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Simulation of Gas Sweetening Process using Extended NRTL and Stages Efficiency as Modeling Approach

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ABSTRACT. Gas sweetening is a process to remove CO_2 and H_2S from natural gas. The current established technology is by using Amine contactor where the solvent used is in form of Amine solution. To simulate the effect of different solvent, electrolyte-NRTL is used to model the equilibrium, and mass transfer-kinetic is used to model the rate-based processes. This modeling approach is rather complex and available only in commercial and proprietary process simulation software. Therefore we propose an alternative modeling approach where we use extended NRTL and stage efficiency to model the acid gas absorption processes. We find that this approach is quite good for describing CO_2 absorption, yet unsuccessful in calculating the H_2S absorption. Inadequate vapor-liquid equilibrium parameter regression for H_2S, specifically at low partial pressure might cause the problem. However, the stage efficiency approach shows good results where it is comparable to the rate-based model and corresponds to the current understanding of the physicochemical phenomenon of acid gas absorption. The simulation of the pilot plant for both the absorption and regenerator unit has been constructed and shows good agreement toward reference.

1. INTRODUCTION

Natural gas produced from gas fields usually contains impurities such as CO_2 and H_2S gas. This gas is called acid gas. The high content of acid gas could impose several problems in processing facilities such as corrosion. It could also reduce the gas heating value thus lowering the sales gas price. Especially for H_2S, it is a very poisonous gas that can inflict a serious health problem or even mortality. Therefore, the removal of acid gas is necessary for natural gas production. The method to lower or remove the acid gas content is called gas sweetening[1].

Current established techniques for gas sweetening is by absorption process using amine solvents such as monoethanolamine (MEA), diethanolamine (DEA), dipropanolamine (DIPA), and dipropanolamine (MDEA) solution. MDEA possesses several advantages such as lower vapor pressure, more stable at high temperatures, lower corrosion risk, and lower heat of reaction [2] The rich solvent is then regenerated in the regeneration unit by introducing heat via a stripper therefore it can be used for the next cycle.

The solvent regeneration is an energy-intensive process. To reduce the energy requirement, a co-solvent with low specific heat is added such as ethylene glycol (EG). The effect of EG addition as a co-solvent had been investigated by a pilot scale experiment and it was found that a significant reduction (25%) of reboiler duty was obtained. The have a better understanding of the effect of EG addition, a model was built by applying an electrolyte-NRTL (Non-Random Two Liquid) thermodynamic package to model the vapor-liquid equilibrium (VLE). Moreover, a rate-based model is developed to accommodate the reaction and mass-transfer phenomena that occur in the absorption tower. Those approaches were successful in meeting the experiment data[3].

The utilization of a rate-based model is found to be computationally demanding and leads to longer simulation time. Therefore another approach is proposed by applying an equilibrium stage model combined with stage efficiency to model the kinetic and mass transfer inside the absorption tower. This approach was applied for the post-combustion CO_2 capture process and gave a good agreement toward experiment data[4]. On the other hand, the electrolyte-NRTL thermodynamics model is only available in a commercial simulator like Aspen Plus™, especially through commercial licensing. Therefore, in this work, we propose a different approach, by applying a rather general Extended-NRTL to model the VLE combined with stage efficiency to model the rate processes in the absorption tower. In current work, we implement this modeling approach by using the pilot plant data and simulation results using Aspen Plus published by Gonzalez, et.al (2023) as a comparison.

2. MATERIALS AND METHODS

2.1 Materials

In this study, we perform modeling and simulation using the Unisim Design 490 academic license, where the license is acquired from the Process Computation Laboratory in the Chemical Engineering Department UPN Veteran Yogyakarta. The experimental data is referred to the work by Gonzalez, et.al. (2023). We also use Microsoft Excel to process and analyze the simulation data.

The hardware that we utilize has the following specifications:

- Processor: Intel Core i5 12th Gen
- RAM: 8 GB DDR5
- Graphic Processor: Nvidia Geforce RTX 3050

2.2 Methods

Our work is started by acquiring gas sweetening experiment data from Gonzalez, et.al. (2023), which consists of the process flow diagram, VLE data, and pilot plant experiment. Then, we develop our VLE modeling to obtain the appropriate model parameter that matches the VLE experiment. We continue by developing the absorption model to match the pilot plant absorption data. After we get our best model, we continue to the simulation of the complete gas sweetening process, followed by result analysis. The flow diagram of our work is shown in Figure 1.

