

Viscosities of diethylene glycol (DEG) + 2-amino-2-methyl-1-propanol (AMP) + water

V Gurylev¹, A N Soriano², A R Caparanga², M-H Li¹, and A P Adornado^{2,3}

¹R & D Center for Membrane Technology and Department of Chemical Engineering, Chung Yuan Christian University, Chung-Li City, Taoyuan, Taiwan, R.O.C

²School of Chemical, Biological, and Materials Engineering and Sciences, Mapúa University, Manila 1002, Philippines

E-mail: apadornado@mapua.edu.ph/adonisadornado@yahoo.com

Abstract. In this study, the viscosities (η) of the aqueous mixed-solvent system of diethylene glycol (DEG) and 2-amino-2-methyl-1-propanol (AMP) were investigated at normal atmospheric pressure over the range of temperature of 303.15 - 343.15 K and across varying component mole fractions range [0.02 to 0.60 for DEG (x_1), 0.02 to 0.60 for AMP (x_2), and 0.20 to 0.80 for H₂O (x_3)]. The calculated properties were fitted to a modified equation originally proposed by Grunberg and Nissan for the viscosity of liquid mixtures to correlate temperature and composition dependency of the measured property. In comparison to literature values, the uncertainty of the kinematic viscosities was approximated to be ± 1.0 %, and that the correlations used satisfactorily represent the effects of temperature and composition on the viscosities of the considered mixed-solvent system as shown by the low overall average absolute deviations obtained in the calculations. The properties obtained and reported in this work are, in general, of adequate correctness for most engineering-design calculations, such as the design of absorption process for carbon dioxide capture using mixed-solvents as absorbents.

1. Introduction

Alkanolamines have been regarded as a suitable solvent choice as absorbent for removal of acidic components such as carbon dioxide (CO₂) and hydrogen sulfide (H₂S) from gas streams due to their high absorption capacities and fast reaction rates [1,2]. The interest in using glycol-amine solutions for acid-gas absorption has dramatically boost when the other considerations in deciding for a suitable absorbent like solvent regeneration, solvent corrosivity, and energy requirement in various processes have been understood. The use of glycol-amine solutions is more favorable due to concurrent purification and dehydration, lesser steam intake, and can almost completely strip the CO₂, resulting to extremely high-purity gas compared to aqueous blended-amine systems [3]. The CO₂ loading capacity and ease of regeneration of 2-amino-2-methyl-1-propanol (AMP) are superior that of primary or secondary amines [2,4].

In order to measure other physical properties of alkanolamines such as liquid diffusivities, free-gas solubility, and reaction rate constants, preceding physical properties such as density, viscosity, and surface tension are needed to be measured first which are also required for the design of acid-gas treatment equipment. Viscosities of aqueous blended amine solutions containing AMP have been made available in the literature [5,6]. In 1996, Song *et al* [7] reported the viscosities of monoethanolamine (MEA) + ethylene glycol (EG) aqueous solutions. Also, Tsierkezos and Molinou



[8], reported the viscosities of EG binary mixtures at 293.15 K, and Braun *et al* [9], the viscosities of mono (ethylene glycol) (MEG) + AMP aqueous solutions. While numerous studies on viscosity measurements have been done on various types of glycols and amines (either focused on pure systems and/or aqueous solutions), viscosity measurements for aqueous mixtures containing diethylene glycol (DEG) + AMP, as far as we know it, are still unavailable.

In line with our systematic studies on the property determination of glycol-amine systems, viscosity measurements of aqueous system containing DEG + AMP were made at normal atmospheric pressure over the range of temperature of 303.15 - 343.15 K. The measurements were likewise made at varying mole fractions of the components: 0.02 to 0.60 for DEG (x_1), 0.02 to 0.60 for AMP (x_2), and 0.20 to 0.80 for H₂O (x_3). The results of the present work are essential data for future works on solvent systems that used the considered components, such as possible absorbent in CO₂ capture. Similarly, the data gathered could add up to the database of the investigated systems which are in-demand information for the process designers.

2. Materials and methods

2.1. Chemicals

The diethylene glycol (DEG) utilized in this study, provided by Tedia Co, had a minimum purity of 99.0%. Likewise, 2-amino-2-methyl-1-propanol (AMP), obtained from ACROS Organics, had a minimum purity of 99.0%. The deionized water (Type I reagent-grade; resistivity $\leq 18.3 \text{ M}\Omega\text{-cm}$; total organic content (TOC) $\leq 15 \times 10^{-9}$) used to prepare all aqueous solutions in the experiments was purified in a Barnstead Thermolyne (model Easy Pure 1052) compact ultrapure water system and degassed under vacuum prior to use. A Mettler-Toledo digital balance (model AL204) having an accuracy of $\pm 1 \times 10^{-4} \text{ g}$ was used in all weight measurements during the preparation of solutions.

