Physical Solubility of N₂O and CO₂ in Aqueous Solutions of Sodium Glycinate at High Pressure

by

Wee Jin Chua

Dissertation submitted in partial fulfillment of the requirements for the Bachelor of Engineering (Hons) (Chemical Engineering)

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CERTIFICATION OF APPROVAL

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A project dissertation submitted to the Chemical Engineering Programme Universiti Teknologi PETRONAS in partial fulfillment of the requirement for the BACHELOR OF ENGINEERING (Hons) (CHEMICAL ENGINEERING)

Approved by,

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May 2012

CERTIFICATION OF ORIGINALITY

This is to certify that I am responsible for the work submitted in this project, that the original work is my own except as specified in the references and acknowledgements, and that the original work contained herein have not been undertaken or done by unspecified sources or persons.

WEE JIN CHUA

ABSTRACT

As a new absorbent for carbon dioxide (CO₂), sodium glycinate was investigated in this work. The physical solubility of nitrous oxide (N₂O) and CO₂ in pure water and of N₂O in aqueous sodium glycinate solutions of various concentrations (1.0, 2.0 and 3.0 kmol/m³) had been measured experimentally at different pressures (20, 40 and 60 bar) over the temperature ranging from 303.15 to 323.15 K. Since CO₂ reacts in the amine group of sodium glycinate, the physical solubility of CO₂ cannot be measured directly and thus N₂O (a non-reacting gas) was used as a surrogate for CO₂. In order to estimate the physical solubility of CO₂ in aqueous solutions of sodium glycinate, N₂O analogy had been applied. The physical solubility increases with a decrease in temperature and with a decrease in concentration, as well as with an increase in pressure. In this work, the optimum condition for physical solubility of CO₂ in aqueous solutions of sodium glycinate of sodium glycinate at high pressure was achieved at low temperature of 30 °C and high pressure of 60 bar.

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CHAPTER 1 INTRODUCTION

1.1 Background of Study

Carbon dioxide (CO₂) is a very important greenhouse gas that is released into the atmosphere during the combustion of fossil fuels such as natural gas, oil, and coal. This increase in atmospheric concentration of anthropogenic CO₂, is responsible for global warming (Kerr, 2006; IPCC, 2001; Houghton, 2004). In CO₂ capture, CO₂ is collected from large point sources such as power plants, oil refineries, petrochemical facilities, fertilizer and gas-processing plants, steel works as well as pulp and paper mills that can be reused.

There are several options for CO_2 separation and capture such as chemical absorption, physical absorption, physico-chemical adsorption, physical adsorption, cryogenic fractionation, and gas-separation membranes. The most likely option for separating CO_2 from flue gases or other gas streams is scrubbing the gas stream using an amine solution. Once the amine leaves the scrubber, it is heated to release high-purity CO_2 and the CO_2 -free amine that is then reused. This technique can also be applied to coal fired power stations, though additional cleaning of the flue gases is required. This process based on the principles of chemical absorption of CO_2 via alkanolamines is considered to be a potential technique for capturing CO_2 emission from flue gas streams. Aqueous alkanolamines such as monoethanolamine (MEA), methyldiethanolamine (MDEA), and 2-amino-2- methyl-1-propanol (AMP) have been widely used as chemical absorbents for removal of acid gases (CO_2 , H_2S) (Sidi-Boumedine *et al.*, 2004; Cullinane *et al.*, 2005; Kamps *et al.*, 2003; Park *et al.*, 2006; Teng *et al.*, 1991).

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1.2 Problem Statement

The removal of acid gas impurities (CO₂, H₂S) from natural, refinery, and synthesis gas streams is a significant operation in gas treatment. Particularly, aqueous MEA solutions have been widely used as an industrially important absorbent because of its rapid reaction rate, the low cost of the solvent, the thermal stability, and low solubility of hydrocarbons as well as high alkalinity. However, the aqueous MEA solutions have disadvantages such as degradation through oxidation of the amine, high enthalpy of reaction, and also it causes operational problems such as foaming, corrosion and fouling of the process equipment (Ma'um et al., 2005; Vaidya et al., 2005). Tertiary alkanolamines such as an aqueous MDEA solutions are better than primary and secondary alkanolamines such as aqueous MEA and diethanolamine (DEA) solutions as they have the properties of high loading capacity, less regeneration energy, and high resistance to thermal degradation, but reaction rates of tertiary alkanolamines are low (Ma'um et al., 2005; Rinker et al., 1995; Li et al., 1992). Sterically hindered amines such as AMP and 2-piperidineethanol (PE) are superior to the others because of CO₂ loading capacity, absorption rate, selectivity, and regeneration energy, but sterically hindered amines have an estimated lower reaction rate than the aqueous MEA solutions (Xu et al., 1996; Yoon et al., 2002; Beak et al., 1998).

