Modelling and Optimization of Natural Gas Sweetening Unit Using Piperazine-Activated Methyldiethanolamine as Solvent

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Abstract

A kinetic model for prediction and performance optimization of an 80-barg high pressure, high capacity, natural gas sweetening columns using the electrical conductivity properties (in conjunction with other process datasets such as concentration, circulation rate, pH, temperature, pressure, and CO² content) has been developed. Findings reveal a strong negative correlation coefficient (-0.9742759) between piperazine-activated methyldiethanolamine (PZ-Activated MDEA) concentration and electrical conductivity. A predictive model expressed as (y=54.8848602+(-0.017323229)x) determines and enhances the optimal operating conditions, enabling real-time monitoring and automatic intervention. The post-optimization results suggest a significant improvement in CO² absorption efficiency which is exemplified in the characteristic reduction in the lean gas average residual CO² content from 1,368.74 ppm to 248.06 ppm commercial quality. Other key improvements include an increase in the PZ+MDEA stream average concentration from 36.47 g/100ml to 42.66 g/100ml and a notable amine outlet stream temperature rise (+6°C) alongside a pH reduction of 2.37 due to enhanced absorption. These adjustments led to a leap in CO² removal efficiency from 93.06% to 98.75% within a 14-day interval, effectively minimizing the production of lean natural gas with high CO² levels.

Keywords: Natural gas sweetening; kinetic modelling; electrical conductivity effect; Piperazine-Activated Methyldiethanolamine.

1.0 Introduction

Gas sweetening plays a crucial role in the removal of acid gases $(H₂S$ and $CO₂)$ from natural gas streams, enhancing its quality and marketability. Among the various amine-based processes, the Piperazine-Activated Methyldiethanolamine (PZ+MDEA) system has gained significant attention due to its high CO_2 absorption capacity, excellent H_2S selectivity, and reduced corrosion potential (Berchiche, *et al.,* 2023). The optimization of PZ-Activated MDEA absorption units is essential to improve process efficiency, reduce operating costs, and meet environmental regulations. This paper presents a detailed kinetic model for the lean PZ+MDEA inlet system, which accurately captures the complex chemical reactions and mass transfer processes involved in the gas sweetening unit. The model is validated against experimental data from industrial-scale PZ+MDEA packed columns, demonstrating its high predictive capabilities. (Wehrung *et al.,* 2023;

Zhan *et al.,* 2023). This work presents results of a predictive model and optimization for the absorption of CO₂ into PZ activated MDEA solvent.

2.0 Methodology

From the bottom tray of the absorption column, and assuming a binary component, the rich natural gas stream is fed and flows upward counter-currently to the lean Piperazine-AQctivated Methyldiethanolamine solution, and when carbon dioxide $(CO₂)$ is absorbed into the mixture of the down-flowing Piperazine ($C_4H_{10}N_2$), Methane based tertiary alkanol amine R1 R2 R3 N and demineralized water blend or PZ+MDEA solution, several reversible proton transfer exothermic reactions take place and MDEA, water, carbamate or bicarbamate products with water are formed. (Valluri, and Kawatra, 2021). Typical chemistry of this reaction is shown below (see reactions 1 to 11).

$$
CO_2 + R_1 R_2 R_3 N + H_2 O \stackrel{K_1 K_{21}}{\longleftrightarrow} R_1 R_2 R_3 N H^+ + HCO_3^- \tag{1}
$$

$$
CO_2 + PZ + H_2O \stackrel{K_2.K_{22}}{\longleftrightarrow} PZCOO^- + H_3O^+
$$
 (2)

$$
CO_2 + R_1 R_2 R_3 N + PZ \stackrel{R_3, R_{23}}{\sim} PZCOO^- + R_1 R_2 R_3 N H^+ \tag{3}
$$

$$
CO_2 + PZCOO^{-} + H_2O \xleftrightarrow{\Lambda_4, \Lambda_2}_V PZ(COO^{-})_2 + H_3O^{+}
$$
\n(4)

