

## Simulation of CO<sub>2</sub> removal with aqueous sodium glycinate solutions in a pilot plant

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**Abstract**—In this paper, density, viscosity, surface tension and vapor pressure of aqueous sodium glycinate solutions of different mass fractions (0.1-0.5) at different temperatures (20-100 °C) were simulated using Pro/II (version 6.01), a commercial process simulator, and compared with corresponding experimentally measured data. It was found that simulated data of physicochemical properties compared well with corresponding experimental data. We have also predicted concentration of CO<sub>2</sub> with each ideal stage in an absorber/stripper tower.

Key words: CO<sub>2</sub> Separation, Sodium Glycinate, Pro/II Software Process, Process Design, Chemical Absorption

### INTRODUCTION

Nowadays, natural disasters such as hurricanes, earthquakes, floods and tidal waves occur frequently in the world. Humankind faces a multitude of challenges as the 21<sup>st</sup> century begins. Compelling scientific evidence implicates anthropogenic CO<sub>2</sub> emissions of greenhouse gases in changing the global climate [1]. Global warming has become a serious challenge that has the potential to affect every part of the globe in the 21<sup>st</sup> century, and cannot be either delayed or avoided. The Kyoto protocol came into effect on February 16, 2005 to prevent climate change. It states that the developed countries must reduce overall greenhouse gas emissions by an average of 5 percent below 1990 levels in its first commitment period from 2008 to 2012 [2].

South Korea is not the country that has to reduce greenhouse gases. However, South Korea is emitting a lot of greenhouse gases and its emission rate is increasing sharply [3]. According to UNFCCC (United Nations Framework Convention on Climate Change) greenhouse gas emission inventory, among South Korean total greenhouse gas emissions, 28% is from the power sector. KEEI (Korea Energy Economics Institute) predicts that the demand for power will increase continuously. Therefore, in South Korea, more action needs to be taken in the next 10 years to prepare beyond Kyoto and to help prevent climate change [4,5]. Most of the greenhouse gases in the power sector are emitted by thermal power plants. The most likely options currently identifiable for CO<sub>2</sub> separation and capture include adsorption, absorption, low temperature distillation, gas separation membranes, mineralization and biomineralization. Researchers over the world are working for the reduction of CO<sub>2</sub> through finding new methods/materials and also for applications of CO<sub>2</sub> in industrial processes [6-14].

Mitigation technology such as chemical absorption is the widely used solution to reduce greenhouse gas emissions from thermal power

plants in a short period [15-20]. In search of a new absorbent, we are studying sodium glycinate (SG) solution as an absorbent for CO<sub>2</sub>. In previous papers, we have studied physico-chemical properties of SG solution, diffusivity, physical solubility and loading capacity of CO<sub>2</sub> and kinetics of CO<sub>2</sub> absorption in SG solution [21-24].

The objective of this work was to simulate the physico-chemical property such as density, viscosity, surface tension and vapor pressure of SG solution from the measured property. We have also predicted the foundation process design in aqueous sodium glycinate solutions using Pro/II (version 6.01), a commercial process simulator. These are necessary for the operation, and optimization of acid gas treatment equipment as well as the economic assessment that would be used in chemical absorption process for the designing of 2 ton CO<sub>2</sub>/day pilot plant in the future.

### EXPERIMENTAL PROCEEDURE

The SG used in this study was obtained from Sigma-Aldrich Chemical Co. with a mass purity of >99%. The aqueous solutions were prepared from doubly distilled water. All solutions were prepared by mass with a balance precision of  $\pm 1 \times 10^{-4}$  g. For the measurements of the vapor pressure and isobaric vapor-liquid equilibrium, a Dr. Sieg & Röck type recirculating glass still was used. For both measurements, approximately 250 mL of aqueous SG solution was introduced into the still and the pressure of the still was controlled by using a Baratron pressure regulating system with an accuracy of  $\pm 0.1$  kPa. The pressure inside the equilibrium cell was also measured with a Wallace & Tiernan precision mercury manometer during the whole measurement. When desired the equilibrium pressure was reached, the liquid phase was heated to boil vigorously, and liquid and vapor phases were circulated in the still for about 2 hours. Then the equilibrium temperature was measured with a Pt-100 temperature probe with accuracy of  $\pm 0.01$  K.