To overcome such problems, various absorbents have been widely investigated. The goal of this study is to identify and evaluate an absorbent called as sodium glycinate that could be utilized for the separation of CO_2 from gaseous streams.

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1.3 Objective and Scope of Study

Amino acid or salts of amino acids are reported to be used for the selective removal of acid gases in some industrial processes (Kohl *et al.*, 1997). The amino acid salts have certain advantages over alkanolamines, such as higher surface tension, an almost nonvolatile nature, resistance to degradation in oxygen-rich flue gas, and better absorption (Hook, 1997; Song *et al.*, 2006).

In the past, sodium glycinate in glycerol was used in an immobilized liquid membrane in a closed loop life support systems, such as in spacecraft or space suits for removal of CO₂ from the atmosphere (Chen *et al.*, 2001; Murdoch *et al.*, n.d.; Chen *et al.*, 2000). This prompted us to study the aqueous sodium glycinate solution as an absorbent for CO₂. Thus, the objective of this research is to measure the physical solubility of CO₂ in aqueous sodium glycinate solutions of various concentrations (1.0, 2.0 and 3.0 kmol/m³) at different pressures (20, 40 and 60 bar) over the temperature ranging from 303.15 to 323.15 K, which is important parameter for the design, operation, and optimization of acid gas treatment equipment and also for measuring other chemical properties such as kinetic reaction, regeneration energy, and enthalpy of reaction. Apart from that, the data will be useful for experimentalists or process design engineers who are interested in the problem of acid gas removal using aqueous solutions of sodium glycinate.

CHAPTER 2 LITERATRE REVIEW

2.1 "N₂O Analogy" Method

For the analysis of the experimental absorption rate and for the rational design of gas absorption units, one of the most important parameters required are the solubility of carbon dioxide (CO₂) in the aqueous amine solutions. But the physical solubility of CO₂ in aqueous amine solutions cannot be measured directly since CO₂ reacts in the amine solutions. As a result, a non-reacting gas such as nitrous oxide (N₂O) is used as a surrogate for CO₂. It is common practice to use the N₂O analogy method originally proposed by Clarke (1964) to estimate the solubility of CO₂. Since N₂O is similar to CO₂ with respect to molecular configuration, electronic structure, molecular weight, and molar volume, Clarke (1964) assumed that the ratios of the solubility of CO₂ and N₂O in water and in aqueous solutions of organic solvents are similar within 5% or better at the same temperature.

This concept was later used by Sada *et al.* (1977; 1978), Versteeg *et al.* (1988), Al-Ghawas *et al.* (1989), and Haimour (1990) to estimate this parameter for CO₂ in various amine solutions. Laddha *et al.* (1981) verified the appropriateness of N₂O analogy method by investigating the solubility of CO₂ and N₂O in various aqueous solutions of organic solvents. From their work, it was found that the solubility of CO₂ and N₂O in the different solutions examined have a constant ratio of 1.37. For a change in temperature between 288 and 303 K, the ratio of solubility in water was found by them to be within 2% of 1.37. Thus, it has been proved that the N₂O analogy may be applied to estimate the solubility of CO₂ in aqueous amine solutions according to the following equation:

Solubility of
$$CO_2$$
 in amine solution = $\frac{1}{A_1}$ (Solubility of N_2O in amine solution)

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 $A_{1} = \frac{Solubility of N_{2}O \text{ in water}}{Solubility of CO_{2} \text{ in water}}$

The value of A_1 is, in general, 1.37. But, for better accuracy of the estimated solubility data of CO₂, it is desirable to find the value of A_1 at the particular temperature at which the CO₂ solubility is required to be estimated.

Sada *et al.* (1977; 1978), Al-Ghawas *et al.* (1989), and Haimour *et al.* (1984) considered that predictions of solubility using the N₂O analogy method can be used to predict absorption rates of CO_2 into aqueous MDEA under conditions of very short contact times where the chemical reaction does not affect the absorption rates. Diaz *et al.* (1988) proved this analogy from measurements on aqueous alcohol solutions.

