Research Journal of Applied Sciences, Engineering and Technology 4(17): 3095-3101, 2012
ISSN: 2040-7467

Submitted: February 25, 2012 Accepted: March 12, 2012 Published: September 01, 2012

# Study on the Change of Refractive Index on Mixing, Excess Molar Volume and Viscosity Deviation for Aqueous Solution of Methanol, Ethanol, Ethylene Glycol, 1-Propanol and 1, 2, 3-Propantriol at T = 292.15 K and Atmospheric Pressure

[1]Fardad Koohyar, [2]Farhoush Kiani, [3]Sasan Sharifi, [4]Meysam Sharifirad and
[5]Seyed Hamed Rahmanpour
[1]Department of Chemistry, Faculty of Science, Islamic Azad University, Babol Branch, Babol, Iran
[2]Department of Chemistry, Faculty of Science, Ayatollah Amoli Branch, Islamic Azad University, Amol, Iran
[3]Department of Chemistry, Faculty of Science, Arak Branch, Islamic Azad University, Arak, Iran
[4]Department of Chemistry, Teacher Research Bojnord, Iran
[5]Faculty of Chemistry, Iran University Science and Technology, Iran

**Abstract:** For aqueous solutions of methanol, ethanol, ethylene glycol, 1-propanol and 1, 2, 3-propantriol the change of refractive indices on mixing, excess molar volumes and viscosity deviations were calculated from the experimental data at 292.15 K. These experimental data (refractive indices, densities and viscosities) were measured over the whole mole fractions range in atmospheric pressure and at T = 292.15 K. For these mixtures, excess thermodynamic properties have been correlated with the Redlich-Kister polynomial equation (and experimental equation) to derive the coefficients and standard errors.

**Keywords:** 1, 2, 3-propantriol, change of refractive indices on mixing, density, excess molar volumes, redlich-kister equation

## INTRODUCTION

For binary mixtures the results of the excess thermodynamic properties data (especially excess molar volumes and viscosities deviation) are discussed in terms of the mentioned intermolecular interactions (Langa *et al*., 2006).

Studies on the refractive index, viscosity and density of binary mixtures along with other thermodynamic properties are being increasingly used as tools for the investigation of the properties of pure components and the nature of intermolecular interactions between liquid mixture constituents (Yang *et al*., 2004). Also these studies find direct applications in chemical and biochemical industry (Kondaiah *et al*., 2011). Viscosity is an important property of a fluid in characterizing its flow through a channel, as well as heat and mass transfer in many food processing processes (Viet Bui and Nguyen, 2004).

Methanol, ethanol, ethylene glycol, 1-propanol and 1, 2, 3-propantriol (glycerol) are alcohols that are very soluble in water. One of the reasons for dissolving of these substrates in water is having OH group and production of hydrogen bonds between water molecules and this group. Hydrogen bonded systems are very interesting because hydrogen bonds play a vital role in chemical, physical and biological processes and water and glycerol have very hydrogen bonds (Zorebski and Lubowiecka, 2009).

The effects of aqueous methanol solutions applied as a foliar spray or via irrigation were investigated in Arabidopsis, tobacco and tomato plants. Methanol applied to roots leads to phototoxic damage in all three species tested (Ramirez *et al*., 2006).

Ethanol is widely used as a solvent both in the home and in industry (Pendlington *et al*., 2001; Lachenmeier, 2008). Consumers may be exposed to ethanol from its application as a constituent of many household and personal products, such as cosmetics, hairsprays, window cleaners, deicers and certain pharmaceutical preparations (Irvine, 2003). Most people have experienced skin contact with alcoholic solutions.

Ethanol or 1-propanol containing hand disinfectants are widely used as surgical hand antisepsis. The primary objective of this study was to investigate transferal absorption of ethanol and 1-propanol from combination of 45% ethanol and 18% 1-propanol with skin protecting ingredients within 1 h after application in comparison to the absorption of these alcohols from the product in the absence of the cosmetic additives. The secondary objective was to evaluate the dermal tolerability (Lang *et al*., 2010).

**Corresponding Author:** Fardad Koohyar, Department of Chemistry, Faculty of Science, Islamic Azad University, Babol Branch, Post code 47188-33348, Babol, Iran, Tel.: +98-111-2267680; Fax: +98-112-2267685

Poisoning with ethylene glycol or methanol can occur through attempted inebriation, unintentional ingestion, or intentional self-harm. In 2007, poison centers in the United States received reports of 5731 possible ethylene glycol exposures and 2283 possible methanol exposures. Because reporting of such exposures is not mandatory, these data undoubtedly underestimate the total number of cases (Brent, 2009).

The 1, 2, 3-propanetriol have tri OH groups and use in pharmacy, hygienic and cosmetic industries (Weast, 1975; Windholz., 1983).

In this work, we have calculated excess molar volumes ($V^E$), change of refractive indices on mixing ($\Delta n_D$) and viscosity deviations ($\Delta\eta$), also measured densities ( $\rho$), refractive indices ($n_D$) and dynamic viscosities ($\eta$) for aqueous solutions of methanol, ethanol, ethylene glycol, 1-propanol and 1, 2, 3-propantriol at T = 292.15 K and over entire mole fraction. By using this data we can observe the effect of increasing number of carbon and –OH on the thermodynamic properties of alcohols aqueous solutions.

