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Viscosities and densities for binary mixtures of 1, 2-ethanediol and water at higher temperature

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Abstract

Viscosities and densities of the binary mixtures of 1,2-ethanediol with water were measured at 318.15, 320.15, 322.15, 324.15, 326.15 and 328.15 K. This experimental data were used to calculate excess molar volume (V^E), viscosity deviations (Δn) and excess Gibbs free energy of activation ΔG^{E*} of viscous flow. These results have been discussed in terms of molecular interaction at different temperatures. The excess molar volume (V^E) and viscosity deviation (Δn) values were found to be negatives at all temperatures. The molecular interactions existing between the components were also discussed.

Keywords: Viscosity, Density, Deviations, Molecular, Interactions.

Introduction

The thermodynamic properties of a binary mixtures such as viscosity and density are important from practical and theoretical points of view to understand molecular interactions and liquid theory. Their properties are extremely useful for the design of types of transport and process equipment in chemical industries[1]. The studies of excess functions of binary liquid mixtures are useful in understanding the nature and strength of molecular interactions between the component molecules. 1, 2-1,2-ethanediol is a simplest homolgues of the diol, largely utilised as thermo regulator fluid, as controlling agent of density viscosity reaction bath, and also used as emulsion coating due to unusual physico chemical properties. It is an alcohol with two -OH groups and widely used as automotive antifreeze[2]. In the present paper, it has been reported density (ρ) and viscosity (η) for the binary system of 1, 2-1,2-ethanediol and water at various temperatures at atmospheric pressure. The calculated excess quantities from such data have been interpreted in terms of molecular interactions and structural effects at higher temperatures.

Material and methods

1,2-ethanediol (A.R. Grade from S.D. Fine Chemicals) was first dried overnight over fused calcium oxide, and then distilled twice under vacuum. The middle fraction collected and the fraction was stored in dark coloured bottle and protected against atmospheric moisture and carbon dioxide. Triple distilled water (specific conductance than 10 S cm^{-1}) was employed in the making of the compositions.

Densities of the pure components and their compositions were measured on a vibrating tube density-meter, reproducible to 1 x 10^{-5} gm cm^{-3} (Anton paar model DMA 5000) measuring with high temperature accuracy (± 0.001 K) in a wide temperature range. The density-meter was calibrated with triple distilled water, the observed density was 0.998212 gm cm^{-3} at 20°C which is closer to literature value[3]. The mixtures were prepared by mixing known masses of pure liquid in air tight, narrow-mouth ground stoppered bottles taking precautions to minimise the evaporation losses. All measurements of mass were performed on an electronic balance with accuracy of 0.1 mg.

Measurement of the dynamic viscosity η viscosities and various compositions was carried out by using a suspended level Cannon-Ubbelohde viscometer. The viscometer was suspended vertically in a constant temperature bath (± 0.05°C). The time given to attain thermal equilibrium for content of viscometer was 15 min. The flow time was measured to an accuracy of 0.1 S till a constant flow time was observed. The viscometer was separately calibrated with benzene and toluene (HPLC grade). At 303.15 K, the reproducibility in the viscosity measurement was ± 0.01 mPaS. Viscosities of the pure component of water were taken from the literature.

Results and discussion

Excess molar volumes V^E were calculated from the measured densities[4,5] (ρ) by using the relation.

$$V^E = \frac{(x_1 M_1 + x_2 M_2)}{\rho} - (x_1 V_1 + x_2 V_2) \quad (1)$$

Where, x_1 and x_2 are mole fractions, M_1 and M_2 the molecular weights and V_1 and V_2 are the molecular volumes of 1,2-ethanediol (1) and water (2) respectively. The experimental densities, excess molar volumes, viscosities and deviation in viscosities of binary mixtures of 1,2-ethanediol (1) + water (2) at six different temperatures are reported in Table-1.

Dynamic viscosities (η) of 1,2-ethanediol (1) and water (2) mixtures of different temperatures were calculated by measuring density and flow time of the mixture (Table-1). The viscosity deviation was calculated[6,7] by the equation (2)

$$\Delta\eta = \eta - \{x_1 \eta_1 + x_2 \eta_2\} \quad (2)$$

The experimental values of Δη are also reported in (Table-1).