Versteeg and Swaaji (1988) suggested, on the basis of the available data of solubility of N_2O and CO_2 in water, the following correlations for the solubility of CO_2 and N_2O in water:

 $H_{CO_2} = 2.8249 \times 10^6 \times \exp(-\frac{2044}{T})$ $H_{N_2O} = 8.5470 \times 10^6 \times \exp(-\frac{2284}{T})$ -

2.2 Solvents/Absorbents

Although N_2O analogy was used by many researchers previously to estimate the physical solubility of CO_2 in aqueous solutions of amines such as MEA, MDEA, MDEA, and di-2-propanolamine (DIPA)., there are limited data in the open literature on this parameter of CO_2 and N_2O in aqueous solutions of sodium glycinate in spite of the emerging importance of this amine acid salt in industrial gas treating processes. The following paragraphs illustrate the researches which have been done during these recent years:

Saha and Bandyopadhyay (1993) measured the solubility of N_2O in four aqueous 2-amino-2-methyl-1-propanol solutions over a range of temperature and atmospheric pressure. In this work, it was found that the solubility of N_2O in aqueous solutions of AMP was an exponential function of temperature at a constant concentration of the amine. Meanwhile, at constant temperature, the solubility changed linearly with the concentration of AMP. Throughout this experiment, the solubility of N_2O in aqueous solutions of AMP was represented by equation below:

$$H_{N_20} = (5.52 + 0.7C) \times 10^6 \exp(-\frac{2166}{T})$$

Kumar *et al.* (2001) measured the solubility of N_2O in aqueous potassium taurate solutions at various temperatures and in aqueous potassium glycinate solutions at 295 K. As a result, the effect of temperature and concentration of the salt on the solubility of N_2O were expressed by the Sechenov equation. The anion specific constant (h-) in the Sechenov equation has been determined for taurate and glycinate ions.

> $^{-}O_{3}SCH_{2}CH_{2}NH_{2}$: $(h_{-}) = 0.0249$ $^{-}O_{2}CCH_{2}NH_{2}$: $(h_{-}) = 0.0276$

Mandal *et al.* (2005) measured the solubility of N_2O in binary mixture (MEA + H_2O) as well as tertiary mixture (MDEA + MEA + H_2O) and (AMP + MEA + H_2O) at various temperatures and at atmospheric pressure. The N_2O analogy was used to estimate the physical solubility of CO_2 in (MEA + H_2O), (MDEA + MEA + H_2O), and (AMP + MEA + H_2O). The N_2O analogy for the physical solubility of CO_2 in amine solutions was given by the equations below, respectively,

$$(H_{CO_2})_{amine} = (H_{N_2O})_{amine} (\frac{H_{CO_2}}{H_{N_2O}})_{water}$$

where $(H_{N_2O})_{amine}$ is the solubility of N₂O in the amine solutions.

Lee *et al.* (2006) measured the solubility of N_2O and CO_2 in water and aqueous sodium glycinate solutions of various mass fractions at various temperatures. The solubility was found to be in decrement when there was an increase in sodium glycinate mass fraction as well as the increment of temperature.

2.3 Henry's Law

Apart from the mole fraction at a standard pressure and the Ostwald coefficient, Henry's law constant is also used to determine the gas solubility (Battino and Clever, 1966). In addition, Henry's law is valid in aqueous solutions of slightly soluble organic liquids (Saylor *et al.*, 1938). In an ideal solution of two liquids, both constituents obey Raoult's law. Nonetheless, for the real solutions at low concentrations, even though the solvent (the major constituent in the solution) commonly obeys Raoult's law, the solute (the minor constituent in the solution) does not (Saylor *et al.*, 1938).

Henry's law is applied to relate the equilibrium concentration of the gas in the liquid phase as a function of its partial pressure in the gas phase. Generally, Henry's law stated that the vapor pressure of the solute is proportional to the mole fraction of a slightly soluble gas dissolved in a liquid, in which the constant of proportionality is not the vapor pressure of the pure constituent. Physical solubility of gas in liquids can be determined from Henry's law, which is defined as:

$$H_A = p_A/C_A$$

where

 C_A = Equilibrium concentration of the absorbed gas (kmol/m³) H_A = Henry's law constant (kPa.m³/kmol)

*A to distinguish solute from solvent (B)

N₂O obeys Henry's law well because of its low solubility.

 p_A = Partial pressure in gas phase (kPa)

CHAPTER 3 . RESEARCH METHODOLOGY

3.1 Project Activities

Aqueous solutions of sodium glycinate were prepared by neutralizing glycine dissolved in distilled water, with an equimolar quantity of sodium hydroxide in a standard flask. The neutralization reaction was carried out with constant cooling.

