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Density and Viscosity Measurement and Correlations of Aqueous Solution of 2-(Diethylamino)ethanol for CO_2 Capture

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CO_2 causes the global issues on climate change and global warming. The main sources of this gas are from energy consumption such as power plants and natural gas processing. The absorption technology with amine solution of 2-(Diethylamino)ethanol (DEAE) is a promising method to remove CO_2 from these processes. To calculate the basic parameters such as diffusivity and the flow of fluids and to design the absorption equipment, the fundamental properties such as the density and the viscosity are needed to be measured. Therefore, the objective of this work is to measure the density and viscosity of the aqueous solution of DEAE over the entire concentration range from 0-1 mole fraction and the temperature ranging from 30-80 °C. The results present that the densities of DEAE decrease with an increase in mole fraction and temperature of solution. Also, the viscosities of all ranges of mole fraction of DEAE decrease as temperature increases. However, the viscosities increase with mole fraction of DEAE up to 0.4 and become lower. Furthermore, the correlations of the Redlich-Kister equation, the Wilson model, the Grunberg and Nissan equation and Chauhan polynomial equation have been applied to represent the results and to predict the data for the equipment design. Among them, the Redlich-Kister equation is the most suitable model to represent the results with the lowest percent average absolute deviation (%AAD) with 0.1590 % and 1.9413 % for density and viscosity.

1. Introduction

Recently, climate change and global warming are the severe global issues which have been received widespread attention from all sectors around the world (IPCC, 2005). The major cause of these phenomena is CO_2 mainly released from the power generation, fuel combustion and gas processing (EIA, 2015). The captured CO_2 can be used for enhanced oil recovery process to produce more oil (Yoosook et al., 2018). The CO_2 capture technologies have been developed and applied to reduce CO_2 released from several processes (IPCC, 2005). Among the CO_2 capture techniques, the chemical absorption is a promising technology for CO_2 from the power plants. There are many chemical solvents which have been researched to use as the effective absorbents. Amines are well-known solvents because of their reversible reactions with CO_2 (Dutcher et al., 2015). Monoethanolamine (MEA) and N-methyldiethanolamine (MDEA) is conventional solvents for capturing CO_2. Nevertheless, these solvents have low absorption capacity and require high energy for regeneration (Kohl and Nielsen, 1997). There are some attempts to overcome these limits by developing the novel amines such as 2-methylaminoethanol to improve the performance of CO_2 mitigation (Maneeintr et al., 2018) for single solvent. Moreover, the mixed amine can be formulated like 3-Amino-1-Propanol (Nimcharoen et al., 2018) for mixed solvents. For this study, 2-(Diethylamino)ethanol (DEAE) is a potential candidate for removing CO_2 due to its superior absorption capacity (Maneeintr et al., 2014). DEAE is an amino alcohol which contains amino group and hydroxyl group as illustrated in Figure 1. Therefore, to design the equipment of absorption process, various physical properties like density and viscosity are required for the whole range of operating conditions. Furthermore, the calculation of some necessary properties such as diffusivity also requires the data of these properties.

Consequently, this work aims to measure the density and the viscosity of the aqueous solution of DEAE over the entire concentration ranging from 0.0 to 1.0 mole fraction and temperature from 30 to 80 °C. In addition, the

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correlations of density and viscosity for binary system of DEAE and water are developed in order to predict the properties for the equipment design.

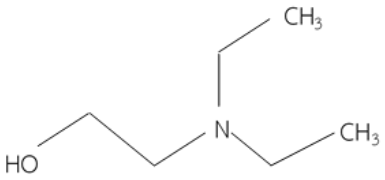

Figure 1: Molecular structure of 2-(Diethylamino)ethanol (DEAE)

2. Experiment

2.1 Materials

DEAE with a purity of ≥99 % is purchased from Merck, Thailand. Distilled water is used to prepare the sample to the desired concentration. Moreover, MEA (99 %purity), used for equipment calibration, is obtained from Sigma-Aldrich.

2.2 Equipment and experimental procedure

Density measurements of binary mixture are executed by using Anton Paar density meter (DMA-4500 model) with 0.00001 g/cm^3 resolution. The accuracies of the equipment for density and temperature are ±0.00005 g/cm^3 and ±0.03 °C. The equipment was calibrated with MEA and distilled water at the temperature from 30 to 80 °C. The measurement results are calibrated with the data of density from the literature (Amundsen et al., 2009).
Kinematic viscosities are measured by means of the appropriate U-tube glass viscometers with uncertainly in absolute viscosity values at ±0.003 mPa•s. The viscometers are operated in a water bath (model CT72/2 from SI analytics) with the temperature accuracy of ±0.20 °C. The viscometer is also calibrated by the same solution and the same temperature range used in the density meter calibration. The calibration results are compared with the data of viscosity from the literature (Li and Lie, 1994).
The DEAE with distilled water samples are prepared with various mole fraction of 0.0 to 1.0. The temperature of solution is varied from 30 to 80 °C. The experiments are repeating for 3 times per each sample and the average results are obtained.

