Novel Design for the Nozzle of a Laminar Jet Absorber

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A novel design for the nozzle of a laminar jet absorber has been produced and used to obtain high-quality absorption-kinetics data devoid of interfacial resistance and interfacial turbulence. The nozzle design consists of a circular hole in a 0.07 ± 0.005 mm thick stainless steel sheet having an inside diameter of between 0.50 and 0.65 mm. The hydrodynamics of the various jets produced from this nozzle were checked by studying the physical absorption of carbon dioxide into water and nitrous oxide into monoethanolamine. Wetting of the orifice face, oscillations in the jet, and interfacial resistance were completely absent. In addition, the new nozzle design was tested by comparing the measured kinetics data with published data for carbon dioxide reactive absorption in monoethanolamine solutions. The measured kinetic data were in excellent agreement with literature data obtained over a period of four decades. This shows that the nozzle design produces high-quality absorption data, and that the kinetic data obtained were accurate and not subject to any interfacial turbulence.

Introduction

The kinetics of the reactive absorption of a gas into liquids are usually obtained in a laminar jet absorber, wetted wall column, one-sphere unit, or stirred cell unit. The use of each apparatus depends mainly on the physical and chemical properties of the absorption system. For fast chemical reactions between absorbed gases and reactive liquids such as carbon dioxide (CO_2) in monoethanolamine (MEA) solutions, the laminar jet absorber has proven to be the best apparatus that can be used to generate reliable absorption data. This is because the contact time between gas and liquid is short, the interfacial area is known accurately, and the physical absorption rates have been shown to agree with penetration theory predictions.[1,2]

The most important and critical part of the laminar jet absorber is the nozzle that produces the liquid jet. This is because the extent of departure of the behavior of the jet from an ideal rodlike flow depends on the shape of the nozzle or orifice at which the liquid-jet is formed.[1] Basically, the liquid jet can be formed with a bell-shaped nozzle[3−7] or an orifice-plate nozzle.[6,8−12] Raimondi and Toor[8] and Clarke[6] tested both design types and concluded that jets produced by orifice-plate nozzles have a flat velocity distribution, whereas the bell-shaped nozzles produce a surface retardation which shows the absorption rate to be lower than theoretical predictions. On this basis, it was decided to base the nozzle design in this work on the orifice-plate nozzle type.

Raimondi and Toor[8] used a square-edged orifice in a plate of 0.1 mm thickness, where the diameter of the orifice was in the range from 0.571 to 0.851 mm. The authors claimed that measurements of the absorption rate of CO_2 into water at 295 K, with contact time between 1 and 20 ms, agreed closely with those predicted theoretically even though they were 1−4% lower than the theoretical. Al-Ghawas et al.[9] also used a square-edged orifice, 0.51 mm in diameter, drilled in a 0.08-mm thick stainless steel sheet. Using this nozzle design, the diffusion coefficient of CO_2 into water at 298 K was reported as 1.95×10^{-5} cm^2/s, which was within the range of values available in the literature[13] (from 1.75×10^{-5} to 2.05×10^{-5} cm^2/s). A similar design for the nozzle was used by Ashour et al.[10] and Rinker et al.,[11] except that the diameter in this case was 0.533 mm instead of 0.51 mm. The measured diffusion coefficients of CO_2 into water at 293 and 313 K were reported[11] as 1.74×10^{-5} and 2.67×10^{-5} cm^2/s, respectively, which are in close agreement with those correlated by the expression of Versteeg and van Swaaij.[13] Good absorption data can be obtained with a square-edged orifice as illustrated above. However, because most theoretical models are predicated on a rodlike jet, it is essential to produce a rodlike jet in the laminar jet absorber as this is key to obtaining good agreement between theoretical and experimental values. It is predicted that a circular-hole orifice might be better at producing a rodlike liquid jet on which the theoretical expressions are based. Consequently, not only are we using the nozzle design in this work, but we are also evaluating the circular-hole orifice in producing a rodlike jet.

In some experiments, unusual operating conditions have been imposed, or modified chemical solvents have been used, to obtain accurate experimental absorption rates relative to theoretical predictions. For example, Clarke[6] conducted the experiments at a reduced pressure to avoid the suspected depletion of unreacted MEA at the interface. Hikita et al.[5] and Hagewiesche et al.[14] added a small amount of surface-active agent to reduce the values of the experimental absorption rates. They indicated that the higher absorption rates observed in the case of the solution without surface active agent could be attributed to interfacial turbulence, presumably