$$
CO_2 + R_1 R_2 R_3 N + PZCOO^{-} \xleftrightarrow{K_5, K_{25}} PZ(COO^{-})_2 + R_1 R_2 R_3 N H^{+}
$$
 (5)

$$
CO_2 + OH^- \xleftrightarrow{\Lambda_{6} \Lambda_{26} \atop K_7} HCO_3^- \tag{6}
$$

$$
HCO_3^- + H_2O \underset{\kappa}{\leftrightarrow} CO_3^{2-} + H_3O^+ \tag{7}
$$

$$
PZ + H_3 O^+ \stackrel{K_8}{\leftrightarrow} PZH^+ + H_2 O \tag{8}
$$

$$
PZCOO^{-} + H_3O^{+} \stackrel{K_9}{\leftrightarrow} PZH^{+}COO^{-} + H_2O
$$
\n
$$
\tag{9}
$$

$$
R_1 R_2 R_3 N + H_3 O^+ \stackrel{K_{10}}{\longleftrightarrow} R_1 R_2 R_3 N H^+ + H_2 O \tag{10}
$$

$$
2H_2O \stackrel{K_{11}}{\leftrightarrow} H_3O^+ + OH^- \tag{11}
$$

2.1 Correlation Coefficient, *r*

Correlation Coefficient, r:

$$
r = \frac{(n(\Sigma xy)) - (\Sigma x)(\Sigma y)}{\sqrt{((n(\Sigma x^2)) - (\Sigma x)^2)(n(\Sigma y^2) - (\Sigma y)^2)}}
$$
(12)

where:

 $n =$ number of observations

 $x =$ Independent continuous level variable (Activated Methyl-di-ethanolamine) a-MDEA concentration.

 $y =$ Dependent continuous level variable (Activated Methyl-di-ethanolamine) a-MDEA electrical conductivity.

$$
r = \frac{n(\Sigma xy) - (\Sigma x)(\Sigma y)}{\sqrt{\left(\left(n(\Sigma x^2)\right) - \left((\Sigma x)^2\right)\right)\left(n(\Sigma y^2) - (\Sigma y)^2\right)}}\tag{13}
$$

$$
=\frac{15(586,363.06) - (526.73)(17,132)}{\sqrt{(15(18,774.3) - (526.73)^2)(15(20,446,358) - (17,132.0)^2}}
$$
(14)

$$
=\frac{8,795,445.9-9,023,938.36}{\sqrt{(281,614.5-277,444.493)(306,695,370-293,505,424)}}
$$
(15)

$$
=\frac{-228,492.46}{\sqrt{(4,170.007)(13,189,946)}} = \frac{-228,492.46}{\sqrt{55,002,167,149.622}} = \frac{-228,492.46}{234,525.4083}
$$
(16)

$$
r = -0.9742759
$$
 (Very strong negative correlation coefficient) (17)

The test statistics are in the critical region at the left-hand end of the correlation coefficient scale range. The test result of -0.9742759 is greater than the critical value, thus there is significant evidence:

where:

y = Line of Regression (Least Squares Line)

 $a =$ The y Intercept *(the value of y or a-MDEA concentration, when x or a-MDEA conductivity is/equals zero).*

 = Slope of the Line *(the increase in y or a-MDEA concentration, for every 1 unit increase in x or a-MDEA conductivity).*

y = Activated Methyl-di-ethanolamine (a-MDEA) *concentration* (Dependent variable)

 $x =$ Activated Methyl-di-ethanolamine (a-MDEA) *conductivity* (Independent variable). \bar{x} = Activated Methyl-di-ethanolamine *conductivity* or mean of Independent variable. \bar{y} = Activated Methyl-di-ethanolamine *concentration* or mean of Dependent variable. $n =$ number of sets of observations.