2.3 Correlations

Various equations which are used for the correlation of the physical and transportation properties such as density and viscosity of binary mixture, are adopted to demonstrate and predict the experimental data. Firstly, the Redlich-Kister (Redlich and Kister, 1984) using for density correlation of binary mixture are presented as shown in Eq(1) to (4) and for viscosity, the equation are illustrated in Eq(5) and (6). Secondly, the equation of the Wilson model (Prausnitz et al., 1999) can be presented in Eq(7). Thirdly, the Grunberg and Nissan Equation (Poling et al., 2001) are shown in Eq(8) and (9) and the last equation, the strictly empirical polynomial correlation by Chauhan (Chauhan et al., 2003) is presented in Eq(10).

$$\Delta P = x_1 x_2 \sum_{i=0}^{n} A_i (x_1 - x_2)^i \quad (1)$$

$$V^E = x_1 x_2 \sum_{j=0}^{n} A_i (x_1 - x_2)^j \quad (2)$$

$$V^E = V_m - \sum x_i V_i \quad (3)$$

$$V_m = \frac{\sum x_i M_i}{\rho_m} \quad (4)$$

$$\Delta\eta = x_1 x_2 \sum_{j=0}^{n} A_i (x_1 - x_2)^j \quad (5)$$

$$\Delta\eta = \eta - x_1\eta_1 - x_2\eta_2 \quad (6)$$

$$\Delta P = RT(-X_1 \ln(X_1 + AX_2) - X_2 \ln(X_2 + BX_1)) \quad (7)$$

$$lnP_m = X_1 lnP_1 + X_2 lnP_2 + X_1 X_2 G_{12} \quad (8)$$

$$G_{ij} = A + B\left(\frac{T}{K}\right) + C\left(\frac{T}{K}\right)^2 + D\left(\frac{T}{K}\right)^3 \quad (9)$$

$$P_m = \sum_{i=0}^{2}\left[A_i x^i + B_i x^i \left(\frac{T}{K}\right) + C_i x^i \left(\frac{T}{K}\right)^2\right] \quad (10)$$

Where ΔP refers to excess molar volume (V^E), A_is refer to coefficients, x_1 and x_2 refer to mole fraction of component 1 and 2 and η is the molar refraction of the mixture. P_i is property of pure component in the mixture whereas P_m refer to the measured mixture properties. A, B, C and D refer to coefficients. Δη refers to viscosity deviation. η, η_1 and η_2 are viscosity of mixture and pure component 1 and 2 and G_{ij} is the interaction parameter while T is the temperature of the mixture in Kelvin.

An average absolute deviation (AADs) between experimental data and calculated values, are illustrated by Eq (11). The purpose of AAD is to measure the statistical dispersion as the average of the absolute deviations from a central point.

$$\%AAD = \frac{100}{n}\sum_{i=1}^{n}\left|\frac{P_{exp}-P_{cal}}{P_{exp}}\right| \quad (11)$$

Where n is the number of data points.

3. Results and discussion

3.1 Density measurement

The density results of the binary mixture of aqueous 2-Diethylamino-ethanol (DEAE) at various mole fraction ranging from 0.0 to 1.0 at the temperature range from 30 to 80 °C are illustrated in Figure 2. The density tends to decrease considerably with an increase in mole fraction and temperature. This phenomenon causes by higher kinetic energy of substance molecule. Then, the volume of substance is expanded. For the effect of concentration, the higher concentration of solution provides lower density because the pure distilled water has higher density than pure DEAE. Thus, increasing water composition in the mixture reduces the mixture density.

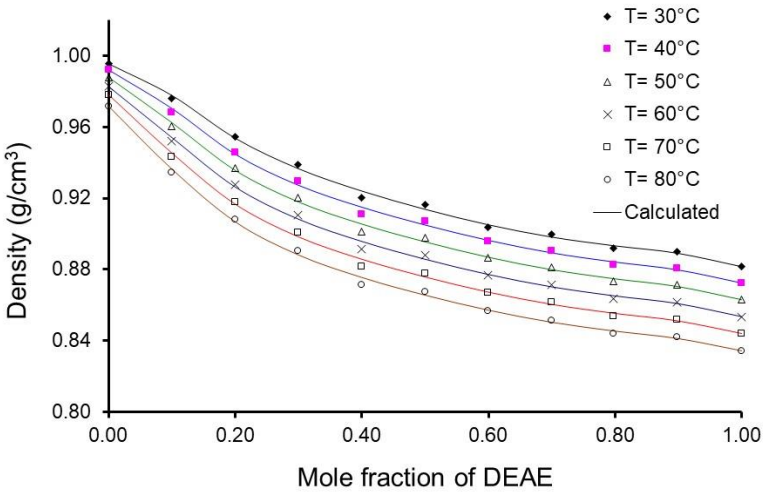

Figure 2: Density values of DEAE + Distilled water at several of mole fraction at the temperature ranging from 30-80 °C

