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Molecular Interaction Studies in Ternary Liquid Mixtures of Quinoline at Varying Temperatures

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ABSTRACT

Density ρ, Viscosity η, and ultrasonic velocity u, have been measured in pure Quinoline, o-cresol, m-cresol, p-cresol and methanol and in their ternary liquid mixtures with quinoline and methanol as common components at 303.15, 308.15, 313.15 and 318.15K over the entire range of composition. From these experimental data, various thermo-acoustic parameters such as excess molar volume V_m^E, excess ultrasonic velocity u^E, viscosity deviation Δη and excess Gibb's free energy of activation for viscous flow ΔG^{*E} have been calculated. The deviation/excess parameters were plotted against the mole fraction of quinoline over the whole composition range. The observed negative and positive values of deviaiton/excess thermo-acoustic parameters were explained on the basis of intermolecular interactions present in these mixtures.

Keywords

Viscosity, quinoline, cresol, thermo-acoustic parameters, ternary mixtures

Academic Discipline And Sub-Disciplines

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SUBJECT CLASSIFICATION

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INTRODUCTION

As a part of ongoing research work on acoustic, thermodynamic and transport properties of binary and ternary liquid mixtures, we report here the results of ternary mixtures of quinoline with o-cresol, m-cresol, p-cresol in methanol at 303.15, 308.15, 313.15, 318.15 K covering the whole composition range. When compared to pure liquids mixed solvents have many applications in chemical and industrial processes [1-3]. When two or more liquids are mixed together, the resulting changes in physical and thermodynamic properties can be considered as a sum of several contributions due to change in energy, free volume change, change in steric hindrance and molecular orientation [4,5]. Knowledge of various thermodynamic properties of mixtures involving quinoline with self associated alcohols and phenols helps in designing an efficient industrial process and also helps in understanding the inter and intra molecular interactions. Several researchers carried out investigations of acoustic and thermodynamical properties of binary mixtures containing phenols [6-9]. As of our knowledge no work is done in ternary mixtures containing quinoline with phenols in methanol.

The liquids used in the present study are important due to their various industrial applications. Quinoline is used in solar cells to increase its efficiency, used in dyes. Phenols are widely used for wood preservatives, selective weed killing, cleaning purpos and phonographic record. Alcohols are self-associated organic liquids and are used as the basic organic compound for the synthesis of other organic compounds. The study of ternary mixtures of quinoline with phenols in methanol would of considerable interest because these mixtures exhibit varying molecular interactions and are important from an industrial point of view.

Here we report the density (ρ), viscosity (η) and ultrasonic velocity (u) of the ternary mixtures of quinoline with o-cresol, or m-cresol, or p-cresol in methanol at varying temperatures of 303.15, 308.15, 313.15 and 318.15 K over the entire range of composition. These values are used to study the thermodynamic properties and molecular interactions in the mixtures choosen for the study. When unlike interactions are equal to like interactions then the solution is said to be an ideal solution. The deviation from ideality is expressed by many thermodynamic variables perticularly by excess properties. The excess properties of the mixtures are useful in the study of molecular interactions and arrangements [10-12]. The experimental values of ρ, η and u were used to calculate the deviation in excess molar volume V_m^E, excess ultrasonic velocity u^E, viscosity deviation Δη and excess Gibb's free energy of activation for viscous flow ΔG^{*E} with the aim of analysing the molecular interactions.

EXPERIMENTAL DETAILS

The analytical grade chemicals obtained from SRL Chemicals, Mumbai were used. They were purified by standard procedure [13]. To prepare the mixtures in the required proportions, Job's method of continuous variation was used. The mixtures were preserved in well-stoppard conical flasks. After mixing the liquids thoroughly, the flasks were left undisturbed to allow them to attain thermal equilibrium.