$$
b = \frac{n(\sum xy) - (\sum x)(\sum y)}{n(\sum x^2) - (\sum x)^2} \tag{21}
$$

$$
b = \frac{15(586,363.06) - (17,132.0)(526.73)}{15(20,446,358.0) - (17,132.0)^2}
$$
(22)

$$
b = \frac{8,795,445.9 - 9,023,938.36}{306,695,370 - 293,505,424} = \frac{-228,492.46}{13,189,946}
$$
(23)

$$
b = -0.017323229 \tag{24}
$$

$$
a = \bar{y} - b\bar{x} \tag{25}
$$

$$
a = 35.1 - (-0.017323229) * 1,142.1
$$
\n⁽²⁶⁾

$$
a = 35.1 - (-19.784860269) = 54.8848602 \tag{27}
$$

 $a = 54.8848602$ (28)

Regression ()

$$
y = a + bx \tag{29}
$$

$$
Regression \t y = 54.8848602 + (-0.017323229)x \t(30)
$$

The equation above can predict with 95% accuracy (5% margin of error) the a-MDEA concentrations (lean or rich) from the amine electrical conductivity or vice-versa. Where:

- (y) = Piperazine activated Methyl-di-ethanolamine (a-MDEA) concentration in (g/100ml).
- (x) = Piperazine-activated Methyl-di-ethanolamine (a-MDEA) conductivity micro S/cm

The a-MDEA **concentration value or when (conductivity) is 694?**

Therefore, **694 micro S/cm** increase in the Piperazine-Activated Methyldiethanolamine (MDEA) electrical conductivity will result in **42.86254 g/100ml** change in the activated Piperazine-Activated Methyldiethanolamine sample concentration.

Conversely to solve for (x) = Piperazine-activated Methyl-di-ethanolamine (a-MDEA) conductivity in micro S/cm:

 $x = 693.999958$ (44)

Piperazine-Activated Methyl-di-ethanolamine (MDEA) conductivity = 694.0 micro S/cm

3.0 Results

1. CO² Absorption Column-A Pre-optimization PZ+MDEA Inlet and Outlet Conductivity and Concentration Difference:

The $CO₂$ Absorption Unit consist of parallel operating column-A/B/C with separate natural gas, and amine inlet and outlet spools from the main Header, while unit-C was on stand-by, the $CO₂$ Absorption Column-A, pre-optimization Piperazine-activated Methyldiethanolamine inlet stream analyzed average electrical conductivity at 25° C 1,112.14 micro S/cm and outlet 11,767.6 micro S/cm at 36.40 g/100ml inlet and 35.01 g/100ml outlet solution concentration within the initial 14 days observational period.

2. CO² Absorption Column-A Pre-optimization PZ+MDEA Inlet and Outlet pH and Temperature Difference:

The Absorption Column-A, pre-optimization Piperazine-activated Methyl-di-ethanolamine inlet and outlet stream analysed pH and temperature difference, there was an average $6^{\circ}C$ gain on the outlet stream with a corresponding decrease of 2 on the pH scale within the 14-day observational period.

3. CO² Absorption Column-A Post-optimization PZ+MDEA Inlet and Outlet pH and Temperature Difference:

The Absorption Column-A, post-optimization Piperazine-activated Methyl-di-ethanolamine inlet and outlet stream analysed pH and temperature difference, there was an average 6.8° C gain on the outlet stream with a corresponding decrease of 2.2 on the pH scale within the final 14-day observational period.

4. CO² Absorption Column-B Pre-optimization PZ+MDEA Inlet and Outlet Conductivity and Concentration Difference:

The Absorption Column-B, pre-optimization Piperazine-activated Methyl-di-ethanolamine inlet stream analyzed average electrical conductivity at 25° C of 1,023.40 micro S/cm and outlet stream conductivity of 9,074.29 micro S/cm at 36.37 g/100ml amine inlet strength, and corresponding 35.16 g/100ml outlet solution concentration within the initial assessment period.