## EXPERIMENTAL SECTION

**Materials:** Methanol, ethanol, ethylene glycol, 1-propanol and 1, 2, 3-propantriol were supplied from Merck Company. The purity of ethylene glycol and 1, 2, 3-propantriol were 98% and purity of methanol, ethanol and 1-propanol were 99%. They are used without further purification.

**Apparatus and procedure:** All solutions were prepared from double-distilled water and reagents by mass using a Scaltec SBA31 analytical balance, with readability of 0.1 mg. All of the experiments are repeated three times and average values are reported. A digital vibrating glass tube densimeter (DA-500E, China.) and Schott-Gerate AVS 350 automatic Ubbelohde viscosimeter were employed to measure density and kinematic viscosity, respectively. Densities were determined with the uncertainty of $\pm 1\times10^{-4}$ (g/cm$^3$). Four Ubbelhode tubs with different capillary size were used in the experiments according to the different viscosity values of the mixtures. For all measurements, a Schott-Gerate CT 1150 digital thermostat was used to control the temperature to ±0.01 K of the desired temperature. The uncertainty of the time measurement in the viscosimeter is ±0.01 s. Refractive indices for the sodium D-line ($n_D$) were measured with an Abbe reftactometer (CARL ZEISS, Model A, Germany). A minimum of three independent readings were taken for each composition. The uncertainties of the refractive index and viscosity are $\pm 1\times10^{-4}$ and ±0.003 mPa·s. Also the uncertainty of the temperature is ±0.01 K.

## RESULTS AND DISCUSSION

The change of refractive indices on mixing ($\Delta n_D$), excess molar volumes ($V^E$), viscosity deviations ($\Delta\eta$) also the experimental data on the densities ( $\rho$), refractive indices ($n_D$) and dynamic viscosities ($\eta$), for aqueous solutions of methanol, ethanol, ethylene glycol, 1-propanol and 1, 2, 3-propanetriol at T = 292.15 K and over entire mole fraction are listed in Table 1. For aqueous solution of ethylene glycol and 1, 2, 3-propantriol, by increasing of mole fractions of solute, the thermodynamic properties of these solutions increases.

The values of $V^E$ for aqueous solutions were calculated from the density data using the following equation (Li *et al*., 2007):

$$V^E = (x_1M_1 + x_2M_2)/\rho - (x_1M_1/\rho_1 + x_2M_2/\rho_2) \quad (1)$$

In this equation $x_1$, $x_2$ are mole fractions. $M_1$, $M_2$ are the molar masses. $\rho_1$, $\rho_2$ are the densities of pure components 1, 2, respectively.

The change of refractive index on mixing ($\Delta n_D$) for aqueous solutions was calculated from following equation (Ku *et al*., 2008):

$$\Delta n = n_D - (x_1n_{D1} + x_2n_{D2}) \quad (2)$$

In this equation $x_1$, $x_2$ are mole fractions. $n_D$, $n_{D1}$, $n_{D2}$ are the refractive index of mixture, pure components 1, 2 respectively.

The viscosity deviation ($\Delta\eta$) for binary mixture was calculated from the viscosity data using the following Equation (Aznarez *et al*., 2006):

$$\Delta\eta = \eta - (x_1\eta_1 + x_2\eta_2) \quad (3)$$

In this Equation $x_1$, $x_2$ are mole fractions. $\eta$, $\eta_1$ and $\eta_2$ are the viscosity of mixture, pure components 1, 2 respectively.

The values of $V^E$, $\Delta n_D$ and $\Delta\eta$ for each mixture were fitted to the Redlich-Kister polynomial equation (Redlich and Kister, 1948):

$$Y = x_1(1-x_1)\sum_{i=0}^{n} A_i(2x_1-1)^i \quad (4)$$

where, Y = $V^E$, $\Delta n_D$ and $\Delta\eta$. $A_i$ are adjustable parameters and $x_1$ is the fraction of component 1.

In each case, the optimum number of coefficients $A_i$ was determined from an examination of the variation of the standard deviation (Ribeiro *et al*., 2006):

$$\sigma(Y) = [\Sigma(Y_{cal} - Y_{exp})^2/(n-m)]^{(½)} \quad (5)$$

For all solutions of this study the values of these parameters, $A_i$, along with the standard errors, $\sigma$, at 292.15 K are listed in Table 2.

The experimental refractive index of aqueous solution ($n_D$) can be fitted by following experimental equation (Koohyar *et al*., 2011):

$$n_D = A_nC + n^*_D \quad (6)$$

Table 1: Densities $\rho$, viscosities $\eta$, refractive indices $n_D$, excess molar volumes $V^E$, change of refractive indices on mixing $\Delta n_D$ and viscosity deviations $\Delta\eta$, for binary mixtures at $T$ = 292.15 K as a function of mole fractions $x_1$