Physical Solubility

The physical solubility of carbon dioxide (CO_2) using nitrous oxide (N_2O) analogy was measured by using the apparatus, as shown in Figure 1. Two pressure vessels were used in this work; one vessel is the mixing vessel, which was used to store N_2O gas, while the other one is the equilibrium cell, where the gas and the solvent were mixed together. Both vessels were thermo-regulated with water bath set at the required temperature.

Firstly, the mixing vessel was pressurized with the N_2O gas to the required pressure. A compressor was used to lift up the pressure. On the other hand, a certain amount of aqueous sodium glycinate solutions was introduced into the equilibrium cell, which has been previously purged with nitrogen (N_2) and vacuumed with the intention of removing the traces of air. The apparatus were then brought to a required temperature, and N_2O was fed to the equilibrium cell. The mixture was continuously stirred with a magnetic stirrer to improve the mass transfer and achieve the equilibrium fast. When the total system pressure reached a constant value and was maintained for at least two hours, the equilibrium was assumed to be achieved.



Figure 1: Solubility apparatus.

Pressure readings taking was repeated for various concentrations (1.0, 2.0 and 3.0 kmol/m^3), pressure (20, 40 and 60 bar) and temperature (303.15, 313.15 and 323.15 K), respectively. In the meantime, the experiments for determining Henry's constant of N₂O and CO₂ in pure water were carried out under a constant pressure of 10 bar and various temperatures of (303.15, 313.15 and 323.15) K. The constant pressure parameter was applied as the pressure gives insignificant influence to the solubility of gas in the distilled water. In other words, Henry's law constant in the water is temperature dependence.

The following pressure readings were taken:

- (1) P_1 , the initial pressure of mixing vessel
- (2) P_2 , the stabilized pressure of mixing vessel and equilibrium cell after the gas is transferred
- (3) P_{eqm} , the equilibrium pressure of equilibrium cell

The schematic diagram of the experimental setup is presented in Figure 2.



Figure 2: Schematic diagram of solubility apparatus.

where;

- MV : Mixing Vessel
- EC : Equilibrium Cell
- V_a : Air Valve
- V_b : Vent Valve
- V_c : N_2 to MV
- V_d : N₂O to MV
- Ve : Solvent to EC
- V_f : MV to EC

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V_g : Drain from EC

V_h : Water Valve

 V_i : N_2 to EC

The detailed procedures were described as follows:

Note: The procedures were repeated for each run except for A (Start-up) and H (Shut Down).

A. Start-up

- 1. The main power sources on computer, temperature and pressure indicator, metering pump, and water bath were turned on.
- 2. The gas cylinders of N_2 and N_2O were opened.

B. EC Purging (*and MV during start-up)

- 1. Valve V_i was opened to let N_2 gas flows into EC.
 - * Both valves V_c and V_f were opened to purge MV with N₂ gas during start-up.
- 2. Valve V_g was opened to let the residue gas flows out.
- 3. Both valves V_i and V_g were closed.
 - * Valves V_c and V_f were closed.

C. Temperature Setting

- 1. Valve V_h was opened to let the water flows into EC.
- 2. The desired temperature for EC was set.

D. MV Pressurizing

- 1. Valve V_d was opened to let N_2O gas flows into MV.
- 2. The flow rate of N_2O gas was set to be in between 1-6 L/min.
- 3. Valve V_a was opened to start pumping the gas into MV. The increment of pressure on the pressure indicator was observed.
- 4. Valve V_a was closed when the desired pressure was achieved.
- 5. The flow rate of N_2O gas was set to be at 0 L/min.
- 6. Valve V_d was closed.

E. Solvent Transfer

- 1. 10 mL of the aqueous solutions of sodium glycinate was inserted.
- 2. Valve Ve was opened. All the other valves were closed.
- 3. The metering pump was switched on to let the solvent flows into EC.
- 4. The solvent are transferred fully. After that, the metering pump was turned off to stop the flow and then valve V_e was closed.
- 5. Once the pressure was stabilized, the pressure reading of MV was taken as the initial pressure, P_1 .

F. Gás Transfer from MV to EC

- 1. Valve V_f was opened to transfer N_2O gas to EC.
- 2. Valve V_f was closed once the pressure for both MV and EC reached similar values.
- 3. The second pressure reading of MV, P_2 was recorded.
- 4. The magnetic stirrer was turned on.
- 5. When there was no or very slight change of the pressure in EC, the equilibrium is said to be achieved. The equilibrium pressure reading of EC, P_{eqm} was taken.