Single crystal ultrasonic pulse echo interferometer (Mittal enterprises, India; Model: F-80X) was used for measuring ultrasonic velocities. It consists of a high frequency generator and a measuring cell. The measurements of ultrasonic velocities were made at a fixed frequency of 3MHz. The calibration of the equipment was done by measuring the velocity in benzene and carbon tetrachloride. The results are in good agreement with the literature values [14]. The ultrasonic velocity has an accuracy of ± 0.1 $m.s^{-1}$. The temperature was controlled by circulating water around the liquid cell from thermostatically controlled constant temperature water bath (accuracy ± 0.01K).

A specific gravity bottle was used for the measurement of densities of pure liquids and liquid mixtures. Weights were measured with an electronic balance (Shimadzu AUY220, Japan) capable of measuring up to 0.1mg. An average of 3-4 measurements were taken for each sample. Ostwald's viscometer was used for measuring viscosities at the desired temperature, which was calibrated using water and benzene. After the mixture had attained bath temperature, flow time has been measured. The flow measurements were made with an electronic stopwatch with a precision of 0.01 s. The viscosity is determined using the relation

$$\eta = k\rho t \qquad (1)$$

Where k, ρ and t are viscometric constant, density of liquid and time of efflux for a constant volume of liquid, respectively.

THEORY

The values of Molar volume, V_m are calculated using standard relation,

$$V_m = M_{eff}/\rho \qquad (2)$$

where $M_{eff} = (X_1M_1+X_2M_2+X_3M_3)$, M_1, M_2 and M_3 are the molecular weights of pure components, and ρ is the density of the mixture.

The strength of interaction between the component molecules of ternary mixtures is well reflected in the deviation of the excess functions from ideality [15]. The excess properties such as V_m^E, u^E, $\Delta\eta$ *and* G^{*E} were calculated using the following equations

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$$V_m^E = \left(\frac{x_1M_1 + x_2M_2 + x_3M_3}{\rho}\right) - \left(\frac{x_1M_1}{\rho_1} + \frac{x_2M_2}{\rho_2} + \frac{x_3M_3}{\rho_3}\right) \quad (3)$$

$$u^E = u_m - [x_1u_1 + x_2u_2 + x_3u_3] \quad (4)$$

$$\Delta\eta = \eta_m - \sum_{i=1}^{n} x_i\eta_i \quad (5)$$

$$G^{*E} = RT\left[\ln\eta V - \{x_1 \ln\eta_1 V_1 + x_2 \ln\eta_2 V_2 + x_3 \ln\eta_3 V_3\}\right] \quad (6)$$

In equations (3) –(6), M_i, ρ_i,η_i, V_{mi} and x_i, respectively represent molecular weight, density, viscosity, molar volume and mole fraction of ith component (i=1,2,3 for ternary mixtures) ρ, u_m, η_m and V are the density, ultrasonic velocity, viscosity and molar volume of the mixture. R is the universal gas constant and T is the absolute temperature.

RESULTS AND DISCUSSION

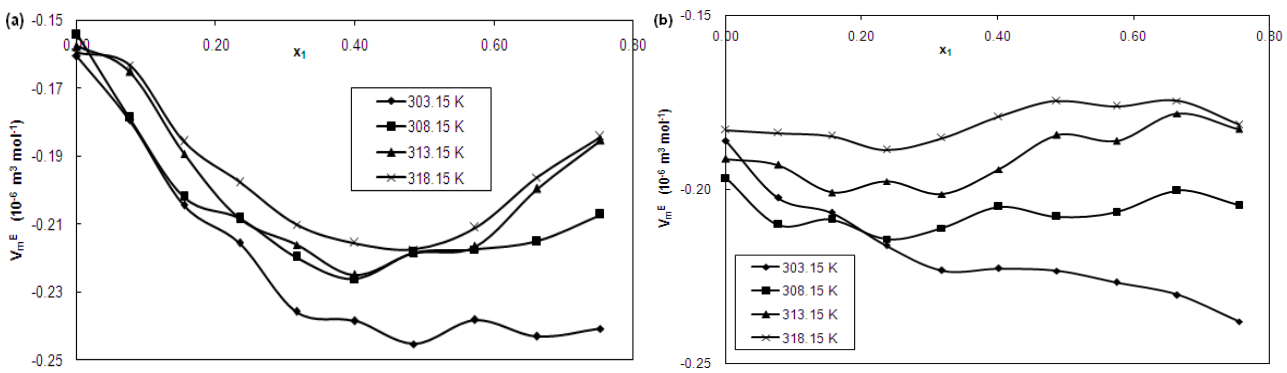
The experimental values of density (ρ), viscosity (η) and ultrasonic velocity (u) for the three ternary mixtures at temperatures 303.15, 308.15, 313.15 and 318.15 K are given as a function of mole fraction in Table 1.

