DESIGN, FABRICATION AND TESTING OF WETTED WALL COLUMN FOR CARBON CAPTURE USING AQUEOUS AMMONIA

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Abstract: Due to the increasing awareness of global warming, carbon mitigation methods are currently being promoted as means of prevention for this impending dilemma. Carbon capture and storage is seen as one of the means of mitigating the effects of global warming. Carbon capture technologies can be applied either at the pre-combustion stage, oxy-fuel combustion and post combustion stage. In this paper, absorption which is classified as a post combustion process was used as a means of capture carbon dioxide since most industries utilize packed columns. Absorbents used for this process are amine solutions or alkaline solutions. In this study, for instance, aqueous ammonia was the absorbent. The highlight of this paper focuses mainly on fabricating a wetted wall column to simulate the performance of CO_2 absorption from simulated flue gas using aqueous ammonia as sorbent. Testing the feasibility of the design parameters is done by varying two levels of each two factors namely concentration (1M & 3M) and temperature (3°C & 10°C) with the overall gas mass transfer coefficient as the response. Analysing the full factorial experiment with the use of Design Expert™, it is found that concentration of the lean absorbent charged into the system has a greater contribution in the performance of absorption. Percentage CO_2 removal is at the very least 72.97% at 1 M aqueous NH_3 and 98.33% at 3 M aqueous NH_3. Also, overall gas mass transfer coefficient (in mmol m^{-2} kPa^{-1} s^{-1}) exhibits values that lie around 0.2 when 1 M aqueous $NH_3$was used and 0.4 when 3 M aqueous NH_3 was used. These results proved that the greater the concentration of the lean absorbent, the greater fraction of carbon dioxide was removed from the simulated flue gas. The temperature, however, is found to have a slight contribution to the variation in the performance of absorption as far as the percentage of absorbed carbon dioxide and overall gas mass transfer coefficient are concerned. Hence, this may need to undergo further observation. The fabricated wetted-wall column reactor was proven to be fully functional albeit requiring some more installation of auxiliary precision devices and major overhaul of the absorbent line.

Key Words: Post-combustion, Carbon capture, Aqueous ammonia, Wetted wall column, Design

1. INTRODUCTION

Carbon dioxide is one of the greenhouse gases (GHG). The amount of carbon dioxide emissions into the atmosphere is so high (NaturalGas.org, 2004) aggravating global warming. Technologies today are emerging in order to capture and store this gas so as to be able to address the issues of CO_2's contribution to global warming.

The three most widely used methods in CO_2 capture are Post-Combustion, Oxyfuel Combustion, and Pre-Combustion. Membrane technology, adsorption, absorption, cryogenics, carbon extraction, biotechnology (cyanobacteria) and energy conversion (Feron & Hendriks, 2005) are some of other methods of Carbon Capture and Storage (CCS). Chemical looping cycle (CLC) which involves the transfer of oxygen with both the gas and fuel separated (Yang et al., 2008). Efficiency of each method will depend on the different parameters applied on the medium, kinds of fuel and the properties of the flue gas produced.

A commonly used method for CCS is absorption, a post-combustion process which relies on the use of absorbent liquid to reduce the amount of CO_2 solute from the flue gas. The monoethanolamine (MEA) is widely used and effective in absorption though it is very costly. According to Yeh et al. (2005), there is a 35% energy penalty for usage of MEA by coal-fired power plants to absorb CO_2. This is a large penalty value and is seen as one of the major problems of using MEA by power plants. Since air qualities imposed by international law cannot be compromised for the sake of cutting cost, measures such as searching for effective and cost-efficient alternatives are necessary. Aqueous ammonia, for instance can be an alternative. There are claims that ammonia can reduce the energy to operate the regeneration of solvent: 62% energy is saved using ammonia with 8% concentration (Yeh, et al., 2005).

Recently, aqueous ammonia was found to have a promising potential as a good absorbent and as an easily regenerated medium for reuse (Yeh, et al., 2005). Most studies regarding this involve the use of wetted wall column which is meant to simulate the packed tower. However due to its simple design, this is used mainly for determining the fraction of solute gas before and after contact in a steady state system.

