Volumetric Properties of Binary Mixtures of 2-Ethoxyethanol and 2-Butoxyethanol with 1,4-Dioxane

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Abstract

Densities, ρ, of 2-Ethoxyethanol (EGMEE) + 1,4-Dioxane (DXN) and 2-Butoxyethanol (EGMBE) + 1,4-Dioxane (DXN) systems have been measured in the whole range of composition at an interval of 5 K ranging from 303.15 to 323.15 K. Excess molar volumes, V_m^E, partial molar volumes, $\overline{V}_i$, thermal expansivities, α, and excess thermal expansivities, α^E, have been estimated from the experimental values of ρ. All of the derived properties have been fitted to appropriate polynomials. Values of V_m^E and α^E were fitted to the Redlich-Kister polynomial equation and their variations with composition and temperature have been discussed in terms of molecular interaction in the mixtures. Values of V_m^E and α^E were all positive and were attributed due to specific interactions.

Keywords: *Excess molar volumes; Partial molar volumes; Excess thermal expansivities; 2-Butoxyethanol; 2-Ethoxyethanol; 1,4-Dioxane*

1. Introduction

Being intermediate between two extremities viz. solid and gaseous state properties of liquids or liquid mixtures are mysterious. Although the physical properties of binary mixtures seem to be simple, they are, in fact, complex as they depend not merely on solute-solvent interactions, but also on their structural and geometric aspects. Volumetric properties data are used to investigate molecular interactions exerted by liquid components in various binary systems and are frequently used for industrial engineering designs and subsequent operations.

To extend our knowledge and contribute new data to the scientific community, we have been investigating continuously the volumetric, viscometric, refractometric, and acoustical properties of various organic liquids and their mixtures [1-12]. Among them, 1,4-dioxane is a colorless, non-polar liquid with a faintly sweet odor primarily used as a stabilizer in solvents, such as the dry cleaning chemical trichloromethane, and as a solvent in lacquers, varnishes, inks, adhesives, paints, plastics, dyes, oils waxes, and resins. On the other hand, alkoxyethanols are widely used as monomers in the production of polymers and emulsion formulations. They are also of considerable interest for studying the heteroproximity effects of the etheric oxygen on the –OH bond and hence their influence on the associated nature of the species in these molecules. More specifically, this work provides a test of empirical equations to correlate density.

Previously, a few researchers have studied the binary systems of a particular solvent with the series of alkoxyethanols, viz., S. Shufeng [13] measured density, viscosity, and excess properties of binary mixtures of 2-(methylamino)ethanol with 2-methoxyethanol, 2-ethoxyethanol, and 2-butoxyethanol from 293.15 to 353.15 K. M.N. Roy [14] measured densities and viscosities for binary mixtures of isoamylalcohol with 2-methoxyethanol, 2-ethoxyethanol and 2-butoxyethanol over the entire range of composition at 303.15 K, 313.15 K and 323.15 K and ultrasonic speeds and refractive indices at 303.15 K under atmospheric pressure. N. V. Sastry and S. R. Patel [15] measured the densities, viscosities, and sound speeds for six binary mixtures of methyl methacrylate (MMA) + 2-methoxyethanol (ME), + ethoxyethanol (EE), + 2-butoxyethanol (BE), + 1-butanol (1-BuOH), + 1-pentanol (1-PeOH), and + 1-heptanol (1-HtuOH) at 298.15 K and 308.15 K. Researchers have also studied on the binary systems of 1,4-dioxane with alcohols/ethane, viz., H. Ohji [16] have measured excess thermodynamic properties of (2-ethoxyethanol + 1,4-dioxane or 1,2-dimethoxyethane) at temperatures between (283.15 and 313.15) K. Contreas [17] determined densities and viscosities for 1,4-dioxane with 1-propanol and 2-propanol mixtures as a function of mole fraction at atmospheric pressure at (25, 30, 35, and 40) °C. Bedare [18] measured sound velocity (u), density (ρ), and viscosity (η) values at 308.15 K in the two binary systems of 1,4-dioxane with ethanol and methanol. A survey of the literature further indicates that not only reports on alkoxyethanols are rare but also any separate or comparative study on binary mixtures of 2-Ethoxyethanol + 1,4-Dioxane and 2-Butoxyethanol + 1,4-Dioxane systems is still not available. Therefore, all of these have prompted us to undertake a systematic study through the measurement of density (ρ), viscosity (η), refractive index (n_D), and ultrasonic velocity (u) at different temperatures on the systems: EGMEE+DXN and EGMBE+DXN. In the present communication, we are going to report volumetric properties for the systems EGMEE + DXN and EGMBE + DXN at different temperatures to gather detailed data on density which will enable us to understand the type of interactions of alkoxyethanols with non-polar liquid DXN and to find out appropriate functional characteristics of density concerning composition as well as temperature.

2. Experimental section

EGMEE (Merck, mass fraction > 0.99), EGMBE (Merck, mass fraction > 0.99) and DXN (Aldrich, mass fraction = 0.998) were used without further purification. The liquids were used after distillation. Solutions were prepared to take the middle portion of distillation. All the chemicals were preserved under molecular sieves for 2 to 3 weeks prior to their use. Their purity was verified by matching the measured density data with the literature as shown in Table 1.

Table 1. Densities, ρ / g.cm^{-3} of the pure liquids at different temperatures along with available data from different works of literature.

Temp. T / K	EGMEE		EGMBE		DXN	
	This Work	Lit.	This Work	Lit.	This Work	Lit.
298.15	0.9298	0.9250[13] 0.9254[15] 0.9253[19]	0.8966	0.8962[13] 0.8966[15]	1.0287	1.026870[20] 1.027882[17] 1.02812[21] 1.0283[22] 1.0279[23]
303.15	0.9253	0.9205[13] 0.9212[24]	0.8924	0.8920[13] 0.8920[14] 0.8923[24]	1.0231	1.0225[25] 1.02221[17] 1.0225[22] 1.0222[21]

						1.02244[26]
308.15	0.9207	0.9159[13] 0.9174[15]	0.8881	0.8878[13] 0.8889[15]	1.0174	1.016595[17] 1.01689[21]
313.15	0.9161	0.9113[13] 0.9120[14] 0.9123[24]	0.8838	0.8835[13] 0.8842[14] 0.8839[24] 0.8842[27]	1.0116	1.0121[25] 1.0110[17]
318.15	0.9114	0.9066[13]	0.8795	0.8792[13]	1.0059	1.005279[21]

A set comprising 21 compositions was prepared by mingling known masses of pure liquids. These were wholly miscible over the full composition range. An electronic balance (SAG 285, Mettler Toledo) was used to measure the mass with an accuracy of $\pm 10^{-7}$ kg. To evade any evaporation or pollution, solutions were always preserved in air-tight glass stopper bottles and controlled cautiously.

Using an automated vibrating-tube density meter (DSA 5000M, Anton Paar, Austria) density (ρ/kg·m^{-3}) was measured. The accuracy of the density meter was ± 0.01 kg·m^{-3}. As the values of ρ are extremely sensitive, the temperature was controlled up to ± 0.01 K by a built-in MKT thermometer. The manufacturer stated repeatability for density and temperature measurement were ± 0.001 kg·m^{-3} and ± 0.001 K, respectively. For all the pure components and mixtures, triplicate measurements were made and their mean was taken.

