Densities of n-tris(hydroxymethyl)methyl-3-aminopropanesulfonic acid (TAPS) + glycol (DEG / TEG / T$_4$EG) + water

Medarlo B. De Jesus$^2$, Allan N. Soriano$^3$, Meng-Hui Li$^1$, and Adonis P. Adornado$^{245}$

1 R & D Center for Membrane Technology and Department of Chemical Engineering, Chung Yuan Christian University, Chung-Li City, Taoyuan, Taiwan, R.O.C
2 School of Chemical, Biological, and Materials Engineering and Sciences, Mapúa University, Manila 1002, Philippines
3 Department of Chemical Engineering, Gokongwei College of Engineering, De La Salle University, 2401 Taft Avenue, Manila 1004, Philippines
4 General Education Department, Colegio de Muntinlupa, Mayor J. Posadas Avenue, Sucat 1770, Muntinlupa City, Metro Manila, Philippines
E-mail: apadornado@mapua.edu.ph / adonisadornado@yahoo.com

Abstract. In this present work, density of ternary system containing n-tris(hydroxymethyl)methyl-3-aminopropanesulfonic acid (TAPS), glycol, and water was presented. The considered glycols are diethylene glycol (DEG), triethylene glycol (TEG), and tetraethylene glycol (T$_4$EG). The measurements were done over a temperature range of 298.15 to 343.15 K at normal atmospheric pressure. Different concentrations (4 to 16 % by weight TAPS or 56 to 44 % water, in a fixed amount of glycol – 40 %) were used. The effects of temperature and compositions on the measured properties were discussed and then correlated based on the equation proposed for aqueous salt-glycol systems. Calculation results show that the applied model was satisfactory in representing the measured property of the considered ternary aqueous solutions containing TAPS and glycol.

1. Introduction
Various alternative solvent systems, such as glycols and carbonate solution have been demonstrated to absorb acid gases, such as CO$_2$ and H$_2$S [1-2]. In recent years, there is a considerable interest to enhance the rate of absorption of CO$_2$ in carbonate solution, thus, some have studied the used of amino acid systems such as amino acid salts and aqueous amino acid salts in enhancing the absorption rate of CO$_2$ in carbonate solution and the results are promising [3-6]. A good example of an amino acid for possible application in CO$_2$ absorption is n-[tris(hydroxymethyl)methyl-3-aminopropanesulfonic acid (TAPS). It is an amino acid that is a derivative of taurine and considered as a biological buffer. The advantage of using TAPS as an absorbent system lies with the zwitterionic species of TAPS to associate with CO$_2$ as predicted by previous work [7]. It is also predicted that CO$_2$ (which usually forms the weak acid H$_2$CO$_3$ in aqueous environment on the basis of the predicted interaction between a weak acid (i.e., HCOOH) and a zwitterion) may be capable of association with the zwitterionic species of TAPS via strong OH–O hydrogen bonding interaction despite the weak acid not being capable of deprotonating the zwitterionic species [7].
A solvent system consisting of glycol together with an aqueous solution of TAPS is a promising absorbent for CO₂ capture via absorption since the added glycol is expected to decrease the vapor pressure of the resulting solvent system and thus, could make the separation of the absorbed gas during solvent recovery easier and eventually the recovery of high-purity gas [1]. Other criteria in selecting the appropriate absorbent have sprung over issues like energy requirement, solvent regeneration and solvent corrosive property.

To exploit fully the favorable properties of this promising solvent system for use in absorption applications, thorough understanding of thermophysical properties, such as refractive index, density, viscosity, heat capacity, electrical conductivity, etc., is very necessary. However, to our knowledge very few have done a thorough thermophysical characterization [8-11], especially of a ternary solvent system consisting of TAPS, glycol, and water. Therefore, in the light of the importance of this solvent system in absorption process as potential absorbent, this study measured and presented new data of density of aqueous solution containing glycol and TAPS for temperatures up to 343.15 K at normal atmospheric pressure. The concentration of glycol was fixed to 40 % (in mass percentage) and the TAPS (4 to 16 %) and water (56 to 44 %) were varied. The considered glycols were diethylene glycol (DEG), triethylene glycol (TEG), and tetraethylene glycol (T₄EG). The temperature and compositional dependency of the considered properties are presented and satisfactorily correlated using a modified form of the equation proposed by Söhnel and Novotný [12], which was empirically designed for aqueous salt-glycol systems.

2. Materials and methods

2.1. Chemicals

Glycols, DEG and TEG, used in this work were supplied by Tedia Co. and both T₄EG and TAPS were supplied by ACROS Organics. All major chemicals used, i.e., glycols and TAPS, all have a minimum purity of 99% (by mass). The water used to prepare all aqueous solutions was Type I reagent-grade water with a resistivity of 18.3 MΩ·cm and total organic carbon of less than 15 ppb. The water was purified using our in-house water purification system (Barnstead Thermodyne, Easy Pure 1052). All weight measurements during the preparation of solutions were performed on a digital balance from Mettler-Toledo (model AL204) having an accuracy of ±1 × 10⁻⁴ g.