| $x_1$ | $\rho$ [g/cm$^3$] | $\eta$ [mPa.s] | $n_D$ | $V^E$[cm$^3$/moL] | $\Delta\eta$ [mPa.s] | $\Delta n_D$ |
|---|---|---|---|---|---|---|
| **Methanol (1) + water (2)** | | | | | | |
| 0 | 0.9987 | 1.0226 | 1.3330 | 0 | 0 | 0 |
| 0.0287 | 0.9892 | 1.1653 | 1.3340 | -0.0635 | 0.1551 | 0.0011 |
| 0.0588 | 0.9821 | 1.3317 | 1.3355 | -0.1741 | 0.3345 | 0.0027 |
| 0.0903 | 0.9742 | 1.4720 | 1.3365 | -0.2716 | 0.4884 | 0.0039 |
| 0.1232 | 0.9663 | 1.6070 | 1.3380 | -0.3701 | 0.6376 | 0.0055 |
| 0.1650 | 0.9575 | 1.7340 | 1.3395 | -0.5075 | 0.7826 | 0.0072 |
| 0.2092 | 0.9486 | 1.8166 | 1.3410 | -0.6461 | 0.8843 | 0.0088 |
| 0.2563 | 0.9379 | 1.8392 | 1.3425 | -0.7458 | 0.9272 | 0.0105 |
| 0.3064 | 0.9275 | 1.8188 | 1.3430 | -0.8538 | 0.9284 | 0.0112 |
| 0.3785 | 0.9111 | 1.7338 | 1.3435 | -0.9279 | 0.8745 | 0.0120 |
| 0.4785 | 0.8904 | 1.5537 | 1.3425 | -1.0002 | 0.7376 | 0.0114 |
| 0.5911 | 0.8664 | 1.3195 | 1.3405 | -0.9335 | 0.5520 | 0.0099 |
| 0.7192 | 0.8419 | 1.0726 | 1.3380 | -0.7887 | 0.3603 | 0.0079 |
| 0.8661 | 0.8146 | 0.8064 | 1.3340 | -0.4352 | 0.1575 | 0.0045 |
| 1 | 0.7918 | 0.5911 | 1.3290 | 0 | 0 | 0 |
| **Ethanol (1) + water (2)** | | | | | | |
| 0 | 0.9987 | 1.0226 | 1.3330 | 0 | 0 | 0 |
| 0.0201 | 0.9896 | 1.2281 | 1.3360 | -0.0732 | 0.2014 | 0.0024 |
| 0.0416 | 0.9822 | 1.5008 | 1.3395 | -0.1829 | 0.4697 | 0.0053 |
| 0.0645 | 0.9749 | 1.8260 | 1.3430 | -0.2991 | 0.7902 | 0.0082 |
| 0.0890 | 0.9684 | 2.1431 | 1.3470 | -0.4388 | 1.1022 | 0.0115 |
| 0.1207 | 0.9605 | 2.4781 | 1.3510 | -0.6144 | 1.4307 | 0.0146 |
| 0.1554 | 0.9506 | 2.7282 | 1.3545 | -0.7546 | 1.6737 | 0.0171 |
| 0.1933 | 0.9389 | 2.8270 | 1.3575 | -0.8540 | 1.7648 | 0.0190 |
| 0.2350 | 0.9271 | 2.8508 | 1.3600 | -0.9525 | 1.7797 | 0.0203 |
| 0.2975 | 0.9105 | 2.7870 | 1.3620 | -1.0579 | 1.7034 | 0.0205 |
| 0.3894 | 0.8875 | 2.4801 | 1.3640 | -1.1007 | 1.3776 | 0.0199 |
| 0.5013 | 0.8631 | 2.1491 | 1.3655 | -1.0445 | 1.0237 | 0.0182 |
| 0.6404 | 0.8385 | 1.8061 | 1.3660 | -0.8976 | 0.6522 | 0.0147 |
| 0.8180 | 0.8123 | 1.4735 | 1.3645 | -0.5263 | 0.2831 | 0.0082 |
| 1 | 0.7905 | 1.2277 | 1.3615 | 0 | 0 | 0 |
| **Ethylene Glycol (1) + water (2)** | | | | | | |
| 0 | 0.9987 | 1.0226 | 1.3330 | 0 | 0 | 0 |
| 0.0059 | 1.0005 | 1.0455 | 1.3350 | -0.0052 | -0.0928 | 0.0014 |
| 0.0151 | 1.0043 | 1.1258 | 1.3375 | -0.0068 | -0.1929 | 0.0030 |
| 0.0246 | 1.0084 | 1.2101 | 1.3405 | -0.0251 | -0.2948 | 0.0050 |
| 0.0381 | 1.0135 | 1.3500 | 1.3445 | -0.0419 | -0.4196 | 0.0077 |
| 0.0524 | 1.0186 | 1.5004 | 1.3485 | -0.0591 | -0.5496 | 0.0103 |
| 0.0676 | 1.0242 | 1.6592 | 1.3525 | -0.0867 | -0.6889 | 0.0127 |
| 0.1014 | 1.0351 | 2.0454 | 1.3605 | -0.1366 | -0.9654 | 0.0174 |
| 0.1403 | 1.0458 | 2.5371 | 1.3685 | -0.1849 | -1.2364 | 0.0215 |
| 0.1857 | 1.0566 | 3.1645 | 1.3770 | -0.2379 | -1.4992 | 0.0254 |
| 0.2392 | 1.0669 | 3.9795 | 1.3850 | -0.2826 | -1.7332 | 0.0281 |
| 0.2697 | 1.0718 | 4.4758 | 1.3890 | -0.2999 | -1.8349 | 0.0290 |
| 0.3033 | 1.0763 | 5.0231 | 1.3930 | -0.3063 | -1.9464 | 0.0297 |
| **1-Propanol (1) + water (2)** | | | | | | |
| 0 | 0.9987 | 1.0226 | 1.3330 | 0 | 0 | 0 |
| 0.0155 | 0.9898 | 1.2679 | 1.3375 | -0.0571 | 0.2258 | 0.0037 |
| 0.0322 | 0.9831 | 1.5788 | 1.3425 | -0.1601 | 0.5156 | 0.0078 |
| 0.0540 | 0.9751 | 1.9872 | 1.3475 | -0.2929 | 0.8964 | 0.0117 |
| 0.0865 | 0.9614 | 2.4333 | 1.3540 | -0.4154 | 1.3015 | 0.0165 |
| 0.1443 | 0.9372 | 2.9016 | 1.3615 | -0.5137 | 1.6969 | 0.0209 |
| 0.2167 | 0.9129 | 3.1705 | 1.3680 | -0.5946 | 1.8744 | 0.0236 |
| 0.3101 | 0.8884 | 3.1877 | 1.3735 | -0.6431 | 1.7737 | 0.0242 |
| 0.3891 | 0.8721 | 3.0826 | 1.3765 | -0.6540 | 1.5689 | 0.0231 |
| 0.4869 | 0.8560 | 2.9167 | 1.3795 | -0.6459 | 1.2795 | 0.0209 |
| 0.6114 | 0.8392 | 2.7274 | 1.3825 | -0.5567 | 0.9331 | 0.0174 |
| 0.6873 | 0.8308 | 2.5971 | 1.3835 | -0.4858 | 0.7070 | 0.0144 |
| 0.7751 | 0.8220 | 2.4734 | 1.3845 | -0.3609 | 0.4725 | 0.0108 |
| 0.8779 | 0.8132 | 2.3510 | 1.3850 | -0.1994 | 0.2203 | 0.0059 |
| 1 | 0.8045 | 2.2848 | 1.3855 | 0 | 0 | 0 |

