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Evaluation of alkanolamine solutions for carbon dioxide removal in cross-flow rotating packed beds

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ABSTRACT

The removal of CO_2 from a 10 vol% CO_2 gas by chemical absorption with 30 wt% alkanolamine solutions containing monoethanolamine (MEA), piperazine (PZ), and 2-amino-2-methyl-1-propanol (AMP) in the cross-flow rotating packed bed (RPB) was investigated. The CO_2 removal efficiency increased with rotor speed, liquid flow rate and inlet liquid temperature. However, the CO_2 removal efficiency decreased with gas flow rate. Also, the CO_2 removal efficiency was independent of inlet gas temperature. The 30 wt% alkanolamine solutions containing PZ with MEA were the appropriate absorbents compared with the single alkanolamine (MEA, AMP) and the mixed alkanolamine solutions containing AMP with MEA. A higher portion of PZ in alkanolamine solutions was more favorable to CO_2 removal. Owing to less contact time in the cross-flow RPB, alkanolamines having high reaction rates with CO_2 are suggested to be used. For the mixed alkanolamine solution containing 12 wt% PZ and 18 wt% MEA, the highest gas flow rate allowed to achieve the CO_2 removal efficiency more than 90% at a liquid flow rate of 0.54 L/min was of 29 L/min. The corresponding height of a transfer unit (HTU) was found to be less than 5.0 cm, lower than that in the conventional packed bed.

1. Introduction

To remove CO_2 using chemical absorption for synthesis-gas production, hydrogen manufacturing, and natural gas processing, alkanolamines including monoethanolamine (MEA), 2-amino-2-methyl-1-propanol (AMP), and piperazine (PZ) were frequently adopted. The reaction rate of these alkanolamines with CO_2 followed the order of PZ > MEA > AMP [1–3]. However, to use these alkanolamine solutions for treating the exhausted gases from power plants induced some problems including their high corrosion and vapor pressure and a huge volume of the exhausted gas streams required to treat. Furthermore, a large space for installation and operation was needed owing to that significant mass transfer limitation existed in the conventional gas–liquid contactors. Consequently, an effective gas–liquid contactor and a proper absorbent formulation were needed to develop.

Ramshaw and Mallinson [4] first presented that an enhancement in the gas–liquid mass transfer could be obtained by setting up a significantly more rapid regeneration of the interface between gas and liquid sides. To carry out this concept, they adopted centrifugal force to offer a contact between gas and liquid in a high centrifugal field by rotating doughnut-shaped packing element. Therefore, the rotating packed bed (RPB) was invented for enhancing the gas–liquid mass transfer in distillation and absorption process. This novel technology was referred to as "Higee" (an acronym for high gravity).

Under RPB operation, thin films and tiny droplets generated owing to a rigorous centrifugal acceleration could provide an enhancement in the gas–liquid mass transfer. Furthermore, the RPB could be operated higher gas and/or liquid flow rates owing to the low tendency of flooding relative to that in the conventional packed bed. Therefore, the gas–liquid mass transfer would frequently be enhanced by a factor of 10–100 and the dramatic reduction in the equipment volume would be achieved, thereby reducing the capital and operating costs [5]. A variety of studies have appeared in which the attention was focused on the applications of the countercurrent-flow RPB in diverse processes such as distillation [6–11], absorption [12–25], stripping [26–30], deaeration [31–35], reactive precipitation [36–47], and ozone oxidation [48–55].

Lin et al. [16] studied the CO_2 removal by chemical absorption from a gas steam containing 1–10% CO_2 in the countercurrent-flow RPB using NaOH, MEA, AMP, and a mixture of MEA and AMP. Their obtained results proposed that the countercurrent-flow RPB could

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replace the conventional packed bed for the CO_2 removal. Tan and Chen [18] obtained more results for chemical absorption of CO_2 by different absorbents including PZ and its mixtures with MEA, AMP, and methlydiethanolamine (MDEA) in the countercurrent-flow RPB.

Recently, Cheng and Tan [19] adopted the countercurrent-flow RPB for reducing CO_2 concentration to 20 ppm in the air, as required by a zinc/air battery, using absorption with PZ and its mixtures with 2-(2-aminoethylamino) ethanol (AEEA) and MEA. Jassim et al. [21] calculated the height of a transfer unit (HTU) for chemical absorption of CO_2 from a gas stream containing 3.5–4.5 vol% CO_2 in the countercurrent-flow RPB. The calculated HTU was found to be 14–27 cm that is much lower than that in the conventional packed bed, implying that a significant reduction in the equipment volume was obtained in the countercurrent-flow RPB. The volume reduction is particularly beneficial to power plants.

To treat a gas containing CO_2 with a huge flow rate, Lin and Chen [56] first adopted the cross-flow RPB for CO_2 absorption using NaOH, suggesting that the cross-flow RPB has a great potential in the removal of CO_2 from the exhausted gases. Also, Lin and Chen [57] presented that the mass transfer efficiency in the cross-flow RPB was comparable to that in the countercurrent-flow RPB for treating a gas containing 1 vol% CO_2 using NaOH. Lin et al. [58] confirmed that the mass transfer efficiency of the cross-flow RPB was higher than that of the countercurrent-flow RPB for treating a gas containing 10 vol% CO_2 using NaOH. However, there are no existing literatures until now to propose the performance of high concentrated alkanolamine solutions in the cross-flow RPB for CO_2 absorption.