G. Waste handling

- 1. The magnetic stirrer was switched off.
- 2. EC was purged with N_2 gas by opening both valves V_b and V_i in order to bring down the pressure to atmospheric pressure and hence avoid spilling.
- 3. Valves V_b and V_i were closed once the atmospheric pressure was reached.
- 4. Valve V_g was opened cautiously to drain out the residue gas.

H. Shut Down

- 1. EC was washed with distilled water or acetone for several times.
- 2. The power sources on the computer, temperature and pressure indicator, metering pump, and water bath were turned off.

3.2 Tools

3.2.1 Materials/Chemicals:

Nitrous oxide (N_2O), carbon dioxide (CO_2), nitrogen (N_2), glycine, sodium hydroxide, distilled water.

3.2.2 Apparatus/Equipment:

2 pressure vessels -- mixing vessel and equilibrium cell, compressor, metering pump, magnetic stirrer, water bath, sample bottles.

3.3 Gantt Chart/Key Milestone

No.	Detail W	/eek	1	2	3	4	5	6	7		8	9	10	11	12	13	14
1	Continuation of Project Work (FYP I)																
2	Submission of Progress Report										٠						
3	Continuation of Project Work																
4	Pre-SEDEX				[i yet Lan a				٠			
5	Submission of Draft Report													٠			
6	Submission of Dissertation Report (soft-bound)						[٠		
7	Submission of Technical Paper															٠	
8	Oral Presentation																•
9	Submission of Project Dissertation (hard-bound)							· · ·									٠

Suggested milestone

Process

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CHAPTER 4 RESULTS AND DISCUSSION

4.1 Instrument Calibration

The solubility apparatus and the experimental procedure of the measurement were first standardized by comparing CO₂-water-MEA equilibrium data obtained on this setup with the literature data published by Jou *et al.* (1995) and Kadiwala *et al.* (2010). In this work, the solubility of aqueous solutions of 30 mass% MEA was studied at 313.15 K for a pressure range of 6 to 60 bar. Table 1 and Figure 3 compare the experimental data for the CO₂ solubility in 30 mass% aqueous MEA solutions at 313.15 K obtained in this work with the data reported in the literature by Jou *et al.* (1995) and Kadiwala *et al.* (2010). The average absolute deviation was found to be less than 10%. This shows that the experimental apparatus and the procedure performed in this work were precise enough to carry out such measurements, even in very high pressure.

Table 1: Comparison between experimental and literature data for 30 mass%MEA at 313.15 K.

	nass% MEA lou <i>et al</i> .)	30 mass% MEA (Kadiwala <i>et al.</i>)		30 ma (Th	ass% MEA ais work)
P (bar)	Loading/ α_{CO2}	P (bar)	Loading/a _{CO2}	P (bar)	Loading/ α_{CO2}
6	0.79	6.59	0.76	6.22	0.75
10	0.84	. 9.01	0.79	10.26	0.88
30	0.97	30.79	0.91	30.78	0.92
_60	1.04	53.29	0.99	62.05	1.08



Figure 3: Solubility of CO₂ in 30 mass% MEA at 313.15 K.

4.2 Physical Solubility of CO₂ and N₂O in Pure Water

As mentioned earlier, the experiments for estimating Henry's constant of CO_2 and N_2O in distilled water were measured at 10 bar for different temperatures of (303.15, 313.15 and 323.15) K. The pressure parameter was set as constant since the pressure shows slight effect on the gas solubility in the pure water. These experimental values were presented in Table 2. The experimental setup can be validated via this work too. Figure 4 compares the literature results and those obtained in this study for the solubility of CO_2 in water. The comparison between the literature results and data obtained in this work for the solubility of N_2O in water was displayed in Figure 5. The results of this work for the solubility of CO_2 and N_2O in pure water were in good agreement with the literature results.

On the other hand, it was found that Henry's constant of CO_2 and N_2O in distilled water increases when the temperature increases. In other words, the physical solubility decreases with an increase in temperature since a higher Henry's constant corresponds to a lower solubility.

	H _{CO2} (kPa.m ³ /kmol)	H _{N2O} (kPa.m ³ /kmol)	Reference
303.15	3192.10	4296.84	This work
	3394.38	4349.88	Al-Ghawas et al.
	3428	4835	Baek et al.
	3289.57	4450.2	Lee et al.
313.15	3706.45	4822.96	This work
	4249.57	5020.65	Al-Ghawas et al.
	4306	6108	Baek et al.
	3965.96	5504.92	Lee et al.
323.15	4872.21	5874.08	This work
	5166.56	5369.21	Al-Ghawas et al.
	5234	7445	Baek et al.
	5053.03	7184.98	Lee et al.