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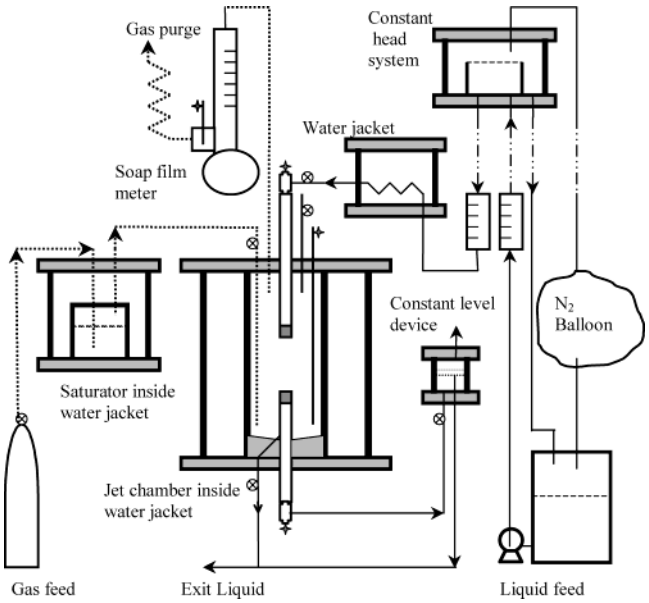

Figure 1. Schematic drawing of the laminar jet apparatus.

produced by a surface tension gradient during the chemical absorption process, as observed by Brian et al.[15] and also reported by Sada et al.[16] The higher absorption rates obtained in these cases could be attributed to the design of the nozzle in addition to both the suspected depletion of unreacted MEA at the interface and interfacial turbulence. In this work, a new nozzle design is proposed and then used to show that it can eliminate the contributions of these parameters (i.e., the depletion of unreacted MEA at the interface and the interfacial turbulence) to the observed absorption rates. It was also used to show that with the new nozzle design it was not necessary to modify the experimental conditions or modify the properties of the liquid to obtain accurate kinetics data.

Experimental Section

The purpose of the laminar jet experiments was to test the reliability of the new nozzle design under several operating conditions. It was also to show that with the new nozzle design there was no need to modify the experimental conditions or modify the properties of the liquid to obtain accurate kinetic data. The main materials used in measuring the rate of gas absorption in the laminar jet absorber are carbon dioxide (CO_2), nitrous oxide (N_2O), and monoethanolamine (MEA). CO_2 with a purity of 99.9% and N_2O with a purity of 99.5% were purchased from PRAXAIR (Regina, SK). The MEA with a purity of 99.0% was purchased from Fisher Scientific (Nepean, ON) and used without any further purification. The experimental kinetic data were obtained by absorption of CO_2 into aqueous MEA solutions. The free MEA concentrations were in the range from 0.193 to 3.158 mol/L with an average CO_2 loading of 1.81 mol/L. All the kinetics experiments were conducted at atmospheric pressure at temperatures in the range of 293−313 K.

Apparatus. The main parts of the laminar liquid jet apparatus used for the measurements reported in this work are schematically presented in Figure 1. The absorbing liquid from a constant-head system was fed by gravity through a temperature-controlled bath to a jet chamber. The constant-head system was used to maintain a constant liquid flow rate. A jet of the liquid issued from a circular nozzle, flowed intact downward through an atmosphere of the absorbed gas, and was collected in a capillary receiver. The absorbed gas was supplied from a pressure cylinder and was saturated with water vapor at the experimental temperature before entering the absorber chamber. In addition to the new nozzle and new receiver design, the apparatus combined various features of the designs of Danckwerts,[1] Astarita et al.,[2] and Al-Ghawas et al.,[9] such as the jet chamber design, the degassing process of the liquid, and the gas flow rate measurement process.

Jet Chamber Assembly. The jet chamber, which is similar to the design of Al-Ghawas et al.,[9] was a 250-mm long and 50-mm i.d. acrylic cylinder enclosed in a constant-temperature jacket constructed from a 250-mm long and 150-mm i.d. acrylic cylinder. Both cylinders were held between two aluminum flanges, and the ends were sealed with rubber gaskets. The nozzle was mounted on the end of a 6.4-mm i.d. acrylic delivery tube, which passed through a Swagelok nut in the aluminum cover flange. The delivery tube could slide vertically to change the jet length, and could also be locked into position by the Swagelok nut.

The jet chamber was provided with a gas entry near the bottom, a gas outlet near the top, a manometer port, and a 6.4-mm i.d. receiver tube fitted into a funnel-shaped base. A hole in the base allowed draining of any liquid overflow through the overflow port. To avoid vibration, the jet absorber was placed on an anti-vibration pad and all the lines leading to the jet absorber were made from flexible tubes.