Excess molar volumes, V_m^E, have been calculated from measured densities using the following equation [28]:

$$V_m^E = \frac{\sum x_i M_i}{\rho} - \sum \frac{x_i M_i}{\rho_i} \tag{1}$$

Where M_i and ρ_i are the molar mass and density respectively of the i-th component and ρ is the density of the mixture at composition x_i.

Partial molar volumes $\bar{V}_i$ of both components 1 and 2 have been calculated Maham et al [29] for the systems EGMEE + DXN and EGMBE + DXN at different temperatures by Eqns. 2-3.

$$\bar{V}_1 = V_1^0 + x_2^2 \sum_{i=0}^n A_i (1 - 2x_2)^i + 2x_2^2 (1 - x_2) \sum_{i=0}^n i A_i (1 - 2x_2)^{i-1} \tag{2}$$

$$\bar{V}_2 = V_2^0 + (1 - x_2)^2 \sum_{i=0}^n A_i (1 - 2x_2)^i - 2x_2 (1 - x_2)^2 \sum_{i=0}^n i A_i (1 - 2x_2)^{i-1} \tag{3}$$

By definition, the average isobaric thermal expansivity, α of any liquid can be expressed as [28]

$$\alpha = -\frac{d \ln \rho}{dT} \tag{4}$$

So, the slope of the plot of ln ρ vs. T yields α. Excess thermal expansivity, α^E of any mixture is then calculated by the following equation [28],

$$\alpha^E = \alpha - (x_1 \alpha_1 + x_2 \alpha_2) \tag{5}$$

Here, α_1 and α_2 are thermal expansivities of pure components 1 and 2, respectively, and α is that of the mixture.

The values of ρ and α were correlated with concentration-dependent polynomials. To correlate measured and some derived properties following general polynomial equations were used:

$$Y = \sum_{i=0}^{n} a_i x_2^i \tag{6}$$

Here, Y represents ρ and α where a_i is the fitting coefficient and x_2 is the mole fraction of DXN.

Excess properties Y^E (V_m^E and α^E) of the mixtures were fitted by the nonlinear least-squares method to a Redlich–Kister polynomial [30].

$$Y^E = (1 - x_2)x_2 \sum_{i=0}^{n} A_i (2x_2 - 1)^i \tag{7}$$

Here, A_i is the fitting coefficient and n is the degree of polynomial expansion.

The standard deviations, $\sigma(Y^E)$, of all excess properties are calculated as:

$$\sigma(Y^E) = \left[\left(Y_{exp}^E - Y_{cal}^E\right)^2 \middle/ (n - p - 2) \right]^{1/2} \tag{8}$$

where n represents the number of experimental data points and p is the number of coefficients.

3. Results and discussion

Density (ρ) for the two binary systems: EGMEE + DXN and EGMBE + DXN was measured between 298.15 K and 318.15 K at an interval of 5 K in the entire range of composition, $0 \leq x_2 \leq 1$, where, x_2 represents the mole fraction of DXN. Table 1 shows the comparison of measured density (ρ) of the pure liquids with several reported data.

All the results ρ are listed in Table 2. Also, the values of ρ as a function of mole fraction of DXN (x_2) are graphically represented by Figure 1(a, b) for the systems EGMEE + DXN and EGMBE + DXN respectively. Fig. 1(c) compares the variation in ρ for the two systems against x_2 at 298.15 K and 318.15 K. In all cases, density is found to follow a five-degree (i.e., n =5) polynomial equation 6. All the coefficients and relevant R^2 values for the two systems at different temperatures were computed and summarized as shown in Table 5.

Table 2: Densities, ρ / kg.m^{-3} and excess molar volumes, $V_m^E.10^6$ / m^3.mol^{-1} of EGMEE + DXN and EGMBE + DXN Systems for different molar ratios at different temperatures.

x_2	ρ	V_m^E	ρ	V_m^E	ρ	V_m^E	ρ	V_m^E	ρ	V_m^E
	298.15 K		303.15 K		308.15 K		313.15 K		318.15 K	
				EGMEE + DXN						
0.0000	929.8	0.0000	925.3	0.0000	920.7	0.0000	916.1	0.0000	911.4	0.0000
0.0496	933.8	0.0372	929.1	0.0536	924.4	0.0598	919.9	0.0444	915.2	0.0404
0.1001	937.8	0.0870	933.4	0.0679	928.7	0.0692	923.9	0.0802	919.1	0.0826
0.1515	942.4	0.0887	937.7	0.0954	933.0	0.0920	928.1	0.1080	923.4	0.0953
0.2001	946.4	0.1305	941.8	0.1227	937.0	0.1249	932.2	0.1254	927.3	0.1295
0.2498	950.9	0.1371	946.0	0.1545	941.2	0.1521	936.4	0.1474	931.5	0.1470
0.3007	955.1	0.1893	950.2	0.2020	945.3	0.2050	940.6	0.1851	935.5	0.2008
0.3506	959.7	0.1980	954.8	0.2057	949.9	0.2036	944.9	0.2085	940.0	0.1993
0.4008	964.2	0.2244	959.4	0.2175	954.4	0.2203	949.4	0.2196	944.4	0.2159
0.4503	968.7	0.2494	963.8	0.2474	958.7	0.2551	953.7	0.2492	948.7	0.2411
0.5005	973.3	0.2763	968.6	0.2505	963.5	0.2529	958.4	0.2511	953.3	0.2481
0.5505	978.2	0.2786	973.4	0.2573	968.3	0.2546	963.2	0.2471	957.9	0.2587
0.6004	983.0	0.2947	978.1	0.2780	972.8	0.2891	967.8	0.2668	962.5	0.2736
0.6503	988.1	0.2888	983.1	0.2762	977.7	0.2912	972.6	0.2728	967.3	0.2748
0.6995	993.2	0.2823	988.1	0.2737	982.6	0.2925	977.5	0.2686	972.2	0.2656
0.7500	998.6	0.2669	993.5	0.2529	988.1	0.2568	982.7	0.2544	977.2	0.2649
0.8000	1003.9	0.2619	998.8	0.2428	993.5	0.2322	988.0	0.2326	982.3	0.2562
0.8500	1009.6	0.2282	1004.1	0.2387	999.0	0.2051	993.3	0.2171	987.7	0.2262
0.9000	1015.4	0.1926	1009.8	0.2059	1004.4	0.1931	998.8	0.1901	993.2	0.1937
0.9499	1021.4	0.1452	1016.0	0.1361	1010.3	0.1432	1004.3	0.1686	999.0	0.1403
1.0000	1028.7	0.0000	1023.1	0.0000	1017.4	0.0000	1011.6	0.0000	1005.9	0.0000
				EGMBE + DXN						
0.0000	896.6	0.0000	892.4	0.0000	888.1	0.0000	883.8	0.0000	879.5	0.0000
0.0506	900.9	0.0176	896.6	0.0259	892.2	0.0344	888.0	0.0130	883.6	0.0216
0.1006	905.1	0.0635	900.8	0.0657	896.5	0.0536	892.0	0.0694	887.7	0.0571
0.1515	909.6	0.1008	905.2	0.1106	900.8	0.1066	896.4	0.1011	892.0	0.0969
0.1993	914.1	0.1198	909.7	0.1232	905.1	0.1404	900.7	0.1286	896.3	0.1182