Therefore, the objective of this work is to present a formulation of the alkanolamine solutions to remove CO_2 from a 10 vol% gas in the cross-flow RPB. The 30 wt% alkanolamine solutions contained the alkanolamines having various reaction rates with CO_2 including the single MEA and singe AMP, and the mixed alkanolamine solutions (PZ/MEA, AMP/MEA). To assess the performance of the single and mixed alkanolamine solutions, CO_2 removal efficiency (E), height of a transfer unit (HTU), and CO_2 loading were calculated. The effects of rotor speed, gas flow rate, and liquid flow rate on E, HTU, and CO_2 loading were investigated. Also, the effects of inlet gas temperature and inlet liquid temperature on E were investigated. From the obtained experimental data, more appropriate formulations of the alkanolamine solutions were then chosen. Moreover, the highest gas flow rate was estimated to be operated in the cross-flow RPB for achieving the desired CO_2 removal efficiency and the corresponding HTU could also be determined.

2. Experimental

MEA, PZ, and AMP with a purity of at least 99% were purchased from Tedia, Seedchem Company, Acors Organics, respectively. Nitrogen gas with a CO_2 concentration of 10 vol% was supplied by Yongen Industrial Gases (Taiwan). The experimental apparatus adopted in this work for CO_2 absorption is shown in Fig. 1. The cross-flow RPB was packed with stainless steel wire mesh having a configuration of interconnected filaments having a mean diameter of 0.22 mm and an average mesh diameter of 3 mm. The packings had a specific surface area of 855 m^2/m^3 and a voidage of 0.95. The cross-flow RPB had an inner radius of 2.4 cm, an outer radius of 4.4 cm, and an axial length of 12 cm.

Under general operation, the CO_2–N_2 stream flowed axially from the bottom of the packing owing to the pressure drop. Simultaneously, the prepared alkanolamine aqueous solution was pumped from the tank into the inner side of the packed bed through a liquid distributor. Alkanolamine aqueous solution traveled radially in the packed bed due to the centrifugal force and, then, exited from the outer side. The liquid distributor consisted of a tube with one vertical set of holes having twelve holes of 0.1 cm diameter with 0.95 cm interval. Both CO_2–N_2 stream and alkanolamine aqueous solution were contacted by the cross-flow mode within the cross-flow RPB, in which CO_2 in the CO_2–N_2 stream reacted with alkanolamine in the liquid stream. To prevent gas from bypassing the packed bed and keep the cross-flow operating mode, both sealings were designed as shown in Fig. 1. The exiting CO_2–N_2 stream, containing low CO_2 concentration, finally left the top of the packed bed, and, then, was expelled from the top of the cross-flow RPB, while the CO_2-rich alkanolamine aqueous solution was discharged from the bottom of the cross-flow RPB.

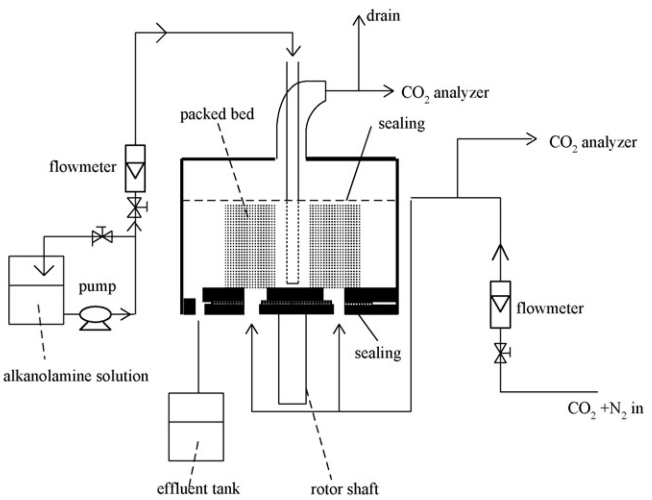

Fig. 1. Experimental setup of a cross-flow RPB for CO_2 absorption process.

The inlet liquid and gas streams could be heated prior to entering the cross-flow RPB. During operation, the gas flow rate (axial direction) was varied at the range of 10–90 L/min and the liquid flow rate (radial direction) was varied at the range of 0.1–0.6 L/min. In general, the cross-flow RPB could be operated at rotor speed of 400–1800 rpm, providing 6–123 times gravitational force based on the arithmetic mean radius. The fed alkanolamine aqueous solution was prepared by adding a predetermined amount of the single or mixed alkanolamines into water. The total alkanolamine concentration was kept at 30 wt% with various combinations.

The CO_2 concentrations in inlet and outlet CO_2–N_2 streams were measured using an infrared (IR) CO_2 analyzer (Polytron, Drager Ltd). The measurement range was from 0 to 15% with resolution of 0.01%. During CO_2 absorption, the CO_2 concentration in outlet CO_2–N_2 stream was observed to drop rapidly and then reached a steady value within 10–15 min. The reproducibility tests under almost all of the operating conditions were carried out in this work. The CO_2 concentration in outlet CO_2–N_2 stream was observed to be reproduced with a deviation of less than 5%, indicating the reliability of the measurement.