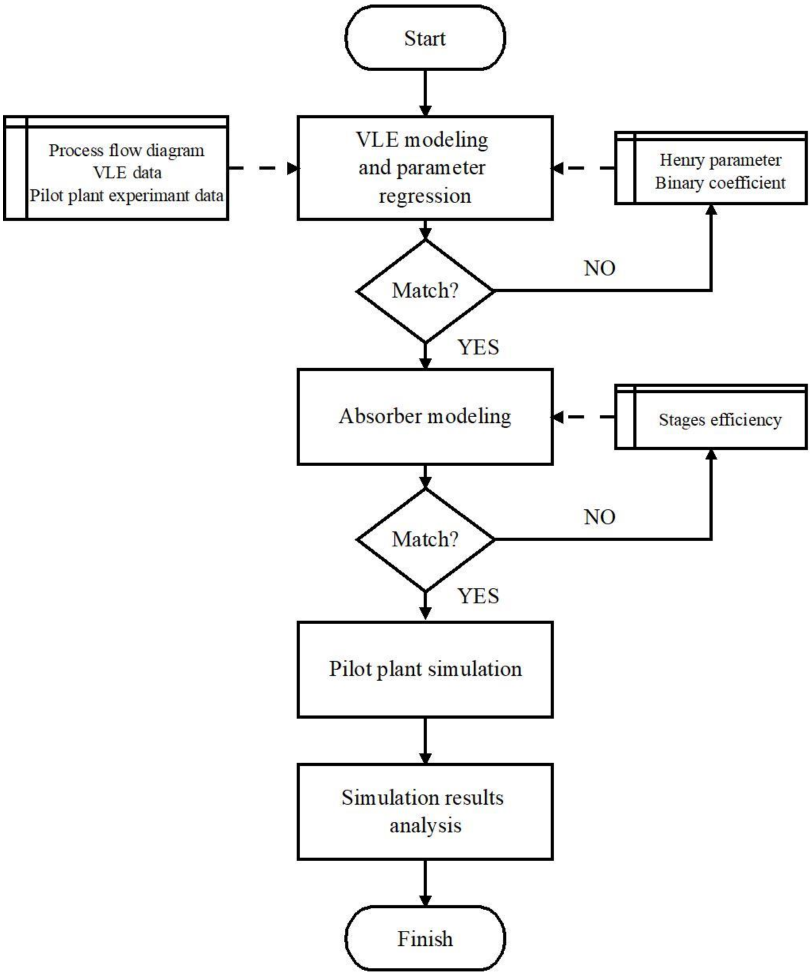
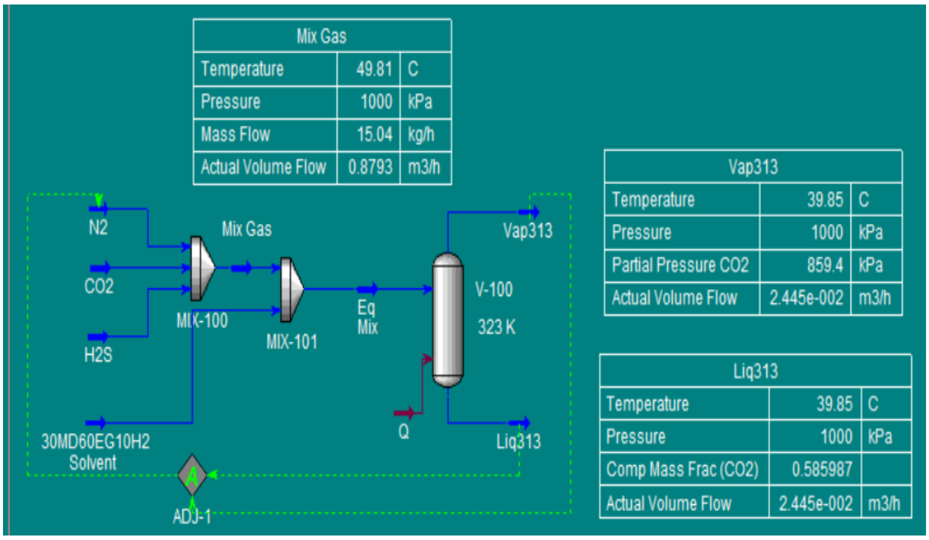


Figure 1. Workflow Diagram

We developed our VLE model in Unisim Design 490 by using separator tools to facilitate an equilibrium calculation. The example for the simulation model is presented in Figure 2. The adjustment tools are used to create the condition where the actual volume of the gas phase and liquid phase is similar, as indicated by the experiment result. The simulation result is presented as the partial pressure of CO_2 and H_2S in the gas phase versus the loading of CO_2 and H_2S in the liquid phase.



Mix Gas		
Temperature	49.81	C
Pressure	1000	kPa
Mass Flow	15.04	kg/h
Actual Volume Flow	0.8793	m3/h

Vap313		
Temperature	39.85	C
Pressure	1000	kPa
Partial Pressure CO2	859.4	kPa
Actual Volume Flow	2.445e-002	m3/h

Liq313		
Temperature	39.85	C
Pressure	1000	kPa
Comp Mass Frac (CO2)	0.585987	
Actual Volume Flow	2.445e-002	m3/h

Figure 2. VLE modeling for solvent mixture 30% MDEA, 60% EG, and 10% H_2O at 323 K

Due to the dilute concentration of the gas in the solvent, the extended Henry's law is used (1):

$$\phi_i^v y_i P = \gamma_i x_i H_i \tag{1}$$

ϕ_i^v is the vapor phase fugacity coefficient, which in our work is calculated by the Peng-Robinson model. H_i is Henry's constant of solute i in the solvent mixture, where it is calculated from Henry's constant of solute in each pure solvent. In Unisim, it is calculated by (2). A, B, C, and D are coefficients where they accommodate the temperature influence of Henry's constant.

$$\ln H_{ij} = A + \frac{B}{T} + C \ln T + DT \tag{2}$$

The pressure correction for H_{ij} is performed by Poynting term as shown in (3)

$$H_{ij}(P) = H_{ij}\left(P_j^{sat}\right) \exp\left(\frac{1}{RT} \int_{P_j^{sat}}^{P} V_{ij}^{\infty}\, dP\right) \tag{3}$$