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0.2504	918.8	0.1783	914.3	0.1886	909.9	0.1724	905.4	0.1671	900.8	0.1775
0.2993	923.5	0.2302	919.1	0.2212	914.6	0.2120	910.0	0.2132	905.4	0.2169
0.3504	928.5	0.2974	924.0	0.2947	919.4	0.2920	914.8	0.2862	910.1	0.2962
0.3998	933.8	0.3288	929.1	0.3441	924.5	0.3349	919.9	0.3220	915.1	0.3376
0.4495	939.1	0.3898	934.4	0.3982	929.7	0.3946	925.0	0.3870	920.3	0.3831
0.4995	944.7	0.4455	940.0	0.4468	935.3	0.4364	930.5	0.4333	925.8	0.4224
0.5495	950.7	0.4828	945.7	0.5110	941.0	0.4939	936.2	0.4833	931.4	0.4771
0.6008	956.9	0.5457	951.9	0.5660	947.1	0.5531	942.2	0.5461	937.3	0.5441
0.6500	963.4	0.5751	958.6	0.5659	953.7	0.5565	948.7	0.5523	943.8	0.5424
0.6997	970.7	0.5595	965.7	0.5634	960.6	0.5673	955.6	0.5545	950.5	0.5583
0.7499	978.5	0.5363	973.4	0.5416	968.3	0.5368	963.2	0.5255	958.1	0.5204
0.7999	986.9	0.4888	981.7	0.4948	976.5	0.4912	971.3	0.4805	966.1	0.4765
0.8498	995.9	0.4233	990.6	0.4294	985.3	0.4261	980.0	0.4153	974.7	0.4116
0.9001	1005.4	0.3618	1000.0	0.3673	994.9	0.3360	989.2	0.3521	983.8	0.3481
0.9500	1016.7	0.1819	1011.2	0.1850	1005.7	0.1792	999.9	0.1920	994.6	0.1680
1.0000	1028.7	0.0000	1023.1	0.0000	1017.4	0.0000	1011.6	0.0000	1005.9	0.0000
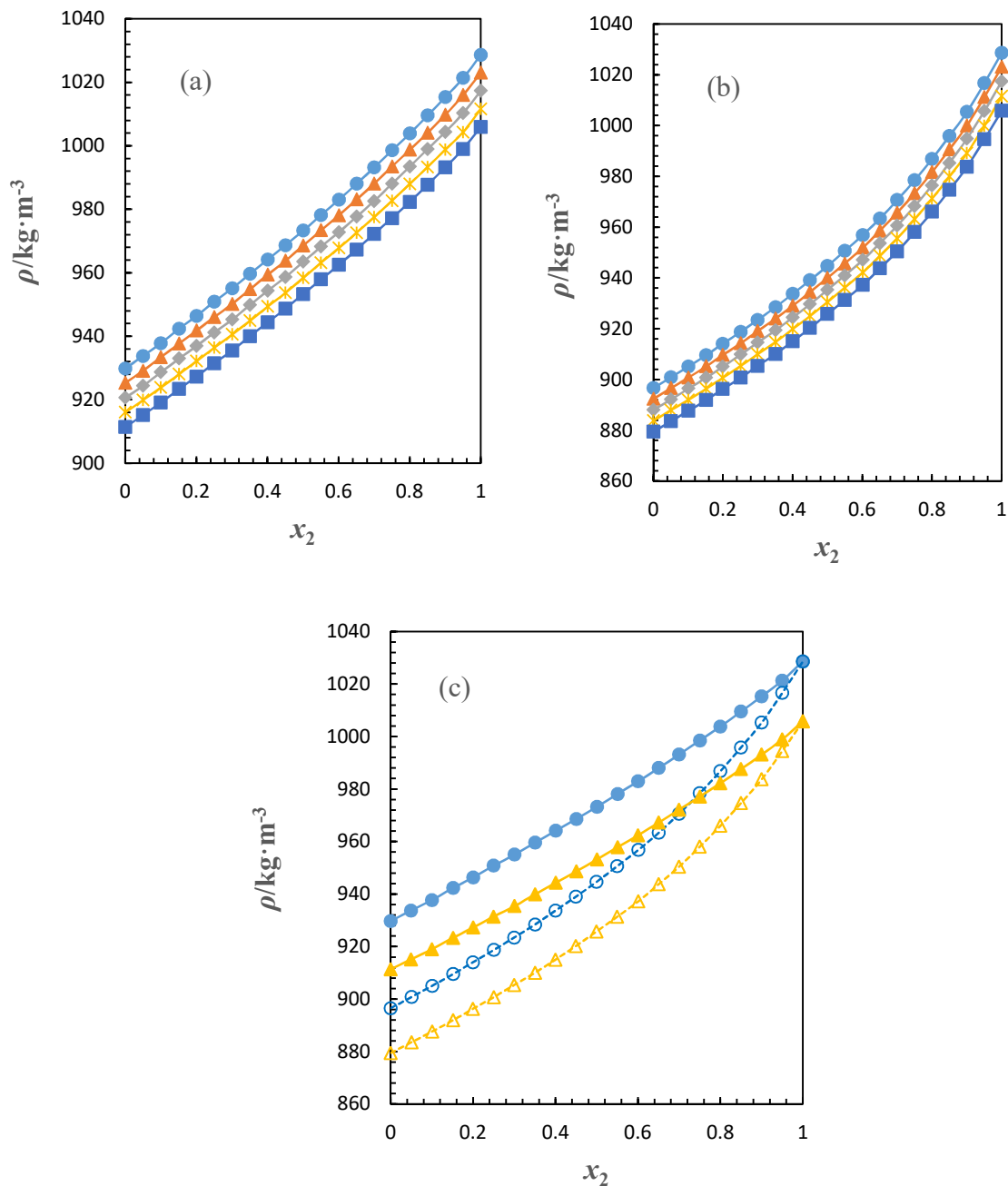
Figure 1. Densities, ρ, against mole fraction, x_2, for (a) EGMEE (x_1) + DXN (x_2) and (b) EGMBE (x_1) + DXN (x_2) systems at: ●, 298.15 K; ▲, 303.15 K; ◆, 308.15 K; ∗, 313.15 K; ■, 318.15 K; and (c) comparative curve for EGMEE (x_1) + DXN (x_2) (●) and EGMBE (x_1) + DXN (x_2) (○) systems at 298.15 K and for EGMEE (x_1) + DXN (x_2) (▲) and EGMBE (x_1) + DXN (x_2) (△) systems at 318.15 K. The solid and dashed lines represent the values calculated from the respective polynomial equation.

Excess molar volume (V_m^E) for both systems at different compositions was calculated at five different temperatures by using Eq. (1). These values are also represented in Table 2. Figs. 2 (a) & (b) show the variation of V_m^E concerning x_2 for EGMEE + DXN and EGMBE + DXN systems respectively and Fig. 2(c) compares the V_m^E of the two systems against the mole fraction x_2 at 298.15 K and 318.15 K.

At a particular temperature, the concentration dependence of V_m^E of each of the systems is fitted to the Redlich-Kister Equation 7. The coefficients of Equation 7 and the standard deviation (σ) [Equation 8] are listed in Table 6. The following are the observations regarding ρ and V_m^E from Figs. 1 and 2:

a) At a particular temperature, ρ of the pure liquids decrease in the order:
DXN > EGMEE > EGMBE

b) The ρ vs. x_2 curves for both systems follow a similar trend: ρ increasing almost linearly with the concentration of DXN.

c) The increment of ρ with respect to x_2 follows the order: EGMEE + DXN > EGMBE + DXN.

d) Effect of temperature on ρ for pure liquids as well as their mixtures is quite large and as usual the more the temperature, the less the density.

e) In the whole range of composition, V_m^E is found to be positive for both EGMEE + DXN and EGMBE + DXN systems.

f) For both EGMEE + DXN and EGMBE + DXN systems, values of V_m^E gradually increase and form a maximum in the DXN-rich region nearly at $x_2 = 0.65$ at stated temperatures.

g) Values of positive V_m^E are greater for the EGMBE + DXN system than that of the EGMEE + DXN system.

h) Variation of V_m^E with the temperature at different molar ratios is very small and irregular for both systems. Hence, d V_m^E /dT is insignificant.

