PRINT ISSN 1119-8362
Electronic ISSN 2659-1502

Full-text Available Online at
https://www.ajol.info/index.php/jasem
http://www.bioline.org.br/ja

J. Appl. Sci. Environ. Manage.
Vol. 27 (6) 1069-1075 June 2023

Modeling and Simulation of CO_2 Capture Unit using Mixed Solvent of Aqueous Methyldiethanolamine and Piperazine for 6.4 MWe Power Plant

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ABSTRACT: Carbon dioxide emissions are the largest greenhouse gases contributor, which is attributed to global warming. Therefore, the capture of CO_2 is necessary, especially from large point sources. Post-combustion capture using chemical solvents is the most matured capture technology. In this paper, mixed solvents of methyldiethanolamine (MDEA) and piperazine (PZ) were studied to potentially address some issues related to the use of conventional monoethanolamine (MEA). Steady-state modeling and simulation of the post-combustion carbon capture using the mixed solvents for a typical 6.4 MWe power plant were done. The model developed was validated against experimented data with relative error between 1 – 4.73% for the absorber on CO_2 capture efficiency and between 2.93 – 10.2% relative errors on CO_2 regeneration efficiency. The model was scaled up to capture CO_2 from the flue gas of a typical 6.4 MWe power plant. The scaled-up results showed packing height and diameter of absorber to be 5.5 m and 1.5 m; while that of the stripper was 5.08 m and 1.45 m respectively. Effect of flue gas temperature on the capture efficiency was studied, it was observed that as the temperature increased from 20 ºC, the capture efficiency increased, but at 70 ºC, the efficiency showed no significant increase which could be associated with the existence of a temperature bulge. Capture efficiency increases with an increase in PZ concentration due to increase in the rate of reaction with concentration. The absorption and regeneration results suggest that an aqueous MDEA/PZ blend could be a prospective absorbent for CO_2 capture.

DOI: **https://dx.doi.org/10.4314/jasem.v27i6.4**

Cite this paper as: UMAR, S. I; JOEL, A. S; HAMZA, U. D. (2023). Modeling and Simulation of CO_2 Capture Unit using Mixed Solvent of Aqueous Methyldiethanolamine and Piperazine for 6.4 MWe Power Plant. *J. Appl. Sci. Environ. Manage.* 27 (6) 1069-1075

Dates: Received: 19 April 2023; Revised: 20 May 2023; Accepted: 06 June 2023
Published: 30 June 2023

Keywords: carbon capture; methyldiethanolamine; piperazine; power plant

Fossil fuels used in thermal power plants have become the foremost source of greenhouse gases related to human activity, primarily due to the discharge of carbon dioxide into the atmosphere (Reddick *et al*., 2016). Continuous efforts, research, and commitments are being made to reduce the severity of future climate change due to the increase in greenhouse gas emissions (Sreedhar *et al*., 2017). Carbon dioxide is one of the primary waste gases in the exhaust of factories, businesses, and transportation vehicles (Babamohammadi *et al*., 2015). The most significant contemporary issue with carbon dioxide is its greenhouse effect on the atmosphere, leading to global warming. However, carbon dioxide capture has been identified as a fundamental solution to significantly address this issue (Sreedhar *et al*., 2017). The three major carbon dioxide capture technologies are pre-combustion, oxy-fuel combustion, and post-combustion. Pre-combustion CO_2 capture entails capturing CO_2 from fossil fuel or biomass prior to the completion of the combustion process (Zhang *et al*., 2020). In oxy-fuel combustion, oxygen is separated from the air prior to combustion, and the fuel is burned in oxygen diluted with recirculated flue gas instead of air. The post-combustion method involves the capture of CO_2 from gas streams resulting from the