Table 1: (Continue)

| $x_1$ | $\rho$ [g/cm$^3$] | $\eta$ [mPa.s] | $n_D$ | $V^E$[cm$^3$/moL] | $\Delta\eta$ [mPa.s] | $\Delta n_D$ |
|---|---|---|---|---|---|---|
| **1, 2, 3-Propantriol (1) + water (2)** | | | | | | |
| 0 | 0.9987 | 1.0226 | 1.3330 | 0 | 0 | 0 |
| 0.0213 | 1.0217 | 1.3174 | 1.3450 | -0.0318 | -35.2572 | 0.0090 |
| 0.0359 | 1.0363 | 1.5708 | 1.3520 | -0.0604 | -59.3728 | 0.0139 |
| 0.0466 | 1.0461 | 1.7769 | 1.3570 | -0.0774 | -77.0262 | 0.0174 |
| 0.0707 | 1.0665 | 2.3407 | 1.3675 | -0.1199 | -116.6879 | 0.0245 |
| 0.0991 | 1.0878 | 3.2093 | 1.3785 | -0.1732 | -163.2219 | 0.0314 |
| 0.1332 | 1.1090 | 4.6115 | 1.3895 | -0.2143 | -218.7363 | 0.0376 |
| 0.1748 | 1.1305 | 6.9124 | 1.4010 | -0.2521 | -285.8703 | 0.0432 |
| 0.2268 | 1.1532 | 11.1324 | 1.4130 | -0.3078 | -368.4440 | 0.0477 |
| 0.3824 | 1.1978 | 43.1006 | 1.4370 | -0.3577 | -596.1891 | 0.0497 |
| 0.4389 | 1.2088 | 64.7962 | 1.4430 | -0.3516 | -668.7981 | 0.0477 |
| 0.5892 | 1.2304 | 193.1452 | 1.4555 | -0.2941 | -778.316 | 0.0388 |
| 0.6922 | 1.2407 | 402.7561 | 1.4615 | -0.2194 | -753.6233 | 0.0302 |
| 0.8244 | 1.2511 | 804.9952 | 1.4675 | -0.1195 | -572.0404 | 0.0175 |
| 1 | 1.2619 | 1670.1310 | 1.4750 | 0 | 0 | 0 |

Table 2: Estimated parameters ($A_i$) of Redlich-Kister equation and standard deviation $\sigma$ for various functions of the binary mixtures at 292.15 K