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Figure 4: Solubility of CO₂ in pure water as a function of temperature at 10 bar.



Figure 5: Solubility of N₂O in pure water as a function of temperature at 10 bar.

4.3 Physical Solubility of N₂O and CO₂ in Sodium Glycinate

Physical solubility of CO_2 in aqueous sodium glycinate solutions of concentration (1, 2 and 3) M at T = (303.15, 313.15 and 323.15) K and P = (20, 40 and 60) bar was estimated using the N₂O analogy. Such H_{N2O} and H_{CO2} values for different concentrations of aqueous sodium glycinate solutions at all temperatures and pressures are reported in Table 3 and presented graphically in Figures 6 to 11. We are unaware of any formerly published report on the physical solubility of N₂O and CO₂ in aqueous solutions of sodium glycinate at high pressure with which to compare our experimental data.

P (bar)	T (K)	SG (kmol/m ³)	H _{N2O} (kPa.m ³ /kmol)	H _{CO2} (kPa.m ³ /kmol)
20	303.15	1.0	603.78	448.54
		2.0	667.21	495.67
		3.0	773.75	574.81
	313.15	1.0	761.99	585.59
	•	2.0	867.72 •	666.84
		3.0	1033.81	794.48
	323.15	1.0	1017.53	843.99
		2.0	1226.89	1017.64
		3.0	1381.23	1145.65
40	303.15	1.0	540.21	401.32
		2.0	608.88	452.33
		3.0	704.44	523.33
	313.15	1.0	661.08	508.04
		2.0	766.40	588.98
_		3.0	930.36	714.98
	323.15	1.0	903.96	749.79
		2.0	1074.46	891.21
		3.0	1229.41	1019.73
60	303.15	1.0	412.09	306.14
		2.0	504.85	375.05
	•	3.0	624.40	463.86
	313.15	1.0	519.17	398.98
		2.0	628.74	483.19
-		3.0	778.48	598.26
	323.15	1.0	769.44	638.21
		2.0	938.29	778.26
		3.0	1178.29	977.33

Table 3: Physical solubility of N₂O and CO₂ in sodium glycinate (SG).



Figure 6, 7 and 8: Solubility of N₂O in sodium glycinate of different concentrations at 20, 40 and 60 bar.



Figure 9, 10 and 11: Solubility of CO₂ in sodium glycinate of different concentrations at 20, 40 and 60 bar.

As stated, a higher Henry's constant value corresponds to a lower solubility and vice versa. It was observed that while physical solubility increases with a decrease in sodium glycinate concentration, it decreases with an increase in temperature. Increased temperature causes an increase in kinetic energy and hence results in greater molecular motion of the gas particles. Consequently, the gas particles dissolved in the liquid are more likely to escape to the gas phase and the existing gas particles are less likely to be dissolved.

On the other hand, the relationship between physical solubility and pressure was described graphically in Figures 12 to 17. It was found that physical solubility increases with a decrease in sodium glycinate concentration and with an increase in pressure. In other words, greater CO_2 absorption in sodium glycinate is achieved at higher pressure. The effect of pressure and concentration on the gas solubility can be explained by Henry's Law, which states that at a constant temperature, the solubility of a gas in a liquid is directly proportional to the pressure of that gas above the surface of the solution. When the pressure increases, the gas particles are "forced" into the liquid. The number of gas particles above the surface of the liquid decreases, whereas the number of gas molecules dissolved in the solution increases.

In this work, the optimum condition for physical solubility of CO_2 in aqueous solutions of sodium glycinate at high pressure is at:

- Low temperature of 30 °C (303.15 K)
- High pressure of 60 bar



Figure 12, 13 and 14: Solubility of N₂O in sodium glycinate of different concentrations at 303.15, 313.15 and 323.15 K.



Figure 15, 16 and 17: Solubility of CO₂ in sodium glycinate of different concentrations at 303.15, 313.15 and 323.15 K.