The density, viscosity and surface tension of the SG solution were measured with a Gay-Lussac pycnometer, RheoStress1 viscometer, and Sigma 70 automated tensiometer in the manner described in

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**Table 1. Physical properties of components of exhaust gas and sodium glycinate solution**

Physical property	H <sub>2</sub> O	CO <sub>2</sub>	O <sub>2</sub>	N <sub>2</sub>	Sodium glycinate
Molecular weight (g)	18.015	44.010	31.999	28.013	97.050
Density (kg/m <sup>3</sup> )	938.566	826.185	1126.378	807.313	1105.309
Normal boiling point (°C)	100.00	-78.48	-182.98	-195.80	268.39
Critical temperature (K)	374.20	31.04	-118.40	-146.90	463.45
Critical pressure (kPa)	221.192	73.815	50.764	33.944	42.700
Critical volume (m <sup>3</sup> /kg mol)	0.0554	0.0940	0.0764	0.0901	0.3490
Acentric factor	0.34800	0.23100	0.01900	0.04500	0.95288
Heat of formation (kcal/kg mol)	-57799.99	-93984.43	0.00	0.00	-89039.36
Free energy of formation (kcal/kg mol)	-54646.99	-94203.21	0.00	0.00	-44986.62

**Table 2. Exhaust gas composition (mol%, dry basis) and its condition from power plant**

N <sub>2</sub>	87
O <sub>2</sub>	2.8
CO <sub>2</sub>	10.2
Temperature (°C)	40
Feed gas pressure (kPa)	200 kPa
Flow rate (Sm <sup>3</sup> /h)	750 Sm <sup>3</sup> /h

our previous paper [21].

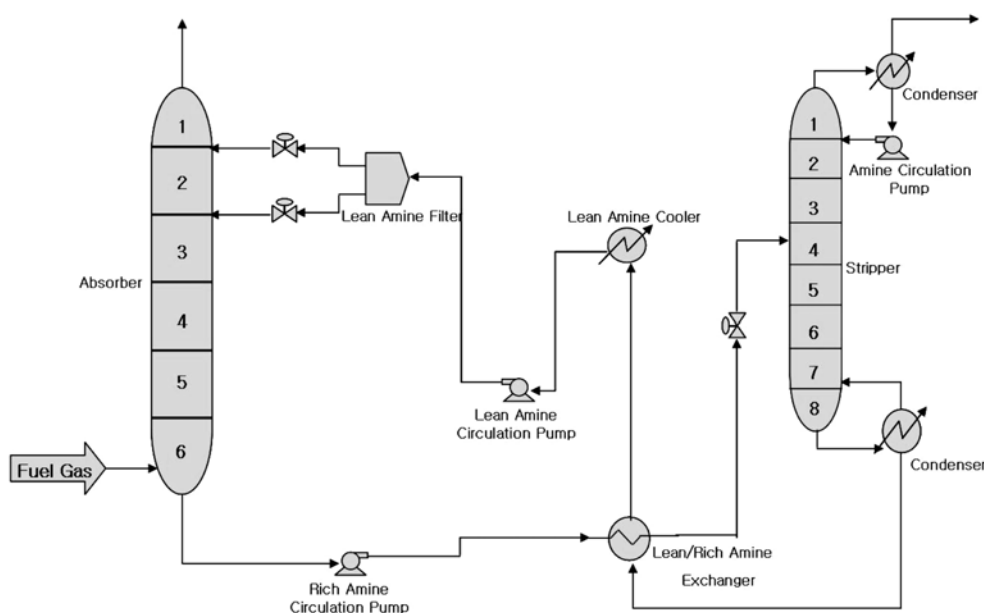
### THE PROCESS DESIGN DESCRIPTION

A Pro/II software process simulation was made with the selection of an appropriate vapor-liquid equilibrium model [25]. The program has the flexibility to model refinery processes in detail. Its modeling capabilities address a wide range of applications from crude oil characterization and preheating to complex reaction and separation units [26,27]. The process simulation of CO<sub>2</sub> separation using Pro/II requires the values of physical properties of components of

exhaust gases and sodium glycinate solution which are reported in Table 1.