| Property | $A_0$ | $A_1$ | $A_2$ | $A_3$ | $\sigma$ |
|---|---|---|---|---|---|
| **Methanol (1) + water (2)** | | | | | |
| $V^E$[cm$^3$/moL] | -4.0033 | -0.6543 | 0.6620 | 1.6851 | 0.01798 |
| $\Delta\eta$[mPa.s] | 2.8888 | 3.7513 | 1.3019 | -1.5078 | 0.04209 |
| $\Delta n_D$ | 0.0458 | 0.0253 | -0.0014 | -0.0334 | 0.00039 |
| **Ethanol (1) + water (2)** | | | | | |
| $V^E$[cm$^3$/moL] | -4.1981 | -3.2341 | -1.0079 | 4.4358 | 0.03653 |
| $\Delta\eta$ (mPa.s) | 3.99731 | 1.7686 | 7.3214 | -11.3113 | 0.06434 |
| $\Delta n_D$ | 0.0719 | 0.0754 | 0.0465 | -0.0599 | 0.00124 |
| **Ethylene Glycol (1) + water (2)** | | | | | |
| $V^E$[cm$^3$/moL] | -1.7358 | 3.0244 | -8.3669 | 6.2598 | 0.00155 |
| $\Delta\eta$[mPa.s] | 2.0175 | -55.8060 | 89.5950 | -49.0925 | 0.04552 |
| $\Delta n_D$ | 0.2396 | -0.6108 | 1.1467 | -0.5685 | 0.00015 |
| **1-Propanol (1) + water (2)** | | | | | |
| $V^E$[cm$^3$/moL] | -2.4512 | -0.5200 | -1.3297 | -2.3992 | 0.04568 |
| $\Delta\eta$[mPa.s] | 4.9671 | 6.1875 | 5.8932 | 3.6892 | 0.16163 |
| $\Delta n_D$ | 0.0384 | 0.0587 | 0.0811 | 0.0830 | 0.00129 |
| **1, 2, 3-Propantriol (1) + water (2)** | | | | | |
| $V^E$[cm$^3$/moL] | -1.3470 | -0.8942 | 0.0940 | 0.4070 | 0.00614 |
| $\Delta\eta$[mPa.s] | -2909.2300 | 1626.4500 | -130.7250 | -259.2000 | 13.14615 |
| $\Delta n_D$ | 0.1782 | 0.1019 | 0.0739 | 0.0877 | 0.00222 |

where, $c$ is the molar concentration of the aqueous solution, $n^*_D$ is the refractive index of solvent (water) and $A_n$ (L/moL) is an experimental parameter that can be named constant of refractive index for aqueous solution of solute.

The Relative Deviations (RD) between experimental and calculated values of $n^*_D$ was calculated according to following Equation (Li *et al*., 2010):

$$RD = \left| \frac{n^*_{D(R)} - n^*_D}{n^*_{D(R)}} \right| \quad (7)$$

where, $n^*_D$ is the calculated value of refractive index of water from Eq. (6) and $n^*_{D(R)}$ is the experimental value of refractive index of water (in literatures) at same temperature.

For mixtures of this study, the values of $A_n$, $n^*_D$ and relative deviations between the experimental values of $n^*_D$ (in literatures) and calculated values of $n^*_D$ Eq. (6) at 292.15 K are listed in Table 3.

Table 3: Values of constants $A_n$, $n^*_D$ and $R^2$ Eq. (6) and Relative Deviation (RD) for different aqueous solutions at 292.15 K

| Systems | $A_n$[L/moL] | $n^*_D$ | $R^2$ | $R.E$[for $n^*_D$](%) |
|---|---|---|---|---|
| Methanol + water | 0.0009 | 1.3328 | 0.9984 | 0.00015 |
| Ethanol + water | 0.0032 | 1.3331 | 0.9962 | 0.00008 |
| Ethylene Glycol + water | 0.0059 | 1.3330 | 0.9999 | 0 |
| 1-Propanol + water | 0.0046 | 1.3346 | 0.9919 | 0.00120 |
| 1, 2, 3-Propantriol + water | 0.0105 | 1.3334 | 0.9999 | 0.00030 |

The thermodynamic property values of these solutions can be affected by two factors. The first factor is the concentration (molar, molal and mole fraction) of solute in mixture. The second factor is the power of bonds between solvent and solute molecules due to the values of polarity (dipole-dipole interaction and the H-bonds) and electrical charge of solute molecules. The second factor is important in excess thermodynamic properties (Jeffery and Sanger, 1991).

Table 4: The mole fractions of different aqueous solutions ($x_1$) that the curve of excess thermodynamic properties (versus mole fraction) on these points is maximum or minimum at 292.15 K

| Methanol (1) + Water (2) | | | Ethanol (1) + Water (2) | | |
|---|---|---|---|---|---|
| $x_1$ ($V^E$ vs $x$) | $x_1$ ($\Delta\eta$ vs $x$) | $x_1$ ($\Delta n_D$ vs $x$) | $x_1$ ($V^E$ vs $x$) | $x_1$ ($\Delta\eta$ vs $x$) | $x_1$ ($\Delta n_D$ vs $x$) |
| 0.48 (Min) | 0.31 (Max) | 0.38 (Max) | 0.39 (Min) | 0.24 (Max) | 0.30 (Max) |
| 1-Propanol (1) + Water (2) | | | 1, 2, 3-Propantriol (1) + Water (2) | | |
| 0.39 (Min) | 0.22 (Max) | 0.31 (Max) | 0.38 (Min) | 0.59 (Min) | 0.38 (Max) |

As seen in Table 1, also Fig. 1 and 5, the maximum values of density, viscosity and refractive index were observed for aqueous solution of 1, 2, 3- propantriol. In this case the second factor is very effective. 1, 2, 3-propantriol molecule have three –OH and it can be reason of the stronger interaction (hydrogen bond) between solute and solvent molecules in aqueous solution of 1, 2, 3-propantriol than aqueous solution of methanol, ethanol, 1-propanol and ethylene glycol.