From Figure 2 and Table 1, the results illustrate that the calculated data provide the good agreement with the measured results for the density. The coefficients of correlation by using Redlich-Kister equation for density of aqueous solution of DEAE are presented in Table 1 and the %AADs of all temperatures is 0.1590 %. The comparison between the measured and calculated data based on effect of mole fraction for DEAE / distilled water mixture at temperature range of 30 to 80 °C is presented in Figure 3.

Table 1: Redlich-Kister equation coefficients of excess molar volume for DEAE + distilled water at temperature ranging from 30-80 °C

T (°C)	A_0	A_1	A_2	A_3	A_4	%AAD
30	-6.1306	2.7606	-0.0473	-1.8729	-9.3494	0.1588
40	-6.1858	2.1812	0.6564	-1.0961	-9.9863	0.1569
50	-6.0896	2.0574	0.7778	-1.2291	-10.1797	0.1589
60	-5.9855	1.945	0.8529	1.2514	-10.3428	0.1595
70	-5.7497	1.872	0.9816	-1.1798	-10.1512	0.1598
80	-5.5922	1.8112	1.024	-1.0992	-10.268	0.1599

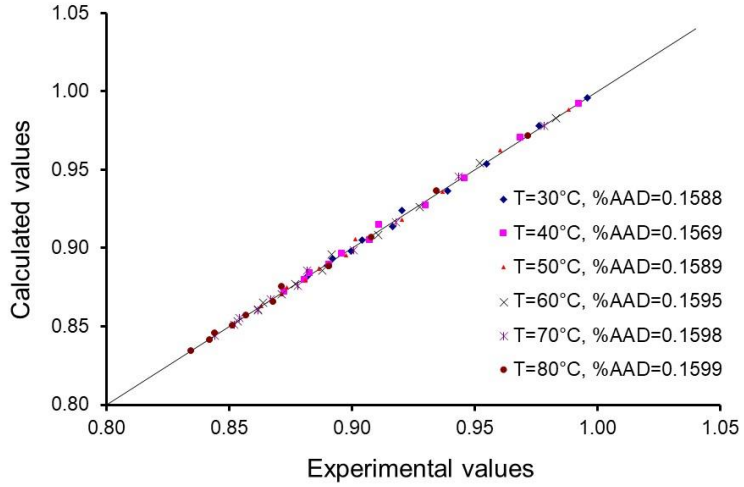

Figure 3: Comparison of measured values and calculated data of density

3.2 Viscosity measurement

The viscosity values of all samples at the varied DEAE concentration of 0.0 to 1.0 and the varied temperature of 30 to 80 °C are presented in the Figure 4.

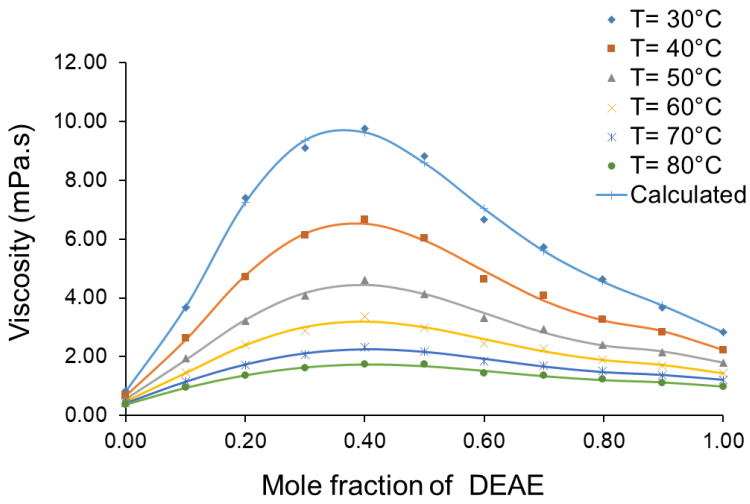

Figure 4: Viscosity values of DEAE + Distilled water at a variety of mole fraction at the temperature ranging from 30-80 °C

The viscosity tends to decrease when the temperature increases for all concentrations. However, the viscosity increases significantly with an increase in mole fraction up to the mole fraction of 0.4 which is the peak of

viscosity for all temperatures. After that, the viscosity of DEAE mixture will decrease with further increasing mole fraction. From this phenomenon, only a specific mole fraction can be reached the maximum viscosity. This is because the hydrogen bonds are formed and has the interaction between amine and water molecules. The mole fraction providing the maximum viscosity is the mole fraction that has the maximum attraction between both components. The Redlich-Kister equation for viscosity also provides the lowest %AADs compared with others and the calculated data also have a good agreement with experimental data as demonstrated in Figure 5. The %AADs at various temperature of the samples are presented in Table 2. Then, the comparison of calculated data and experimental results based on the effect of differential mole fractions is illustrated in Figure 5.