Table 1. Ultrasonic velocities (*u*), densities (ρ) and viscosities (η) for the ternary mixtures of quinoline at T = 303.15, 308.15, 313.15, and 318.15 K

x_1	x_3	u (m s^{-1})	ρ (Kg m^{-3})	η x10^3 (Kg m^{-1} s^{-1})	x_1	x_3	u (m s^{-1})	ρ (Kg m^{-3})	η x10^3 (Kg m^{-1} s^{-1})
quinoline (1) + methanol (2) + o-cresol (3)									
T = 303.15 K									
0.0000	0.7765	1482.16	1030.46	6.7070	0.3996	0.3645	1504.52	1063.35	4.7409
0.0765	0.6977	1487.78	1037.46	6.4127	0.4849	0.2764	1509.00	1068.87	4.2840
0.1546	0.6171	1491.16	1045.16	5.9417	0.5722	0.1864	1515.30	1072.80	3.6529
0.2345	0.5347	1495.52	1051.22	5.5147	0.6615	0.0943	1520.26	1078.08	3.0764
0.3161	0.4505	1500.52	1058.30	5.2090	0.7530	0.0000	1527.10	1082.54	2.5834
T = 308.15 K									
0.0000	0.7765	1460.00	1021.66	5.5977	0.3996	0.3645	1499.31	1054.66	3.8353
0.0765	0.6977	1469.70	1029.44	5.3276	0.4849	0.2764	1504.31	1058.72	3.5192
0.1546	0.6171	1478.05	1037.08	4.9588	0.5722	0.1864	1510.60	1063.56	3.0119
0.2345	0.5347	1485.68	1042.75	4.5740	0.6615	0.0943	1515.89	1068.24	2.5174
0.3161	0.4505	1492.89	1048.99	4.1273	0.7530	0.0000	1520.94	1072.32	2.0114
T = 313.15 K									
0.0000	0.7765	1443.15	1016.90	3.9098	0.3996	0.3645	1488.00	1050.40	2.6320
0.0765	0.6977	1453.50	1022.86	3.6617	0.4849	0.2764	1495.43	1054.40	2.4641
0.1546	0.6171	1464.05	1030.72	3.3695	0.5722	0.1864	1500.47	1059.28	2.0456
0.2345	0.5347	1474.16	1037.98	3.1722	0.6615	0.0943	1505.78	1062.52	1.7551
0.3161	0.4505	1481.52	1043.94	2.9141	0.7530	0.0000	1511.47	1066.08	1.4592
T = 318.15 K									
0.0000	0.7765	1421.05	1011.86	2.3176	0.3996	0.3645	1479.15	1044.31	1.6801
0.0765	0.6977	1434.50	1017.53	2.2703	0.4849	0.2764	1488.31	1049.75	1.5922
0.1546	0.6171	1449.05	1025.29	2.1257	0.5722	0.1864	1494.30	1054.28	1.4256
0.2345	0.5347	1460.68	1031.87	2.0064	0.6615	0.0943	1500.78	1057.90	1.2614
0.3161	0.4505	1469.36	1038.49	1.8341	0.7530	0.0000	1505.10	1061.86	1.0698
quinoline (1) + methanol (2) + m-cresol (3)									
T = 303.15 K									
0.0000	0.7769	1460.32	1023.94	7.2862	0.4019	0.3633	1495.21	1057.45	4.8669
0.0772	0.6975	1469.09	1031.69	6.7052	0.4874	0.2754	1502.37	1063.38	4.3087
0.1559	0.6165	1476.54	1038.05	6.2833	0.5747	0.1855	1509.71	1069.60	3.7283
0.2362	0.5338	1482.67	1045.00	5.7997	0.6639	0.0938	1519.42	1075.85	3.1825
0.3182	0.4495	1489.00	1051.67	5.2968	0.7550	0.0000	1527.10	1082.54	2.5834
T = 308.15 K									
0.0000	0.7769	1444.32	1017.94	6.7614	0.4019	0.3633	1485.47	1048.58	4.3968
0.0772	0.6975	1455.28	1025.39	6.3549	0.4874	0.2754	1494.31	1054.84	3.8336