The excess thermodynamic properties of liquid mixtures are important to study of interaction between molecules in solution (Rathnam *et al*., 2008). The value of excess molar volume may be affected by three factors. The first factor is the specific forces between molecules, such as hydrogen bond, charge-transfer complex, breaking of hydrogen bonds (weaker hydrogen bonds) and complexes giving positive excess molar volumes and forming hydrogen bonds (stronger hydrogen bonds) and complexes bringing negative values of $V^E$. The second factor is the physical intermolecular forces, including electrostatic forces between charged particles and between permanent dipoles and so on, induction forces between a permanent dipole and an induced dipole and forces of attraction (dispersion forces) and repulsion between nonpolar molecules. Physical intermolecular forces are usually weak and the sign of $V^E$ values may be positive or negative, but the absolute values are small. The third factor is the structural characteristics of the component, arising from geometrical fitting of one component into the other's structure, due to the differences in shape and size of component and free volume (Al-Azzawl and Allos, 1992; Yang *et al*., 2003). The negative $V^E$ values for binary mixtures indicate a decrease in the overall volume of the mixture. Similar to the excess molar volumes, viscosity is related to the molecular interaction between the components of mixtures as well as of the size and shape of molecules. Positive values of $\Delta\eta$ are indicative of strong interactions whereas negative values indicate weaker interactions (Yang *et al*., 2004; Yang *et al*., 2006).

For aqueous solution of methanol, ethanol, 1-propanol and 1, 2, 3-propantriol, the excess thermodynamic properties have maximum or minimum values at 292.15 K. The points (mole fraction of solute) related to these maximum or minimum values are listed in Table 4.

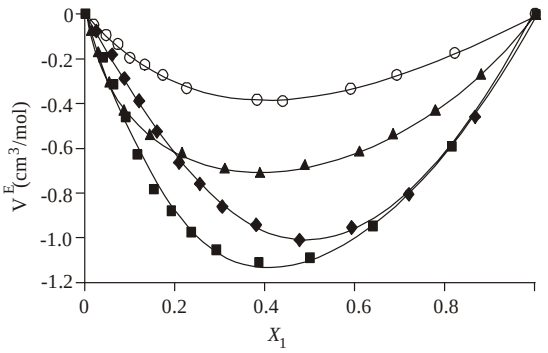


Fig. 1: Excess volume $V^E$ plotted against mole fraction $x_1$ for mixtures: (-■-), (ethanol (1) + water (2)), (-♦-), (methanol (1) + water (2)), (-▲-), (1-propanol (1) + water (2)), (-○-), (1, 2, 3-propantriol (1) + water (2)) at 292.15 K

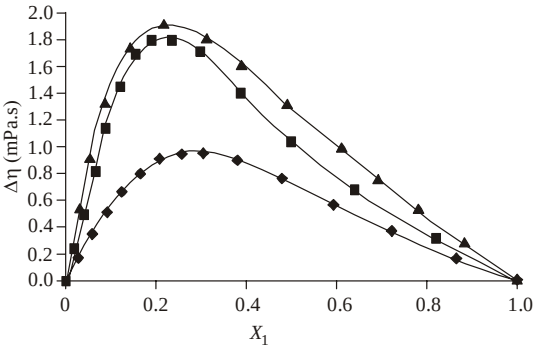


Fig. 2: Viscosity deviation $\Delta\eta$ plotted against mole fraction $x_1$ for mixtures: (-♦-), (methanol (1) + water (2)), (-■-), (ethanol (1) + water (2)), (-▲-), (1-propanol (1) + water (2)) at 292.15 K

It can be observed from the experimental data in Table 1 and Fig. 2, 3 and 4 that over the whole composition range for all solutions of this study at T = 292.15 K the values of $V^E$ and $\Delta n_D$ is negative and positive, responsively. Also for aqueous solution of methanol, ethanol and 1-propanol the values of $\Delta\eta$ is positive at the same temperature. In these cases the interaction between solute and solvent molecules in solution stronger than solute or solvent (pure components). We can mention two reasons for this case. The first reason is the less space (closer distance) between molecules in solution than solute or solvent (pure

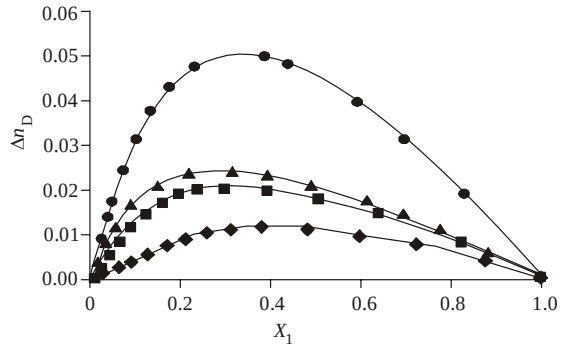


Fig. 3: Change of refractive index on mixing $\Delta n_D$ plotted against mole fraction $x_1$ for mixtures: (-■-), (ethanol (1) + water (2)); (-♦-), (methanol (1) + water (2)); (-▲-), (1-propanol (1) + water (2)); (-•-), (1, 2, 3-propantriol (1) + water (2)) at 292.15 K

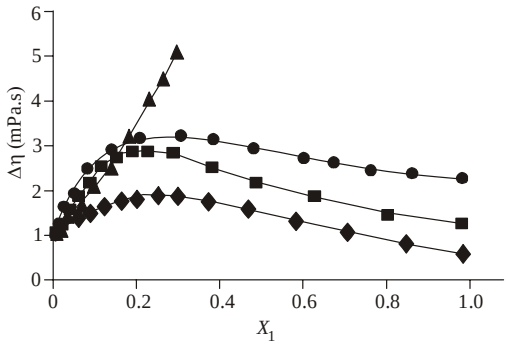


Fig. 4: Viscosities $\eta$ plotted against mole fraction $x_1$ for mixtures: (-■-), (ethanol (1) + water (2)); (-♦-), (methanol (1) + water (2)); (-▲-), (ethylene Glycol (1) + water (2)); (-•-), (1-propanol (1) + water (2)) at 292.15 K