On the basis of theory of absolute reaction rates, the excess Gibb's energies (ηG^{*E}) of activation was calculated[8,9] from equation (3)

$$\frac{\Delta G^{*E}}{RT} = \left\{ \ln\left(\frac{\eta V}{\eta_2 V_2}\right) - x_1 \ln\left(\frac{\eta_1 V_1}{\eta_2 V_2}\right) \right\} \quad (3)$$

where v, v_1 and v_2 are the molar volumes of the binary mixture and pure components and calculated values of ΔG^{E*} are shown in Table-1.

Discussion: The results of densities of binary systems 1,2-(1,2-ethanediol + water), at different compositions were measured at six different temperatures at an interval of 2 K (318.15 K — 328.15 K). The experimental densities (ρ), excess molar volumes (V^E) are furnished in Table-1 respectively.

The sign and magnitude of excess molar volume gives a good estimate of the strength of unlike molecular interactions in the solution phase. It is explicit that when large negative values of excess molar volume are found, then interactions are strong. It is observed that V^E is negative for all the systems studied over the entire range of mole fraction at the measured temperatures.

Figure-1 shows that, maximum deviation in the excess molar volumes of 1,2-ethanediol was observed at 0.38 mole fraction in the present work. The largest value of V^E is — 0.283692 cm^3 $mole^{-1}$ in temperature range 318.15 K to 328.15 K at an interval of 2 K. These large negative value of V^E arises due to increased interactions between unlike molecules[10] (1,2-ethanediol and water) or very large differences in the molar volume of pure components at all temperature. The V^E becomes more negative with decreasing temperature. It would appear that the temperature coefficient of excess molar volume depends on the increase in molar volume complex formation. The magnitude suggesting that there is weak interaction of hydrophobic hydration between the polar hydroxyl group of the 1,2-ethanediol and water.

The graphical presentation of the experimental Δη values plotted against mole fraction are as shown in Figure-2.