2.2. Density measurements

“The densities of the considered solutions were measured using an integrated density measuring cell, which used the principle of oscillating U-tube and is supplied by Anton Paar Stabinger (model SVM 3000). The repeatability of the density measurement was ±0.0002 g·cm⁻³. The estimated uncertainties for temperature and density were ±0.002 K and ±0.0004 g·cm⁻³, respectively. The density meter used the principle of oscillating U-tube. The sample is introduced into a U-shaped tube that is electronically excited to oscillate at its characteristic frequency. The characteristic frequency changes depending on the density of the sample. Through precise determination of the characteristic frequency and an appropriate adjustment, the density of the sample is determined. But, due to the high temperature dependency of the density, the measuring cell has to be accurately thermostatted. For each concentration, 5 mL was placed in a syringe, and 1.5 mL of it was used per trial [13-14].”

3. Results and discussion

The results of the present measurements for the ternary systems containing the glycol DEG or TEG or T₄EG are presented in table 1. As presented in this table, at fixed composition (representatively shown in figure 1) the value of the density decreases as the temperature increases while at fixed temperature (representatively shown in figure 2), its value increases as the TAPS composition increases. With respect to the kind of glycol use, the measured density data showed an increasing trend in the order: DEG < TEG < T₄EG.

| Table 1. Densities of the studied ternary systems (TAPS + glycol + water). |
A. TAPS (1) + DEG (2) + Water (3): $x_1/x_2$  

| $T / K$ | $\rho / g \cdot cm^{-3}$ | $\rho / g \cdot cm^{-3}$ | $\rho / g \cdot cm^{-3}$ |
|--------|--------------------------|--------------------------|--------------------------|
| 298.15 | 1.0724                   | 1.0929                   | 1.1226                   |
| 303.15 | 1.0697                   | 1.0900                   | 1.1198                   |
| 308.15 | 1.0669                   | 1.0872                   | 1.1167                   |
| 313.15 | 1.0640                   | 1.0842                   | 1.1137                   |
| 318.15 | 1.0610                   | 1.0812                   | 1.1106                   |
| 323.15 | 1.0579                   | 1.0780                   | 1.1074                   |
| 328.15 | 1.0548                   | 1.0748                   | 1.1042                   |
| 333.15 | 1.0515                   | 1.0716                   | 1.1009                   |
| 338.15 | 1.0482                   | 1.0682                   | 1.0975                   |
| 343.15 | 1.0447                   | 1.0648                   | 1.0941                   |

B. TAPS (1) + TEG (2) + Water (3): $x_1/x_2$  

| $T / K$ | $\rho / g \cdot cm^{-3}$ | $\rho / g \cdot cm^{-3}$ | $\rho / g \cdot cm^{-3}$ |
|--------|--------------------------|--------------------------|--------------------------|
| 298.15 | 1.0758                   | 1.0963                   | 1.1266                   |
| 303.15 | 1.0730                   | 1.0934                   | 1.1235                   |
| 308.15 | 1.0701                   | 1.0904                   | 1.1204                   |
| 313.15 | 1.0671                   | 1.0874                   | 1.1173                   |
| 318.15 | 1.0640                   | 1.0842                   | 1.1141                   |
| 323.15 | 1.0608                   | 1.0810                   | 1.1108                   |
| 328.15 | 1.0575                   | 1.0777                   | 1.1075                   |
| 333.15 | 1.0542                   | 1.0744                   | 1.1041                   |
| 338.15 | 1.0507                   | 1.0709                   | 1.1006                   |
| 343.15 | 1.0472                   | 1.0674                   | 1.0971                   |

C. TAPS (1) + $T_4$EG (2) + Water (3): $x_1/x_2$  

| $T / K$ | $\rho / g \cdot cm^{-3}$ | $\rho / g \cdot cm^{-3}$ | $\rho / g \cdot cm^{-3}$ |
|--------|--------------------------|--------------------------|--------------------------|
| 298.15 | 1.0776                   | 1.0986                   | 1.1295                   |
| 303.15 | 1.0747                   | 1.0956                   | 1.1264                   |
| 308.15 | 1.0718                   | 1.0925                   | 1.1232                   |
| 313.15 | 1.0687                   | 1.0894                   | 1.1200                   |
| 318.15 | 1.0656                   | 1.0862                   | 1.1167                   |
| 323.15 | 1.0624                   | 1.0829                   | 1.1133                   |
| 328.15 | 1.0590                   | 1.0795                   | 1.1099                   |
| 333.15 | 1.0556                   | 1.0761                   | 1.1064                   |
| 338.15 | 1.0521                   | 1.0726                   | 1.1029                   |
| 343.15 | 1.0498                   | 1.0690                   | 1.0993                   |