0.1559	0.6165	1463.66	1031.16	5.8696	0.5747	0.1855	1502.12	1060.65	3.2729
0.2362	0.5338	1470.94	1037.73	5.4038	0.6639	0.0938	1510.30	1065.91	2.6781
0.3182	0.4495	1478.58	1043.32	4.8428	0.7550	0.0000	1520.94	1072.32	2.0114
T = 313.15 K									
0.0000	0.7769	1433.46	1012.57	5.0748	0.4019	0.3633	1478.84	1043.13	3.1958
0.0772	0.6975	1446.17	1018.81	4.6565	0.4874	0.2754	1486.16	1048.11	2.8040
0.1559	0.6165	1455.32	1025.73	4.2961	0.5747	0.1855	1495.15	1054.35	2.3578
0.2362	0.5338	1463.41	1031.42	3.9499	0.6639	0.0938	1502.28	1059.55	1.9521
0.3182	0.4495	1470.52	1037.87	3.5887	0.7550	0.0000	1511.47	1066.08	1.4592
T = 318.15 K									
0.0000	0.7769	1416.25	1007.63	4.3075	0.4019	0.3633	1468.15	1037.42	2.5022
0.0772	0.6975	1427.19	1013.77	3.9387	0.4874	0.2754	1476.68	1042.97	2.1756
0.1559	0.6165	1440.27	1019.90	3.5688	0.5747	0.1855	1485.67	1049.18	1.8232
0.2362	0.5338	1450.37	1026.39	3.2080	0.6639	0.0938	1494.06	1055.06	1.3956
0.3182	0.4495	1460.21	1032.05	2.8562	0.7550	0.0000	1505.10	1061.86	1.0698
quinoline (1) + methanol (2) + p-cresol (3)									
T = 303.15 K									
0.0000	0.7756	1475.32	1024.76	6.9880	0.4030	0.3616	1501.15	1060.41	4.9919
0.0776	0.6960	1481.15	1034.10	6.6442	0.4884	0.2739	1506.69	1065.37	4.5224
0.1566	0.6148	1486.80	1040.95	6.1827	0.5755	0.1844	1512.56	1071.35	3.8995
0.2371	0.5320	1491.30	1048.06	5.7814	0.6643	0.0931	1518.58	1075.81	3.3819
0.3193	0.4476	1496.47	1054.81	5.3935	0.7550	0.0000	1527.10	1082.54	2.5834
T = 308.15 K									
0.0000	0.7756	1451.21	1019.60	5.8979	0.4030	0.3616	1493.42	1051.34	4.0080
0.0776	0.6960	1462.44	1027.33	5.5675	0.4884	0.2739	1499.84	1056.84	3.5814
0.1566	0.6148	1470.89	1033.98	5.1697	0.5755	0.1844	1506.12	1061.32	3.1633
0.2371	0.5320	1478.31	1039.65	4.7147	0.6643	0.0931	1513.29	1066.11	2.6592
0.3193	0.4476	1486.05	1045.83	4.3531	0.7550	0.0000	1520.94	1072.32	2.0114
T = 313.15 K									
0.0000	0.7756	1440.15	1015.20	4.1748	0.4030	0.3616	1486.48	1045.87	2.8418
0.0776	0.6960	1450.19	1021.30	3.8981	0.4884	0.2739	1492.48	1050.80	2.5630
0.1566	0.6148	1460.89	1028.24	3.6093	0.5755	0.1844	1499.31	1055.84	2.2463
0.2371	0.5320	1470.83	1034.58	3.3408	0.6643	0.0931	1504.38	1060.19	1.9253
0.3193	0.4476	1479.15	1040.79	3.0784	0.7550	0.0000	1511.47	1066.08	1.4592
T = 318.15 K									
0.0000	0.7756	1418.58	1009.35	2.5837	0.4030	0.3616	1476.63	1040.50	1.9821
0.0776	0.6960	1430.98	1015.22	2.4877	0.4884	0.2739	1484.84	1045.06	1.8313
0.1566	0.6148	1444.69	1021.61	2.3858	0.5755	0.1844	1491.82	1050.80	1.6034
0.2371	0.5320	1457.15	1028.50	2.2945	0.6643	0.0931	1498.22	1055.79	1.4104
0.3193	0.4476	1466.99	1034.74	2.1767	0.7550	0.0000	1505.10	1061.86	1.0698