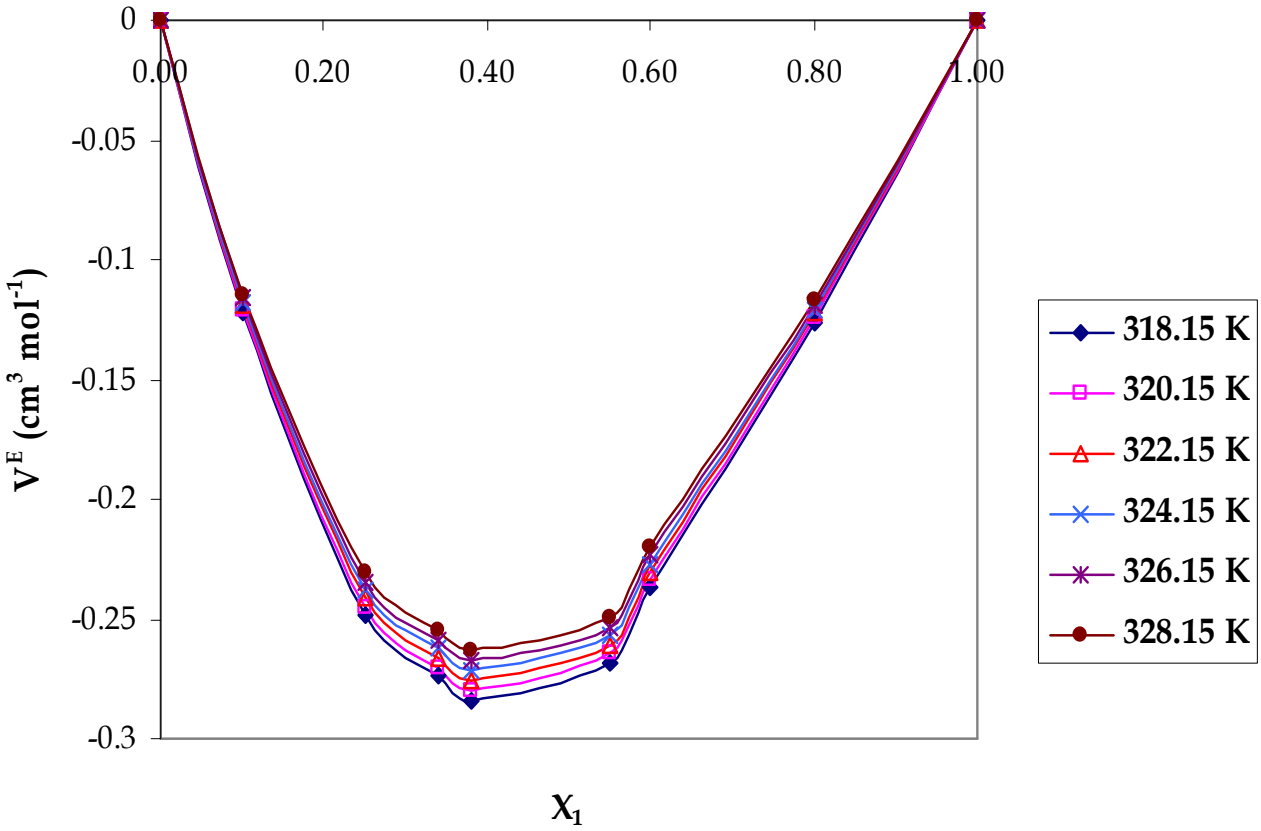

Figure-1: Excess molar volumes (V^E) for the system 1,2-ethanediol (1) + water (2) from 318.15 K to 328.15 K.

Graphical representation of the experimental viscosity deviations (Δη) plotted against the fitted curve for 1, 2-ethanediol + water system are shown in Figure-2. The Δη is negative and its values decrease with increasing temperature for binary aqueous systems of 1,2-ethanediol. The curves are skewed toward the water rich region[11]. Increased temperature enhance the mixture viscosity contribution to the overall viscosity deviation. The values decrease with increasing temperature due to weakening of the interactions.

Not only the strong intermolecular hydrogen bonds but also molecular shapes and sizes of the component influence the deviation in viscosity of binary mixtures. Thus, negative values of Δη indicate the presence of dispersion forces. The large viscosity of diol in the pure state may be due to self associations by dipole—dipole interactions.

The value of Δη are negative over the entire range of mole fraction and maximum negative value at mole fraction 0.38 at six temperatures. Figure-3 shows the graphical variation of ΔG^{E*} for the binary mixtures of 1, 2-ethanediol with water at different six temperatures.

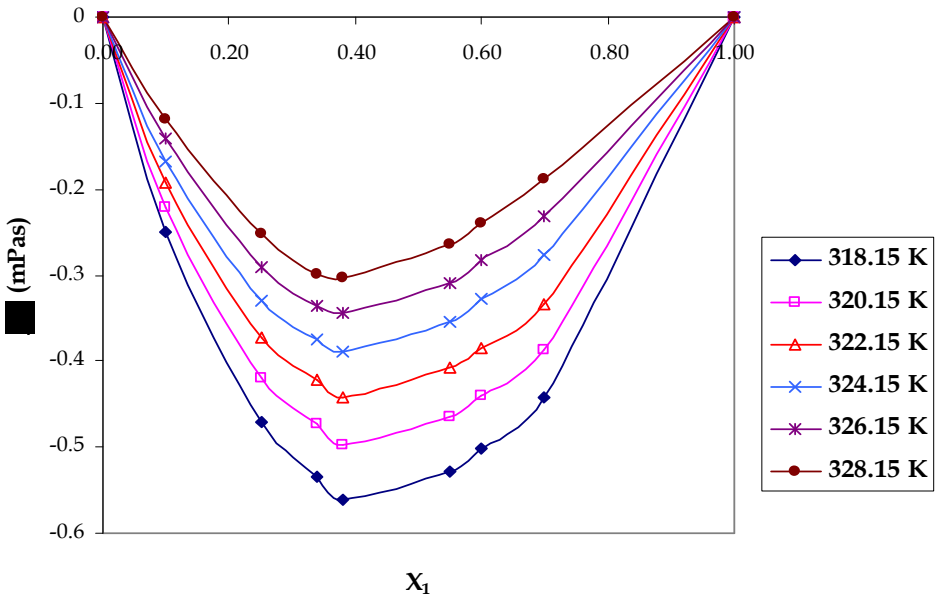

Figure-2: Viscosity deviation (Δη) for the system ethanediol (1) + water (2) from 318.15 K to 328.15K.