Table 2 illustrates the composition of exhaust gases as well as its temperature, feed gas pressure and flow rate. The exhaust gases (40 °C, 200 kPa, 7,500 Sm<sup>3</sup>/hr), which were supplied by the plant, were reacted counter-currently with a lean solvent in an absorber tower that usually operates at a low temperature. Treated gas leaves overhead from the absorber. The rich solvent is heated against the returning hot regenerated lean solvent in a heat exchanger. The stripper is a low pressure countercurrent tower in which the remaining dissolved acid gases are stripped off by heating with steam generated by reboiler. Fig. 1 shows the simulation for a pilot plant consisting of an absorber tower, a stripper tower, a reboiler, a reclaimer, a pump, a condenser, and a lean/rich amine exchanger. The design for process modeling is given below:

The CO<sub>2</sub> recovery ratio of one of exhaust gases is above 90% and the purity of recovered CO<sub>2</sub> is above 99.5% (dry basis). The characteristic absorber/stripper tower and operating conditions are shown in Table 3. The number of ideal stages required for an absorption/stripper tower can be found by a design program such as Pro/II software process simulation, ASPEN PLUS and HYSIS used

**Fig. 1. Schematic diagram of simulation modeling for aqueous sodium glycinate solutions.**

involving material and enthalpy. The numbers of ideal stages in the absorption/stripper tower are 6 plates and 8 plates, respectively. Operating pressure in absorption and stripper tower is approximately 200 to 1,000 kPa and 175 kPa, respectively. The pressure drop per unit packing depth comes from fluid friction [28]. We assumed that pressure drop is 20 kPa in the absorption/stripper tower.

The process modeling of physicochemical properties of the aqueous SG solution such as density, viscosity, surface tension, and vapor pressure was operated over a wide range of concentration (10 to 50 wt%) of SG and temperature (30 to 80) °C.

The following equations were used for the regression of measured data:

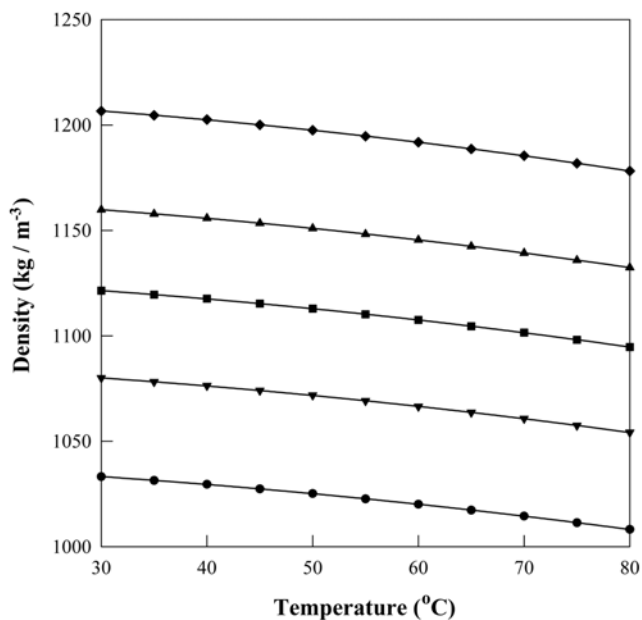
$$Y = A_1 + A_2(T/^{\circ}\text{C}) + A_3(T/^{\circ}\text{C})^2 \quad (1)$$

$$\eta/\text{mPa} = \exp\left[A_1 + \frac{A_2}{(T/^{\circ}\text{C}) + A_3}\right] \quad (2)$$

$$\ln P^{\text{sat}} = A_1 - \frac{A_2}{(T/^{\circ}\text{C}) + A_3} \quad (3)$$

**Table 3. Characteristics of absorption and stripper tower and operating conditions for process modeling**

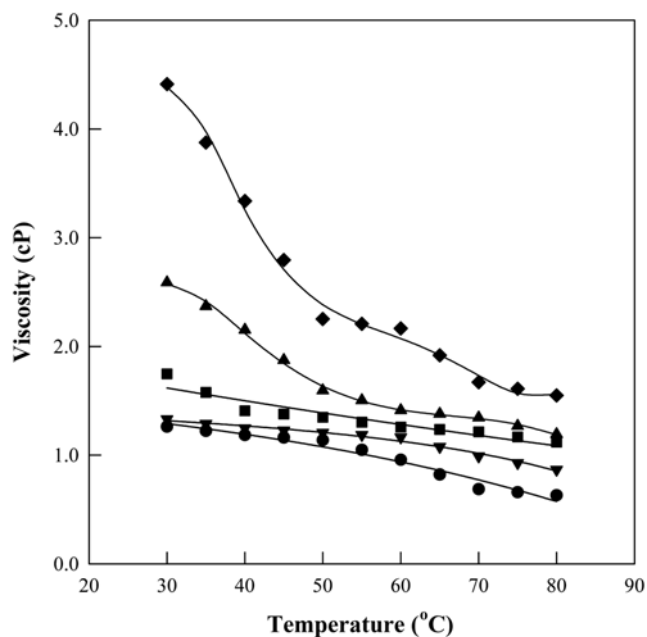
	Absorption tower	Stripper tower
Number of ideal stage	6	8
Stages of rich SG solution	-	3
Operating pressure (kPa)	200-1,000	175
Pressure drop (kPa)	20	20
Foaming factor	0.8	0.85
Temperature (°C)	40	120