γ_i is liquid phase activity coefficient, where the extended-NRTL is used to calculate it. Extended-NRTL is a more flexible form of NRTL model where we can specify $\alpha_{ij} \neq \alpha_{ji}$. For this model, we insert the parameter in the form of $A_{ij}, B_{ij}, C_{ij}, \alpha_{1ij}$, and α_{2ij}

$$\ln \gamma_i = \frac{\sum_{j=1}^{n} \tau_{ji} x_j G_{ji}}{\sum_{k=1}^{n} x_k G_{ki}} + \sum_{j=1}^{n} \left(\tau_{ij} - \frac{\sum_{m=1}^{n} \tau_{mi} x_m G_{mj}}{\sum_{k=1}^{n} x_k G_{kj}}\right) \tag{4}$$

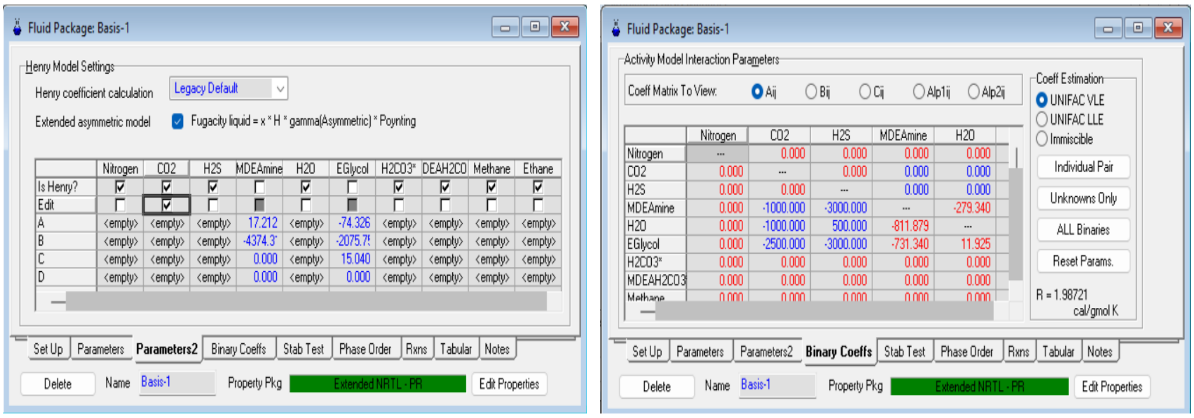
$$G_{ij} = \exp\left(-\tau_{ij}\alpha_{ij}\right) \tag{5}$$

$$\tau_{ij} = A_{ij} + B_{ij} t + \frac{C_{ij}}{T} \tag{6}$$

$$\alpha_{ij} = \alpha_{1ij} + \alpha_{2ij} \tag{7}$$

The input of the above parameters in Unisim is illustrated in Figure 3. The VLE parameter is regressed [5]. For the modeling of the rate process, we specify the stage efficiency as shown in

Figure 4. The stage efficiency is a parameter that is provided in Unisim to accommodate how much the solution in the stages approaching equilibrium. Equilibrium is achieved by mass transfer of solute from gas to solvent.



Fluid Package: Basis-1

Henry Model Settings

Henry coefficient calculation: Legacy Default

Extended asymmetric model: ☑ Fugacity liquid = x * H * gamma(Asymmetric) * Poynting

	Nitrogen	CO2	H2S	MDEAmine	H2O	EGlycol	H2CO3*	DEAH2CO	Methane	Ethane
Is Henry?	☑	☑	☑	☐	☑	☐	☑	☑	☑	☑
Edit	☐	☑	☐	■	☐	■	☐	☐	☐	☐
A	<empty>	<empty>	<empty>	17.212	<empty>	-74.326	<empty>	<empty>	<empty>	<empty>
B	<empty>	<empty>	<empty>	-4374.3	<empty>	-2075.7	<empty>	<empty>	<empty>	<empty>
C	<empty>	<empty>	<empty>	0.000	<empty>	15.040	<empty>	<empty>	<empty>	<empty>
D	<empty>	<empty>	<empty>	0.000	<empty>	0.000	<empty>	<empty>	<empty>	<empty>

Name: Basis-1

Property Pkg: Extended NRTL - PR

Fluid Package: Basis-1

Activity Model Interaction Parameters

Coeff Matrix To View: ● Aij ○ Bij ○ Cij ○ Alp1ij ○ Alp2ij

	Nitrogen	CO2	H2S	MDEAmine	H2O
Nitrogen	---	0.000	0.000	0.000	0.000
CO2	0.000	---	0.000	0.000	0.000
H2S	0.000	0.000	---	0.000	0.000
MDEAmine	0.000	-1000.000	-3000.000	---	-279.340
H2O	0.000	-1000.000	500.000	-811.879	---
EGlycol	0.000	-2500.000	-3000.000	-731.340	11.925
H2CO3*	0.000	0.000	0.000	0.000	0.000
MDEAH2CO3	0.000	0.000	0.000	0.000	0.000
Methane	0.000	0.000	0.000	0.000	0.000