Nozzle. It is our hypothesis that a circular-hole orifice on a very thin sheet should be better at producing a rodlike liquid jet than a square-edged orifice design, which previous literature has recommended.[8−11] In this work, several nozzles were designed as circular holes in 0.07 ± 0.005 mm-thick stainless steel sheets and used to evaluate their ability to produce a smooth-surfaced uniform "rod-like" flow. Very thin sheets were used to minimize boundary-layer effects and to approach a flat velocity distribution at the charge point. The circular holes had diameters ranging from 0.50 to 0.65 mm. These nozzles were used without any further treatments such as the paraffin-wax coating employed by Scriven and Pigford[3] to prevent wetting. With our design, no wetting of the orifice faces and no oscillations in the jets were observed. Photographs of typical jet nozzle components and dimensions are shown in Figure 2. The hydrodynamics of the various jets were checked by studying the physical absorption of CO_2 into water within the temperature range of 288−333 K. Testing the apparatus within this temperature range is required to ensure that the behavior of the laminar jet as a function of temperature is correct.

Receiver. Usually the receiver is made from a glass capillary tube having an internal diameter 20−40% greater than the jet diameter.[3,6,9] This capillary tube is capped by a Teflon plug into which a hole is drilled according to the capillary tube diameter. This design is complex, difficult to produce, and thus expensive to obtain. To overcome these shortcomings, the receiver in this work was designed as a capillary hole drilled into an acrylic rod as shown in Figure 3. The length and diameter of the hole were 20.0 mm and 1.0 mm, respectively. This design is simpler, cheaper, and much easier to produce than the ones reported in the litera-

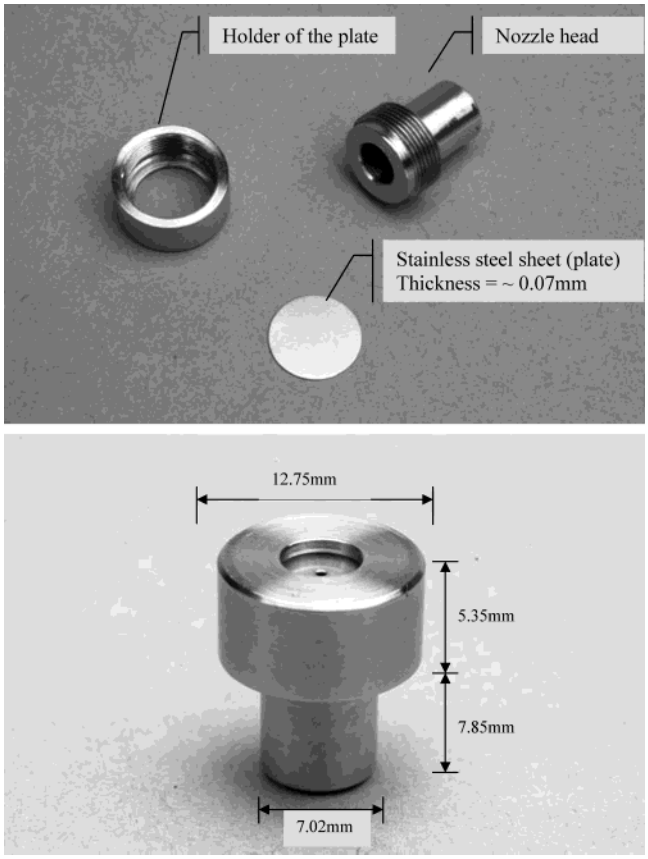

Figure 2. Jet nozzle components and dimensions.

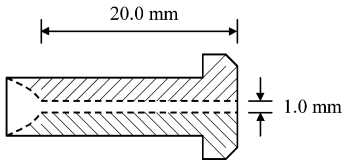

Figure 3. Receiver of the liquid jet.

ture.[3,6,9] The receiver assembly was fitted onto the end of the receiver tube by two O-rings. The downstream end of the receiver was connected to a constant-level device, which was mounted on a vertical slide rod and provided with a fine-adjustment leveling screw (see Figure 1). This receiver design assisted in the collection of the liquid jet without any liquid spillover or gas entrainment inside the receiver for several hours of continuous operation.