combustion of fossil fuels or other carbonaceous materials. However, research and development activities concerning solvent-based post-combustion CO_2 capture, with a focus on reducing the energy consumption of the system, have been demonstrated worldwide through pilot-scale post-combustion carbon dioxide capture (Ali *et al*., 2016). Ali *et al*. (2016) used MEA for the techno-economic process design of a commercial-scale amine-based CO_2 capture system for a natural gas combined cycle power plant with exhaust gas recirculation. Also, Lawal *et al*., (2010) reported an absorber for demonstrating full-scale post-combustion CO_2 capture for a coal-fired power plant through modeling and simulation, which was also huge. Zhang *et al*., (2017) used aqueous piperazine to model an absorber pilot plant's performance for CO_2 capture. They reported that at high concentrations of PZ, it crystallized, which is a great setback for the solvent. Borhani *et al*., (2015) used MDEA for a modeling study on the simultaneous removal of CO_2 and H_2S. They reported a slower reaction of MDEA with CO_2, which is a disadvantage of the solvent. The objective of this paper is therefore to report the modeling and simulation of CO_2 capture unit using mixed solvent of aqueous methyldiethanolamine and piperazine for 6.4 MWe power plant.

MATERIALS AND METHOD

*Motivation***:** The choice of a suitable solvent for absorption is essential to lowering the energy penalty of the CO_2 capture process. Most of the research carried out using individual solvents was reported to have some setbacks. Borhani *et al*., (2015) uses MDEA as a solvent and reported a lower reaction rate of MDEA with CO_2. Zhang *et al*., (2017) uses PZ as a solvent and reported that at higher concentrations of PZ, it crystallized. However, using the blend of MDEA and PZ as solvent will enhance CO_2 capture since PZ acts as a promoter in speeding up the reaction while MDEA contributes its advantages of being chemically stable and requiring low heat for regeneration.

Model Development: The two major units in the CO_2 capture process are an absorber and a stripper. The unit operation model used to develop the absorber and the stripper is the RadFrac rate-based model. The rate-based model is an extension of the RadFrac equilibrium distillation model, and its computations provide more reliable results compared to the equilibrium-based model (Otitoju *et al*., 2020). Essential data for pure components and binary interaction parameters for species such as CO_2, H_2O, CO, and hydrocarbons were all available in Aspen Plus Databanks.

Thermodynamic and Kinetic Model: The thermodynamic model adopted for the CO_2 absorption process was the electrolyte non-random two-liquid activity coefficient (ELECTNRTL) because of its versatility and appropriate properties for aqueous and mixed solvent systems (Mudhasakul *et al*., 2013). However, Equations 1 through 8 provide the chemical equilibrium reactions for the electrolyte system in conjunction with E-NRTL (Hemmati *et al*., 2019).

$$2H_2O \leftrightarrow H_3O^+ + OH^- \quad (1)$$
$$CO_2 + 2H_2O \leftrightarrow HCO_3^- + H_3O^+ \quad (2)$$
$$HCO_3^- + H_2O \leftrightarrow CO_3^{2-} + H_3O^+ \quad (3)$$
$$MDEAH^+ + H_2O \leftrightarrow MDEA + H_3O^+ \quad (4)$$
$$PZH^+ + H_2O \leftrightarrow PZ + H_3O^+ \quad (5)$$
$$PZ + HCO_3^- \leftrightarrow PZCOO^- + H_2O \quad (6)$$
$$PZH^+COO^- + H_2O \leftrightarrow PZCOO^- + H_3O^+ \quad (7)$$
$$PZCOO + HCO_3^- \leftrightarrow PZCOO_2^- + H_2O \quad (8)$$