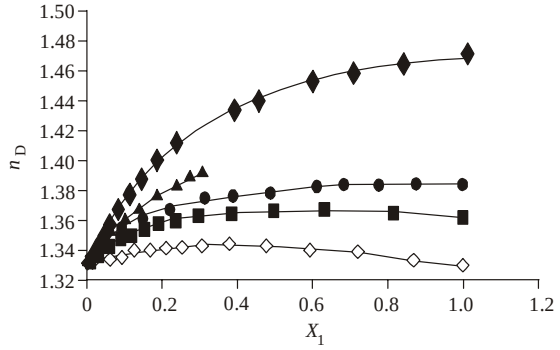


Fig. 5: Refractive indices $n_D$ plotted against mole fraction $x_1$ for mixtures: (-■-), (ethanol (1) + water (2)); (-◇-), (methanol (1) + water (2)); (-▲-), (ethylene Glycol (1) + water (2)); (-•-), (1-propanol (1) + water (2)); (-♦-), (1, 2, 3-propantriol (1) + water (2)) at 292.15 K

components) due to smaller size of solvent (water) molecules than solute molecules. The second reason is the electron donor effect of $-CH_3$, $-C_2H_5$, $-C_3H_7$, $-C_2H_4$ and $-C_3H_5$. In R–OH oxygen have partial negative electrical charge (-δ) and hydrogen have partial positive electrical charge (+δ) due to having more electronegative effect of oxygen than hydrogen. The absolute value of (-δ) for oxygen (in R-OH) increase as the electron donor effect of (–R) group (or number of carbon) increases.

As seen in Table 1, at 292.15 K and over the whole composition range for aqueous solutions of ethylene glycol and 1, 2, 3-propantriol the values of $\Delta\eta$ are negative. In this case, the negative deviations in viscosity support the main factor of breaking of the self-associated alcohols (through hydrogen bonding of solute molecules) and weak interactions between unlike molecules. Moreover, the hydrogen bond between molecules in ethylene glycol and 1,2,3-propantriol is stronger than water due to the number of –OH in ethylene glycol and 1, 2, 3-propantriol.

Also Fig. 2, 3 and 4 shows that for aqueous solution of methanol, ethanol, 1-propanol and 1, 2, 3-propantriol in the entire rang of mole factions at 292.15 K the values of excess thermodynamic property lie in the order of:

$$V^E (C_2H_5OH) < V^E (CH_3OH) < V^E (C_3H_7OH) < V^E (C_3H_8O_3)$$
$$\Delta\eta (CH_3OH) < \Delta\eta (C_2H_5OH) < \Delta\eta (C_3H_7OH)$$
$$\Delta n_D (CH_3OH) < \Delta n_D (C_2H_5OH) < \Delta n_D (C_3H_7OH) < \Delta n_D (C_3H_8O_3)$$

These equations show that the values of excess thermodynamic properties for solutions of this study may be affected by the main factor. This factor is the electron donor effect of agents $CH_3$-, $CH_3CH_2$-, $CH_3CH_2CH_2$- and $CH_2CHCH_2$- or size of solute molecules (R-OH) in mixture. (because electron donor effect value of agents is related to size of these agents). For these solutions the values of $\Delta n_D$ and $\Delta\eta$ increase by increasing of the electron donor effect of (–R) group (or size of –R).

## CONCLUSION

From the experimental viscosities, densities and refractive indices, the value of viscosity deviations, excess molar volumes and change of refractive indices on mixing were calculated at $T$ = 292.15 K. Experimental data of densities ($\rho$), refractive indices ($n_D$) and dynamic viscosities ($\eta$) for aqueous solutions of methanol, ethanol, ethylene glycol, 1-propanol and 1, 2, 3-propantriol at T = 292.15 K were measured over the entire mole fractions. It was found that for all solutions of this study at 292.15 K the values of $V^E$ and $\Delta n_D$ are negative and positive, respectively. Also the values of $\Delta\eta$ for aqueous solutions of methanol, ethanol and 1-propanol are positive and for aqueous solutions of ethylene glycol and 1, 2, 3-propantriol are negative. The Redlich-Kister polynomial

equation was applied successfully for the correlation of $\Delta n_D$, $\Delta\eta$ and $V^E$.

## REFERENCES


Al-Azzawl, S.F. and E.I. Allos, 1992. Density, viscosity and refractivity data of solution of potassium iolde in *N*-formylmorpholine-water at 298.15 K, 308.15 K and 318.15 K. J. Chem. Eng. Data., 37: 158-162

Aznarez, S., M.M.E.F. De Ruiz Holgado and E.L. Arancibia, 2006. Viscosities of mixtures of 2-alkanols with tetraethyleneglycol dimethyl ether at different temperatures. J. Mol. Liqs., 124: 78-83.

Brent, J., 2009. Fomepizole for ethylene glycol and methanol poisoning. New England J. Medicent., 360: 2216-2223.

Irvine, L.F., 2003. Relevance of the developmental toxicity of ethanol in the occupational setting: A review. J. Appl. Toxicol., 23: 289-299.