2.2. Viscosity measurements

The kinematic viscosities of the considered ternary systems were measured by means of Cannon-Fenske routine viscometers with uncertainty of $\pm 0.2\%$ in a water bath isothermally held to $\pm 0.05 \text{ K}$. An electronic stopwatch with 0.01s precision was used to measure the efflux time. These measurements followed the standard test method and specs of the ASTM D 445 (Test Method for Kinematic Viscosity of Transparent and Opaque Liquids) [10]. In the calculation of the kinematic viscosity, end effect corrections were neglected as in the method of Rinker *et al* [11]. The product of the kinematic viscosity and density gives the absolute viscosity of the fluid. In the present work, the density data for the considered ternary solution were adapted from our published work [12]. In comparison with the literature values, the uncertainty of the kinematic viscosities was approximated to be $\pm 1.0 \%$. The assessments of the viscosities were carried out over the range of temperature of 303.15 - 343.15 K. All measurements performed in this study were carried out in three to five replicate runs and the mean values were reported.

3. Results and discussion

The viscosity measurements for the ternary system are summarized below. Table 1 shows the experimental viscosities of the aqueous solutions containing diethylene glycol (DEG) and 2-amino-2-methyl-1-propanol (AMP). The results are reported over a range of temperature of 303.15 - 343.15 K, at varying concentrations of the components, expressed as mole fraction (x_i).

Table 1. Experimental viscosities (η) of DEG (1) + AMP (2) + H₂O (3) solutions a range of temperature of 303.15 - 343.15 K, at varying mole fractions of the components: 0.02 to 0.60 for DEG (x_1), 0.02 to 0.60 for AMP (x_2), and 0.20 to 0.80 for H₂O (x_3).

viscosity, η / (mPa·s)	
x_1/x_2 ($x_3 = 0.20$)	

<i>T</i> / K	0.60/0.20	0.50/0.30	0.40/0.40	0.30/0.50	0.20/0.60
303.15	31.625	41.825	55.252	63.082	72.499
313.15	18.729	25.385	31.440	38.822	50.278
323.15	13.437	15.855	19.867	22.682	24.968
333.15	8.553	10.585	12.017	13.714	14.950
343.15	6.222	6.851	8.081	8.716	9.204
<i>x</i>₁/<i>x</i>₂ (<i>x</i>₃ = 0.40)					
<i>T</i> / K	0.50/0.10	0.40/0.20	0.30/0.30	0.20/0.40	0.10/0.50
303.15	21.330	29.077	39.849	51.830	61.801
313.15	13.914	18.183	22.585	29.559	38.477
323.15	9.055	11.685	14.434	18.943	21.376
333.15	6.433	7.923	9.442	10.677	12.149
343.15	4.804	5.454	6.388	6.889	8.167
<i>x</i>₁/<i>x</i>₂ (<i>x</i>₃ = 0.60)					
<i>T</i> / K	0.35/0.05	0.30/0.10	0.20/0.20	0.10/0.30	0.05/0.35
303.15	12.952	19.673	25.969	31.885	36.397
313.15	8.481	12.712	16.115	19.944	22.952
323.15	6.097	8.092	10.944	13.606	16.209
333.15	4.463	5.672	6.819	7.964	9.064
343.15	3.474	3.835	4.549	5.005	5.882
<i>x</i>₁/<i>x</i>₂ (<i>x</i>₃ = 0.80)					
<i>T</i> / K	0.18/0.02	0.14/0.06	0.10/0.10	0.06/0.14	0.02/0.18
303.15	8.424	9.021	9.493	9.948	10.678
313.15	4.666	5.457	6.178	6.702	7.022
323.15	3.056	3.752	4.158	4.537	4.986
333.15	2.365	2.578	3.082	3.432	3.937
343.15	2.057	2.165	2.390	2.422	2.464

The studied ternary system exhibited an increase in viscosity with a decrease in the mole fraction of DEG (*x*₁), while keeping the mole fraction of water (*x*₃) constant (see figure 1(a)). Such observed behavior could be attributed to the significant difference of the viscosities of pure DEG and AMP at a particular temperature (see table 2). The AMP, being a much viscous fluid than DEG, could influence heavily the viscosity of the investigated ternary mixture. Furthermore, a decrease in the viscosity measurements was observed with the increase in temperature of the systems (see figure 1(b)).

Table 2. Viscosities of the pure fluids under studied at a range of temperature of 303.15 - 343.15 K.