3. Results and discussion

The CO_2 removal efficiency (E) and height of a transfer unit (HTU) were calculated from the experimental data using the following equations,

$$E = \frac{C_i - C_o}{C_i} \times 100 \qquad (1)$$

$$\mathrm{HTU} = \frac{Z_b}{\ln(C_i/C_o)} \qquad (2)$$

In Eqs. (1) and (2), C_i and C_o are the CO_2 concentrations in the inlet and outlet gas streams, respectively, and Z_b is the axial height

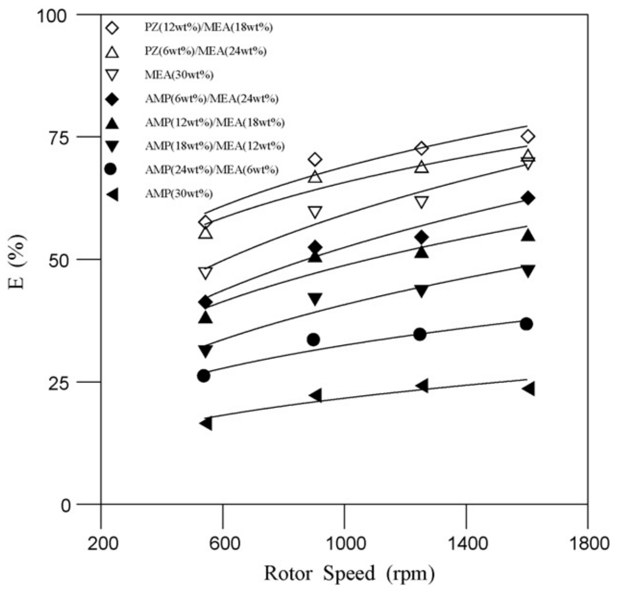

Fig. 2. Effect of rotor speed on *E* at inlet gas temperature of 30 °C and inlet liquid temperature of 30 °C for liquid flow rate of 0.54 L/min and gas flow rate of 86 L/min.

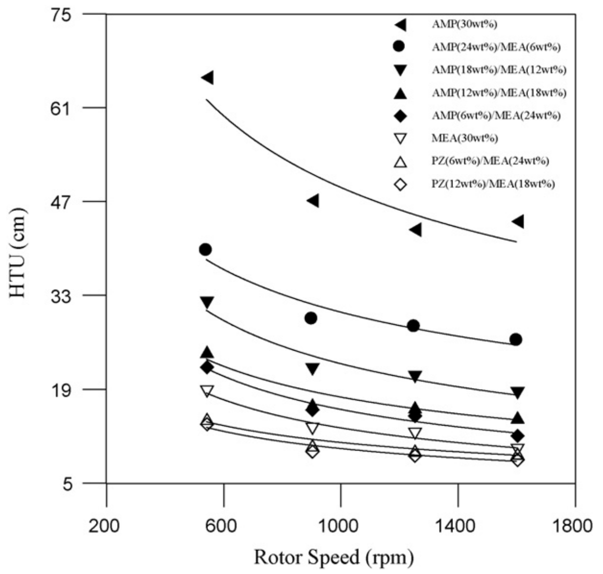

Fig. 3. Effect of rotor speed on HTU at inlet gas temperature of 30 °C and inlet liquid temperature of 30 °C for liquid flow rate of 0.54 L/min and gas flow rate of 86 L/min.

of the cross-flow RPB. A higher *E* represents the better CO_2 removal efficiency, and a lower HTU represents the smaller absorber volume required to obtain the desired removal efficiency. The CO_2 loading, representing the CO_2 moles absorbed per mole of absorbent fed to the cross-flow RPB, was also estimated in this work. The maximum CO_2 loading of each absorbent depends on its thermodynamic limitation, for example, the maximum CO_2 loading is 0.5 mole for one mole of MEA and 1.0 mole for one mole of AMP. A higher CO_2 loading for an absorbent therefore represents the closer to its maximum loading.

Fig. 2 shows the *E* values as functions of rotor speed from 540 to 1600 rpm at a fixed liquid flow rate of 0.54 L/min and a fixed gas flow rate of 86 L/min for 30 wt% alkanolamine solutions. These rotor speeds provided the corresponding centrifugal acceleration varied from 109 to 955 m/s^2. A higher rotor speed could provide higher mass transfer rate and gas–liquid interfacial area that were beneficial to CO_2 absorption. However, the reduced contact time between gas and liquid by an increase in the rotor speed limited CO_2 absorption. Based on the observed result, it was evident that the *E* values increased with an increasing rotor speed for all alkanolamine solutions, implying that the reduction of the contact time had an insignificant effect on CO_2 absorption.

Moreover, the *E* values for the 30 wt% mixed alkanolamine solutions containing PZ with MEA were higher than those for the single 30 wt% MEA solution and the mixed alkanolamine solutions containing AMP with MEA. Also, the *E* values for the single 30 wt% AMP solution were the lowest. The reason for the mixed alkanolamine solutions containing APM with MEA not comparable to the mixed alkanolamine solutions containing PZ with MEA was owing to the lowest reaction rate of AMP with CO_2. The obtained results therefore indicate that a choice of an alkanolamine having high reaction rate with CO_2 is essential to achieve a satisfactory CO_2 removal in the cross-flow RPB.