The focus of this study was to fabricate a functional wetted wall column for gas absorption. To test the functionality of the column runs were conducted for the observation of the differences of CO_2 fractions in the solute (simulated flue gas) with aqueous ammonia as the absorbent. Also, significance of the effects of varying temperatures (3 &10^0C) and concentrations (1 & 3M) on the performance of absorption with the overall gas mass transfer coefficient and % CO_2 absorption were also assessed by design of experiments.

2. METHODOLOGY

2.1 Mass Transfer with Chemical Reaction

The rate of absorption can be described using Equation 1 (Liu, et al., 2009; Puxty, et al., 2010).

$$\boldsymbol{N_{CO_2} = K_G(P_{CO_2,b} - P^*)} \qquad \textbf{(Eq. 1)}$$

where: $N_{CO_2} = absorption\ flux, mmol/m^2 \cdot s$

$K_G = overall\ gas\ mass\ transfer\ coefficient, mmol/kPa \cdot m^2 \cdot s$

$P_{CO_2,b} = log\ mean\ average\ pressure, kPa$

$P^* = equilibrium\ partial\ pressure$

The equilibrium partial pressure is constant for a certain loading (Puxty, et al., 2010). A N_{CO_2} vs. $P_{CO_2,b}$ graph is plotted to obtain K_G as the slope and $K_G P^*$ as the constant. In order to do this, CO_2 composition has to be varied to obtain a different flux. Aside from the fact that the CO_2 inlet concentration cannot be varied, it is also found that the equilibrium pressure exerted at the rich absorbent portion ranges from 0.2 to 1.2 kPa (where factors in this study fall within that range) which can be neglected (Puxty, et al., 2010). Therefore the overall gas mass transfer coefficient is obtained by simply dividing the absorption flux with the log mean average CO_2 pressure as shown in Equation 2.

$$\boldsymbol{K_G = N_{CO_2}/P_{CO_2,b}} \qquad \textbf{(Eq. 2)}$$

2.2 Process Flow Diagram

The processes involved in this study are illustrated in the schematic diagram shown in Figure 1.