Coeff Estimation: ● UNIFAC VLE ○ UNIFAC LLE ○ Immiscible

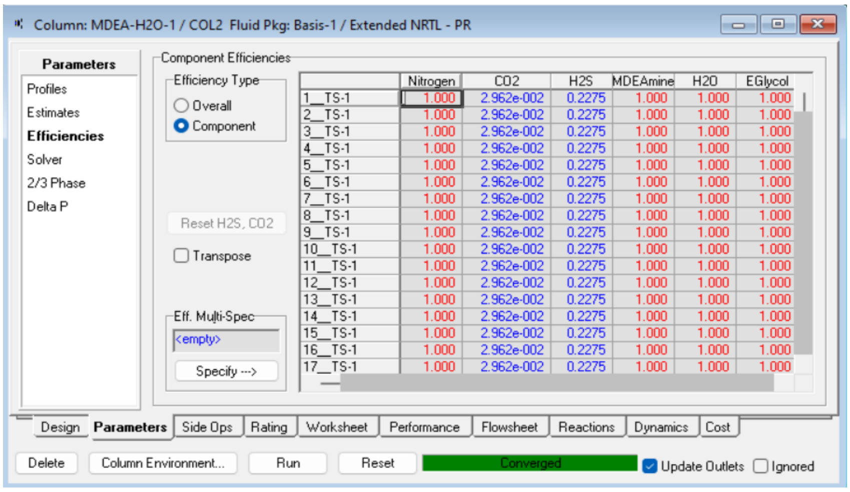
R = 1.98721 cal/gmol K

Name: Basis-1

Property Pkg: Extended NRTL - PR

(a) (b)

Figure 3. Input VLE parameters in Unisim (a) Henry Constant (b) Extended NRTL



Column: MDEA-H2O-1 / COL2 Fluid Pkg: Basis-1 / Extended NRTL - PR

Component Efficiencies

Efficiency Type: ○ Overall ● Component

☐ Transpose

Eff. Multi-Spec: <empty>

	Nitrogen	CO2	H2S	MDEAmine	H2O	EGlycol
1__TS-1	1.000	2.962e-002	0.2275	1.000	1.000	1.000
2__TS-1	1.000	2.962e-002	0.2275	1.000	1.000	1.000
3__TS-1	1.000	2.962e-002	0.2275	1.000	1.000	1.000
4__TS-1	1.000	2.962e-002	0.2275	1.000	1.000	1.000
5__TS-1	1.000	2.962e-002	0.2275	1.000	1.000	1.000
6__TS-1	1.000	2.962e-002	0.2275	1.000	1.000	1.000
7__TS-1	1.000	2.962e-002	0.2275	1.000	1.000	1.000
8__TS-1	1.000	2.962e-002	0.2275	1.000	1.000	1.000
9__TS-1	1.000	2.962e-002	0.2275	1.000	1.000	1.000
10__TS-1	1.000	2.962e-002	0.2275	1.000	1.000	1.000
11__TS-1	1.000	2.962e-002	0.2275	1.000	1.000	1.000
12__TS-1	1.000	2.962e-002	0.2275	1.000	1.000	1.000
13__TS-1	1.000	2.962e-002	0.2275	1.000	1.000	1.000
14__TS-1	1.000	2.962e-002	0.2275	1.000	1.000	1.000
15__TS-1	1.000	2.962e-002	0.2275	1.000	1.000	1.000
16__TS-1	1.000	2.962e-002	0.2275	1.000	1.000	1.000
17__TS-1	1.000	2.962e-002	0.2275	1.000	1.000	1.000

Converged

☑ Update Outlets ☐ Ignored

Figure 4. Input stages efficiency in Unisim

The process flow diagram for gas sweetening pilot plant is illustrated in Figure 5, while the simulation model is shown in Figure 6.

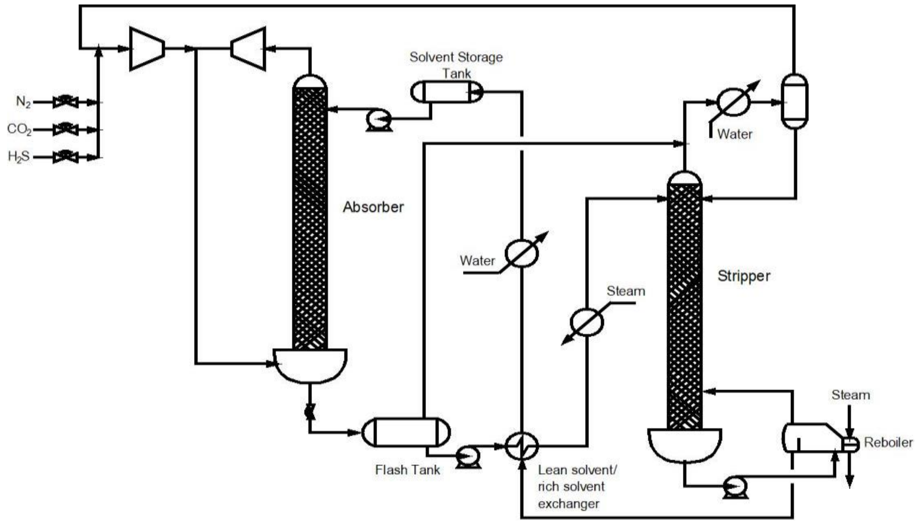
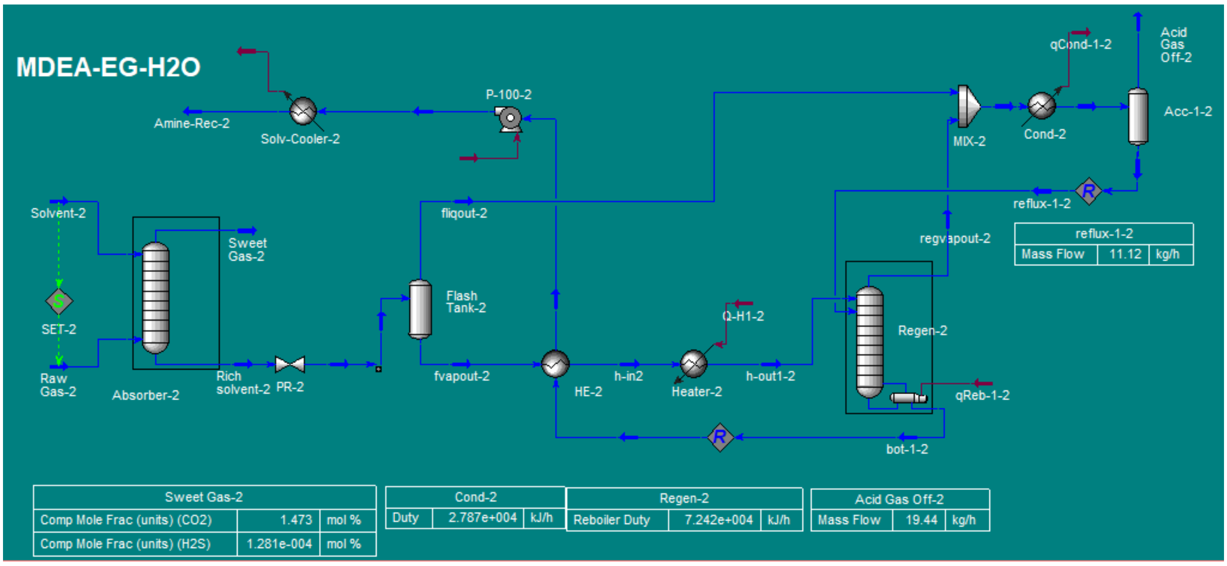