Fig. 3 shows the effect of rotor speed on the HTU values at a fixed liquid flow rate of 0.54 L/min and a fixed gas flow rate of 86 L/min for 30 wt% alkanolamine solutions. As expected, the HTU values decreased with rotor speed for all alkanolamine solutions. The mixed alkanolamine solution containing 12 wt% PZ and 18 wt% MEA provided the lowest HTU values that were varied from 13.9 to 8.6 cm when rotor speed was increased from 540 to 1600 rpm. However, the HTU values at rotor speed of 1600 rpm were changed to 9.5 and 10.1 cm for the mixed alkanolamine solution containing 6 wt% PZ and 24 wt% MEA and the single 30 wt% MEA solution, respectively. The HTU values for the mixed alkanolamine solutions containing AMP with MEA increased with an increase in the concentration of AMP in the alkanolamine solutions. Also, the highest HTU values (44.2–65.6 cm) were offered by the single 30 wt% AMP solution, implying that the alkanolamine solutions containing AMP are not suggested to be used in the cross-flow RPB.

Fig. 4 shows the effect of rotor speed on the CO_2 loading at a fixed liquid flow rate of 0.54 L/min and a fixed gas flow rate of 86 L/min for 30 wt% alkanolamine solutions. The CO_2 loading increased with rotor speed for all alkanolamine solutions. The mixed alkanolamine solution containing 12 wt% PZ and 18 wt% MEA offered the highest CO_2 loadings varied from 0.089 to 0.115 while rotor speed was increased from 540 to 1600 rpm. However, the CO_2 loadings at rotor speed of 1600 rpm were changed to 0.102 and 0.095 for the mixed alkanolamine solution containing 6 wt% PZ and 24 wt% MEA

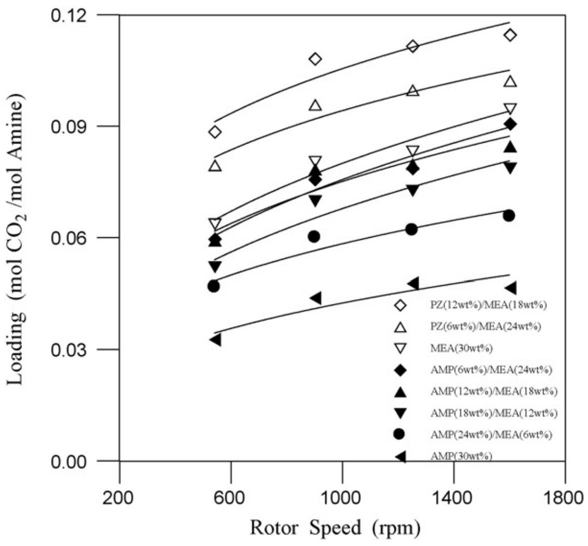

Fig. 4. Effect of rotor speed on CO_2 loading at inlet gas temperature of 30 °C and inlet liquid temperature of 30 °C for liquid flow rate of 0.54 L/min and gas flow rate of 86 L/min.

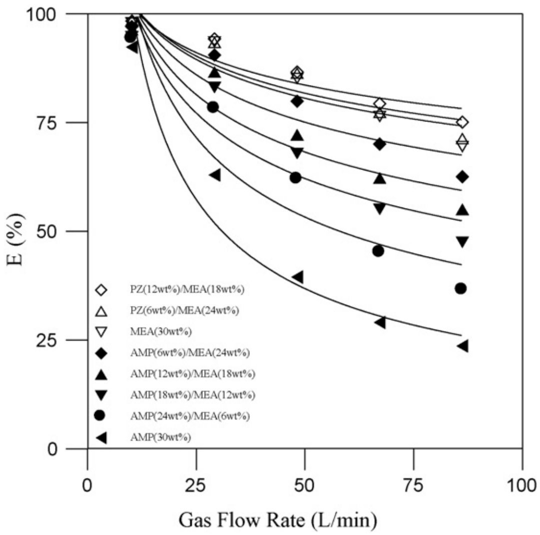

Fig. 5. Effect of gas flow rate on *E* at inlet gas temperature of 30 °C and inlet liquid temperature of 30 °C for liquid flow rate of 0.54 L/min and rotor speed of 1600 rpm.

and the single 30 wt% MEA solution, respectively. The CO_2 loadings for the mixed alkanolamine solutions containing AMP with MEA decreased with an increase in the concentration of AMP in the alkanolamine solutions. Also, the lowest CO_2 loadings (0.033–0.047) were provided by the single 30 wt% AMP solution. These low CO_2 loadings were attributed to the fact that the reaction rate of AMP with CO_2 was the lowest among the alkanolamine solutions used in this work.