The variations of V_m^E, u^E, Δη and G^{*E} with the mole fraction of quinoline for o-cresol, m-cresol, p-cresol in methanol at 303.15, 308.15, 313.15, 318.15 K are plotted in Figure 1(a-c), Figure 2(1-c), Figure 3(a-c) and Fig 4(a-c).



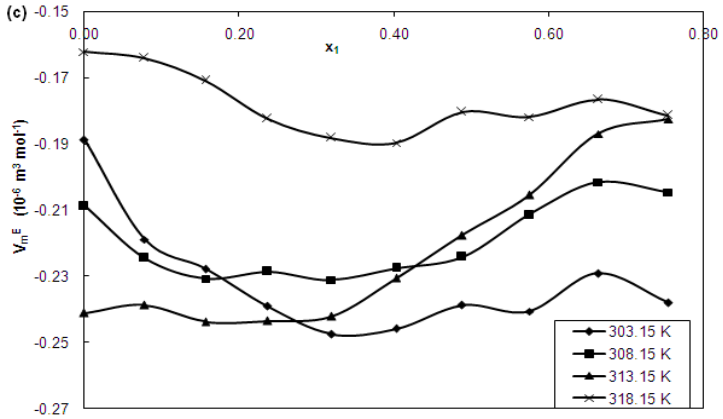


Fig. 1: (a) Variation of excess molar volume, V_m^E, with mole fraction, x_1, for Quinoline+ methanol+o-cresol mixtures at different temperatures:♦,303.15K; ■,308.15K; ▲, 313.15K; and ×, 318.15K. (b) Variation of excess molar volume, V_m^E, with mole fraction, x_1, for Quinoline+ methanol+m-cresol mixtures at different temperatures: ♦,303.15K; ■,308.15K; ▲, 313.15K; and ×, 318.15K. (c) Variation of excess molar volume, V_m^E, with mole fraction, x_1, for Quinoline+ methanol+p-cresol mixtures at different temperatures: ♦,303.15K; ■,308.15K; ▲, 313.15K; and ×, 318.15K

The results shown in Figures 1(a-c) indicate that V_m^E values are negative over the entire mole fraction range and at all temperatures reported. Generally, dispersive forces and improper interstitial accommodation of molecules of a ternary mixture will be reflected in positive V_m^E and negative $\Delta\eta$ values [16,17]. However, strong interactions taking place between unlike molecules through charge transfer forces, the formation of new hydrogen bonds and proper interstitial accommodation will yield negative V_m^E and positive $\Delta\eta$ values.