Figure 5. PFD of pilot scale gas sweetening (modified from Gonzales et.al, 2023)



reflux-1-2		
Mass Flow	11.12	kg/h

Sweet Gas-2		
Comp Mole Frac (units) (CO2)	1.473	mol %
Comp Mole Frac (units) (H2S)	1.281e-004	mol %

Cond-2		
Duty	2.787e+004	kJ/h

Regen-2		
Reboiler Duty	7.242e+004	kJ/h

Acid Gas Off-2		
Mass Flow	19.44	kg/h

Figure 6. Simulation model of gas sweetening pilot plant

The operating condition for pilot plant is listed in Table 1.

Table 1. Pilot plant operating condition

Variables	Min	Max
Operating Condition		
Solvent mass rate [kg/hr]	821.4	1502.1
Absorber temperature [K]	303	323
Absorber pressure [bar]	31	40.1
Parameter		
Feed gas impurities	CO_2 2.5%mol and H_2S 2.5%mol	
Solvent 1	MDEA 50%wt - H_2O 50%wt	
Solvent 2	MDEA 34%wt - EG 44.5%wt - H_2O 21.5%wt	

3. RESULTS AND DISCUSSION

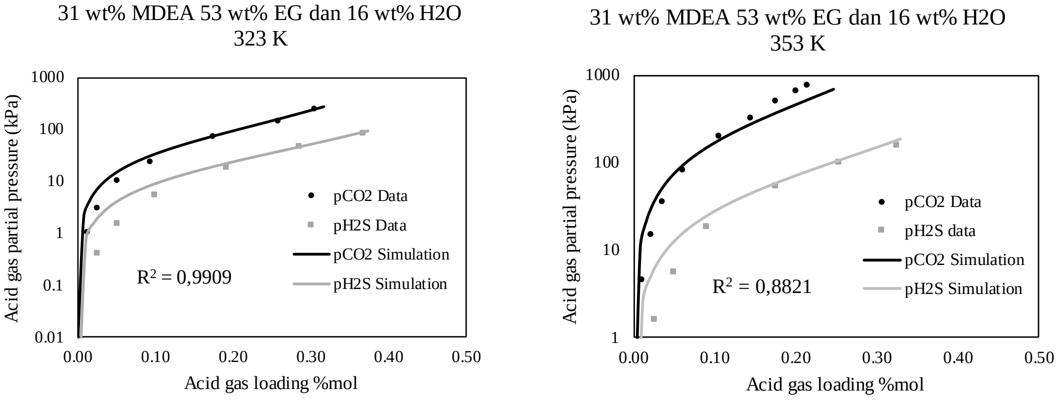
3.1 Vapor-Liquid Equilibrium Modeling (VLE)

The model validation for VLE is first using the parameter regressed by Gonzalez et.al., 2023. Due to the difference in fluid package, the calculated partial pressure of CO_2 and H_2S is still deviated from the data. Therefore, a parameter adjustment is executed by manually setting Henry's constant and binary NRTL parameters. The obtained adjusted parameters that best fit the experimental data are presented in Table 2. The parameters for CO_2-H_2O and H_2S-H_2O is using the default parameters in Unisim.

Table 1. Adjusted Henry's constant and NRTL binary coefficient

Parameters	Binary System					
	CO_2-MDEA	CO_2-EG	CO_2-H_2O	H_2S-MDEA	H_2S-EG	H_2S- H_2O
Henry's constant						
A	17.212	-74.326	Default	16.260	-73.455	Default
B	-4374.319	-2075.759		-4000.000	-5031.564	
C	0.000	15.040		0.000	19.480	
D	0.000	0.000		0.000	0.000	
NRTL binary coefficient						
A_{ij}	-1000.000	-1000.000	-2500.000	-3000.000	500.000	-3000.000

The comparison between our VLE model and the experimental data is shown in Figure 7. From the comparison, the average R^2 for 323 K is 0.9909 while for 353 K is 0.8821. The parameter adjustment we put here, on average showed fairly good result. However the deviation is still found especially for low partial pressure.