Fig. 5 shows the effect of varying gas flow rate from 10 to 86 L/min on the *E* values at a fixed liquid flow rate of 0.54 L/min and a fixed rotor speed of 1600 rpm for 30 wt% alkanolamine solutions. The gas flow rate influenced the *E* values; that is, the *E* values decreased with gas flow rate for all alkanolamine solutions. For the desired removal in treatment of a gas with 10 vol% CO_2 using 30 wt% alkanolamine solutions, as shown in Fig. 5, the highest gas flow rates allowed to treat depended on alkanolamine formulation. For example, all the alkanolamine solutions could be used to achieve the CO_2 removal efficiency more than 90% at a liquid flow rate of 0.54 L/min for treating a gas with flow rate of 10 L/min. However, the mixed alkanolamine solutions containing PZ with MEA and the single 30 wt% MEA solution could be used to achieve to the same removal efficiency at the same liquid flow rate for treating a gas with flow rate of 29 L/min. Moreover, the differences of *E* values between the mixed alkanolamine solutions containing PZ with MEA and the single 30 wt% AMP solution increased with gas flow rate. This characteristic could be explained by the fact that the contact time induced by gas flow rate decreased with gas flow rate and the reaction rate of AMP with CO_2 was the lowest among the alkanolamine solutions used in this work.

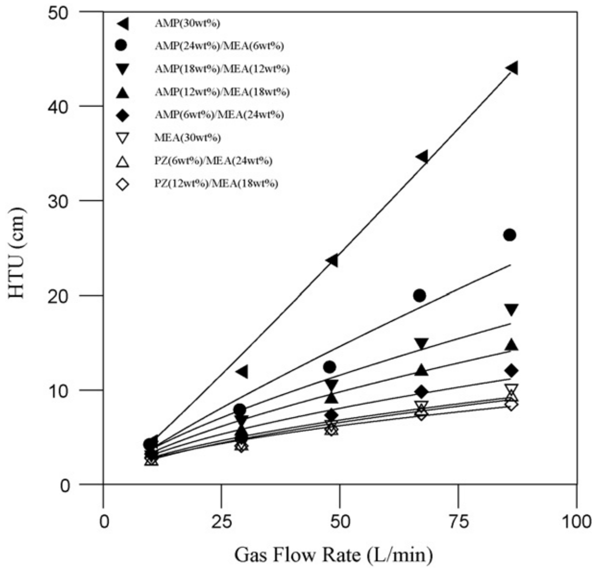

Fig. 6. Effect of gas flow rate on HTU at inlet gas temperature of 30 °C and inlet liquid temperature of 30 °C for liquid flow rate of 0.54 L/min and rotor speed of 1600 rpm.

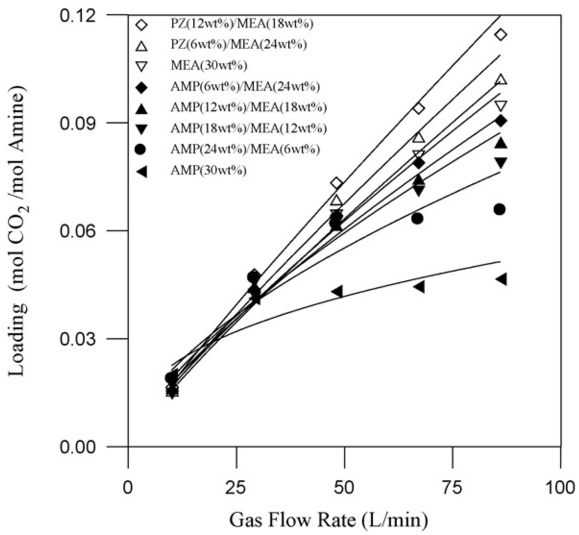

Fig. 7. Effect of gas flow rate on CO_2 loading at inlet gas temperature of 30 °C and inlet liquid temperature of 30 °C for liquid flow rate of 0.54 L/min and rotor speed of 1600 rpm.

Fig. 6 shows the effect of gas flow rate on the HTU values at a fixed liquid flow rate of 0.54 L/min and a fixed rotor of 1600 rpm for 30 wt% alkanolamine solutions. As expected, the HTU values increased with gas flow rate for all alkanolamine solutions. It was noted that the calculated HTU values corresponding to the CO_2 removal efficiency higher than 90% were all less than 5 cm, much less than those in the conventional packed bed. This observation confirms the conclusions presented by Lin et al. [16] and Jassim et al. [21] that the size of an absorber can be significantly reduced if

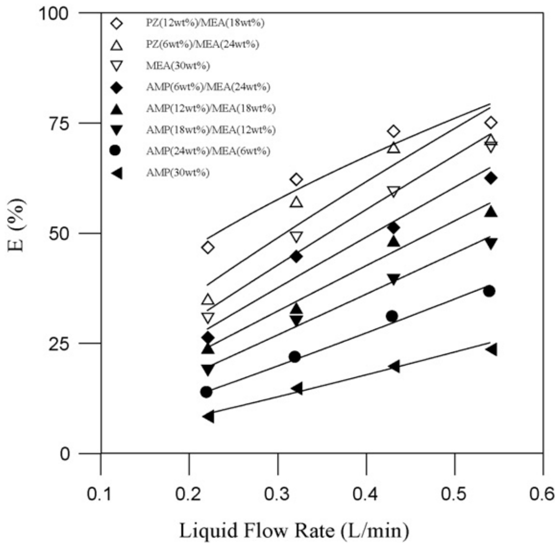

Fig. 8. Effect of liquid flow rate on *E* at inlet gas temperature of 30 °C and inlet liquid temperature of 30 °C for gas flow rate of 86 L/min and rotor speed of 1600 rpm.