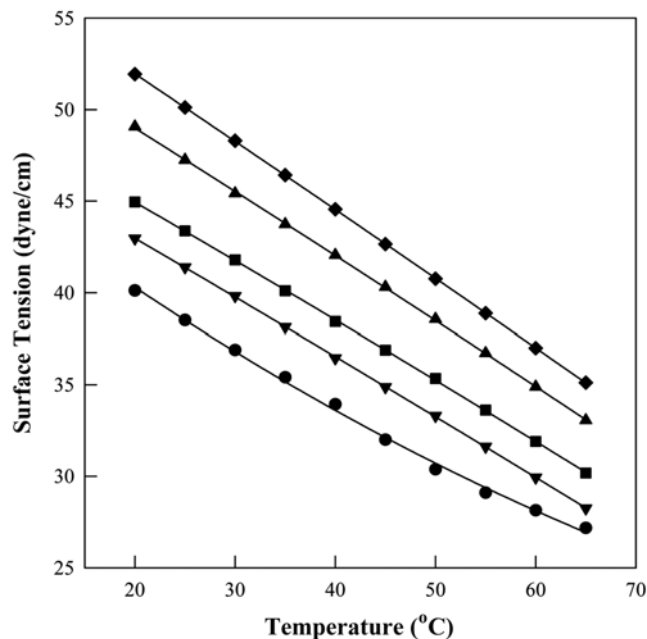
**Fig. 2.** Comparison of experimental data with predicted data of the density as a function of temperature for aqueous sodium glycinate solution of different concentrations (wt%); ●, 10; ▼, 20; ■, 30; ▲, 40; ◆, 50; line represents simulated values.

$$\text{AAD} = \frac{1}{N} \sum_{i=1}^N |A_{i,\text{exp}} - X_{i,\text{calcd}}| \quad (4)$$

where  $Y$  ( $=\rho$  or  $\sigma$ ) is density or surface tension,  $\eta$  is viscosity,  $P^{\text{sat}}$  is vapor pressure,  $T$  is temperature,  $N$  is the number of data points,



**Fig. 3.** Comparison of experimental data with predicted data of the viscosity as a function of temperature for aqueous sodium glycinate solution of different concentrations (wt%); ●, 10; ▼, 20; ■, 30; ▲, 40; ◆, 50; line represents simulated values.



**Fig. 4.** Comparison of experimental data with predicted data of the surface tension as a function of temperature for aqueous sodium glycinate solution of different concentrations (wt%); ●, 10; ▼, 20; ■, 30; ▲, 40; ◆, 50; line represents simulated values.

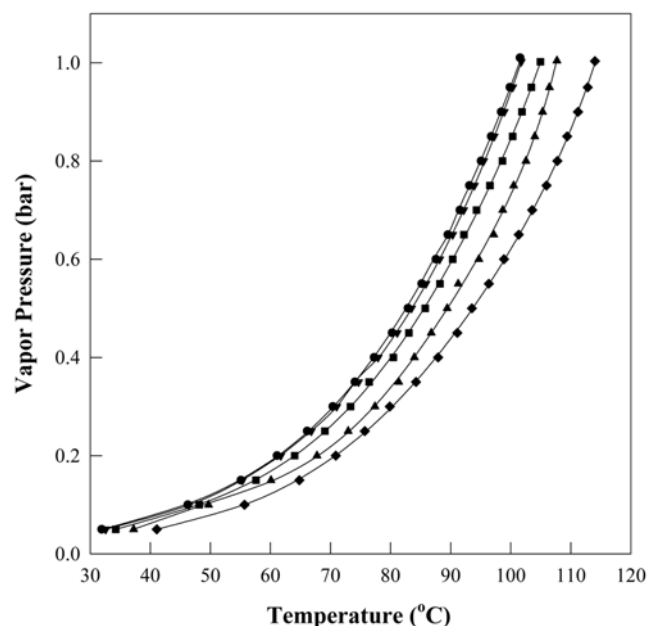


Fig. 5. Comparison of experimental data with simulated data of the vapor pressure as a function of temperature for aqueous sodium glycinate solution of different concentrations (wt%); ●, 10; ▼, 20; ■, 30; ▲, 40; ◆, 50; line represents simulated values.