(a) (b)

Figure 7 Comparison between VLE data and model (a) 323 K (b) 353 K

3.2 Matching of Stages Efficiency

The mechanism of acid gas absorption in an amine contactor tower involves the transfer of acid gas from bulk gas into the interface where the equilibrium occurs between the vapor and liquid phases. Next, the acid gas dissolved in the amine solution diffuses and reacts with the amine molecule by chemical reaction [6] below:

$$MDEA + H_2O + CO_2 \rightleftharpoons MDEAH^+ + HCO_3^- \tag{8}$$

$$MDEA + H_2S \rightleftharpoons MDEAH^+ + HS^- \tag{9}$$

The MDEA-CO_2 reaction is slower, while the MDEA-H_2S is considered instantaneous. Therefore, the CO_2 absorption kinetic is controlled by the reaction, while the H_2S by the mass transfer[7].

The stage efficiency is utilized to accommodate the rate process that involves acid gas absorption as explained previously. In Unisim Design, the ideal stage is defined to have 100% stage efficiency where it is assumed that the stage achieves vapor-liquid equilibrium. This condition is plausible if the rate process such as reaction kinetics and mass transfer does not become the rate-determining step. However, for the real stage, where the reaction kinetic and/or mass transfer is limited, only some portion of the gas phase achieves an equilibrium, while the rest is bypassed to next stage. The schematic of the application of stages efficiency in Unisim Design is depicted in Figure 8[4].

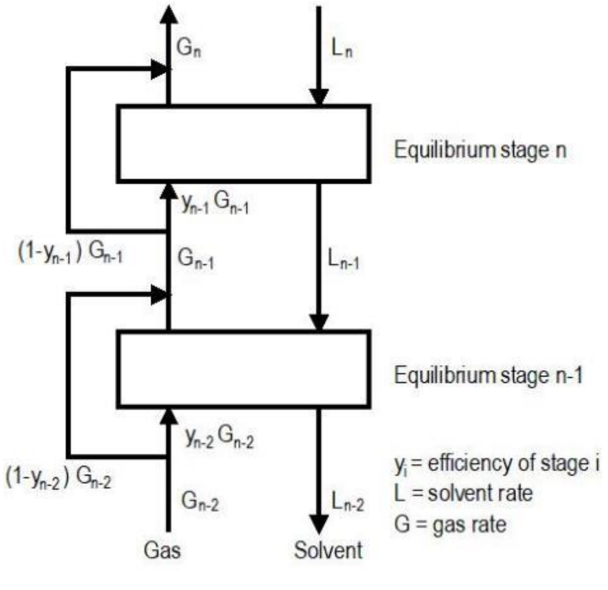


Figure 8. Schematic of stage efficiency application

By applying stage efficiency, we match the impurities concentration in sweet gas from the pilot plant experiment data. The matching result is listed in Table 2.

Table 2. Matching stages efficiency for pilot plant experiment data

Solvent: MDEA 50%wt -H_2O 50%wt

L/G	Average T [K]	Pressure Abs [bara]	CO_2 (%mol)	CO_2 (%mol)	H_2S (ppmv)	H_2S (ppmv)	Matching Stage Efficiency	
			Data	Sim	Data	Sim	CO_2	H_2S
3.67	319	31	1.000	1.001	7	7.002	0.0296	0.2275
2.45	319	32	1.000	1.002	6	6.001	0.0282	0.2347
2.46	318	40	0.900	0.900	5	4.999	0.0328	0.2348
2.45	323	40.1	0.600	0.600	6	6.001	0.0452	0.2320
2.46	313	40.1	1.000	1.000	3	3.000	0.0283	0.2476
1.34	319	40	0.900	0.900	7	7.000	0.0301	0.2253

Solvent: MDEA 34%wt - EG 44.5%wt - H_2O 21.5%wt

L/G	Average T [K]	Pressure Abs [bara]	CO_2 (%mol)	CO_2 (%mol)	H_2S (ppmv)	H_2S (ppmv)	Matching Stage Efficiency	
			Data	Sim	Data	Sim	CO_2	H_2S
2.46	318	31.1	1.100	1.101	6	6.000	0.0241	0.2312
2.46	324	39.4	0.600	0.600	8	8.000	0.0446	0.2249
2.45	329	39.2	0.600	0.601	13	13.007	0.0480	0.2163
2.01	318	39.4	0.800	0.800	4	4.001	0.0354	0.2412
3.67	323	39.4	0.500	0.500	12	12.001	0.0524	0.2141

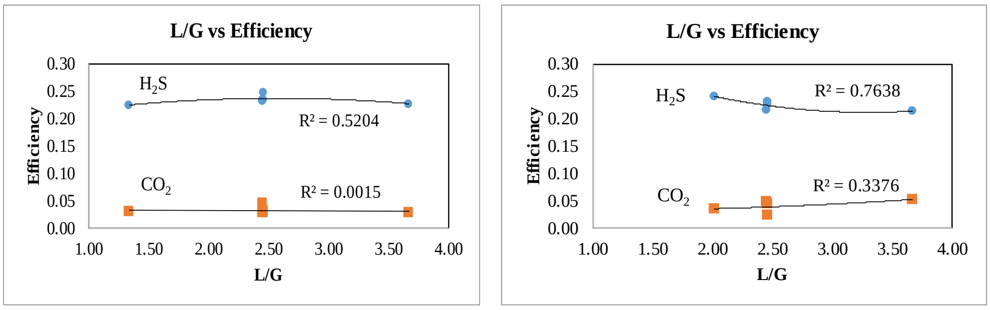
Note:
L/G : ratio of solvent to gas mass rate
Impurities concentration of sweet gas