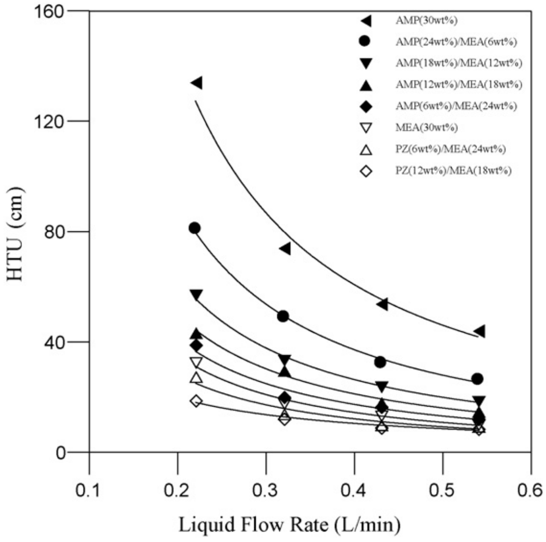

Fig. 9. Effect of liquid flow rate on HTU at inlet gas temperature of 30 °C and inlet liquid temperature of 30 °C for gas flow rate of 86 L/min and rotor speed of 1600 rpm.

the RPB instead of the conventional packed bed is used to remove CO_2 from the gas stream. Moreover, the differences of HTU values between the mixed alkanolamine solutions containing PZ with MEA and the single 30 wt% AMP solution increased with gas flow rate.

Fig. 7 shows the effect of gas flow rate on the CO_2 loading values at a fixed liquid flow rate of 0.54 L/min and a fixed rotor of 1600 rpm for 30 wt% alkanolamine solutions. The CO_2 loading increased with gas flow rate for all alkanolamine solutions. Moreover, the gas flow rate had a significant effect on the CO_2 loading for the mixed alkanolamine solutions containing PZ and MEA, implying that the less contact time resulting from gas flow rate could be overcome by fast reaction rate of alkanolamine with CO_2. The mixed alkanolamine solution containing 12 wt% PZ and 18 wt% MEA had the highest CO_2 loadings of from 0.017 to 0.115 as gas flow rate was increased from 10 to 86 L/min. However, the CO_2 loadings at gas flow rate of 86 L/min were changed to 0.102 and 0.095 for the mixed alkanolamine solution containing 6 wt% PZ and 24 wt% MEA and the single 30 wt% MEA solution, respectively. The CO_2 loadings at gas flow rate of 86 L/min for the mixed alkanolamine solutions containing AMP with MEA decreased from 0.091 to 0.066 as the concentration of AMP in the alkanolamine solutions was increased from 6 to 24 wt%. Also, the lowest CO_2 loadings (0.020–0.047) were provided by the single 30 wt% AMP solution, implying that the

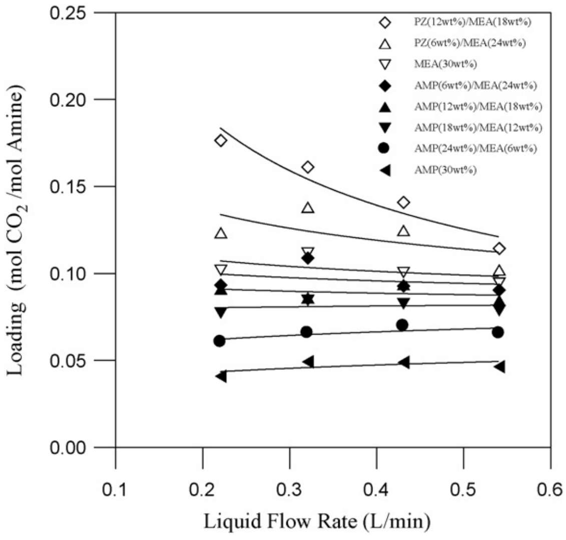

Fig. 10. Effect of liquid flow rate on CO_2 loading at inlet gas temperature of 30 °C and inlet liquid temperature of 30 °C for gas flow rate of 86 L/min and rotor speed of 1600 rpm.