Equations 1-4 present the chemical equilibrium for the MDEA-CO_2 system, while for the PZ-CO_2 system, it is presented in Equations 5-8. PZ can associate with H^+ to form an ion (PZH^+), reaction 4, and react with CO_2 to form a carbamate ion ($PZCOO^-$), reactions 6 and 7, and a dicarbamate ion ($PZCOO_2^-$), reaction 8. The reaction mechanism for CO_2 with a tertiary amine (e.g. MDEA) is a base-catalyzed hydration reaction. It can associate with H^+ from $MDEAH^+$ reaction 4. Equations 9–11 are bicarbamate reactions for MDEA (Hemmati *et al*., 2019). Another set of chemical reactions (9–21) were implemented separately for the PZ + MDEA + CO_2 + H_2O system, as the reactions in the absorber are both in equilibrium and rate-controlled. All ionic reactions were assumed to have chemical equilibrium, except those between CO_2 and OH^-, CO_2 and $PZCOO^-$, and CO_2 and MDEA which are considered as kinetics-controlled reactions (Li *et al*., 2020).

$$2H_2O \leftrightarrow H_3O^+ + OH^- \quad (9)$$
$$HCO_3^- + H_2O \leftrightarrow CO_3^{2-} + H_3O^+ \quad (10)$$
$$PZH^+ + H_2O \leftrightarrow PZ + H_3O^+ \quad (11)$$
$$H^+PZCOO^- + H_2O \leftrightarrow PZCOO^- + H_3O^+ \quad (12)$$
$$MDEAH^+ + H_2O \leftrightarrow MDEA + H_3O^+ \quad (13)$$
$$CO_2 + OH^- \rightarrow HCO_3^- \quad (14)$$
$$HCO_3^- \rightarrow CO_2 + OH^- \quad (15)$$
$$PZ + CO_2 + H_2O \rightarrow PZCOO^- + H_3O^+ \quad (16)$$
$$PZCOO^- + H_3O^+ \rightarrow PZ + CO_2 + H_2O \quad (17)$$
$$PZCOO^- + CO_2 + H_2O \rightarrow PZ(COO^-)_2 + H_3O^+ \quad (18)$$
$$PZ(COO^-)_2 + H_3O^+ \rightarrow PZCOO^- + CO_2 + H_2O \quad (19)$$
$$MDEA + CO_2 + H_2O \rightarrow MDEAH^+ + HCO_3^- \quad (20)$$
$$MDEAH^+ + HCO_3^- \rightarrow MDEA + CO_2 + H_2O \quad (21)$$

The power law expressions used to form the rate-controlled reactions (14-21) is presented in Equation 22 adopted from (Otitoju *et al*., 2021)

$$r_j = k_j exp\left(-\frac{E_j}{R_c}\left[\frac{1}{T}-\frac{1}{298.15}\right]\right)\prod_{i=1}^{N} a_i^{\alpha_{ij}} \quad (22)$$

The kinetic parameters for reactions in Equations 14 – 21 were listed in Table 1

Table 1. Kinetic parameters of CO_2 absorption by MDEA (Li *et al*., 2020)

Reaction NO.	K	E, Cal/mol
14	1.33e+17	13,249
15	6.63e+16	25,656
16	1.70e+10	319
17	3.40e+23	14,160
18	1.04e+14	8,038.3
19	3.20e+20	8,692
20	6.85e+10	9,029
21	6.22e+17	22,131

Model Implementation Procedure: The step-by-step method employed for the process simulation in this paper is depicted in Figure 1. The model implementation in Aspen Plus® is shown in Figure 2.

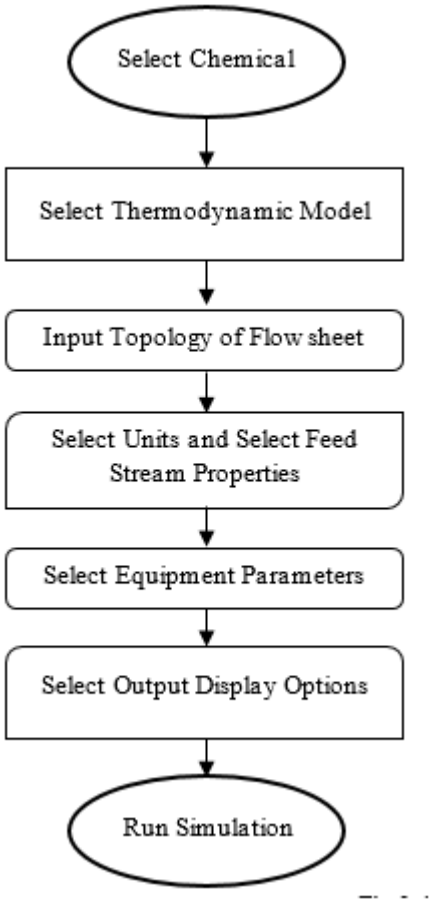

Fig. 1: Steps in Process Simulation (Turton *et al*., 2019).