Generally, it is accepted that excess properties like excess molar volume (V_m^E) of binary liquid systems are the results of positive and negative contributions due to specific effects or interactions. The sign of V_m^E of the solution depends upon the relative magnitude of expansion and contraction in the mixing of two liquids. When the factors causing expansion outweigh the factors causing contraction, the values of V_m^E become positive, but when the contraction factors are dominant over the expansive factors, V_m^E becomes negative.

Some of the important factors that are thought to be responsible for expansion in volume upon mixing leading to a positive contribution towards V_m^E are:

a) Dispersive forces occur in systems involving the non-polar + non-polar or self-associated (polar) + non-polar components. Here, molecular associations either through physical forces (e.g., dipole-dipole interaction) or chemical forces (e.g., H-bonding in alkanols) are destroyed either partially or fully [31,32].

b) Unfavorable geometric fitting due to size and shape differences between solute and solvent molecules.

c) Steric hindrance due to branching in chains or due to the presence of bulky substituents.

d) Electrostatic repulsion opposes the closer approach of component molecules with opposite poles.

Similarly, factors that influence the mixture volumes to be contracted [33], i.e., contributing negatively towards V_m^E are:

a) Specific chemical interactions between component molecules through H- bonding, π-π or π-n interactions, most of which lead to forming hetero molecular complexes of different stoichiometry.

b) Physical forces such as dipole-dipole, dipole-induced dipole, or even Van der Waals forces between components.

c) Favorable geometric fitting due to differences in the size of the two components.

d) Interstitial accommodation effect due to incorporation of smaller entities into the structural network of an associated component and hence the relevant packing effect.

e) Hydrophobic hydration promotes the structuring of water around a hydrophobic (non-polar) molecule or its hydrophobic moiety leading to the formation of 'cage' structures with reduced volume. This phenomenon is called 'hydrophobic hydration.
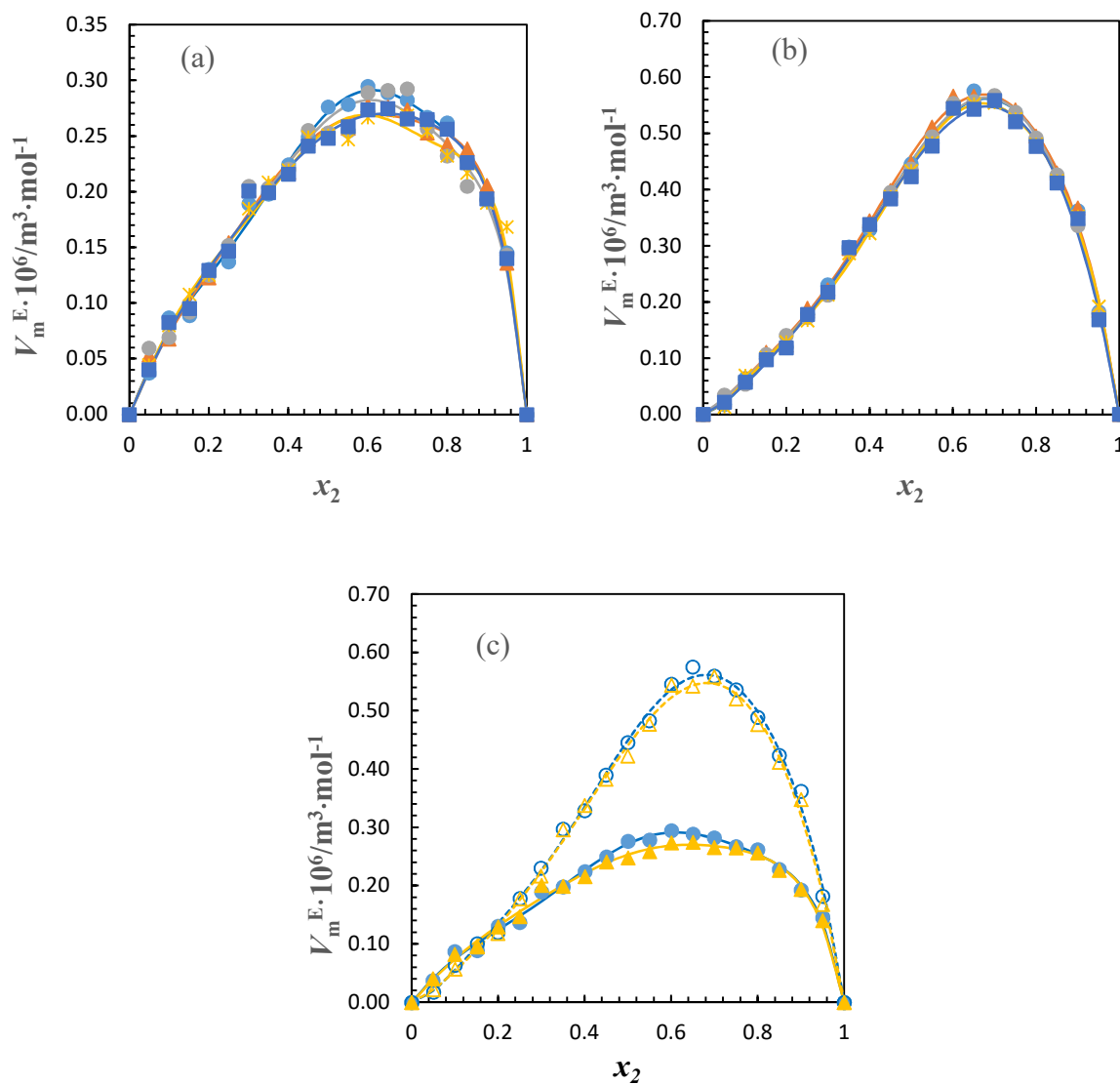

Figure 2. Excess molar volume, V_m^E, against mole fraction, x_2, for (a) EGMEE (x_1) + DXN (x_2) and (b) EGMBE (x_1) + DXN (x_2) systems at: •, 298.15 K; ▲, 303.15 K; ♦, 308.15 K; *, 313.15 K; ■, 318.15 K; and (c) comparative curve for EGMEE (x_1) + DXN (x_2) (•) and EGMBE (x_1) + DXN (x_2) (○) systems at 298.15 K and for EGMEE (x_1) + DXN (x_2) (▲) and EGMBE (x_1) + DXN (x_2) (△) systems at 318.15 K. The solid and dashed lines represent the values calculated from the respective polynomial equation.