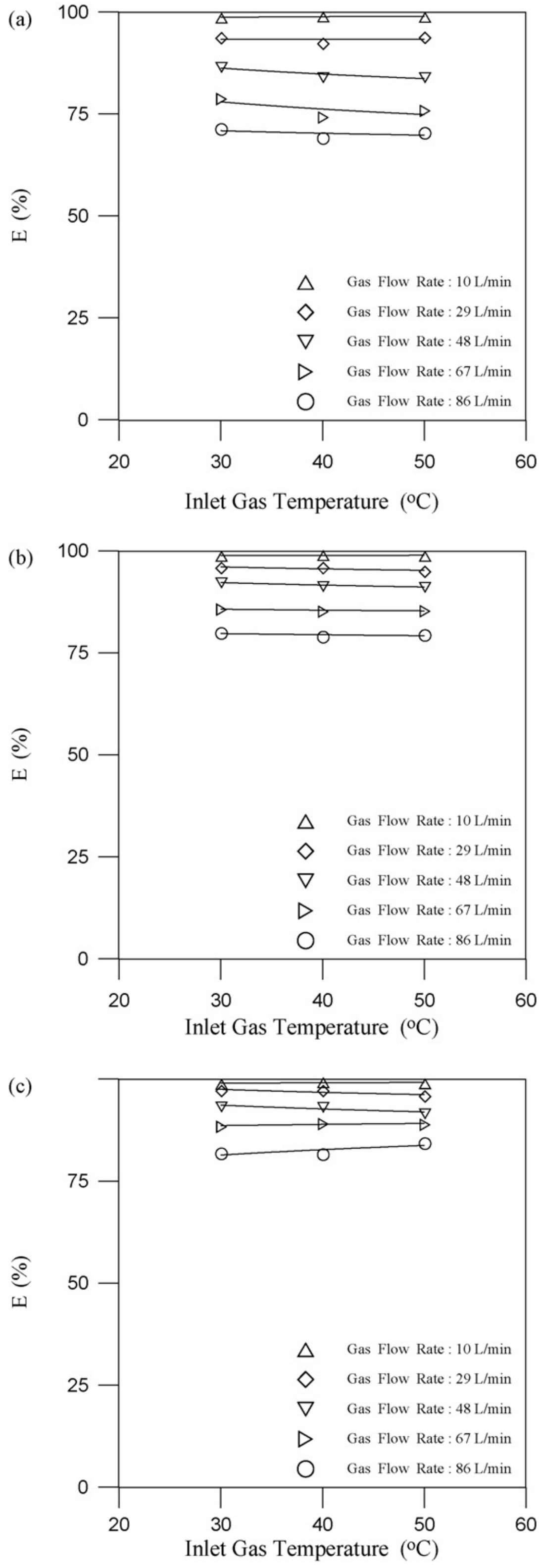

Fig. 11. Effect of inlet gas temperate on *E* for gas flow rate of 86 L/min, liquid flow rate of 0.54 L/min and rotor speed of 1600 rpm (a) inlet liquid temperature of 30 °C; (b) inlet liquid temperature of 40 °C; (c) inlet liquid temperature of 50 °C.

alkanolamine solutions containing AMP was not applicable to the cross-flow RPB.

Fig. 8 shows the effect of liquid flow rate ranging from 0.22 to 0.54 L/min on the *E* values at a fixed gas flow rate of 86 L/min and a fixed rotor speed of 1600 rpm for 30 wt% alkanolamine solutions. The liquid flow rate had an influence on the *E* values; that is, an increase in liquid flow rate yielded an increase in the *E* values for all alkanolamine solutions. This behavior was attributed to the fact that more alkanolamine solutions were allowed to absorb CO_2 and the liquid-side mass transfer resistance was reduced by the reaction of alkanolamines with CO_2. Fig. 8 also shows that more PZ was present in the alkanolamine solutions, more CO_2 could be removed from the gas stream. The enhancement of removal efficiency by PZ was mainly attributed to fastest reaction rate of PZ with CO_2 compared with other alkanolamines used in this work.