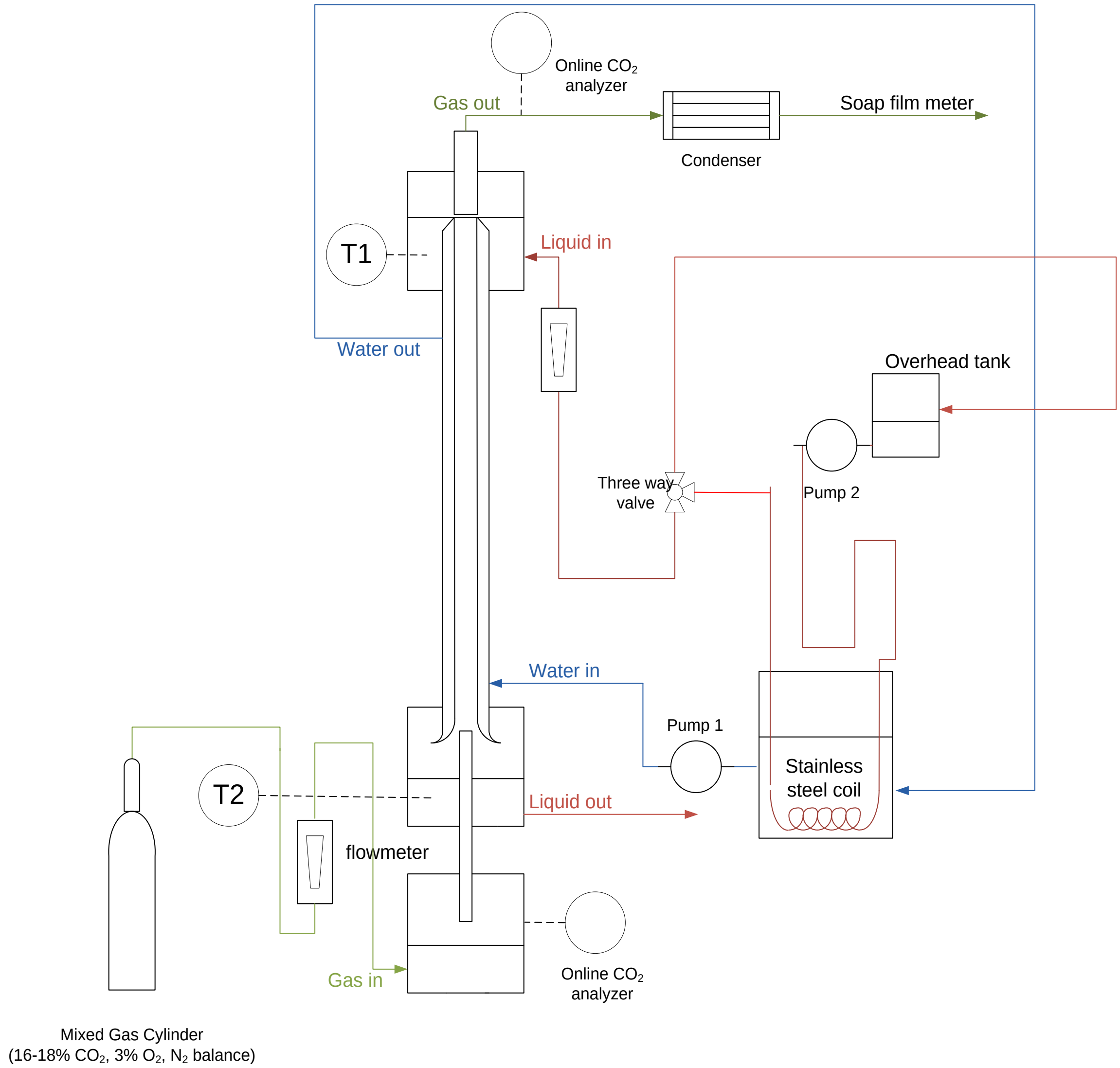

Figure 1. Schematic Process Flow Diagram

2.3 Design of Experiment

The experiment consists of 2 factors to be tested: concentration and temperature. Each of these factors has two numerical levels classified as high and low. Values are based upon what have been considered in different literatures.

Parameters for this experiment were chosen as prescribed by Darde et al. (2009) who used Eli Gal's patent (2006) as basis. As described in the patent, the process requires low operating temperatures at nearly atmospheric pressure due to the volatile nature of ammonia. Optimum temperature ranges from 0 to 10°C. The parameters for testing interactions with varying temperatures were therefore set to a minimum of 3°C and maximum of 10°C.

For the concentration of the absorbent, any concentration would be valid for as long as the solution could perform absorption efficiently. Darde et al. (2009) suggested that the concentration of the ammonia should be no higher than 22.33 M in order to avoid the formation of persistent precipitates at higher loading. Although no initial CO_2 loading was applied, it was also necessary to prevent the vaporization of ammonia for safety. Most literatures made use of concentrations ranging from 1 to 8M (Puxty, et al., 2010; Qin, et al., 2010; Rivera-Tinoco & Bouallou, 2010; Yeh, et al., 2005) with the latter found to be the largest. Hence, the levels for concentration were set at a minimum of 1 M and a maximum of 3 M.

Other parameters to be set constant are the gas flow rate which is at 1 LPM since further increase compromises the safety experiment. The flow rate of aqueous ammonia will vary slightly from 36~42 mL/min as long as ripples are eliminated for safety and stability as well. Temperature set for the cooling bath may not necessarily be stationary (due to the lack of equipment). Internal pressure inside of the column was kept constant at 100 kPa because this, considering the ambient place, is enough pressure to make the gas move up. The gas flow rate and liquid volume will also be kept constant. Liquid flow rate needed for each run is dependent upon the physical properties of the varying composition of the liquid solvent.

Table 1. Design of Experiment (General Factorial)

Factors:		Temperature (°C)	Concentration of solvent (mol/L or M)
Levels:	(low)	3	1
	(high)	10	3
Constant variables:			Number of factors: 2
Internal column pressure: 100 kPa			
Gas flow rate: 1 LPM			Number of levels for each factor: 2
Liquid flow rate: 36~42 mL/min			Number of replications: 3
Height: 28 in			Number of runs: 12
Responses: Overall gas mass transfer coefficient K_G and % CO_2 absorbed			

Table 1 shows the summary of the general factorial design. The runs total to 12: two factors with two levels each total to 4 single runs then multiplied by three replications. The response variables are the overall gas mass transfer coefficient and the %CO_2 absorbed.