Model Validation: The model validation was carried out using the experimental data of Ali *et al*. (2017). The experimental parameters used for the model validation are: packing type: heli-pack with the following specifications: size (inch) 0.175, material type: nonreactive metal, surface area of 1500 m^2/m^3, void fraction of 84%, packing density of 1450 k/m^3, solvent flowrate of 3 x 10^{-3} m^3 min^{-1}, and the blend of (MDEA 20 wt% + PZ 10 wt%).

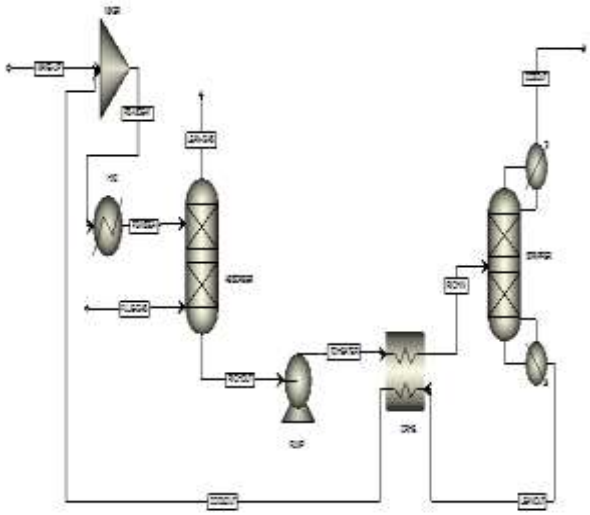

Fig. 2: Model implementation flow sheet in this study.

$$\text{CE} = \left(\frac{\text{kg } CO_2 \text{ in flue gas} - \text{kg}CO_2 \text{ in outlet gas}}{\text{kg } CO_2 \text{in flue gas}}\right) \times 100 \quad (23)$$

$$\%\ \text{Error} = \left(\frac{\text{ED} - \text{SD}}{\text{ED}}\right) \times 100 \quad (24)$$

$$\text{RE} = \left[\left(1 - \frac{y_{CO_2,out}}{y_{MEA,out} + y_{PZ,out}}\right) \Big/ \left(\frac{y_{CO_2,in}}{y_{MEA,in} + y_{PZ,in}}\right)\right] \times 100 \quad (25)$$

Where CE = Capture Efficiency; ED = Experimental Data; SD = Simulation Data; RE = Removal Efficiency

Table 2: The stream conditions and specification.

Parameter	Component	Description	Unit
Flue gas flow rate		5, 6, 7 & 8	lit/min
Solvent flow rate		0.3	lit/min
Flue gas compositions	CO_2	15	mol %
	O_2	10	mol %
	N_2	75	mol %
Flue gas pressure		1	atm
Inlet solvent temperature		40	°C
Inlet flue gas temperature		25	°C
Absorber & Stripper column's packed height		0.84	m
Absorber & Stripper column's diameter		0.04	m

Source: Ali et al., 2017

Table 3 presents the simulation results compared to the experimental results for four different gas flow rates of 5, 6, 7, and 8 lit/min. The results showed that CO_2

capture efficiency for experimental results is in agreement with the model results with a relative error of 1.73 – 4.73%. Therefore, the model developed has predicted the absorber unit quite well since the relative error is quite small.

Table 3: Absorber experimental result compared to validated result.