and  $A_i$  ( $i=1$  to 3) are the regression parameters. The average of the average absolute deviation (AAD) between experimental and simulated values for density, viscosity, surface tension, and vapor pressure was 0.049, 0.558, 0.061, and 0.003%, respectively. Figs. 2-5 show that simulated values compared well with the experimental values of density, viscosity, surface tension and vapor pressure of the SG solution. The average absolute deviation (AAD) between experimental and simulated values for density, viscosity, surface tension, and vapor pressure was calculated from Eq. (1)-(4) and reported along with the regression parameters in Table 4.

## RESULTS AND DISCUSSION

Table 5 shows the results of process modeling in aqueous SG solutions. All the cases are satisfied with the operating condition for foundation design such as  $\text{CO}_2$  recovery and  $\text{CO}_2$  purity. The SG solution concentration increases with a decrease in amine circulation rate and reboiler duty. Similar trends were also reported in

Table 4. Regression parameters and AADs for density, viscosity, surface tension and vapor pressure correlations of aqueous sodium glycinate solution (wt%)

$X_{SG}$	$A_1$	$A_2$	$A_3$	AAD%
Density				
10	1040.553	-0.1476	-0.0032	0.046
20	1087.455	-0.146	-0.0034	0.052
30	1128.962	-0.1448	-0.0035	0.045
40	1167.757	-0.159	-0.0035	0.042
50	1214.829	-0.1611	-0.0037	0.051
Surface tension				
10	48.204	-0.4254	0.0015	1.613
20	49.282	-0.3119	-1.74E-04	0.271
30	51.151	-0.3053	-2.58E-04	0.333
40	55.7685	-0.3348	-2.17E-04	0.426
50	59.2476	-0.3614	-1.55E-04	0.149
Viscosity				
10	0.869	54.0696	-117.951	0.036
20	0.5304	20.294	-109.464	0.015
30	6.3002	4540.124	-810.184	0.046
40	7.8634	3163.709	-485.035	0.099
50	21.4349	17811.24	-920.715	0.151
Vapor pressure				
10	17.534	4691.22	261.5778	0.003
20	20.7312	7109.329	339.1636	0.002
30	14.6983	3023.084	194.6092	0.003
40	25.0877	11436.85	450.3008	0.004
50	14.6726	3199.887	203.4762	0.003

the literature for reboiler heat duty [29-31].

The simulated density, and viscosity data of aqueous SG solution at (30 to 80) °C for various concentration (10 to 50 wt%) are shown in Figs. 2 and 3. It was found that while density increases linearly with increase in concentration of SG and decreases linearly with decrease in temperature, viscosity decreases with increase in temperature and the decrease is more sharp in case of higher concentration. The surface tension data of aqueous SG solution at (20 to 65) °C for various concentration (10 to 50) wt% are shown in Fig. 4. The surface tension was found to increase linearly with increase in concentration of SG and decrease with decrease in temperature. Next, we compared the vapor pressure of aqueous sodium glycinate at (30 to 120) °C for various concentrations (10 to 50) wt%

Table 5. The results of process modeling in aqueous sodium glycinate (SG) solutions

Design parameter	Case 1	Case 2	Case 3	Case 4	Case 5
SG concentration (wt%)	10	20	30	40	50
Feed gas pressure (kPa)	100	100	100	100	100
$\text{CO}_2$ Recovery (wt%)	99.981	99.911	93.892	90.822	90.792
$\text{CO}_2$ Purity (mol %)	99.76	99.779	99.783	99.771	99.765
Amine circulation rate ( $\text{m}^3/\text{h}$ )	4.189	2.555	1.599	1.235	1.043
Rich amine loading ( $\text{CO}_2$ mole/amine mole)	0.537	0.429	0.413	0.392	0.367
Lean amine loading ( $\text{CO}_2$ mole/amine mole)	0.03	0.03	0.03	0.05	0.05
Reboiler duty (kJ/h)	$6.49 \times 10^5$	$5.28 \times 10^5$	$3.98 \times 10^5$	$2.93 \times 10^5$	$2.76 \times 10^5$