A large number of previous investigators have demonstrated that in addition to dipole-dipole and Van-der Waals interactions, alcohols are strongly self-associated through H-bonding (O-H......O-H) interaction forming clusters or networks with restricted rotations about the H-bonds and hence show variable degrees of polymeric aggregates. Alkoxyethanols undergo less self-association in contrast to the high tendency of 1-alcohols to undergo self-association via intermolecular hydrogen bonding. Nevertheless, the presence of etheric oxygen in alkoxyethanols facilitates the formation of intramolecular hydrogen bonding between the etheric oxygen and hydrogen of the -OH group of the

same molecule. Infrared, microwave and calorimetric studies [34,35] have also shown the presence of mostly ten-membered dimers and linear intramolecular associates in $R-O-C_2H_5OH$ (where $R= -C_2H_5$, -C_4H_9, etc.). It is well known that alcohols form a variety of species with different degrees of association in the pure state, however, polymeric linear associates are expected to be predominant in the pure state. Therefore, mixing up an alkanol with a dissimilar component whether it is polar, non-polar, or associative initially leads to dissociating the multimeric structures of the alkanol. Depending on the counter species, sometimes new H-bonds are formed between unlike molecules; the process however depends on the proton-accepting ability of the other component. Thus, the addition of non-polar 1,4-dioxane to an alkoxyethanol causes the dissociation of the H-bonded structure of alkoxyethanols and the subsequent formation of (new) H-bonds [C=O ......H-O] between the proton acceptor oxygen atom (with lone pair of electrons) of the C=(O) group of DXN and the proton of the -OH group of alkoxyethanols. The first (dissociation) effect leads to an increase in volume, resulting in positive V_m^E, whereas, the latter effect leads to a reduction in volume, resulting in negative V_m^E. The positive values of V_m^E for both systems over the entire composition range suggest that the effect due to the disruption of H-bonded associations of alkoxyethanols dominates over the effect of H- bonding between unlike molecules, i.e., the DXN-alkoxyethanol interaction is weaker than that of the DXN-DXN or alkoxyethanol- alkoxyethanol interactions.

This type of variation pattern was also observed for the system isoamyl alcohol with 2-butoxyethanol by M. N. Roy et al [2], Again, from the comparative diagram as in Fig. 2(c), it is observed that the values of V_m^E for EGMBE + DXN system are more positive than those for EGMEE+ DXN system. This leads to suggest that, as the size of the substituent group in the alkoxyethanol molecule increases possibly due to steric hindrance, the DXN - EGMBE interaction becomes weaker than DXN-EGMEE interaction and this effect also reinforces the effect of volume expansion due to disruption of H-bonding in the case of DXN- EGMBE system.

The partial molar volumes of alkoxyethanols ($\bar{V}_1$) and DXN ($\bar{V}_2$) in each of the binary systems are calculated from measured ρ by Eq. 2 & 3 and the results are listed in Table 3 Figs. 3 (a) & (b), and Figs. 4 (a) & (b) show the graphical representation $\bar{V}_1$ and $\bar{V}_2$ within the range $0 \leq x_2 \leq 1$ at different temperatures respectively. Accordingly, Fig. 3 (c) and Fig. 4 (c) compare their variations against x_2. The following are the observations:

a) Partial molar volumes ($\bar{V}_1$) of EGMEE and EGMBE vary steadily with respect to the mole fraction of solute showing almost similar fashion at different concentrations and temperatures. And the variation of $\bar{V}_2$ against x_2 for DXN in EGMEE + DXN is found to decrease gradually though slight maxima and minima formed at stated temperatures. In the case of $\bar{V}_2$ DXN in the EGMBE + DXN system, broad maxima are observed at $x_2 \sim 0.40$.

b) At any composition, the increasing order of both $\bar{V}_1$ and $\bar{V}_2$ is EGMBE + DXN > EGMEE + DXN.

c) The effect of temperature on both $\bar{V}_1$ and $\bar{V}_2$ is quite large.

Table 3. Partial molar volumes of component 1 (EGMEE / EGMBE), $\bar{V}_1 . 10^6 / m^3 . mol^{-1}$ and component 2 (DXN), $\bar{V}_2 . 10^6 / m^3 . mol^{-1}$ in EGMEE + DXN and EGMBE + DXN systems respectively for different molar ratios at different temperatures.

x_2	$\bar{V}_1$	$\bar{V}_2$	$\bar{V}_1$	$\bar{V}_2$	$\bar{V}_1$	$\bar{V}_2$	$\bar{V}_1$	$\bar{V}_2$	$\bar{V}_1$	$\bar{V}_2$
	298.15 K		303.15 K		308.15 K		313.15 K		318.15 K	
					EGMEE + DXN					
0.0496	96.92	86.40	97.37	86.81	97.83	87.32	98.30	87.83	98.79	88.25
0.1001	96.93	86.24	97.36	86.72	97.80	87.19	98.26	87.66	98.73	88.09
0.1515	96.94	86.16	97.36	86.62	97.80	87.08	98.25	87.51	98.70	87.98
0.2001	96.95	86.14	97.36	86.58	97.81	87.01	98.25	87.44	98.69	87.91
0.2498	96.95	86.15	97.36	86.58	97.81	87.00	98.25	87.41	98.69	87.88
0.3007	96.93	86.18	97.36	86.59	97.80	86.99	98.24	87.41	98.69	87.85
0.3506	96.91	86.18	97.36	86.59	97.79	86.99	98.23	87.40	98.68	87.82
0.4008	96.89	86.14	97.36	86.55	97.77	86.95	98.21	87.37	98.68	87.77
0.4503	96.87	86.06	97.36	86.47	97.75	86.88	98.20	87.30	98.67	87.70
0.5005	96.88	85.95	97.36	86.37	97.76	86.78	98.21	87.19	98.67	87.62
0.5505	96.90	85.82	97.35	86.24	97.77	86.66	98.23	87.08	98.66	87.53
0.6004	96.93	85.69	97.35	86.12	97.81	86.55	98.26	86.97	98.67	87.45
0.6503	96.96	85.58	97.33	86.02	97.84	86.45	98.29	86.88	98.68	87.38
0.6995	96.98	85.50	97.31	85.94	97.87	86.37	98.29	86.82	98.69	87.32
0.7500	96.99	85.44	97.31	85.88	97.89	86.32	98.27	86.80	98.73	87.29
0.8000	97.02	85.41	97.37	85.86	97.93	86.30	98.26	86.80	98.82	87.27
0.8500	97.16	85.40	97.58	85.85	98.07	86.30	98.37	86.81	99.04	87.26
0.9000	97.58	85.41	98.08	85.87	98.47	86.32	98.82	86.83	99.54	87.28
0.9499	98.57	85.45	99.09	85.92	99.41	86.39	100.00	86.89	100.51	87.36
					EGMBE + DXN					
0.0506	131.81	86.28	132.44	86.82	133.09	87.28	133.73	87.78	134.39	88.18
0.1006	131.80	86.42	132.44	86.87	133.08	87.33	133.72	87.90	134.39	88.31
0.1515	131.78	86.44	132.43	86.87	133.07	87.35	133.72	87.88	134.37	88.38
0.1993	131.77	86.46	132.41	86.90	133.06	87.38	133.72	87.86	134.35	88.44
0.2504	131.75	86.50	132.38	86.97	133.03	87.45	133.70	87.91	134.33	88.50
0.2993	131.72	86.56	132.34	87.06	133.00	87.53	133.66	87.99	134.31	88.56