CONCLUSIONS AND RECOMMENDATIONS

Throughout this work, we studied the physical solubility of nitrous oxide (N₂O) and carbon dioxide (CO₂) in aqueous sodium glycinate solutions of various concentrations (1.0, 2.0 and 3.0 kmol/m³) at the temperature of (303.15, 313.15 and 323.15) K and the pressure of (20, 40 and 60) bar. The N₂O analogy was used to estimate the solubility of CO₂ in aqueous solutions of sodium glycinate. The solubility data for CO₂ in aqueous solutions of 30 mass% MEA was found to in good agreement with the literature results, therefore supporting the reliability of the present solubility apparatus and experimental method. Furthermore, it was also validated through the study of physical solubility of N₂O and CO₂ in pure water, which the results showed that there is a good agreement between literature data and those of the present work.

On the one hand, it was observed that the physical solubility of CO_2 and N_2O in distilled water decreases when the temperature increases since a higher Henry's constant corresponds to a lower solubility. For the physical solubility of N_2O and CO_2 in aqueous solutions of sodium glycinate, it decreases with an increase in sodium glycinate concentration, with an increase in temperature and with a decrease in pressure. In our study, the optimum condition for physical solubility of CO_2 in aqueous solutions of sodium glycinate at high pressure is accomplished at low temperature of 30 °C and high pressure of 60 bar. As a result, sodium glycinate could be recommended as an appropriate solvent for the separation of CO_2 from gaseous streams.

In future work, the solubility measurement for sodium glycinate can be carried out at higher pressure under safety concern. Additionally, the comprehensive behaviors and characteristics of sodium glycinate need to be studied thoroughly since sodium glycinate is still considered as a new absorbent industrially. Aside from that, more researches on physical solubility of N_2O and CO_2 in various solvents should be done at high pressure to make a comparison between the solubility data using different solvents.

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APPENDIX

Sample of Calculation

Volume of mixing vessel, V_{MV}	= 3 L
Volume of equilibrium cell, V_{EC}	= 0.1 L
Volume of water/solvent, V _{water/solvent}	= 0.01 L
Universal gas constant, R	= 0.083145 bar.L/mol.K

(A) CO2 Loading in 30 mass% MEA at 313.15 K and 6 bar

Density of MEA, ρ_{MEA}	$= 1.012 \ g/cm^3$
Molecular weight of MEA, MW _{MEA}	= 61.08 g/mol
Compressibility factor, Z	= 0.975
P_I	= 6.41 bar
P_2	= 6.02 bar
P_{eqm}	= 2.45 bar
T_{MV}	$= 39.6 \ ^{\circ}C = 312.75 \ K$
T_{EC}	$= 39.8 \ ^{\circ}C = 312.95 \ K$

$$\Delta P = P_1 - P_2$$

= (6.41 - 6.02) bar
= 0.39 bar

$$n_{co_2} = \frac{\Delta P V_{MV}}{ZRT_{MV}}$$

= $\frac{0.39 \ bar \times 3 \ L}{0.975 \times 0.083145 \ \frac{bar \cdot L}{mol \cdot K} \times 312.75 \ K}$
= 0.046147 mol

$$n_{CO_2}^g = \frac{P_{eqm}(V_{EC} - V_{solvent})}{ZRT_{EC}}$$
$$= \frac{2.45 \ bar \ (0.1 - 0.01) \ L}{0.975 \times 0.083145 \ \frac{bar \cdot L}{mol \cdot K} \times 312.95 \ K}$$
$$= 0.008691 \ mol$$

$$n_{CO_2}^l = n_{CO_2} - n_{CO_2}^g$$

= 0.037456 mol

 $n_{MEA}^{l} = \frac{w_{/W} \cdot \rho_{MEA} \cdot V_{solvent} \cdot 1000}{MW_{MEA}}$ $= \frac{0.3 \times 1.012 \frac{g}{cm^{3}} \times 0.01 L \times 1000}{61.08 \frac{g}{mol}}$ = 0.049705 mol

$$\alpha = \frac{n_{CO_2}^l}{n_{MEA}^l}$$
$$= 0.753561$$

* The same calculation procedures were applied to determine CO_2 loading in the same MEA concentration at the same temperature but different pressures (10, 30 and 60 bar).