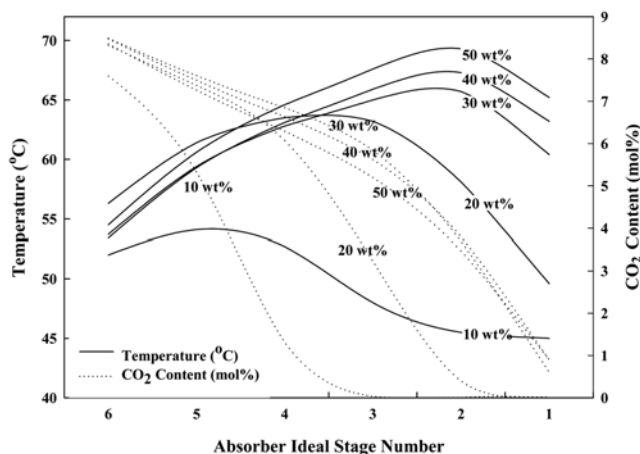


Fig. 6. Effect of Pro/II modeling with temperature and CO<sub>2</sub> concentration in absorber.

in Fig. 5. It was observed that vapor pressure increased with increase in temperature and decreased in concentrations. All of the experimental data were regressed by using customized simulation of the modeling process. It was found that data predicted from the simulation of modeling process agrees well with the experimental data.

Various changes in the temperature and concentration of CO<sub>2</sub> with each ideal stage in absorber tower were shown in Fig. 6. It is clear from Fig. 6 that concentration of absorbed CO<sub>2</sub> decreases as we move up the tower (stage 6 to 1 in Fig. 1). As for as absorber temperature is concerned, as we go up the tower it increases up to stage 2 (for 30, 40, 50 wt% SG solution), then it starts decreasing. As the temperature increases, the solubility decreases. CO<sub>2</sub> absorption is exothermic in nature and an increase in temperature should decrease the extent of chemical absorption in accordance with Le Chatelier's principle. This is indeed true in this case where the absorption of carbon dioxide in aqueous SG solution decreases with increasing temperature. It was observed that the concentration of CO<sub>2</sub> increases with increase reaction temperature at the top of the absorption tower.

Fig. 7 shows the various changes of the temperature and concentration of CO<sub>2</sub> with each ideal stage in the stripper tower. The de-

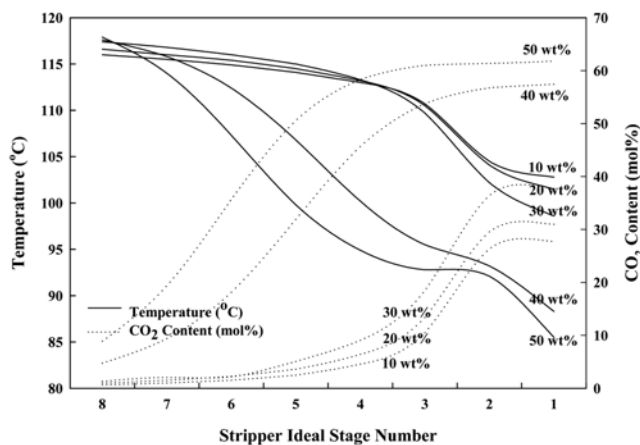


Fig. 7. Effect of Pro/II modeling with temperature and CO<sub>2</sub> concentration in stripper.

sorption (stripper reaction) of CO<sub>2</sub> from SG is an endothermic reaction in the stripper tower. Therefore, it should be opposite to what we have simulated in Fig. 6. Thus, the concentration of CO<sub>2</sub> stripped off the SG solution increases with increase in absorbent temperature which is found to be increasing as we move down the stripper. The maximum regenerated CO<sub>2</sub> is approximately 62% at 1 stage when the concentration of SG is 50 wt%. The reaction temperature decreases with increase in the concentration of CO<sub>2</sub> with flow rate of CO<sub>2</sub> of bottom stage (8 stage) to top stage (1 stage). Similar trends for concentration of absorbents at different stages were reported in literatures [32,33].

## CONCLUSIONS

We studied the simulation of physicochemical properties such as density, viscosity, surface tension, and vapor pressure of aqueous SG solution of various concentrations at different temperatures. The simulated data of physicochemical properties using Pro/II software process simulation were found to compare well with our experimental data. We have simulated the process design in aqueous SG solutions with the temperature of each ideal stage and distributed concentration of CO<sub>2</sub> in the absorber/stripper tower. The process was optimized for different concentrations of SG, amine circulation rate, rich and amine loading, reboiler duty, and CO<sub>2</sub> recovery. These results would be used in designing a 2 ton/day pilot plant.

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