|									
0.3504	131.68	86.63	132.29	87.14	132.95	87.60	133.61	88.08	134.28	88.60
0.3998	131.65	86.66	132.26	87.18	132.92	87.64	133.57	88.14	134.26	88.61
0.4495	131.64	86.65	132.25	87.17	132.91	87.63	133.55	88.15	134.26	88.59
0.4995	131.67	86.59	132.29	87.10	132.94	87.57	133.57	88.08	134.29	88.53
0.5495	131.75	86.49	132.39	86.98	133.02	87.46	133.67	87.97	134.36	88.43
0.6008	131.90	86.35	132.56	86.82	133.17	87.32	133.84	87.81	134.49	88.30
0.6500	132.10	86.20	132.77	86.66	133.38	87.17	134.06	87.64	134.67	88.16
0.6997	132.36	86.05	133.04	86.52	133.65	87.02	134.33	87.49	134.92	88.02
0.7499	132.68	85.93	133.34	86.40	133.98	86.88	134.62	87.37	135.24	87.89
0.7999	133.05	85.83	133.68	86.30	134.38	86.77	134.94	87.28	135.63	87.78
0.8498	133.51	85.75	134.10	86.23	134.85	86.69	135.32	87.22	136.10	87.69
0.9001	134.13	85.69	134.71	86.18	135.44	86.63	135.89	87.16	136.71	87.63
0.9500	135.06	85.65	135.72	86.13	136.25	86.60	136.91	87.12	137.52	87.59
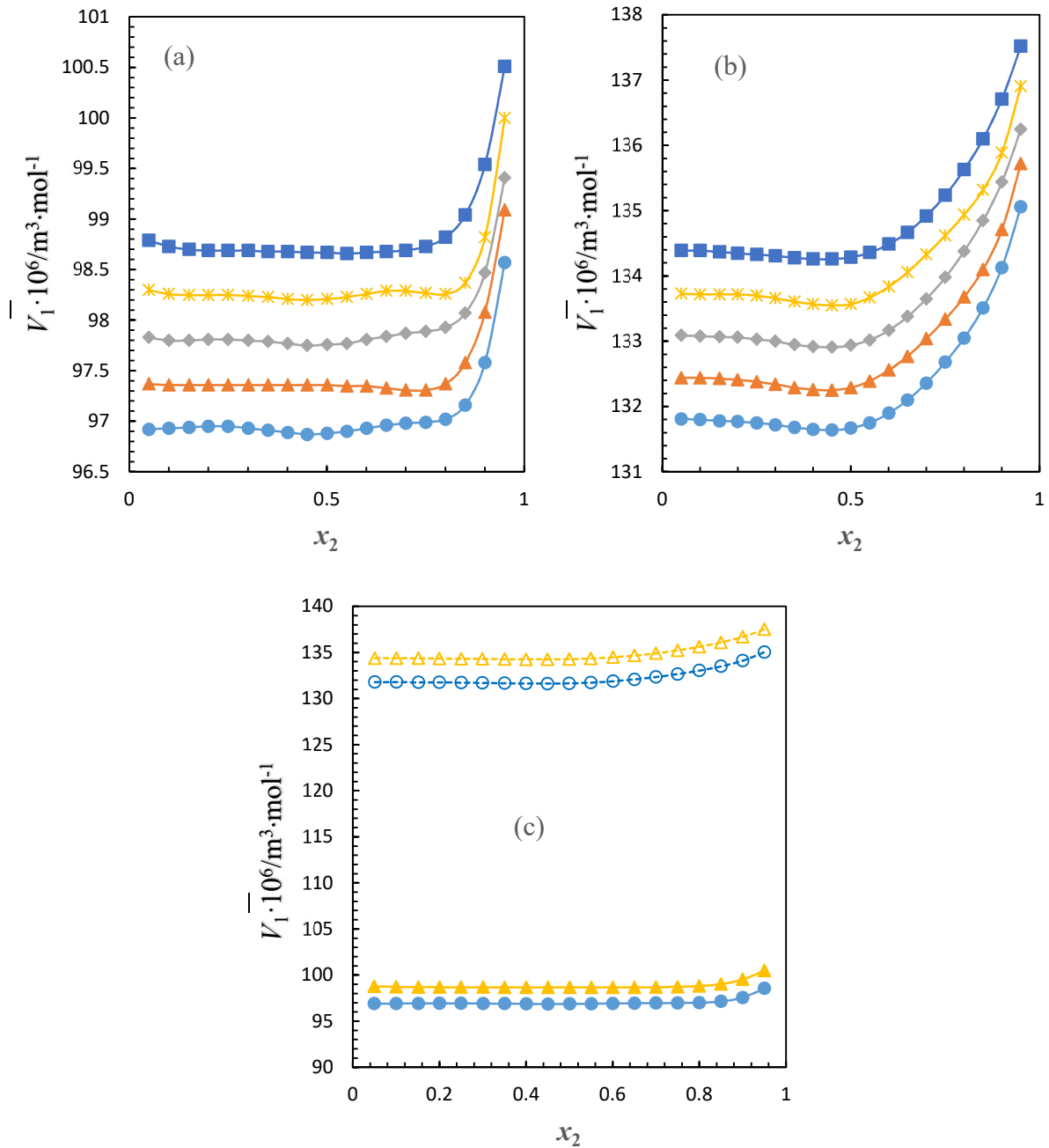
Figure 3. Partial molar volumes of component 1, $\overline{V}_1$, against mole fraction, x_2, for (a) EGMEE (x_1) + DXN (x_2) and (b) EGMBE (x_1) + DXN (x_2) systems at: •, 298.15 K; ▲, 303.15 K; ♦, 308.15 K; *, 313.15 K; ■, 318.15 K; and (c) comparative curve for EGMEE (x_1) + DXN (x_2) (•) and EGMBE (x_1) + DXN (x_2) (○) systems at 298.15 K and for EGMEE (x_1) + DXN (x_2) (▲) and EGMBE (x_1) + DXN (x_2) (△) systems at 318.15 K. The solid and dashed lines represent the values calculated from the respective polynomial equation.
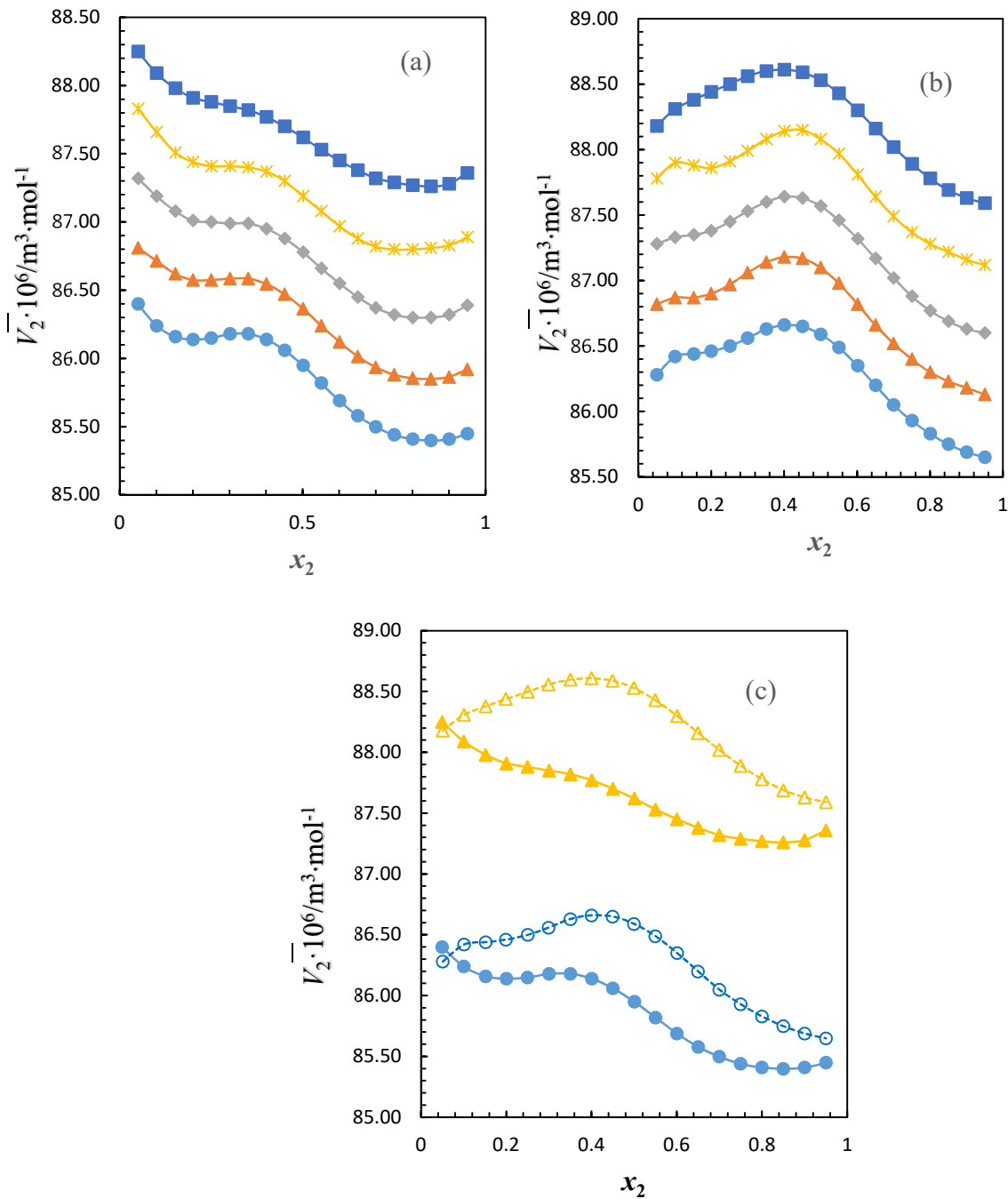
Figure 4. Partial molar volumes of component 2, $\overline{V}_2$, against mole fraction, x_2, for (a) EGMEE (x_1) + DXN (x_2) and (b) EGMBE (x_1) + DXN (x_2) systems at: ●, 298.15 K; ▲, 303.15 K; ♦, 308.15 K; ✳, 313.15 K; ■, 318.15 K; and (c) comparative curve for EGMEE (x_1) + DXN (x_2) (●) and EGMBE (x_1) + DXN (x_2) (○) systems at 298.15 K and for EGMEE (x_1) + DXN (x_2) (▲) and EGMBE (x_1) + DXN (x_2) (△) systems at 318.15 K. The solid and dashed lines represent the values calculated from the respective polynomial equation.