5. CO² Absorption Column-B Pre-optimization PZ+MDEA Inlet and Outlet pH and Temperature Difference:

The Absorption Column-B, pre-optimization Piperazine-activated Methyl-di-ethanolamine inlet and outlet stream analysed pH and temperature profiles relationship, there was an average 6^oC increase on the outlet stream with a corresponding decrease of 1.9 on the pH scale within the initial 14-day period.

6. CO² Absorption Column-B Post-optimization PZ+MDEA Inlet and Outlet pH and Temperature Difference:

The Absorption Column-B, post-optimization Piperazine-activated Methyl-di-ethanolamine inlet and outlet stream analysed pH and temperature assessment, there was an average $6.1^{\circ}C$ improvement on the outlet stream with a corresponding decrease of 2.23 on the pH scale within the final 14-day period.

7. CO² Absorption Column-A/B Pre and Post-optimization PZ-MDEA Inlet Conductivity Average Difference:

The Absorption Column-A/B, pre-optimization Piperazine-activated Methyl-di-ethanolamine inlet stream electrical conductivity variable analyzed at 25° C of 1,069.1 and 1,142.1 micro S/cm relationship, with an analyzed average amine concentration inlet stream of 36.47 g/100ml declining in the opposite directions, occasionally crossing path near the optimal values and drifting wider apart from the mean.

8. CO² Absorption Column-A/B Pre and Post-optimization PZ-MDEA Inlet Concentration Average Difference:

The Absorption Column-A/B, average pre-optimization Piperazine-activated Methyl-diethanolamine intlet stream analyzed concentration of 36.47 g/100ml is relatively lower than the post-optimization 42.66 g/100ml outlet stream average, figure-1.

9. CO² Absorption Column-A/B Pre and Post-optimization PZ-MDEA Inlet Conductivity Average Response:

Absorption Column-A/B, Pre-optimization Piperazine-activated Methyl-di-ethanolamine inlet stream analysed average electrical conductivity at 25° C of 697.85 micro S/cm relationship with the post-optimization PZ+MDEA inlet stream average concentration of 42.66 g/100ml converges more at the optimal values near the centre, same response holds for the PZ+MDEA outlet average electrical conductivity of 1,293.5 micro S/cm at 41.0 g/100ml strength.

10. CO² Absorption Column-A/B Pre-optimization PZ-MDEA Inlet Conductivity and Concentration, Correlation, Regression and Mathematical Model:

Absorption Column-A/B pre-optimization Piperazine-activated Methyl-di-ethanolamine inlet stream analyzed electrical conductivity and concentration average data analysis resulted in a very strong negative correlation coefficient of -0.9742, a regression linear relationship of 0.9491 and a mathematical model expressed as $y = 54.8848602 - 0.017323229x$.

11. CO² Absorption Column-A/B Post-optimization PZ-MDEA Inlet Conductivity and Concentration, Correlation and Regression:

Absorption Column-A/B post-optimization Piperazine-activated Methyl-di-ethanolamine inlet stream analyzed electrical conductivity and concentration average results data analysis reported an improved very strong negative correlation coefficient of -0.992447762 and a linear regression relationship of 0.985.

12. CO² Absorption Column-A/B Post-optimization PZ-MDEA Inlet Conductivity Model Data Validation Differential:

Absorption Column-A/B post-optimization Piperazine-activated Methyl-di-ethanolamine inlet stream average electrical conductivity model data was validated against analyzed data and closely matched the laboratory observed values at an average differential of 7.843 micro S/cm.