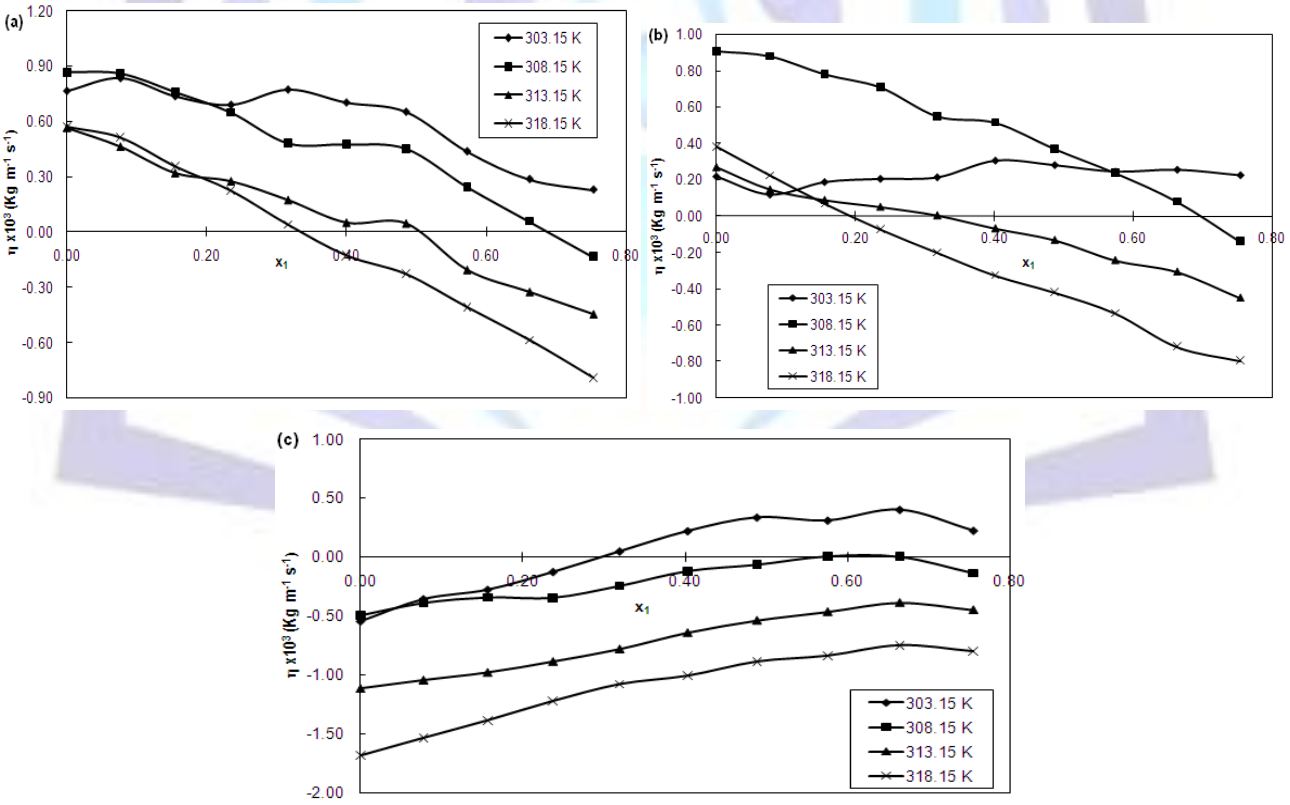


Fig. 2: (a) Variation of deviation in viscosity, $\Delta\eta$, with mole fraction, x_1, for Quinoline+ methanol+o-cresol mixtures at different temperatures:♦,303.15K; ■,308.15K; ▲, 313.15K; and ×, 318.15K. (b) Variation of deviation in viscosity, $\Delta\eta$, with mole fraction, x_1, for Quinoline+ methanol+m-cresol mixtures at different temperatures: ♦,303.15K; ■,308.15K; ▲, 313.15K; and ×, 318.15K. (c) Variation of deviation in viscosity, $\Delta\eta$, with mole fraction, x_1, for Quinoline+ methanol+p-cresol mixtures at different temperatures: ♦,303.15K; ■,308.15K; ▲, 313.15K; and ×, 318.15K

From Figures 2(a,b) it is observed that as the mole fraction of quinoline increases the Δη values becomes negative and at higher temperatures. From Figure 2(c) it is observed that the Δη values are negative at lower mole fraction of quinoline and becomes positive at higher mole fractions and at temperature 303.15K only.