Flue gas flow rate (lit/min)	CO_2 Capture Efficiency (%)		Relative Error (%)
	Experimental (Ali *et al*., 2017)	Simulation	
5	76.86	76.13	1
6	73.46	74.35	1.2
7	71.22	72.82	2.25
8	68.34	71.57	4.73

Table 4: Regenerator experimental result compared to model result.

Temperature K	Regeneration Efficiency (%)		Relative Error (%)
	Experimental	Simulation	
374	80.50	82.90	2.98
371	75.2	80.90	7.58
368	72.50	79.90	10.2

Table 4 presented the regenerator simulation result compared to experimental results for three different temperature conditions of 374, 371 and 368 K and the modeling results are in agreement with the experimental results with relative error ranging from 2.98 – 10.2%. It was observed from the prediction that as the temperature decreased, the relative error increased. This was attributed to the poor performance of the regenerator at lower temperatures, leading to lower CO_2 capture efficiency predictions.

Model Scale-up: Based on the model prediction, which showed that the relative error is less than 5% for the absorber and less than 10.5 % relative error for the stripper. The model is now scaled up for 90% capture of CO_2 from a 6.4 MWe power plant. The scale-up is centered on the design of the packed column because the diameter of a packed column is a fundamental factor that must be determined in the design of a packed bed absorber or stripper. Therefore, the 6.4 MWe power plant used has flue gas composition of CO_2 16.3%, O_2 6.4%, N_2 63%, and H_2O 14.3%. To determine the size of the absorber and the stripper, scale-up calculations were done. Table 5 presents the scaled-up target values for the 6.4 MWe power plant with an absorption capture efficiency of 90 % and a stripper regeneration efficiency of 85%.

Evaluation of lean solvent flowrate: The lean solvent flow rate required to capture 90% of the CO_2 from a 6.4 MWe power plant was estimated based on the absorption capacity, lean solvent MDEA/PZ concentration, CO_2 mass fraction, and flue gas mass flow rate. The equation relating the lean solvent flow rate to the amount of CO_2 recovered from the flue gas stream, the mass fraction of the amine in the unloaded solution (Amine), and the lean amine solution CO_2 loading was given by Equation 26 Agbonghae *et al*., (2014).

$$L_{Lean} = \frac{Gx_{CO2}\varphi_{CO2}}{100Z(\alpha_{Rich}-\alpha_{Lean})}\left[\frac{M_{MDEA/Pz}}{44.009}\left(1+\frac{1-\omega_{MDEA/Pz}}{\omega_{MDEA/Pz}}\right)+Z\alpha_{Lean}\right] \quad 26$$

The lean solvent flow rate was calculated to be 0.065 m3/s.

*Evaluation of columns diameter***:** The diameter of the absorber and the stripper was estimated using Eqs. 27–30 adopted from Otitoju *et al*., (2020)

$$V_{G.fl} = 0.3048\left[\left(\frac{\rho_G}{\rho_L-\rho_G}\right)^{-0.5}\upsilon^{-0.05}F_P^{-0.5}\left\{A\left(log\left(\frac{L}{G}\sqrt{\frac{\rho_G}{\rho_L}}\right)\right)^2+ B\left(log\left(\frac{L}{G}\sqrt{\frac{\rho_G}{\rho_L}}\right)\right)+C\right\}\right] \quad 27$$

$$V_G = 0.7V_{G.fl} \quad 28$$

$$D = \sqrt{\frac{4G}{\pi V_G \rho_G}} \quad 29$$

$$\Delta P_{fl} = 0.115F_P^{0.7} \quad 30$$

Data regarding the density and kinematic viscosity of the MDEA/PZ solvent was attained from the validated model. The absorber and the stripper were packed with Metal Heli-Pak packing type (0.175). The values obtained for the absorber and stripper diameters are 1.5 m and 1.45 m, respectively.