Operating Procedures. The absorbing liquid was degassed by spraying it into a vacuum, similar to the Danckwerts[1] procedures, and kept under a nitrogen blanket. This degassing process was found to be more effective, easier, and safer than the degassing process by heating while the solution was under vacuum as implemented by other workers,[9,11] especially when hazardous chemical solutions are used. The degassed solution was fed by gravity to the jet chamber from a constant-head system, located about 3.0 m above the absorption chamber (see Figure 1). This was to provide sufficient head for the required rate of flow. The constant-head system is a 200-cm^3 capacity cup provided with a liquid-inlet port and a liquid-outlet port on its bottom. During the liquid feed operation, the inlet flow rate was kept higher than the outlet flow rate and the overflow from the cup was returned to the feed tank. The liquid in both the constant-head system and the feed tank was kept under a blanket of nitrogen during the operation. The N_2 supply to replace the used liquid was stored in a balloon connected to both the feed tank and the constant-head system. The flow rate of the liquid was controlled with a rotameter. This technique did prove to give a stable and constant flow rate for several hours of operation and was found to be better than the surge-tank technique applied in the liquid line between the pump and the rotameter to eliminate the pump pulses, as implemented by other workers.[9,11] The actual flow rate of liquid was determined by weighing the liquid discharged in a timed interval during each experiment. A 1-L volumetric flask was used to collect the discharged liquid. The constant-level device was used to adjust the liquid level in the receiver. When the level was slightly too high, not all the liquid-jet was collected in the receiver; some of the jet spilled over. When the level was too low, the free jet continued several mm down into the receiver and small gas bubbles were entrained in the rapidly moving liquid inside the receiver. The level was adjusted to give the condition in which there was neither spillover of liquid nor entrainment of gas. A two-dimensional traveling microscope was used to measure the jet-length to within 0.01 mm and the jet-diameter to within 0.001 mm. The gas from a pressure cylinder was saturated with water vapor at the experimental temperature before entering the jet chamber as practiced by many researchers.[9,11] Temperatures of the liquid entering and leaving the jet-chamber, the gas entering the jet-chamber, and the gas entering the gas-flow meter were measured. The temperatures of the liquid and the gas entering the jet chamber were controlled to within ±0.3 °C. This was achieved by having two separate heating/cooling circulator units, one for controlling the gas-stream temperature and the other for controlling the liquid-stream temperature.

The best method for measuring the mass transfer rate in terms of the absorption rate is the volumetric technique, as reported by other authors.[9,11,17] As shown in Figure 1, after obtaining a stable jet-flow, a gaseous stream consisting of only the gas to be absorbed was forced to flow through the absorption chamber of the laminar jet and purged through a digital flow meter (soap-film meter). The used flow meter was designed to measure flows from 0.1 to 50 cm^3/minute with an accuracy of ±3% of readings. After sufficient time had elapsed to make sure that the jet chamber, the soap meter, and the long-purge tube had been filled with the gas, the gas-feed valve was closed. At this point, the flow rate of gas that was indicated by the soap-film meter was equal to the volumetric rate of absorption. Experiments were repeated several times and an average was taken. In all cases, the purge gas was vented into a fume hood.

The choice of the operating conditions of the laminar jet is based on a number of design constraints (mainly the design of the nozzle and the receiver). First, the length of the jet cannot be varied over a wide range. If the jet is too short, end effect becomes too important; if it is too long, operation of the receiver becomes difficult. Second, the liquid flow rate can be changed only over a limited range: a lower limit is imposed by the requirement of avoiding dripping and an upper limit is imposed by the need to avoid turbulence. This behavior has been reported by Astarita et al.,[2] and has been observed and confirmed in this experimental work. As a result, the

first constraint restricts the range of the interfacial area and the latter constraint limits the range of the mass-transfer coefficient.[2]

Results and Discussion

The experimental data generated in this work were obtained by a dynamic method in which a jet of liquid moves continuously through the gas for a known contact time. The absorption rate was measured volumetrically with a digital soap-film meter. The absorption rates were utilized to determine molecular diffusivity and kinetics of reaction. The physical absorption data, from the CO_2−water and N_2O−MEA systems, were used to test the apparatus and to calculate the molecular diffusivities. The chemical absorption data, from the CO_2−MEA system, were used to calculate the kinetics. These results were compared with the published data to confirm the reliability of the nozzle and the receiver design in this work.

CO_2−Water Absorption. The laminar jet absorber designed in this work was first tested by absorbing CO_2 into water, which is a standard method for testing this type of apparatus because the solubility and the diffusivity of CO_2 in water are well known.[1,2] In the absence of reaction, the total rate of absorption into the jet can be calculated theoretically from eq 1, which was based on a rodlike jet.[1,18]

$$R_A = 4C_e^*(DLh)^{1/2} \quad (1)$$

where R_A is the total rate of absorption of gas, C_e^* is the equilibrium concentration of gas at the interface, D is the diffusivity of gas in liquid, L is the liquid flow rate, and h is the jet length. A plot of R_A against $(Lh)^{1/2}$ at various flow rates and jet lengths, should give a straight line through the origin with a slope equal to $4C_e^*D^{1/2}$.

The raw experimental data for the absorption rate of CO_2 into water are documented elsewhere.[19] The absorption experiments were carried out over the temperature range of 288−333 K using seven different nozzle diameters ranging from 0.50 to 0.65 mm. All experiments were done under atmospheric pressure. The proper partial pressure of carbon dioxide that was used in calculating the absorption rate was taken to be equal to the total pressure less the vapor pressure of water at the temperature of the experiment. The water vapor pressure was calculated from the Antoine equation at the experimental temperature.