The above results show, $\bar{V}_1$, of EGMEE and EGMBE gradually increase in the whole region. Considering the significance of $\bar{V}_1$, rising $\bar{V}_1$ for both systems indicates that in the overall volume expansion, EGMEE and EGMBE contribute significantly making the corresponding V_m^E values positive for the respective system. On the other hand, at $x_2 < 0.40$ for EGMBE + DXN system DXN contributes more positively to the excess molar volume rather than at compositions $x_2 > 0.40$, In the case of the EGMEE+ DXN system with the falling trend of $\bar{V}_2$ (DXN) throughout the whole composition, the axis indicates that its contribution toward volume expansion is relatively higher, especially in the EGMEE-rich region.

The thermal expansivity of the liquids (α) and their excess (α^E) were calculated using Equations 4 & 5 respectively. All the values of α and α^E are represented in Table 4. Fig. 5(a) and Fig. 5 (b) show α vs. x_2 and α^E vs. x_2 for the two systems, respectively. For each system, α^E is also fitted well into the Redlich-Kister type equation.

Table 4. Thermal expansivities, α, and excess thermal expansivities, α^E, in K^{-1}, for the systems of EGMEE + DXN and EGMBE + DXN at different molar ratios

x_2	$\alpha \cdot 10^4$	$\alpha^E \cdot 10^4$	x_2	$\alpha \cdot 10^4$	$\alpha^E \cdot 10^4$
EGMEE + DXN			**EGMBE + DXN**		
0.0000	10.13	0.000	0.0000	9.68	0.000
0.0496	10.18	0.013	0.0506	9.74	0.018
0.1001	10.22	0.023	0.1006	9.80	0.041
0.1515	10.27	0.033	0.1515	9.86	0.062
0.2001	10.32	0.040	0.1993	9.91	0.086
0.2498	10.37	0.054	0.2504	9.95	0.131
0.3007	10.41	0.067	0.2993	10.01	0.153
0.3506	10.46	0.078	0.3504	10.08	0.158
0.4008	10.51	0.088	0.3998	10.14	0.182
0.4503	10.57	0.086	0.4495	10.20	0.202
0.5005	10.62	0.093	0.4995	10.26	0.226
0.5505	10.66	0.109	0.5495	10.30	0.260
0.6004	10.71	0.117	0.6008	10.39	0.258
0.6503	10.78	0.100	0.6500	10.45	0.275
0.6995	10.83	0.107	0.6997	10.53	0.271
0.7500	10.89	0.108	0.7499	10.62	0.263
0.8000	10.99	0.066	0.7999	10.71	0.257
0.8500	11.04	0.075	0.8498	10.82	0.230
0.9000	11.12	0.052	0.9001	10.94	0.193
0.9499	11.21	0.025	0.9500	11.09	0.119
1.0000	11.29	0.000	1.0000	11.29	0.000
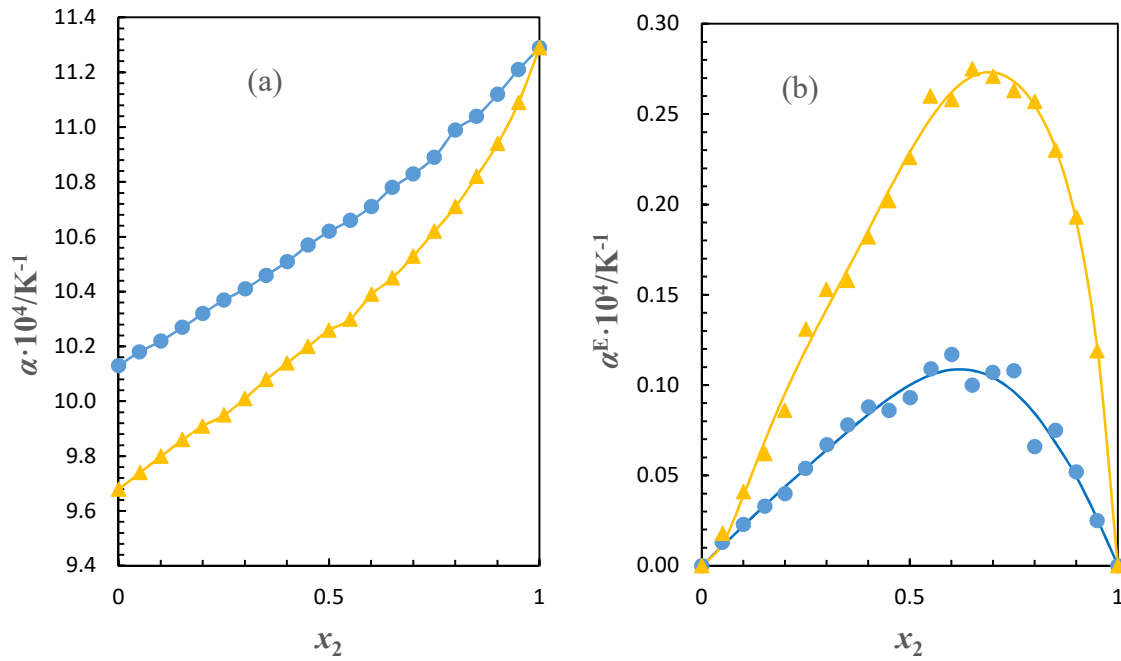
Figure 5. Comparative diagrams of (a) α vs. x_2 and (b) α^E vs. x_2 for the systems of EGMEE (x_1) + DXN (x_2) (•) and EGMBE (x_1) + DXN (x_2) (▲). Solid lines represent fittings for the polynomial and Redlich-Kister equation respectively.