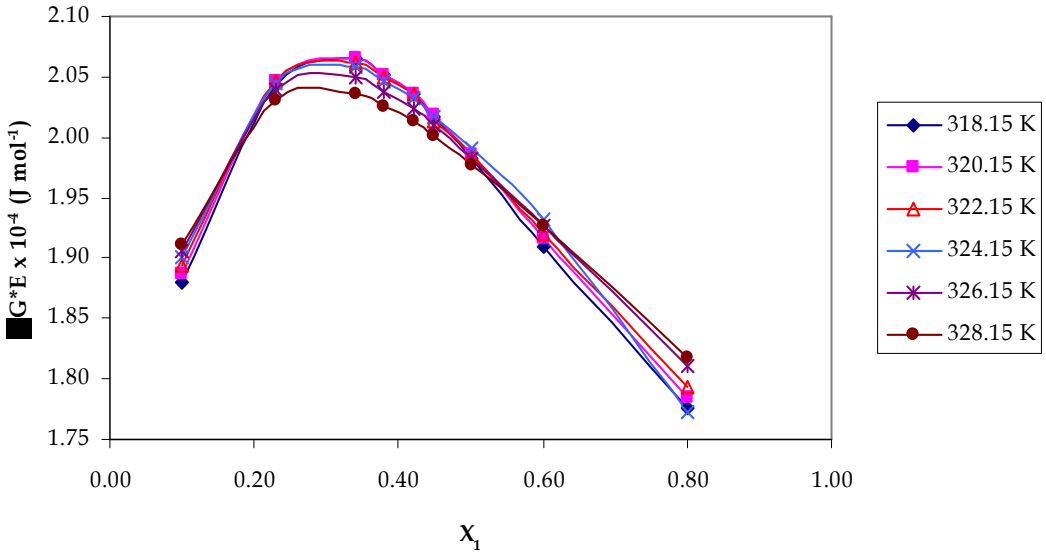

Figure-3: Variation of Excess energy of activation for viscous flow (ΔG^{E*}) against mole fraction (x_1) of 1,2-ethanediol with water from 318.15 K to 328.15 K.

Excess energy of activation for viscous flow is shown in Figure-3. The observed ΔG^{E*} values are positive[11,12] for entire mole fraction of 1,2-ethanediol (1) + water (2). Large positive values indicates the specific interaction due to complex formation through intermolecular hydrogen bonding interaction between 1,2-ethanediol (1) and water (2) molecules (unlike molecules) compared to like molecules[13,14].

Table-1: Experimental Densities, Viscosities and calculated values of excess molar volume, viscosity deviations and excess energy of activation for viscous flow of 1,2-ethanediol with water at 318.15 K

X_1	ρ (g cm^{-3})	η (mPas)	V^E (cm^3 mol^{-1})	$\Delta\eta$ (mPas)	ΔG^{*E} x 10^{-4} (J mol^{-1})
0.00	0.990188	0.598680			
0.10	1.023149	1.058990	-0.122463	-0.249623	1.637271
0.25	1.053463	1.902990	-0.248650	-0.470522	1.706504
0.34	1.064606	2.477459	-0.273809	-0.534992	1.719528
0.38	1.068760	2.736010	-0.283692	-0.560414	1.719479
0.55	1.081183	3.976000	-0.267915	-0.527311	1.700992
0.60	1.083381	4.355740	-0.236778	-0.502538	1.689230
0.80	1.090721	5.126026	-0.125953	-0.442185	1.657918
1.00	1.095746	7.698010			

Table-2: Experimental Densities, Viscosities and calculated values of excess molar volume, viscosity deviations and excess energy of activation for viscous flow of 1,2-ethanediol with water at 320.15 K

X_1	ρ (g cm^{-3})	η (mPas)	V^E (cm^3 mol^{-1})	$\Delta\eta$ (mPas)	ΔG^{*E} x 10^{-4} (J mol^{-1})
0.00	0.989419	0.576286			
0.10	1.022112	1.013970	-0.120862	-0.220186	1.645425
0.25	1.052201	1.800910	-0.245096	-0.420051	1.710246
0.34	1.063283	2.339011	-0.270062	-0.474035	1.721661
0.38	1.067408	2.577980	-0.279679	-0.498213	1.720612
0.55	1.079778	3.730020	-0.264296	-0.464554	1.698832
0.60	1.081971	4.083727	-0.233490	-0.439781	1.686235
0.80	1.089298	4.794001	-0.124161	-0.387379	1.653696
1.00	1.094320	7.154990			

Table-3: Experimental Densities, Viscosities and calculated values of excess molar volume, viscosity deviations and excess energy of activation for viscous flow of 1,2-ethanediol with water at 322.15 K