The experimental data as well as the theoretical predictions of the absorption rate of CO_2 into water are shown in Figure 4. The points are the experimental data and the line in this figure is calculated from eq 1. The diffusivity and solubility of carbon dioxide in water used in this equation at 298 K and atmospheric pressure were taken as 1.96×10^{-5} cm^2/s and 3.29×10^{-5} mol/cm^3, respectively.[27] The agreement between the measured and calculated absorption rates is excellent over various flow rates and jet lengths with an average absolute deviation of 0.8% from the calculated rates. This excellent agreement shows the reliability of the experimental apparatus design and arrangement to produce a rodlike jet, as well as indicates the absence of any interfacial resistance.

In addition, the average experimental value found for the diffusion coefficient of CO_2 in water at 298 K and atmospheric pressure was 1.96×10^{-5} cm^2/s. This value

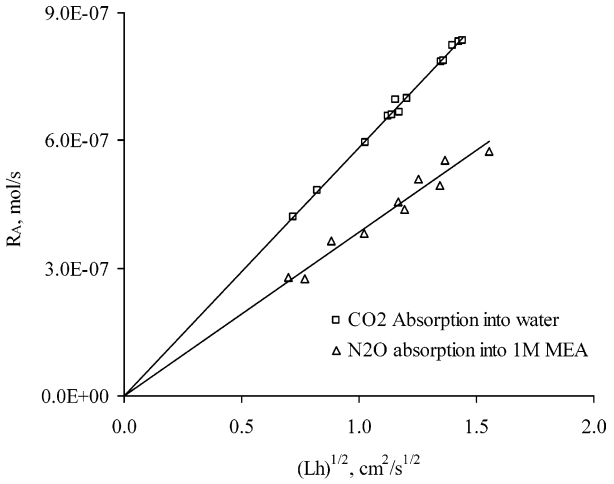

Figure 4. Physical absorption rates of CO_2 into water and N_2O into 1 M MEA solution at 298 K and atmospheric pressure in laminar jet absorber: experimental (points), and theoretical (line).

Table 1. Diffusivity of CO_2 in Water at 298 K and Atmospheric Pressure

D, cm^2/s	reference
1.75×10^{-5}	Tamann and Jessen, 1929[a]
1.90×10^{-5}	Taniguchi and Sakurada, 1955[a]
1.87×10^{-5}	Scriven, 1956[a]
1.94×10^{-5}	Davidson and Cullen, 1957[a]
2.00×10^{-5}	Vivian and King, 1964[a]
1.85×10^{-5}	Unver and Himmelbau, 1964[a]
2.05×10^{-5}	Clarke, 1964[a]
1.95×10^{-5}	Thomas and Adams, 1965[a]
1.87×10^{-5}	Tang and Himmelbau, 1965[a]
1.98×10^{-5}	Duda and Vrentas, 1968[a]
1.93×10^{-5}	Haimour and Sandall, 1984[28]
1.98×10^{-5}	Tang and Sandall, 1985[29]
1.95×10^{-5}	Al-Ghawas et al., 1989[9]
1.94×10^{-5}	Tamimi et al., 1994[30]
1.96×10^{-5}	Perry and Green, 1997[27]
1.96×10^{-5}	**this work**

[a] Data reported in Versteeg and van Swaaij, 1988[13].

is calculated by using the slope of the experimental data, which is 5.83×10^{-7}, and the solubility of CO_2 into water. The experimental diffusivity that was obtained in this work shows good agreement with that available in the literature, as shown in Table 1.

It should be mentioned that the data presented in Figure 4 were produced by using the 0.63-mm diameter nozzle. The other six nozzles designed in this work gave similar satisfactory results. The measured absorption rates at 298 K from all nozzles and the predicted absorption rates calculated from eq 1 were compared in the parity plot shown in Figure 5. This shows excellent agreement between the measured rates of CO_2 absorption into water using the seven different nozzle diameters and the rates of CO_2 absorption predicted from the theoretical model. The average absolute deviation of the measured rates from the predicted rates is 1.7%. On the basis of this result, any nozzle diameter within the range of 0.5−0.65 mm will give satisfactory absorption rates if it is as well-designed as the nozzle in this work. In addition, these excellent agreements again show that the experimental apparatus, especially the nozzle design, is excellent. It also shows the absence of any interfacial resistance.