The resulting stage efficiency is then analyzed by plotting it toward the absorber operating condition as illustrated in Figure 9 to Figure 11. For the three operating conditions (L/G, average T, and absorber pressure), we also apply quadratic regression to show the effect of each operating condition on stage efficiency. Temperature displays a larger effect on stage efficiency, followed by L/G and the last is absorber pressure. It is also noted that generally, the stage efficiency for H_2S is higher than CO_2 for all operating conditions and solvent compositions. If we take the average stage efficiency for CO_2 and H_2S, we find that for MDEA-H_2O solvent, the efficiency ratio of H_2S over CO_2 is 7.2, while for MDEA-EG-H_2O solvent, the efficiency ratio is 5.5. The ratio of mass transfer coefficient of H_2S over CO_2 that is obtained by Gonzales, et.al., 2023 is 7.1 for MDEA-H_2O solvent and 6.4 for MDEA-EG-H_2O solvent. It shows that our simulation result by using the stage efficiency approach is comparable to rate-based modeling by previous work.

We also find that the stage efficiency of CO_2 is proportional to average absorption temperature, while for H_2S, we find quite a different trend where it quite insensitive toward temperature. The explanation for this result is by assuming that the gas sweetening process is a physico-chemical process that involves mass transfer and reaction. CO_2 absorption is reaction-limited, therefore the absorption rate have a similar trend with the reaction rate where it is proportional toward temperature. While the H_2S absorption is determined by the mass transfer rate due to the instantaneous reaction rate. Therefore it is less sensitive to temperature. A similar explanation is also highlighted by several authors [3], [8], [9]. This result also shows the validity of the stage efficiency approach to model the rate processes in acid gas absorption towers compared to a more complex physicochemical model (rate-based model).

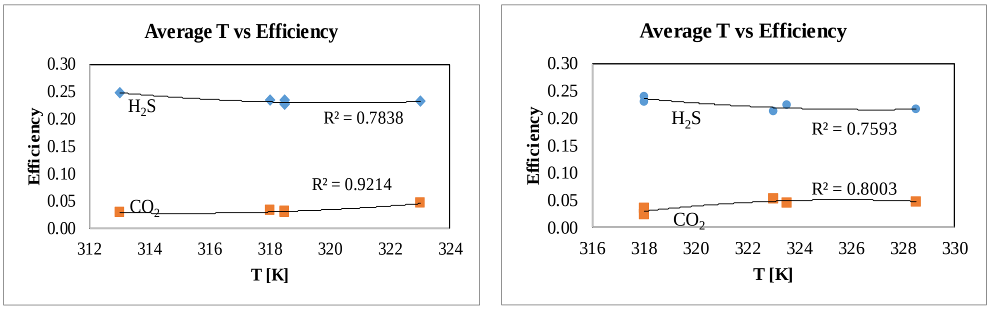
For the scale-up application, we develop a multiple quadratic regression model that connects stage efficiency toward operating conditions, in the form of (10). The coefficient for each gas and solvent is presented in Table 3, while the comparison of stage efficiency between the data and model is displayed in Figure 12.

$$y = a_0 + a_1\left(\frac{L}{G}\right) + a_2T + a_2P + a_4\left(\frac{L}{G}\right)T + a_5\left(\frac{L}{G}\right)P + a_6TP + a_7\left(\frac{L}{G}\right)^2 + a_8T^2 + a_9P^2 \tag{10}$$



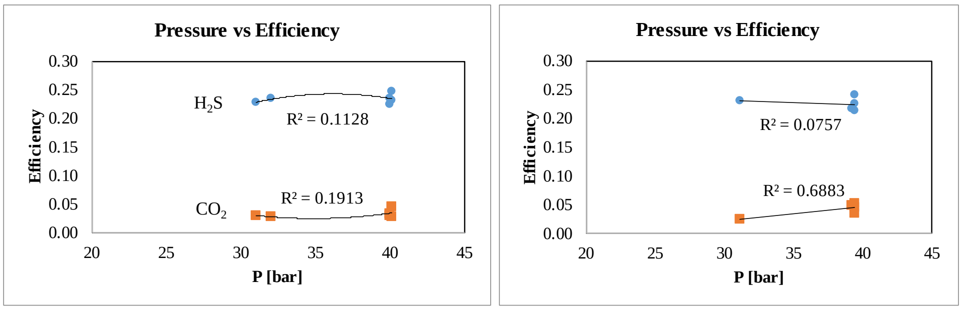
(a) (b)

Figure 9. Stage efficiency vs L/G for (a) MDEA-H_2O Solvent and (b) MDEA-EG-H_2O Solvent



(a) (b)

Figure 10. Stage efficiency vs average absorber temperature for (a) MDEA-H_2O Solvent and (b) MDEA-EG-H_2O Solvent



(a) (b)

Figure 11. Stage efficiency vs absorber pressure for (a) MDEA-H2O Solvent and (b) MDEA-EG-H2O Solvent

Table 3. Stage efficiency regression coefficient for each gas and solvent.