Evaluation of packing height: The height of packing required for a given separation in a packed column is most often expressed in terms of the overall gas-phase mass transfer coefficient and the gas composition. Based on this, the packing height of the column can be calculated with the expression adopted from Otitoju *et al*., (2020);

$$Z_H = H_{OG.}N_{OG}$$

$$N_{OG} = \int_{yCO_{2,in}}^{yCO_{2,out}}\frac{dy}{y-y^*} = \ln\left(\frac{y_{CO2,in}}{y_{CO2,out}}\right)$$

$$H_{OG} = \frac{G_i}{K_G aP}$$

Based on the above, the packing height (Z_H) of the absorber was calculated to be 5.5m and that of stripper was 5.08m.

Table 5: Scaled Up Targeted Values.

	Value	Unit
Absorption Efficiency	90	%
Removal Efficiency	85	%
Amine Blend	20/10	%

Process Analysis: In this section, process analysis for the scaled-up model was carried out with the absorber and the stripper packing height and diameter of 5.5 m and 5.08 m, 1.5m and 1.45m respectively. The analysis was carried out in order to look at the effect of some parameters on the performance of the power plant. The parameters considered are the effect of flue gas temperature on the capture efficiency, the effect of piperazine concentration on the capture efficiency, the effect of liquid-to-gas ratio on the capture efficiency, the effect of PZ concentration on the reboiler duty, and the effect of L/G ratio on the reboiler duty as well.

Effect of flue gas temperature on the capture efficiency: Temperature is one of the significant factors for gas-liquid absorption in a packed column (Li *et al*., 2020). From Figure 3, it was observed that with an increase in the temperature of flue gas in the column, the capture efficiency increases. The increase in the capture efficiency is in the range of 90.299 to 94.429 with the temperature varying from 20 to 80 ^{0}C, when considering the blend of (MDEA 20 wt% + PZ 10 wt%).

However, at 60 °C, the capture efficiency eventually stabilizes and starts decreasing as the flue gas temperature exceeds 70 °C. This is associated with the presence of a temperature bulge, which drops the absorption performance. This is in agreement with the work of Kvamsdal *et al*., (2011); Rochelle, GT; Kvamsdal, HT (2008) where it was found that CO_2 capture efficiency increases with an increase in temperature and decreases at higher temperatures due to the existence of a temperature bulge. Also, Joel *et al*., (2021) carried out a process analysis on the effect of lean solvent temperature on the capture efficiency for an intensified absorber using MEA solvent. The finding also demonstrated that the effect of temperature bulge reduces the CO_2 absorption rate since the reaction between CO_2 and the solvents is exothermic.

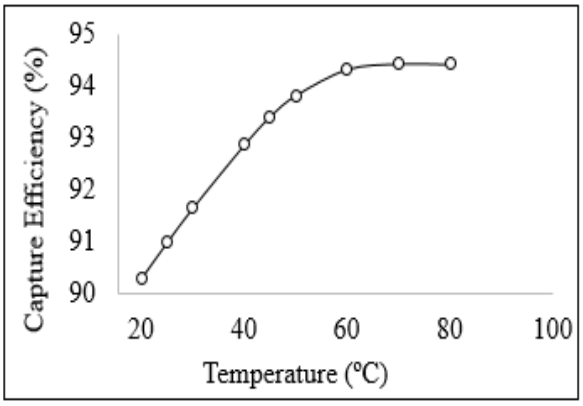

Fig. 3: Effect of Flue Gas Temperature on the Capture Efficiency.

Effect of PZ concentration on the capture efficiency: Pz is one of the most significant parameters that mainly influences the mass transfer force driving the CO_2 capture process (Mudhasakul *et al*., 2013). According to Figure 4, the capture efficiency increases with an increase in PZ concentration. This is because PZ is a promoter and also enhances the capturing performance of CO_2. The gradual addition in the amount of PZ from 2 wt% to 10 wt% to the solution of MDEA results in considerable improvement in the capture efficiency because of structurally two nitrogen atoms in the molecule of PZ and higher rate of reaction with CO_2. However, there is a limitation to using higher concentrations of PZ because at higher concentrations it crystallizes, which reduces the surface area and capture efficiency. In this work, the highest percentage of PZ used was 10% to tackle the issue of crystallization. Figure 4 showed a similar trend to what was reported by Ali *et al.* (2016), who used PZ and AMP solvents for CO_2 capture and found that the percentage of CO_2 absorbed increased with an increase in PZ concentration.