3. RESULTS AND DISCUSSION

3.1 Performance of the Wetted Wall Column

It is among one of the primary objectives of this study to be able to successfully fabricate the wetted wall column and most of all ensure that it serves its function as a reactor for absorption. Aside from the fact that column should be able to eliminate ripples in the film, the basis for determining its capabilities were none other than the use of aqueous ammonia as the absorbent and simulated flue gas with carbon dioxide as the solute. The differences of the CO_2 fractions at the top and the bottom of the column during the execution of 12 runs were tabulated as shown in Table 2.

Table 2. Carbon Fraction Differences at the Top and Bottom of the Column

Setting	Run Number	CO_2 in (bottom of column, v/v)	CO_2 out (top of column, v/v)	CO_2Absorbed (%)
1M & 10°C	1	0.1480	0.0310	79.05
	2	0.1480	0.0340	77.01
	3	0.1480	0.0400	72.97
1M & 3°C	4	0.1410	0.0270	80.85
	5	0.1460	0.0250	82.88
	6	0.1490	0.0240	83.89
3M & 10°C	7	0.1135	0.0040	96.48
	8	0.1450	0.0060	95.86
	9	0.1490	0.0055	96.31
3M & 3°C	10	0.1500	0.0025	98.33
	11	0.1500	0.0030	98.00
	12	0.1500	0.0095	93.67

According to the data presented, CO_2 absorbed generally ranges from 73-98% which indicates that the performance of absorption is astoundingly significant. Furthermore, it can also be observed that the amount of CO_2increases as the concentration of ammonia increases, proving that the rate of absorption is proportional to the concentration of the solvent. However, for the temperature, the change cannot be clearly distinguished especially when the concentration of ammonia was increased. Nevertheless, it can be claimed that the fabricated wetted wall column is fully functional.

3.2 Overall Gas Mass Transfer Coefficients, K_G

Using Equation 2 and assuming that the internal pressure in the column is maintained 100 kPa, the overall gas mass transfer coefficients for each run were calculated. The average gas mass transfer coefficients were computed for each combination and are compared to the overall gas mass transfer coefficients of available combinations from other sources. The average gas mass transfer coefficients found for each combination are tabulated in Table 3.

Table 3. Average K_G (mmol m^{-2} kPa^{-1} s^{-1}) for Each Combination

Temperature \ Concentration	1M	3M
3° C	0.223	0.426
10° C	0.180	0.458

In order to verify whether the order of magnitude is valid, the data obtained is compared with the results from other literatures as shown in Table 4. Since this study does not cover the same temperature parameter as that of the other two sources, nearest temperature values are adopted. It can be seen that the values obtained in this experiment is smaller compared to the data presented from other sources. This is due to the fact that the height of the wetted wall columns used are usually of the heights ranging from 8-11 cm whereas the column used is about 71.12 cm . As mentioned earlier, the height affects the absorption rate in such way that a longer path tends to make the solute approach saturation and decrease the driving force as well. In other words, this also decreases the value of K_G. But the order of magnitude of the overall gas mass transfer coefficients obtained is the same as far as the variation of concentration is concerned. For temperature however, the effects cannot be clearly evaluated at this point.

Table 4. Comparison of Experimental Data on K_G from Other Literature

	Liu et. al	Puxty et. al	This Work
1M NH_3	0.698 (T=293°K)	-	0.180 (T=283°K)
3M NH_3	0.978 (T=293°K)	0.83 (T=283°K)	0.458 (T=283°K)

4. CONCLUSIONS

This study shows that the fabricated wetted wall column is fully functional albeit the fact that there still several flaws observed in terms of design and methodology.

Both the percentage of absorbed CO_2 and the overall gas mass transfer coefficients were found to be affected mainly by concentration. Percentage CO_2 removal is at the very least 72.97% at 1 M aqueous NH_3 and 98.33% at 3 M aqueous NH_3. Also, overall gas mass transfer coefficient (in mmol m-2 kPa-1 s-1) exhibits values that lie around 0.2 when 1 M aqueous NH_3 was used and 0.4 when 3 M aqueous NH_3 was used. For temperature however, slight significance was found in its effects on the percentage of absorbed CO_2 but there's ultimately none for the overall gas mass transfer coefficient. This may be due to the disturbance caused by the weir temperature value.

5. ACKNOWLEDGEMENTS

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