<i>T</i> / K	viscosity, <i>η</i> / (mPa·s)		
	DEG	AMP^a	H₂O^b
303.15	21.963	99.475	0.815
313.15	14.445	46.926	0.668
323.15	10.067	24.211	0.559
333.15	7.320	13.998	0.477
343.15	4.443	8.642	0.412

^a Taken from Li and Lie [5].

^b Taken from Miller *et al* [14].

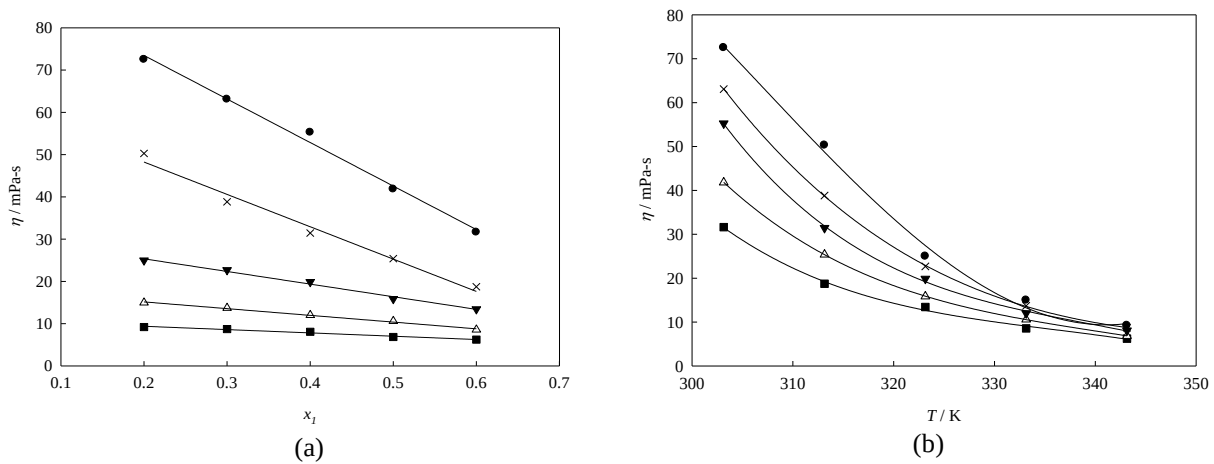


Figure 1. Plot of viscosity (η) data for DEG (1) + AMP (2) + H₂O (3) systems: (a) at fixed temperatures and different DEG-AMP concentration (x_1/x_2), and fixed water concentration ($x_3 = 0.20$): $\bullet$, $T = 303.15$ K; $\times$, $T = 313.15$ K; $\blacktriangledown$, $T = 323.15$ K; $\triangle$, $T = 333.15$ K; $\blacksquare$, $T = 343.15$ K; and lines are calculated using equation (4); (b) at different temperatures and DEG-AMP concentration (x_1/x_2), and fixed water concentration ($x_3 = 0.20$): $\bullet$, (x_1/x_2) = 0.60/0.20; $\times$, (x_1/x_2) = 0.50/0.30; $\blacktriangledown$, (x_1/x_2) = 0.40/0.40; $\triangle$, (x_1/x_2) = 0.30/0.50; $\blacksquare$, (x_1/x_2) = 0.20/0.60; and lines are calculated using equation (4).

The empirical formula of Grunberg and Nissan, shown in equation (1), presented by Reid *et al* [13] in *The Properties of Gases and Liquids* for the viscosity of liquid mixtures is utilized to correlate the viscosity data.

$$\ln \eta_m = \sum_i x_i \ln \eta_i + \sum_{i \neq j} x_i x_j G_{ij}, \quad (1)$$

where η_m is the viscosity of the mixtures and η_i the viscosity of the i th pure fluid. In this work, the viscosity data for the involved pure fluids were taken from the available literature data whenever possible. Of the involved pure fluids, DEG has no available viscosity data, thus the viscosity for pure DEG was also measured for the considered range of temperature of. Table 2 presented the viscosity data for the investigated pure fluids.

For a ternary system, equation (1) has

$$\ln \eta_m = x_1 \ln \eta_1 + x_2 \ln \eta_2 + x_3 \ln \eta_3 + x_1 x_2 G_{12} + x_1 x_3 G_{13} + x_2 x_3 G_{23}. \quad (2)$$

In equation (1), G_{ij} is the binary pair-interaction parameters which is assumed as temperature-dependent and has the form

$$G_{ij} = a + b(T/K) + c(T/K)^2 + d(T/K)^3. \quad (3)$$

In this work, we have modified equation (2) by incorporating another parameter to describe the ternary interaction. The new form of equation (2) is written as

$$\ln \eta_m = x_1 \ln \eta_1 + x_2 \ln \eta_2 + x_3 \ln \eta_3 + x_1 x_2 G_{12} + x_1 x_3 G_{13} + x_2 x_3 G_{23} + x_1 x_2 x_3 G_{123}, \quad (4)$$

where G_{123} is the ternary interaction parameter which is also assumed as temperature-dependent and has a similar form as that of equation (3).