Table 1.0: Absorption Column-A/B PZ-Activated MDEA Conductivity Vs Concentration Inlet (equation 14)

Date	\boldsymbol{n} (Observations)	PZ+MDEA (Inlet) Conductivity Analyzed (micro S/cm) χ	PZ+MDEA (Inlet) Concentration Analyzed (g/100ml) \mathcal{Y}	y_i^2	x_i^2	$(x_i * y_i)$
$Day-1$	1	1,460.0	28.82	2,131,600.0	830.6	42,077.20
$Day-2$	$\overline{2}$	1,425.0	29.52	2,030,625.0	871.4	42,066.00
$Day-3$	$\overline{3}$	1,414.0	30.23	1,999,396.0	913.9	42,745.22
$Day-4$	$\overline{4}$	1,383.0	30.69	1,912,689.0	941.9	42,444.27
Day-5	5	1,351.0	31.86	1,825,201.0	1,015.1	43,042.86
Day-6	6	1,320.0	33.04	1,742,400.0	1,091.6	43,612.80
Day-7	$\overline{7}$	1,289.0	34.21	1,661,521.0	1,170.3	44,096.69
Day-8	8	1135.00	34.96	1,288,225.0	1,222.2	39,679.60
Day-9	9	1,048.0	35.38	1,098,304.0	1,251.7	37,078.24
$Day-10$	10	994.00	35.85	988,036.0	1,285.2	35,634.90
$Day-11$	11	961.0	39.34	923,521.0	1,547.6	37,805.74
$Day-12$	12	924.0	39.58	853,776.0	1,566.6	36,571.92
$Day-13$	13	922.0	40.30	850,084.0	1,624.1	37,156.60
$Day-14$	14	812.0	40.54	659,344.0	1,643.5	32,918.48
Day- 15	15	694.00	42.41	481,636.0	1,798.6	29,432.54
TOTAL: Σ		17,132.0	526.73	20,446,358.0	18,774.3	586,363.06
Mean (μ) :		1,142.1	35.1			

The study found that Piperazine-Activated Methyldiethanolamine (PZ+MDEA) exhibits a strong negative correlation (-0.9742) between electrical conductivity and concentration, which shifts to a highly negative correlation (-0.992447762) after optimization (figure-7), as validated by comprehensive experiments including linear regression and sensitivity analysis of process variables (Abdulkhaleq, 2016). Absorption Column-A pre-optimization showed an inlet and outlet electrical conductivities of 1,112.14 and 11,777.86 micro S/cm (figure-6), respectively, with slight decreases in solution concentration from inlet to outlet and a notable 6°C temperature increase in

the outlet stream, alongside a 2-point pH decrease (figure-2&3). These findings relationship pattern were consistent with unit-B response over the 28-day investigation period, and highlight the significant impact of PZ+MDEA on the post- optimization.

Figure 1.0: Column-A/B: Pre-Post-Optimization PZ+MDEA Inlet Concentration

Figure 2.0: Column-A: Post-Optimization PZ+MDEA Inlet/Outlet pH

The observed elevation in Absorption Column-A/B pre-optimization PZ+MDEA outlet electrical conductivity average from 1,112.14 micro S/cm at the inlet to 11,777.86 micro S/cm at the outlet (figure-6), alongside the decline in lean amine inlet concentration from $36.47 \frac{\text{g}}{100 \text{ml}}$ to 35.04 $g/100$ ml (figure-1&5), a decrease in pH (figure-2&3), and a rise in outlet temperature, can be attributed to the $CO₂$ absorption by the PZ+MDEA solution through a base-hydration mechanism. This process, an exothermic reaction, not only strips the $CO₂$ from the incoming rich natural gas

Unit-B: Post-Optimization PZ+MDEA Inlet Vs Outlet pH (Decrease Δ in pH = 2.23) 12.0 11.5 11.0 PZ+MDEA pH 10.5 Unit-B: Post-Optimization 10.0 PZ+MDEA Inlet pH 9.5 Unit-B: Post-Optimization 9.0 PZ+MDEA Outlet pH 8.5 8.0 **12-13-09**

but also increases PZ+MDEA outlet acidity, as evidenced by the lower pH values observed (Ullah, 2022).

