved compared with that in the axial flow of gas. The $K_g a$ deceases with the increase in the tower height. The spray nozzle producing finer droplets provides enhanced mass transfer rates. The counter-current gas–droplets flow provides higher mass transfer rates compared with those in co-current flow. Further investigations would be useful for the optimisation of the gas/spray contact arrangement in order to maximise the enhancement of mass transfer rate using swirling flow and to examine the CO_2 capture performance using aqueous alkanolamine solutions.

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