The viscosity data of the ternary system considered in this work is for DEG + AMP + H₂O. From the ternary system's viscosity data, the pair-interaction parameters for DEG-AMP; and ternary interaction parameters for DEG-AMP-H₂O, could be determined using equation (4). Since there is

already an available literature value [5] for the pair-interaction parameters for AMP–H₂O, these data were simply used and the other interaction parameters were determined using equation (4). Table 3 presented the determined interaction parameters. As presented in this table, equation (4) acceptably evaluated the considered pair-interaction parameters as shown by an acceptable overall average absolute difference (AAD) of 0.005 for a total of 100 data points. Here, AAD is evaluated as

$$\text{AAD} = \frac{1}{n} \times \sum_{i=1}^n \left| \left(\varepsilon_{\text{cald}} - \varepsilon_{\text{expt}} \right) \right|_i, \quad (5)$$

where n is the number of data points and $(\varepsilon_{\text{cald}}$ and $\varepsilon_{\text{expt}})$ are calculated and experimental values, respectively.

Table 3. Determined binary parameters of viscosity using the Grunberg and Nissan equation.

Interacting System	parameters			
	<i>a</i>	<i>b</i>	<i>c</i>	<i>d</i>
AMP–H ₂ O ^c	1,961.80	-17.2300	0.050803	-5.0036×10^{-5}
DEG–AMP	-469.95	5.9696	-0.023330	2.9036×10^{-5}
DEG–H ₂ O	682.72	-5.0066	0.011723	-8.3694×10^{-6}
DEG–AMP–H ₂ O	-21,314.00	184.1600	-0.528720	5.0325×10^{-4}

^c Taken from Li and Lie [5].

4. Conclusion

The viscosities of the ternary system of mixed-solvent consisting of diethylene glycol (DEG) + 2-amino-2-methyl-1-propanol (AMP) + water (H₂O) are reported over the range of temperature of up to 343.15 K at normal atmospheric pressure. The varying mole fractions of the components studied are: 0.02 to 0.60 for DEG (x_1), 0.02 to 0.60 for AMP (x_2), and 0.20 to 0.80 for H₂O (x_3). The values of the viscosity were found to generally increase with a decrease in the glycol component. Likewise, a decrease in the viscosity measurements was reported with an increase in temperature. Our values of viscosity as a function of temperature and composition were well represented by a modified empirical equation originally proposed by Grunberg and Nissan for the viscosity of liquid mixture systems as shown by the overall average absolute difference (OAAD) of 0.50% and the result shows that the systems examined in this paper are potential absorbents for carbon dioxide capture process.

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References

- [1] Kohl A and Riesenfeld F 1979 *Gas Purification* (Texas: Gulf Publishing Company)
- [2] Sartori G and Savage D 1983 *Ind. Eng. Chem. Fund.* **22** 239-49
- [3] Kohl A and Nielsen R 1997 *Gas Purification* (Texas: Gulf Publishing Company)
- [4] Zhang P, Shi Y, Wei J, Zhao W and Ye Q 2008 *J. Environ. Sci.* **20** 39-44
- [5] Li M-H and Lie Y-C 1994 *J. Chem. Eng. Data* **39** 444-7
- [6] Hsu C-H and Li M-H 1997 *J. Chem. Eng. Data* **42** 714-20
- [7] Song J, Park S, Yoon J, Lee H and Lee K 1996 *J. Chem. Eng. Data* **41** 1152-4
- [8] Tsierkezos N and Molinou I 1994 *J. Chem. Eng. Data* **44** 955-8
- [9] Braun N, Persson U and Karlsson H 2001 *J. Chem. Eng. Data* **46** 805-8
- [10] American Society for Testing and Materials 1984 *Annual Book of ASTM Standards* (Philadelphia)
- [11] Rinker E, Oelschlager D, Colussi A, Henry K and Sandall O 1994 *J. Chem. Eng. Data* **39** 392-5
- [12] Gurylev V, Soriano A, Eleazar E, Caparanga A and Li M-H 2014 *PICChE J.* **15** 21-9

- [13] Reid R, Prausnitz J and Poling B 1988 *The Properties of Gases and Liquids* (New York: McGraw-Hill)
- [14] Miller J, Schorr G and Yaws C. 1976 *Chem. Eng.* **83** 157-9