Jeffery, G.A. and W. Sanger, 1991. Hydrogen Bonding in Biological Structures. Springer-Verlg, Berlin.

Kondaiah, M., D. Sravana Kumar, K. Sreekanth and D. Krishna Rao, 2011. Ultrasonic velocities, densities and excess molar volumes of binary mixtures of N,N-dimethyl formamide with methyl acrylate, or ethyl acrylate, or butyl acrylate, or 2-ethyl hexyl acrylate at T = 308.15 K. J. Chem. Thermodyn., 43: 1844-1850.

Koohyar, F., A.A. Rostami, M.J. Chaichi and F. Kiani, 2011. Refractive indexes, viscosities and densities for L-cysteine hydrochloride monohydrate + D-sorbitol + water and glycerol + D-sorbitol + water in the temperature range between *T* = 303.15 K and *T* 323.15 K. J. Solution Chem., 40: 1361-1370.

Ku, H.C., C.C. Wang and C.H. Tu, 2008. Density, viscosity, refractive index and surface tension for binary and ternary mixtures of tetrahydofuran, 2-propanol and 2,2,4-trimethylpentane. J. Chem. Eng. Data., 53: 566-573.

Lachenmeier, D.W., 2008. Safety evaluation of topical applications of ethanol on the skin and inside the oral cavity. J. Occupational. Med. Toxicol., 42: 3-26.

Langa, L., A.M. Mainar, J.I. Pardo and J.S. Urieta, 2006. Excess enthalpy, density and speed of sound for the mixtures *â*-Pinene + 1-Butanol or 2-Butanol at (283.15, 298.15 and 313.15) K. J. Chem. Eng. Data., 51: 392-397.

Lang, R.A., D. Egli-Gany, F.H.H. Brill, J.G. Böttrich, M. Breuer, B. Breuer and M.H. Kirschner, 2010. Transdermal absorption of ethanol- and 1-propanol-containing hand disinfectants. *Langenbecks* Arch. *Surg*., 103: 734-747.

Li, Q.S., M.G. Su and S. Wang, 2007. Densities and excess molar volumes for binary glycerol +1-propanol, + 2-propanol, + 1,2-propanediol and + 1,3-propanediol mixtures at different temperatures. J. Chem. Eng. Data., 52: 1141-1145.

Li, H., X. Chen, F. Guo, L. Zhao, J. Zhu and Y. Zhang, 2010. Measurement and correlation of densities and viscosities of thiourea in triglycol + water at temperatures from (302.85 to 341.45) K. J. Chem. Eng. Data., 55: 1659-1662.

Pendlington, R.U., E. Whittle, J.A. Robinson and D. Howes, 2001. Fate of ethanol topically applied to skin. Food. Chem. Toxicol., 39: 169-174.

Ramirez, I., F. Dorta, V. Espinoza, E. Jimenez, A. Mercado and H. PenaCortes, 2006. Effects of foliar and root applications of methanol on the growth of arabidopsis, tobacco and tomato plants. J. Plant. Growth. Regul., 25: 30-44.

Rathnam, M.V., R.K.R. Singh and M.S. Kumar, 2008. Excess volumes of binary solutions of methyl formate, ethyl formate, propyl formate and benzyl acetate with bromo-, chloro- and nitrobenzenes at (303.15, 308.15 and 313.15). K. J. Chem. Eng. Data., 53: 265-270.

Redlich, O. and A.T. Kister, 1948. Algebraic representation of thermodynamic properties and the classification of solutions. Ind. Eng. Chem., 40: 345-348.

Ribeiro, A.F., E. Langa, A.M. Mainar, J.I. Pardo and J.S. Urieta, 2006. Excess molar enthalpy, density and speed of sound for the mixtures *â*-Pinene +1- or 2-Pentanol at (283.15, 298.15 and 313.15) K. J. Chem. Eng. Data., 51: 1846-1851.

Viet Bui, A. and M.H. Nguyen, 2004. Prediction of viscosity of glucose and calcium chloride solutions. J. Food. Eng., 62: 345-349.

Weast, R.C., 1975. CRC Handbook of Chemistry and Physics. 59th Edn., CRC Press Inc., New York.

Windholz, M., 1983. The Merck Index, An Encyclopedia of Chemical, Drugs and Biological. 10th Edn., Merck and Co Inc., Rahway.

Yang, C., H. Lai, Z. Liu and P. Ma, 2006. Density and viscosity of binary mixtures of diethyl carbonate with alcohols at (293.15 to 363.15) K and Predictive Results by UNIFAC-VISCO Group Contribution Method. J. Chem. Eng. Data., 51: 1345-1351.

Yang, C., P. Ma, F. Jing and D. Tang, 2003. Excess molar volume, viscosity and heat capacities for the mixtures of ethylene glycol-water from 273.15 K to353.15 K. J.Chem. Eng. Data., 48: 836-840.

Yang, C., W. Xu and P. Ma, 2004. Thermodynamic properties of binary mixtures of *p*-Xylene with cyclohexane, heptane, octane and *N*-methyl-2-pyrrolidone at several temperatures. J. Chem. Eng. Data., 49: 1794-1801.

Zorebski, E. and B. Lubowiecka, 2009. Thermodynamic and transport properties of (1, 2-ethanediol + 1-nonanol) at temperatures from (298.15 to 313.15) K. J. Chem. Thermodyn., 41: 197-204.