Figure 3.0: Column-B: Post-Optimization PZ+MDEA Inlet/Outlet pH

Figure 4.0: Column-A/B: Pre-Post-Optimization PZ+MDEA Inlet Conductivity

An analysis of the Piperazine-activated Methyl-di-ethanolamine (PZ-MDEA) mixture in an absorption column pre-optimization study revealed a strong negative correlation (-0.9742) between electrical conductivity and concentration (figure-7). Utilizing a regression model (Famoye & Singh, 2021) and a mathematical formula (Peng *et al.,* 2021), the optimal concentration range for PZ-MDEA was identified as $40 - 45$ g/100ml. This concentration ensures optimal $CO₂$ removal efficiency, monitored and regulated via inline sensors (Steinhauser *et al.,* 2020) and a feedback control system (Chen *et al.,* 2021). The PZ+MDEA inlet stream average electrical conductivity model predicted data was validated against the analyzed data, and showed a very close match at an average differential of 7.843 micro S/cm thus highlighting (figure-8) the reliability and significance of the model and inline monitoring automated system. (Pan *et al.,* 2022).

Figure 5.0: Column-A/B: Pre-Post-Optimization PZ+MDEA Outlet Concentration

Figure 6.0: Column-A/B: Pre-Post-Optimization PZ+MDEA Outlet Conductivity -Average

After optimizing the absorption column-A/B, the rich natural gas inlet flow-rate average was maintained at 155,000.0 std m3/hr, with a $CO₂$ concentration of 19,675.01ppm threshold limit (Shankar *et al.,* 2022). Post-optimization, the average concentration of the PZ+MDEA solution

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inlet stream increased from 36.47 g/100ml to 42.66 g/100ml, improving its efficiency (Khan *et al.,* 2017). Additionally, there was a notable increase in the amine outlet stream's temperature by $+6^{\circ}\text{C}$ and a decrease in pH by 2.37, facilitated by an automatic inline monitoring system (Huang *et al.,* 2017; Penn & Camberato, 2019). These adjustments helped reduce the $CO₂$ content in the produced gas from 1,368.74ppm to 248.06ppm average (figure-9&10), effectively preventing the production of off-specification sweet natural gas with high CO₂ levels (Sahl *et al.*, 2021). As a result, the unit's performance was optimized from 93.06% to 98.75%, marking a 5.69% improvement within a 14-days post-optimization period (Zhang *et al.,* 2023).

Figure 7.0: Column-A/B: Pre Vs Post-optimization PZ+MDEA Inlet variables scatter plot

Figure 8.0: Column-A/B: Post-optimization PZ+MDEA inlet conductivity model Vs analyzed data

Figure 9.0: Column-A/B Pre-optimization averages: PZ+MDEA inlet conductivity Vs concentration and outlet residual CO² content

Figure 10.0: Column-A/B: Post-optimization average performance assessment

Figure 11.0: Column-A/B Pre and Post-optimization outlet gas residual CO² content

Figure 12.0: Column-A/B Pre and Post-optimization CO² removal performance average

4.0 Conclusion

The study demonstrated that Piperazine-Activated Methyldiethanolamine (PZ+MDEA) significantly influences electrical conductivity, $CO₂$ absorption, and other chemical properties in an absorption column, both before and after optimization processes. Initially, a strong (-0.9742) negative correlation between electrical conductivity and concentration of PZ+MDEA was observed, which further intensified to (-0.992447762) post-optimization. The model predicting the electrical conductivity of the PZ+MDEA inlet stream was validated and found to closely align with analyzed data, with an average difference of just 7.843 micro Siemens per centimetre. This highlights the model and the automated inline monitoring system's reliability and importance. Before optimization, measurements showed a substantial increase in electrical conductivity and temperature alongside a decrease in pH and solution concentration from the column inlet to the outlet, indicating CO_2 absorption by the PZ+MDEA solution. After optimization, the efficiency of the absorption process improved, evidenced by better $CO₂$ removal and adjusted chemical concentrations, leading to a 5.69% enhancement in unit performance. This process optimization, supported by regression analysis and online monitoring, significantly increases the removal efficiency of $CO₂$ from natural gas, ensuring the production of gas within the desired 250.0ppm residual CO² specifications.

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