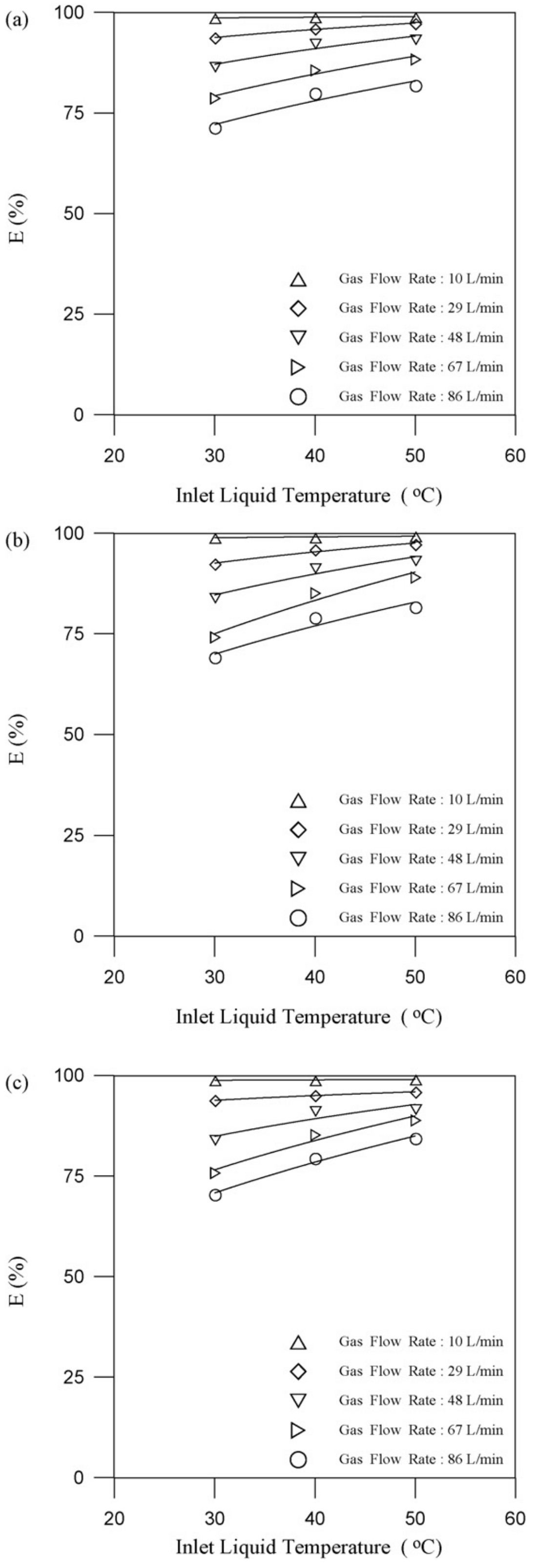

Fig. 12. Effect of inlet liquid temperate on *E* for gas flow rate of 86 L/min, liquid flow rate of 0.54 L/min and rotor speed of 1600 rpm (a) inlet gas temperature of 30 °C; (b) inlet gas temperature of 40 °C; (c) inlet gas temperature of 50 °C.

Fig. 9 shows the effect of liquid flow rate on the HTU values at a fixed gas flow rate of 86 L/min and a fixed rotor of 1600 rpm for 30 wt% alkanolamine solutions. As expected, the HTU values decreased with liquid flow rate for all alkanolamine solutions. The mixed alkanolamine solution containing 12 wt% PZ and 18 wt% MEA provided the lowest HTU values that were varied from 18.9 to 8.6 cm when liquid flow rate was increased from 0.22 to 0.54 L/min. The mixed alkanolamine solutions containing AMP with MEA had the HTU values of more than 12 cm at the range of liquid flow rate used in this work. Also, the highest HTU values (44.2–134.2 cm) were offered by the single 30 wt% AMP solution, further verifying that the alkanolamine solutions containing AMP are not suggested to be used in the cross-flow RPB.

Fig. 10 shows the effect of liquid flow rate on the CO_2 loading at a fixed gas flow rate of 86 L/min and a fixed rotor of 1600 rpm for 30 wt% alkanolamine solutions. The CO_2 loading decreased with liquid flow rate for the mixed alkanolamine solutions containing PZ with MEA. This may be attributed to the less contact time induced by higher liquid rates, thus leading to the fewer enhancements in the *E* values at higher liquid flow rates. However, this behavior was not found for other alkanolamine solutions, indicating that the CO_2 loading at different liquid flow rates were nearly identical. Based on the more amounts of alkanolamine at higher liquid flow rates and nearly the same loading at different liquid flow rates, a more removal efficiency therefore was observed at higher liquid flow rates, as shown in Fig. 8.

Fig. 11 shows the effect of inlet gas temperature on the *E* values at a fixed liquid flow rate of 0.54 L/min and a fixed rotor speed of 1600 rpm under various gas flow rates and inlet liquid temperatures for the mixed alkanolamine solution containing 12 wt% PZ and 18 wt% MEA. It was evident that the *E* values did not change significantly when inlet gas temperature was increased from 30 to 50 °C for all gas flow rates and inlet liquid temperatures. This observation indicated that the reaction of alkanolamines with CO_2 was not affected by inlet gas temperatures. However, the effect of inlet liquid temperature on the *E* values was different from that of inlet gas temperature on the *E* values, as shown in Fig. 12, presenting that the *E* values increased with inlet liquid temperature for all inlet gas temperatures and gas flow rates of more than 29 L/min. This behavior implied that the CO_2 removal could be increased by increasing inlet liquid temperature.

4. Conclusions

The removal of CO_2 from a gas containing 10 vol% of CO_2 by chemical absorption with 30 wt% alkanolamine solutions over wide ranges of rotor speed, gas flow rate, liquid flow rate, inlet liquid temperature, and inlet gas temperature in the cross-flow RPB was investigated. The results were considered in relation with the CO_2 removal efficiency (*E*), height of a transfer unit (HTU), and CO_2 loading. The *E* values were found to increase with rotor speed, liquid flow rate and inlet liquid temperature. However, the *E* values decreased with gas flow rate. Also, the *E* values were found to be independent of inlet gas temperature. The HTU values were found to decrease with rotor speed and liquid flow rate. Moreover, the HTU values increased with gas flow rate. The CO_2 loadings were found to increase with rotor speed and gas flow rate. Moreover, the CO_2 loadings decreased with liquid flow rate for the mixed alkanolamine solutions containing PZ with MEA but were independent of liquid flow rate for other alkanolamine solutions used in this work.

Based on the measured *E* values, HTU values and CO_2 loadings, the 30 wt% alkanolamine solution containing 12 wt% PZ and 18 wt% MEA were found to be very effective absorbents to remove CO_2. For treatment of a gas with flow rate of 29 L/min by this solution, the *E* value more than 90% could be achieved. The HTU value corresponding to the *E* values more than 90% was less than 5 cm, lower than that in the conventional packed bed. The less HTU was mainly attributed to the proper combination of the alkanolamines having high reaction rates with CO_2 and the increase in contact between gas and liquid in the cross-flow RPB. Owing to that high CO_2 removal efficiency can be achieved with less HTU, the cross-flow RPB is an effective gas–liquid contactor for treating the flue gases from power plants.

Acknowledgements

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