Table 2: Redlich-Kister equation coefficients of viscosity deviation for DEAE+water at 30-80 °C

T (°C)	A_0	A_1	A_2	A_3	A_4	%AAD
30	27.0414	-32.6797	2.0657	34.102	-18.2513	1.9094
40	17.9995	-20.8789	-5.4006	22.3019	-1.1405	1.8914
50	11.8048	-13.0554	-5.3409	12.8612	2.6331	1.9236
60	8.1684	-8.4947	-2.4916	7.9953	0.7519	2.3947
70	5.3776	-5.1552	-2.0638	3.6717	2.1233	1.7739
80	3.9939	-3.618	-1.2362	2.355	1.7882	1.7548

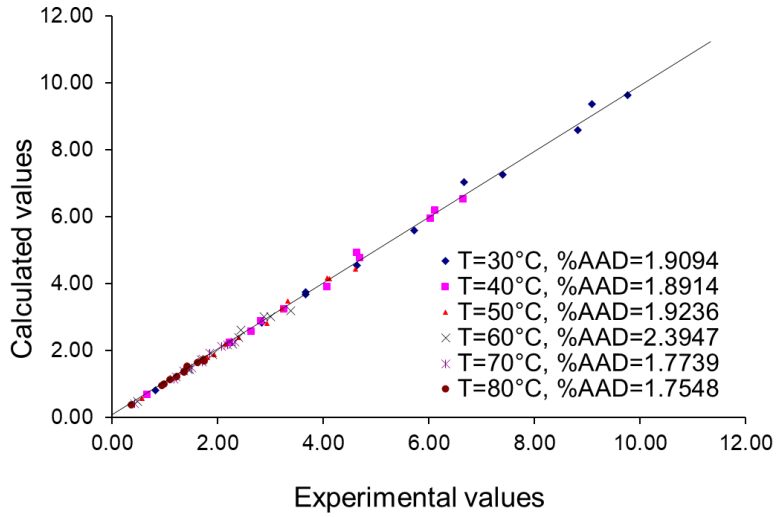

Figure 5: Comparison of measured values and calculated data of viscosity

3.3 Correlation comparison

Although the various equations, the Redlich-Kister equation, the Wilson model, the Grunberg and Nissan equation and Chauhan polynomial equation, are developed for binary mixture to calculate and predict the physical properties data such as density and viscosity values, the calculated data from The Redlich-Kister equation is the most suitable equation to use because this offers the lowest percent absolute deviation between measured data and experimental data. The difference of %AADs of each model are presented in Table 3 as shown below. Properties such as density and viscosity at several temperatures both for pure chemicals and their binary liquid mixtures over the whole concentration range are useful for understanding of the thermodynamic and transport properties associated with heat and fluid flow. From the table, the Grunberg-Nissan equation and empirical polynomial correlation have higher average absolute deviation because they are concerned about the temperature in the non-linear equation with power 3 for the Grunberg-Nissan equation and power 2 for empirical polynomial correlation.

Table 3: Comparison of percentage of average absolute deviation between various equations

Properties	Redlich-Kister	Wilson	Grunberg-Nissan	Polynomial
Density (g/cm^3)	0.1590	0.3381	0.4451	0.3925
Viscosity (mPa•s)	1.9413	5.5255	27.3262	20.6635

4. Conclusion

The measurement of physical and transport properties such as density and viscosity of 2(Diethylamino)ethanol or DEAE is for carbon dioxide absorption process which is one of the technologies for carbon capture, storage and utilization process. The sample is measured at the varied mole fraction from 0.0 to 1.0 and the temperature from 30 to 80 °C and to investigate the effects of these parameters on density and viscosity. The results present that the density of aqueous solution of DEAE tends to decrease with an increase in temperature in all different concentrations. Nevertheless, the results of viscosity are quite different. The viscosity increases as mole fraction increases up to 0.4 of mole fraction and decreases with mole fraction. This is because of the hydrogen bonds between the molecules. For temperature, the viscosity decreases as temperature increases. For the correlations, among these 4 equation models, the Redlich-Kister model is the most suitable model for binary mixture with the lowest percentage of average absolute deviation between experimental data and calculated values. This model provides the %AADs for the density and the viscosity of aqueous solution of DEAE at 0.1590 % and 1.9413 %. This work can be applied to predict these properties and other basic parameters such as diffusivity and the flow of fluids and to design the equipment of absorption.

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