(B) Henry's Constant of N2Oand CO2 in Pure Water at 303.15 K and 10 bar

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B1. Henry's constant of CO2 in pure water at 303.15 K and 10 bar
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Compressibility factor, Z	= 0.94
P_1	= 10.22 bar
P_2	$= 9.92 \ bar$
P_{eqm}	= 9.41 bar
T_{MV} .	$= 24.1 \ ^{\circ}C = 297.25 \ K$
T_{EC}	$= 29.6 \ ^{\circ}C = 302.75 \ K$

$$\Delta P = P_1 - P_2$$
$$= 0.3 \, bar$$

$$n_{CO_2} = \frac{\Delta P V_{MV}}{ZRT_{MV}}$$
$$= 0.038740 \ mol$$

$$n_{CO_2}^g = \frac{P_{eqm}(V_{EC} - V_{solvent})}{ZRT_{EC}}$$
$$= 0.035792 \ mol$$

$$n_{CO_2}^l = n_{CO_2} - n_{CO_2}^g$$

= 0.002948 mol

$$C_{CO_2} = \frac{n_{CO_2}^l}{V_{solvent}}$$
$$= 0.294790 \ mol/L$$

$$(H_{CO_2})_{water} = \frac{P_{eqm}}{C_{CO_2}}$$
$$= 31.921042 \frac{bar \cdot L}{mol}$$
$$= 3192.104197 \frac{kPa \cdot m^3}{kmol}$$

B2. <u>Henry's constant of N₂O in pure water at 303,15 K and 10 bar</u>

Compressibility factor, Z	= 0.94
P_{I}	= 10.26 bar
P_2	= 9.95 bar
P _{eqm}	= 9.73 bar
T_{MV}	$= 30.4 \ ^{\circ}C = 303.55 \ K$
T_{EC}	$= 30.2 \ ^{\circ}C = 303.35 \ k$

$$\Delta P = P_1 - P_2$$
$$= 0.31 \ bar$$

$$n_{N_2O} = \frac{\Delta P V_{MV}}{ZRT_{MV}}$$

= 0.039200 mol
$$n_{N_2O}^g = \frac{P_{eqm}(V_{EC} - V_{solvent})}{ZRT_{EC}}$$

= 0.036936 mol

$$n_{N_20}^l = n_{N_20} - n_{N_20}^g$$

= 0.002264 mol

$$C_{N_2O} = \frac{n_{N_2O}^l}{V_{solvent}}$$
$$= 0.226445 \ mol/L$$

$$(H_{N_2O})_{water} = \frac{P_{eqm}}{C_{N_2O}}$$
$$= 42.968448 \frac{bar \cdot L}{mol}$$
$$= 4296.844775 \frac{kPa \cdot m^3}{kmol}$$

$$(\frac{H_{CO_2}}{H_{N_2O}})_{water} = \frac{3192.104197}{4296.844775} = 0.742895$$

* The similar calculation procedures were applied to determine Henry's constant of N_2O and CO_2 in distilled water at the same pressure but different temperatures (40 and 50 °C).

(C) Henry's Constant of N2O and CO2 in 1 M Sodium Glycinate (SG) at 303.15

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K and 20 bar

Compressibility factor, Z	= 0.89
P_1	= 20.28 bar
P_2	= 19.47 bar
P _{eqm}	= 19.08 bar
T_{MV}	$= 30.7 \ ^{\circ}C = 303.85 \ K$
T_{EC}	$= 30.3 \ ^{\circ}C = 303.45 \ K$

$$\Delta P = P_1 - P_2$$
$$= 0.81 \ bar$$

$$n_{N_2O} = \frac{\Delta P V_{MV}}{ZRT_{MV}}$$
$$= 0.108074 \ mol$$

$$n_{N_2O}^g = \frac{P_{eqm}(V_{EC} - V_{solvent})}{ZRT_{EC}}$$
$$= 0.076473 \ mol$$

$$n_{N_2O}^l = n_{N_2O} - n_{N_2O}^g$$

= 0.031601 mol

$$C_{N_2O} = \frac{n_{N_2O}^l}{V_{solvent}}$$
$$= 3.160101 \ mol/L$$

$$(H_{N_2O})_{SG} = \frac{P_{eqm}}{C_{N_2O}}$$
$$= 6.037782 \frac{bar \cdot L}{mol}$$
$$= 603.7782 \frac{kPa \cdot m^3}{kmol}$$

$$(H_{CO_2})_{SG} = (H_{N_2O})_{SG} \times \left(\frac{H_{CO_2}}{H_{N_2O}}\right)_{water}$$
$$= 603.7782 \frac{kPa \cdot m^3}{kmol} \times 0.742895$$
$$= 448.544 \frac{kPa \cdot m^3}{kmol}$$

* The same calculation procedures were used to find out Henry's constant of N_2O and CO_2 in sodium glycinate with other concentrations (2 and 3 M) at different temperatures (40 and 50 °C) as well as different pressures (40 and 60 bar).