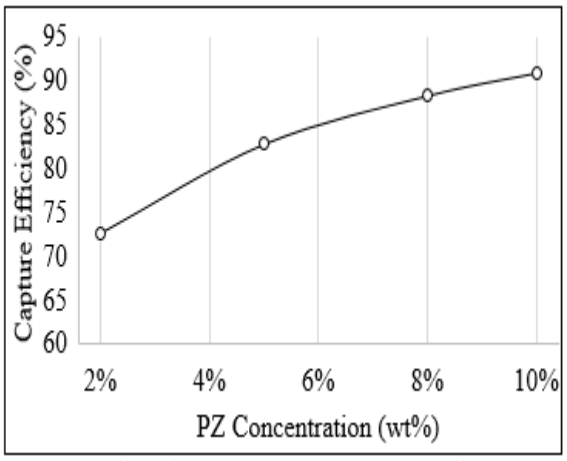

Fig. 4: Effect of PZ Concentration on Capture Efficiency.

Effect of PZ concentration on the Reboiler duty: Reboiler heat duty measurement is one of the dynamic studies essential for the regeneration of CO_2-loaded amine solvent for CO_2 capture through an absorption-desorption system (Aghel *et al*., 2019). Figure 5 shows that reboiler heat duty increases with increase in PZ because heat of absorption (kJ/molCO_2) of PZ is higher than that of MDEA and more heat will be required to break up the MDEA-PZ composite formation during regeneration process. That is, the higher the PZ concentration, the lower the MDEA concentration, and the higher the reboiler heat duty. Mudhasakul *et al*. (2013) reported that any PZ concentration beyond 5 wt% causes the reboiler duty per captured CO_2 to increase significantly. For example, as the PZ concentration increases from 5

wt% to 7.5 wt%, the reboiler duty per captured CO_2 goes up from 74.66 kJ/mol to 199.88 kJ/mol. Also, Ali *et al*. (2016) reported that reboiler heat duty gradually increases with an increase in concentration of PZ, i.e., 2 wt%, 5 wt%, 8 wt%, or 10 wt%, in the aqueous blend of AMP and PZ because the heat of absorption (kJ/mol CO2) of PZ is higher than that of AMP. Therefore, the trend obtained in Figure 5 is in agreement with previous studies (Ali *et al*., 2016; Mudhasakul *et al*., 2013).

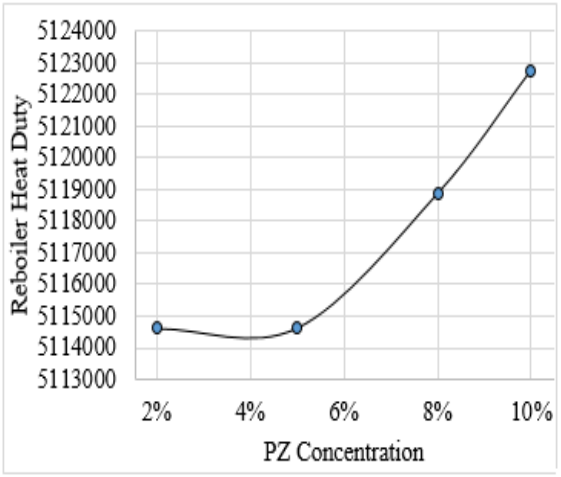

Fig. 5: Effect of PZ Concentration on Reboiler Heat Duty.