To test the behavior of the laminar jet absorber as a function of temperature, the temperature effect on the diffusivity of CO_2 in water was determined experimen-

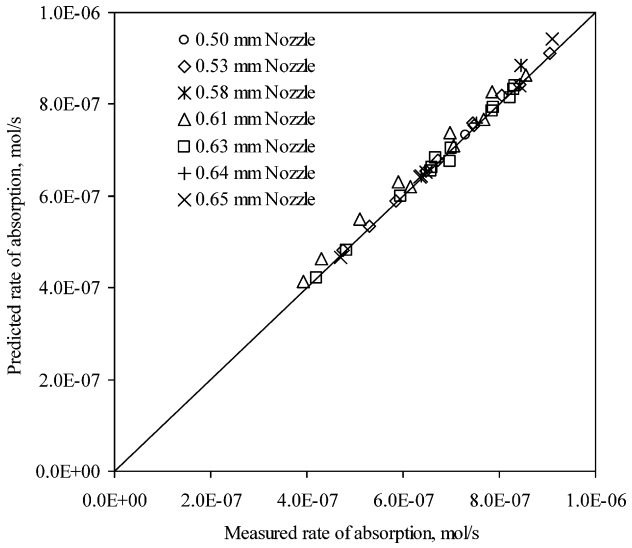

Figure 5. Comparison of measured and predicted CO_2 absorption rate into water at 298 K in the laminar liquid jet absorber.

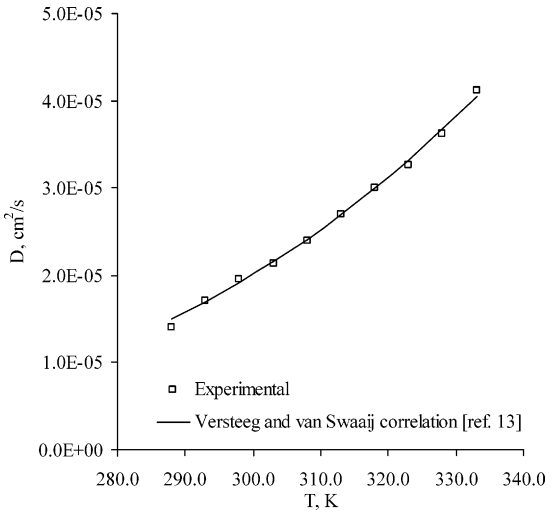

Figure 6. Diffusivity of CO_2 in water as a function of temperature.

tally within the temperature range from 288 to 333 K, the typical temperature range found in gas absorbers. Figure 6 shows this experimental diffusivity as well as the diffusivity from eq 2 based on the correlation developed by Versteeg and van Swaaij[13] (diffusivity is in cm^2/s and temperature is in Kelvin).

$$D = 2.35 \times 10^{-2} \exp(-2119/T) \qquad (2)$$

Excellent agreement between the measured and predicted diffusivity was obtained. The average absolute deviation of the measured diffusivity from the predicted diffusivity was 1.7%. This indicates that the behavior of the laminar jet is excellent within the temperature range of 288−333 K.

It is well-known that there could be a small departure from the assumption of idealized rodlike jet. The departure from ideal behavior is caused by several factors: first, an entrance effect caused by the nonflattened velocity profile at the nozzle; second, a gravity effect caused by the acceleration and shrinkage under the influence of gravity; and third, an exit effect caused mainly by rippling and surface stagnation induced by the jet receiver.[2] The results obtained from our nozzle

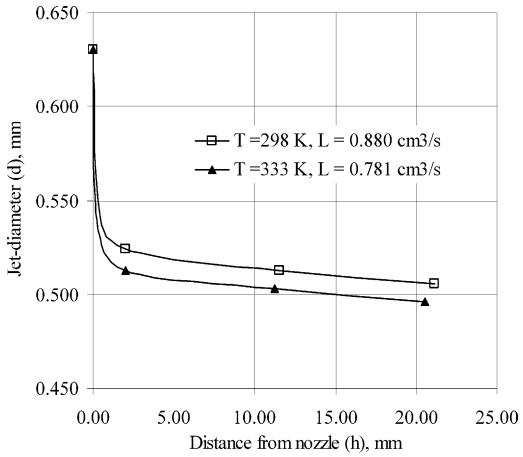

Figure 7. Water-jet diameter for a 0.630-mm i.d. nozzle in a stainless steel plate 0.07 mm thick.

design illustrate very clearly that the departure from the assumption of idealized rodlike cylindrical jet can be kept to a minimum by using a well-designed nozzle.