Coefficient	MDEA-H_2O CO_2	MDEA-H_2O H_2S	MDEA-EG-H_2O CO_2	MDEA-EG-H_2O H_2S
a_0	0.000E+00	0.000E+00	0.000E+00	0.000E+00
a_1	0.000E+00	0.000E+00	0.000E+00	0.000E+00
a_2	-1.645E-03	2.973E-03	-1.481E-04	2.047E-03
a_3	0.000E+00	0.000E+00	0.000E+00	0.000E+00
a_4	-3.042E-05	-1.893E-04	-1.649E-03	2.601E-03
a_5	3.767E-04	1.779E-03	1.369E-02	-2.157E-02
a_6	-2.375E-11	-3.293E-11	-3.335E-19	5.527E-19
a_7	2.164E-11	3.001E-11	2.581E-19	-4.279E-19
a_8	5.375E-06	-6.378E-06	7.390E-06	-1.518E-05
a_9	9.817E-07	-5.574E-05	-4.478E-04	7.541E-04

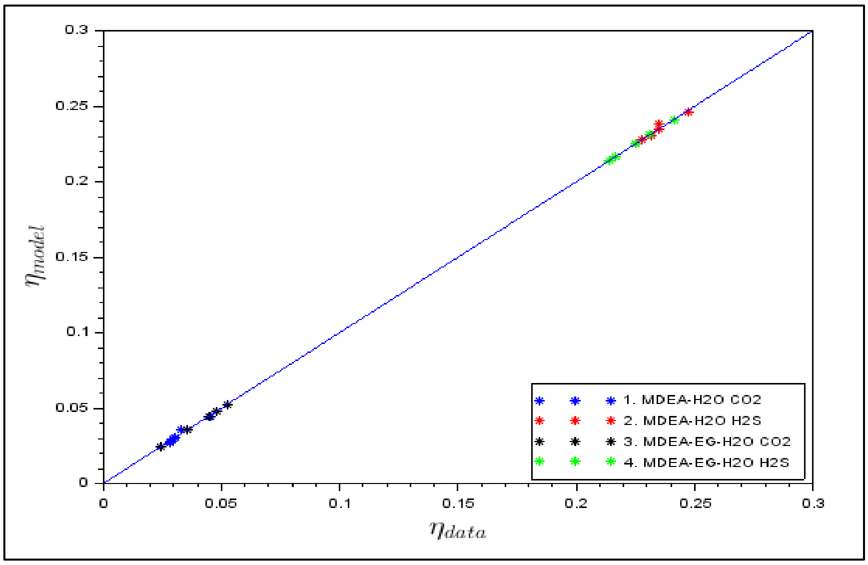


Figure 12. Stage Efficiency data vs model

3.3 Simulation of Gas Sweetening Pilot Plant

The simulation is performed using a Unisim Design as shown in Figure 6 and then the result is compared to those by Gonzales, et.al, 2023. The comparison is presented in Table 4. From the table, several parameters have approached the literature. However, the H_2S in both lean solvent and sweet gas still shows a discrepancy. This problem occurs due to inadequate H_2S VLE model in current work, especially for low H_2S partial pressure. For current work, the VLE model and matching parameter are quite good for describing the process in the absorber. However, the process in the amine regenerator whose temperature is higher might not be properly described. Therefore, for this work, we apply different VLE parameters for the regenerator, where we use the default NRTL fluid package provided in the Unisim Design. Somehow, the performance indicator (such as reboiler temperature, reboiler duty, duty/mass of CO_2 captured, and reflux flow rate) for the regenerator unit still shows a good match toward the reference result.

Table 4. Simulation result comparison with that by Gonzalez, et.al., 2023.

Operating Variables	Units	Gonzalez et.al	This work	Gonzalez et.al	This work
		MDEA-H_2O		MDEA-EG-H_2O	
Set parameter					
Lean solvent flow rate	kg/h	340	340	602	602
Lean solvent T	K	313	313	313	313
P absorber	bar	40	40	40	40
P regenerator	bar	1.9	1.9	1.9	1.9
T raw gas	K	323	323	323	323
H_2S in lean solvent	wt%	0.044	0.044	0.011	0.011
CO_2 in lean solvent	wt%	0.0001	0.0001	0.0004	0.0004
MDEA in lean solvent	wt%	50	50	34	34
EG in lean solvent	wt%	0	0	45	45
Simulation result					
CO_2 in sweet gas	mol%	1.1	1.1	1.1	1.4
H_2S in sweet gas	ppmv	12	18	12	4
Reboiler temperature	K	397	396.2	413	403.7
Reboiler duty	kW	30	34.66	23	20.11
Duty/mass of CO_2 captured	kJ/kg	12,500	13,761	9,400	10,632
Reflux flow rate	kg/h	42	41.92	11	11.12

4. CONCLUSION

The simulation of the gas sweetening process using the extended-NRTL VLE model and stage efficiency approach has been conducted. On average, our regressed parameter shows good fit toward experiment data. However, the H_2S equilibrium for low partial pressure is not properly described. The stage efficiency approach could be used to validate the absorber simulation toward pilot plant data. The obtained trend is also comparable with the rate-based model and in accordance with the physicochemical phenomena of the CO_2 and H_2S absorption process. The simulation using the above methods can properly calculate several parameters, however, it poorly described the H_2S both in sweet gas and lean solvent, as well as reboiler duty. Further work to get better regression parameters in the current range of operating conditions and especially in higher temperatures is necessary to overcome those problems.

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