Effect of Liquid to Gas ratio concentration on the capture efficiency: The increase in liquid to gas ratio leads to the increase in energy of regeneration for a close loop capture system because of the high amount of solvent to be regenerated in the regenerator (Samanta & Bandyopadhyay, 2011). Examining the liquid-to-gas ratio is important so as to get the optimum point of high capture efficiency with less regeneration energy in the regenerator. Figure 6 shows that the liquid-to-gas ratio increases rapidly from 3.66 kg/kg to 6.36 kg/kg due to the high amount of solvent available for CO_2 capture, but when the L/G ratio is increased beyond 6.36 kg/kg, there is no substantial increase in the rate of CO_2 absorption. This is due to the fact that the amount of solvent is sufficient to capture most of the CO_2. Joel *et al*. (2021) also carried out a process analysis on the effect of the liquid-to-gas ratio on the capture efficiency and observed a similar trend.

Effect of Liquid to Gas ratio on the reboiler duty: The figure below shows that reboiler heat duty increases with an increase in the liquid-to-gas ratio. This is because increasing the liquid-to-gas ratio leads to higher CO_2 loading, which in turn leads to higher boiler heat duty. That is, the volume of the liquid matters for the amount of energy consumption. However, the effect is small at low concentration of the liquid but becomes more at higher concentration of the liquid. Mudhasakul *et al*., (2013) carried out a similar study but on effect of solvent to feed ratio in his work and reported that as solvent to feed ratio increase the reboiler heat duty increase. From his work, the trend on the graph looks similar to the trend in this study.

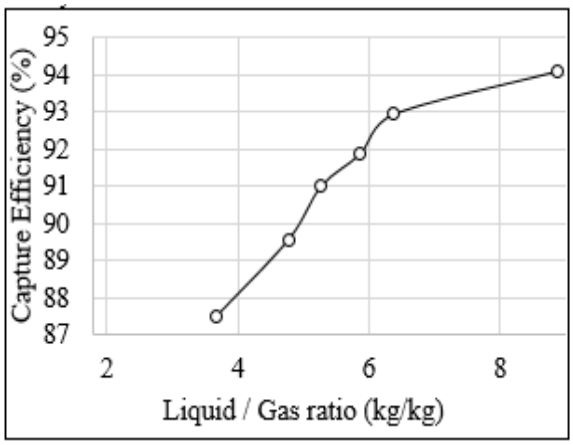

Fig. 6: Effect of Liquid to Gas Ratio on Capture Efficiency.

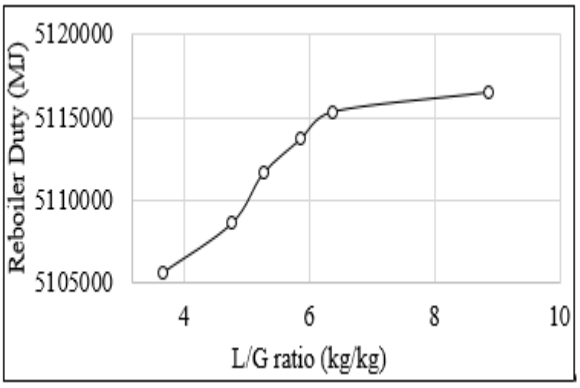

Fig. 7: Effect of Liquid to Gas Ratio on Reboiler Duty.

Conclusions: Modeling and simulation of post-combustion carbon dioxide capture using mixed solvents of MDEA and PZ was done in Aspen Plus®. Model validation for both absorber and regenerator showed small error and it closely agreed with experimental results. The validated model was scaled up to capture CO_2 from the flue gas of a typical 6.4 MW power plant. The scale-up result showed that the absorber have a packing height of 5.5 m and diameter of 1.5 m; and the stripper have a packing height of 5.08 m and diameter of 1.45 m. Increase in temperature of the flue gas entering the column increases the capture efficiency for temperature range 25 ^{0}C to 60 ^{0}C, but at 70 ^{o}C, the efficiency began to show no significant increase due to temperature bulge. The results of this study on absorption and

regeneration demonstrated that that aqueous (MDEA + PZ) blend could be a prospective absorbent for CO_2 capture associated to conventional amine.

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