Regarding the measurements of the jet diameter, it is found that the measurements taken at the middle of the jet length were sufficient for the mass transfer calculation. Diameter measurements for a 0.630-mm i.d. nozzle in a stainless steel plate of ~0.07-mm thick are shown in Figure 7. It can be seen that the diameter of the water jet is reduced from 0.630 to 0.506 mm within the first 2.0 mm of the downstream flow mainly because of the influences of viscous drag in the nozzle and gravitational acceleration. At a distance downstream of the nozzle of more than 2.0 mm, the change in diameter with length is somewhat negligible. This was achieved by using a very thin stainless steel plate for manufacturing the nozzle. It can also be seen from the figure that the diameter is a function of temperature and the liquid flow rate. Thus, it is recommended to take new measurements for the diameter whenever there are extreme changes in these operating conditions. From this it can be concluded that any absorption calculation should be based on the measured diameter of the liquid jet and not on the nozzle diameter. Basing calculations on nozzle diameter instead of the actual measured diameter may explain the high experimental absorption rates reported by Hagewiesche et al.[14]

N_2O−MEA Absorption. An additional test for the laminar jet apparatus was performed by the physical absorption of N_2O into MEA solution, from which the diffusivity (D) of N_2O in 1 M MEA solution at 298 K was measured. The absorption rates (R_A) of N_2O shown in Figure 4 were measured at different gas−liquid contact times ranging from 1.3 to 6.0 ms by varying both the jet length and liquid flow rate. Each absorption rate value reported in this figure was an average of 5−7 measurements. In the absence of reaction, the total rate of absorption into the jet can be represented theoretically by eq 1. In this case, C_e^* is the interfacial-equilibrium concentration of the N_2O gas in the liquid phase, which is known as the solubility of N_2O in MEA solution. This solubility was calculated from the semiempirical model of Wang et al.[20] The enhanced parameters for this model by Tsai et al.[21] were adapted in this solubility calculation. As shown in Figure 4, a plot of the experimental data of R_A against $(Lh)^{1/2}$, at various flow rates and jet lengths, should give a straight line through the origin with a slope equal to $4C_e^*D^{1/2}$. From this figure, the value of the slope of the experimental

data was found to be 3.844×10^{-7}. This slope and the solubility of N_2O in 1 M MEA solution at 298 K, which is 2.33×10^{-5} mol/cm^3, were used to determine the experimental diffusivity. The average experimental value found from this work for the diffusion coefficient of N_2O in 1 M MEA solution at 298 K was 1.70×10^{-5} cm^2/s. This is in excellent agreement with the value of 1.69×10^{-5} cm^2/s reported under the same conditions by Sada et al.[22]

In conclusion, the preceding critical analysis of all the physical experimental results, and the absorption rates observed, show that the jets were evidently similar to an ideal rodlike cylindrical jet thereby yielding accurate results from the laminar jet absorber designed in this work. On the basis of the range of dimensions used in our work, it is recommended that the nozzle be a circular hole in a 0.07 ± 0.005-mm thick stainless steel sheet and that the inside diameter of this nozzle should be between 0.50 and 0.65 mm. We have confirmed that with this design, no wetting of the orifice face, no oscillations in the jets, and no interfacial resistance are observed. It is also recommended to feed the liquid by gravity to the jet chamber using a constant-head system with a blanket of inert gas. This technique gives a stable and constant liquid flow rate for several hours of continuous operation.

CO_2−MEA Absorption. The reaction between CO_2 and MEA solutions was investigated in the laminar jet absorber to further test the reliability of the new nozzle design and receiver of this work. The absorption experiments were carried out within the temperature range of 293−313 K because the CO_2−MEA kinetics is well defined within this temperature range in the literature where a second-order kinetics (eq 3) for CO_2 reaction with MEA solution has been reported.[23]

$$r = -k_2[\mathrm{MEA}][\mathrm{CO_2}] = k_{\mathrm{app}}[\mathrm{CO_2}] \qquad (3)$$

It is well-known that the accuracy of the deduced kinetic data decreases with increasing depletion of free amine toward the gas−liquid interface because the absorption flux is increasingly determined by diffusion.[24] Therefore, to minimize the effect of the diffusion limitation, we used loaded MEA solutions in our work to generate experimental kinetics data. This resulted in the difference between bulk and interfacial concentrations of free amine being kept as low as possible during the experiment. In addition, all the experimental data were interpreted with the aid of a developed numerically solved absorption-rate/kinetics model reported elsewhere[25] to obtain reliable kinetics or reaction-rate data.[11,23,24] This absorption-rate/kinetics model takes into account the coupling between chemical equilibrium, mass transfer, and chemical kinetics of all possible chemical reactions. The mathematical model is capable of predicting gas absorption rates and enhancement factors from the system hydrodynamics and the physicochemical properties, as well as predicting the kinetics of reaction from experimental absorption data.[25]