X_1	ρ (g cm^{-3})	η (mPas)	V^E (cm^3 mol^{-1})	$\Delta\eta$ (mPas)	ΔG^{*E} x 10^{-4} (J mol^{-1})
0.00	0.988647	0.553880			
0.10	1.021076	0.970690	-0.119297	-0.193305	1.658117
0.25	1.050939	1.706890	-0.241543	-0.372277	1.725193
0.34	1.061959	2.205813	-0.266283	-0.422458	1.738036
0.38	1.066056	2.430100	-0.275662	-0.442216	1.738063
0.55	1.078372	3.501980	-0.260635	-0.407533	1.720158
0.60	1.080562	3.828582	-0.230234	-0.385988	1.708593
0.80	1.087876	4.491030	-0.122410	-0.333655	1.678630
1.00	1.092894	6.655030			

Table-4: Experimental Densities, Viscosities and calculated values of excess molar volume, viscosity deviations and excess energy of activation for viscous flow of 1,2-ethanediol with water at 324.15 K

X_1	ρ (g cm^{-3})	η (mPas)	V^E (cm^3 mol^{-1})	$\Delta\eta$ (mPas)	ΔG^{*E} x 10^{-4} (J mol^{-1})
0.00	0.987876	0.531482			
0.10	1.020040	0.930491	-0.117726	-0.167242	1.669430
0.25	1.049677	1.616900	-0.237975	-0.330210	1.734987
0.34	1.060635	2.083009	-0.262488	-0.373727	1.747809
0.38	1.064703	2.294000	-0.271597	-0.389238	1.747898
0.55	1.076967	3.292000	-0.256993	-0.353866	1.730129
0.60	1.079152	3.602064	-0.226925	-0.326928	1.719124
0.80	1.086453	4.219236	-0.120607	-0.276008	1.689396
1.00	1.091468	6.194000			

Table-5: Experimental Densities, Viscosities and calculated values of excess molar volume, viscosity deviations and excess energy of activation for viscous flow of 1,2-ethanediol with water at 326.15 K

X_1	ρ (g cm^{-3})	η (mPas)	V^E (cm^3 mol^{-1})	$\Delta\eta$ (mPas)	ΔG^{*E} x 10^{-4} (J mol^{-1})
0.00	0.987107	0.509084			
0.10	1.019003	0.892080	-0.116112	-0.142292	1.681195
0.25	1.048415	1.533990	-0.234382	-0.290813	1.745461
0.34	1.059312	1.962452	-0.258696	-0.336013	1.757139
0.38	1.063351	2.165970	-0.267535	-0.343009	1.758294
0.55	1.075561	3.095010	-0.253291	-0.308659	1.740514
0.60	1.077742	3.383655	-0.223594	-0.283150	1.729578
0.80	1.085031	3.960864	-0.118838	-0.232239	1.700225
1.00	1.090042	5.771970			

Table-6: Experimental Densities, Viscosities and calculated values of excess molar volume, viscosity deviations and excess energy of activation for viscous flow of 1,2-ethanediol with water at 328.15 K

X_1	ρ (g cm^{-3})	η (mPas)	V^E (cm^3 mol^{-1})	$\Delta\eta$ (mPas)	ΔG^{*E} x 10^{-4} (J mol^{-1})
0.00	0.986373	0.486686			
0.10	1.017967	0.853669	-0.114533	-0.119344	1.692873
0.25	1.047153	1.451080	-0.230787	-0.251423	1.755825
0.34	1.057988	1.841895	-0.254871	-0.298301	1.766427
0.38	1.061998	2.037940	-0.263436	-0.303615	1.768685
0.55	1.074155	2.898020	-0.249580	-0.263462	1.750899
0.60	1.076333	3.165186	-0.220294	-0.239459	1.740028
0.80	1.083609	3.702490	-0.117064	-0.188482	1.711044
1.00	1.088616	5.349951			

Conclusion

Experimental data of the density and viscosity of 1,2-ethanediol and water mixtures have been measured over the entire composition range at 328.15 K, 320.15 K, 322.15 K, 324.15 K, 326.15 K and 328.15 K. It has been observed that negative deviations were observed for excess molar volume (V^E) viscosity deviation ($\Delta\eta$) and positive values of excess energy of activation for viscous flow (ΔG^{E*}) the existence strong molecular interaction between the molecules of 1,2-ethanediol and water of the mixtures taken up for study.

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