For the general purpose of application, the temperature and compositional dependence of the measured properties were correlated based on the equation proposed by Söhnel and Novotný [12], which was originally used for estimating the density of aqueous glycol-salt system, which is written as

$$\rho / (g \cdot cm^{-3}) = \rho_a + A_1(w) + A_2(w)^2 + A_3(w)^3 (1)$$

where $w$ is the mass percentage of salt, $\rho_a$ the density of the salt-free glycol solution, and $A_i$ are temperature-dependent parameters which is assumed as

$$A_i = a_{i,1} + a_{i,2}(T/K) + a_{i,3}(T/K)^2 (2)$$

In the present work, the salt component was replaced with the glycol component, (DEG or TEG or $T_4$EG) and the mass percentage, $w$, was changed to mole fraction of the glycol, which is denoted by $x_1$. The salt was replaced by the glycol (instead of the TAPS) due to scarcity of data for binary system of
TAPS + water. Thus, the modified form of equation (1) was now written as

\[ Y = Y_{bm} + A_1(x_1) + A_2(x_1)^2 + A_3(x_1)^3 \]  

(3)

where \( Y \) is the measured ternary data containing TAPS + glycol + water, \( x_1 \) the mole fraction of glycol and \( Y_{bm} \) the measured data of the binary system containing glycol + water.

**Figure 1.** Plot of density data for (TAPS + TEG + water) systems at different temperatures (298.15 to 343.15 K) and fixed glycol concentration (40 wt %) [■, 4% TAPS / 40% TEG / 56% H\(_2\)O; ●, 9% TAPS / 40% TEG / 51% H\(_2\)O; ▲, 16% TAPS / 40% TEG / 44% H\(_2\)O; and lines, calculated using Eqn. (3)].

**Figure 2.** Plot of the density data for (TAPS + TEG + Water) system as a function of temperature, \( T \) and mole fraction of glycol, \( x_1 \): ■, \( T = 298.15 \) K; ●, \( T = 308.15 \) K; ▲, \( T = 318.15 \) K; ◇, \( T = 328.15 \) K; ●, \( T = 338.15 \) K; and lines, calculated using equation (3).

The results of the present calculation are presented in table 2. The overall AAD value for the measured density is 0.0049 %, which is satisfactory for the design calculations. This acceptable AAD values concluded that the measured property of the studied ternaries can be well correlated using equation (3). Here, AAD is evaluated as
\[
\text{AAD} = \frac{1}{n} \sum_{i=1}^{n} [\text{e}_{\text{cald}} - \text{e}_{\text{exp}}] 
\]

where \( n \) is the number of data points and \((\text{e}_{\text{cald}} \text{ and } \text{e}_{\text{exp}})\) are calculated and experimental values, respectively. The good agreements of the measured data with that of the calculated values were also graphically represented in figures 1 and 2 via the smoothed curves.

Table 2. Determined parameters of equation (3) for the density calculations.

| Parameters / Systems          | TAPS + DEG + Water | TAPS + TEG + Water | TAPS + T\(_4\)EG + Water |
|-----------------------------|--------------------|--------------------|--------------------------|
| \( a_{1,1} \)               | 8.3105             | 8.3741             | 51.222                   |
| \( a_{1,2} \)               | -3.0428 \times 10^2| -3.4215 \times 10^2| -3.0863 \times 10^4       |
| \( a_{1,3} \)               | 4.7421 \times 10^5 | 5.3653 \times 10^5 | 4.9051 \times 10^4       |
| \( a_{2,1} \)               | -307.0300          | -289.1000          | -5.465.2000              |
| \( A_1 \)                   | 1.3635             | 1.5137             | 34.660                   |
| \( a_{2,2} \)               | -2.0434 \times 10^3| -2.1965 \times 10^3| -5.5017 \times 10^2       |
| \( a_{2,3} \)               | 6.052.3000         | 4.469.5000         | 1.4571 \times 10^5       |
| \( A_2 \)                   | -26.3760           | -23.3550           | -927.48                  |
| \( a_{3,1} \)               | 3.9306 \times 10^2 | 3.1301 \times 10^2 | 1.4721                   |
| \( A_3 \)                   | 0.0040             | 0.0023             | 0.0084                   |

4. Conclusion
Density measurements were made for three ternary glycol systems namely TAPS + DEG + Water, TAPS + TEG + Water, and TAPS + T\(_4\)EG + Water. The measurements were made over a temperature range of (298.15 to 343.15) K at normal atmospheric pressure. The measurements were done at varying compositions of TAPS and water but fixed composition of glycol. The density data of the considered systems were found to generally decrease as the temperature increases at fixed composition and at fixed temperature, its value increases as the TAPS composition increases (water composition decreases). The temperature and compositional dependency of the measured property were successfully correlated using the model used by other researchers as shown by overall AAD value of 0.0049 %. Results of the present work could be used in the design of unit operations that would utilize data of density of the studied system.

5. References
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