Details of the kinetics calculation from absorption data are documented elsewhere.[25,26] In brief, for each absorption rate obtained experimentally, the calculated enhancement factor by the absorption-rate/kinetics model was fitted to the experimentally observed enhancement factor with the apparent reaction-rate constant, k_{app}, as an adjustable parameter. The experimental results of these apparent reaction-rate constants are presented in Figure 8. A definite temperature dependence of the overall reaction rate can be observed for the three temperatures studied (293, 303, and 313 K). The data presented in Figure 8 were regressed to obtain the orders of reaction and the reaction rate constant at each temperature as presented in Table 2. From this table it can be seen that the obtained order of reaction is approximately one with respect to MEA within this temperature range and MEA concentrations. This is in agreement with the data reported in the literature.[23]

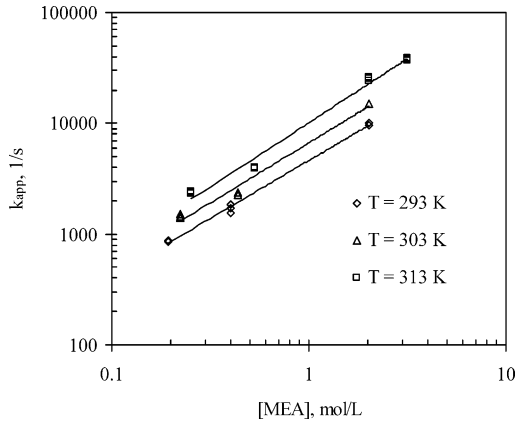

Figure 8. Experimental results of the apparent reaction-rate constants for CO_2 reaction with aqueous MEA solutions.

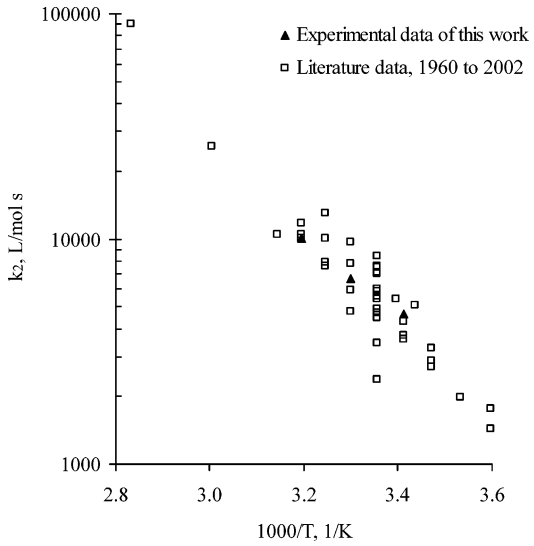

Figure 9. Arrhenius plot for the reaction between CO_2 and aqueous MEA solutions.

Table 2. Apparent Reaction Order and Second-Order Rate Constant for CO_2 Absorption into MEA Solutions, When Assuming $R = k_{app}[CO_2]$ and $k_{app} = k_2[MEA]^n$

T (K)	[MEA] (mol/L)	P_{CO2} (kPa)	av. $[CO_2]_{tot}$ (mol/L)	n (reaction order)	k_2 (L/mol·s)
293	0.193−2.016	91.59−92.97	∼1.81	1.04	4615
303	0.223−2.021	89.84−91.08	∼1.81	1.08	6674
313	0.254−3.158	86.97−88.54	∼1.83	1.16	10119

An Arrhenius plot was obtained using the second-order rate constants, k_2, presented in Table 2. The results are shown in Figure 9 as solid points. Also included in this figure are the literature kinetic data reported over a period of four decades (1960−2002) as reported elsewhere.[26] This figure shows that the reaction rate constants obtained in this work are in agreement with the kinetic data reported in the literature. Thus, the overall results confirm that the absorption

rates and the kinetic data obtained in the laminar jet apparatus are accurate. Again, this indicates the absence of any interfacial turbulence during the experimental work.

Conclusions

A novel design for the nozzle of a laminar jet absorber has been introduced. The nozzle is a circular hole in a 0.07 ± 0.005-mm thick stainless steel sheet, and the inside diameter of this nozzle is between 0.50 and 0.65 mm. A very thin sheet is used to minimize boundary-layer effects and to approach a flat velocity distribution at the charge point. In addition, a new design for the receiver of the liquid jet has been introduced. The receiver is a capillary-hole, 20.0 mm long with 1.0 mm diameter, drilled in an acrylic rod. This receiver design assisted in the collection of the liquid jet without any liquid spillover or gas entrainment inside the receiver for several hours of continuous operation. The nozzle and receiver designs gave high-quality absorption data obtained without interfacial resistance and without interfacial turbulence. The test results show that the agreement between the measured and calculated absorption rates is excellent over various liquid flow rates and jet lengths for two physical absorption systems, CO_2−water and N_2O−MEA. In addition, the diffusivities of CO_2 into water and N_2O into MEA solution, as well as the kinetics of the CO_2−MEA system at low concentrations and temperatures, were found to be in agreement with published data. These agreements indicate the reliability of the experimental apparatus design and configuration.

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