Table 5. Coefficients, $a_{i,}$ of Equation 6 expressing densities, ρ; thermal expansivities, α; and regression indices, R^2 of EGMEE + DXN and EGMBE + DXN systems for different molar ratios at different temperatures.

Property	T(K)	a_0	a_1	a_2	a_3	a_4	a_5	R^2
				EGMEE + DXN				
$\rho.10^3$ (kg m^{-3})	298.15	0.929	0.083	-0.008	0.063	-0.080	0.041	1.000
	303.15	0.925	0.084	-0.029	0.132	-0.168	0.078	1.000
	308.15	0.920	0.081	-0.010	0.068	-0.085	0.041	1.000
	313.15	0.916	0.081	-0.020	0.106	-0.137	0.064	1.000
	318.15	0.911	0.079	0.013	0.090	-0.121	0.060	1.000
$\alpha.10^4$ (K^{-1})		10.132	0.814	1.051	-3.087	3.847	-1.466	1.000
				EGMBE + DXN				
$\rho.10^3$ (kg m^{-3})	298.15	0.896	0.082	0.022	0.000	0.007	0.018	1.000
	303.15	0.892	0.081	0.024	-0.006	0.017	0.014	1.000
	308.15	0.888	0.079	0.036	-0.041	0.057	-0.002	1.000
	313.15	0.883	0.079	0.027	-0.016	0.026	0.009	1.000
	318.15	0.879	0.079	0.020	0.001	0.008	0.016	1.000
$\alpha.10^4$ (K^{-1})		9.680	1.305	-1.596	4.966	- 6.491	3.422	1.000

Table 6. Coefficients, A_i, of Equation 7, expressing excess molar volumes, V_m^E; excess thermal expansivities, α^E; and standard deviation, σ (Eqn. 8) of EGMEE + DXN and EGMBE + DXN systems for different molar ratios at different temperatures.

Property	T(K)	A_0	A_1	A_2	A_3	A_4	A_5	Σ
				EGMEE + DXN				
$V_m^E . 10^6$ ($m^3 . mol^{-1}$)	298.15	1.0877	0.6896	0.2816	0.4955	1.5175	1.2310	0.0093
	303.15	1.0324	0.4200	-0.0081	0.4473	1.2376	0.4810	0.0101
	308.15	1.0627	0.6101	-0.1492	-0.5723	1.2269	1.3535	0.0164
	313.15	1.0278	0.5203	-0.2559	-0.5576	1.7148	1.7467	0.0123
	318.15	1.0131	0.4891	0.1654	0.1461	0.9716	0.6860	0.0090
$\alpha^E . 10^4$ (K^{-1})		0.4007	0.2650	0.0043	-0.1834	-0.0129	0.0982	0.0080
				EGMBE + DXN				
$V_m^E . 10^6$ ($m^3 . mol^{-1}$)	298.15	1.7990	2.1379	0.4272	-1.1858	0.2398	1.3675	0.0140
	303.15	1.8459	2.1985	0.1696	-1.4358	0.7170	1.5744	0.0133
	308.15	1.7988	2.1719	0.4219	-1.1573	0.2135	0.9470	0.0099
	313.15	1.7744	2.2296	0.2389	-1.9092	0.6092	2.2014	0.0112
	318.15	1.7637	2.0046	0.4348	-0.6468	0.0933	0.6407	0.0150
$\alpha^E . 10^4$ (K^{-1})		0.9164	0.8051	0.4152	-0.3378	0.2232	1.1268	0.0076

It has been found that for both EGMEE + DXN and EGMBE + DXN systems α increases slowly up to the pure state of DXN. Values of α for the pure liquids decrease in the order: DXN > EGMEE > EGMBE, whereas, increment of α with respect to x_2 for the solutions vary as: EGMEE + DXN > EGMBE + DXN. However, for both systems, values of α^E are positive in the full range of composition forming maxima nearly at $x_2 = 0.65$, indicating that DXN acts as structure breakers [11] towards alkoxyethanols i.e., towards EGMBE and EGMEE and the variation follows: EGMBE+DXN > EGMEE+DXN, which is due to greater steric hindrance in EGMBE compared to that in EGMEE.

4. Conclusion

The density of EGMEE + DXN and EGMBE + DXN systems have been measured and various derived parameters, viz., V_m^E, $\bar{V}_1$, $\bar{V}_2$, α and α^E, have been calculated from the experimental values of density. ρ, $\bar{V}_1$, $\bar{V}_2$, and α have been fitted to appropriate polynomials. V_m^E and α^E have been fitted to the Redlich-Kister polynomial equation. Variations with the composition of 1,4-Dioxane of all the properties have also been shown. Here, it is found that with the addition of DXN in alkoxyetanol, ρ increases linearly for both the systems and the increment follows the order: EGMEE+ DXN > EGMBE+ DXN. And the effect of temperature on ρ is quite large. Values of V_m^E are found to be positive for both systems and form a maximum in the DXN-rich region at $x_2 \sim 0.65$. Values of positive V_m^E follow the order: EGMBE + DXN > EGMEE + DXN. $d V_m^E /dT$ is insignificant.

Partial molar volumes ($\bar{v}_1$) against x_2 of EGMEE and EGMBE vary steadily showing almost similar fashion at different concentrations and temperatures. And the variation of $\bar{v}_2$ against x_2 for DXN in EGMEE+ DXN is found to decrease gradually though slight maxima and minima formed at stated temperatures. In the case of $\bar{v}_2$ of DXN in the EGMBE + DXN system, broad maxima are observed at $x_2 \sim 0.40$. Increasing order of both $\bar{v}_1$ and $\bar{v}_2$ is: EGMBE + DXN > EGMEE + DXN. The effect of temperature on both $\bar{v}_1$ and $\bar{v}_2$ is quite large.

For both systems α increases slowly with increasing x_2. Pure liquids decrease in the order: DXN > EGMEE > EGMBE, whereas, the solutions vary as: EGMEE + DXN > EGMBE + DXN. However, for both systems values of α^E are positive. And its variation follows: EGMBE+DXN > EGMEE+DXN.

Analysis of all these results revealed that being a less self-associated compound and the presence of etheric oxygen in alkoxyethanols facilitates the formation of intramolecular hydrogen bonding; later the addition of non-polar 1,4-dioxane to an alkoxyethanol causes the dissociation of the H-bonded structure of alkoxyethanols and DXN. Positive V_m^E is observed and reinforced due to steric hindrance. Being α^E positive, indicates that DXN act as structure breakers toward alkoxyethanols. These findings will be useful for future works on liquid-liquid mixtures and chemical processing.

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