The liquid mixture volume depends upon intermolecular interactions as well as structural arrangement [18]. An increase in the strength of the heteromolecular forces manifests in a decrease in adiabatic compressibility and volume and, hence the size of cluster. Thus, negative V_m^E values account for the strong interactions between the unlike molecules. Thus excess molar volume and deviation in viscosity complements the presence of strong interactions in the first two systems and at lower mole fraction of quinoline.

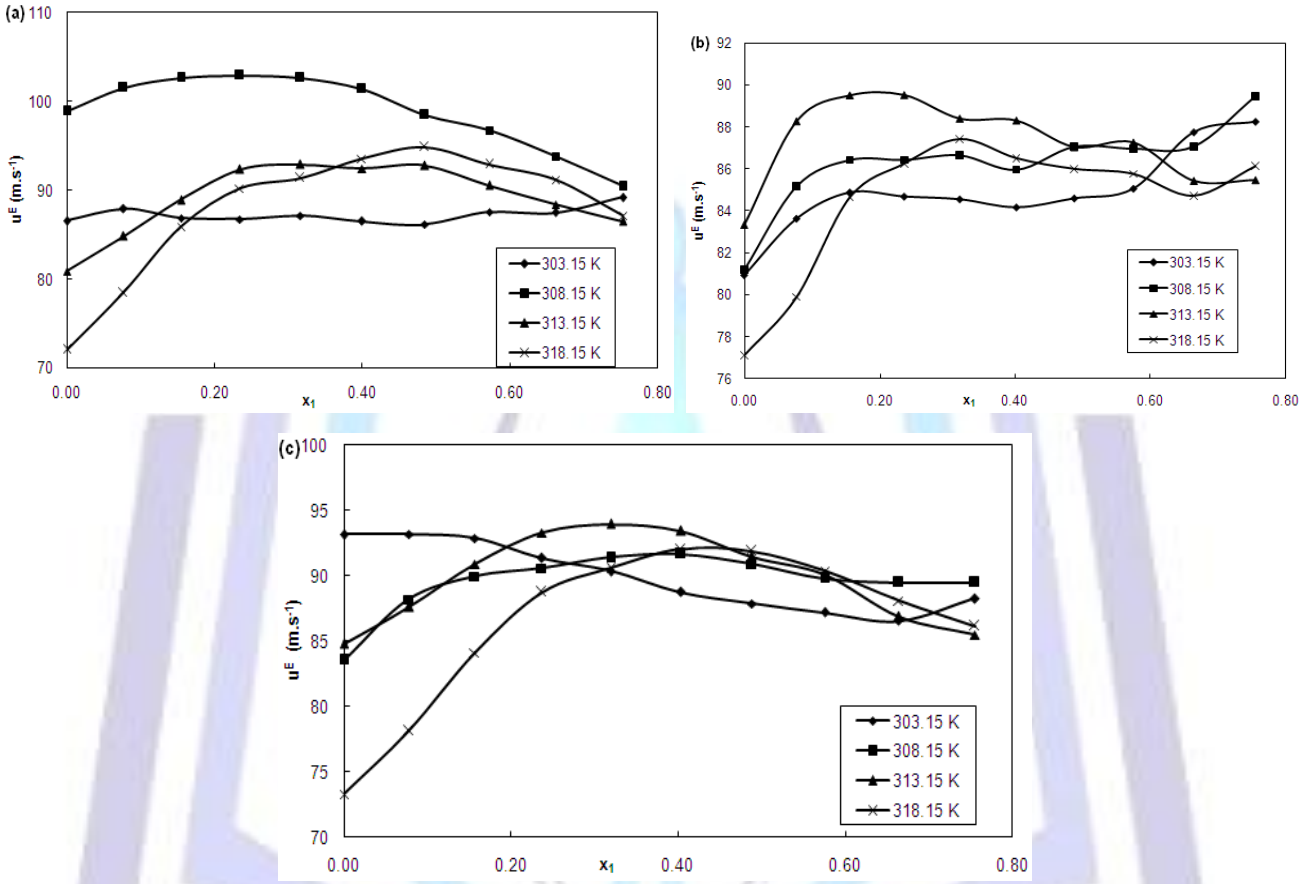
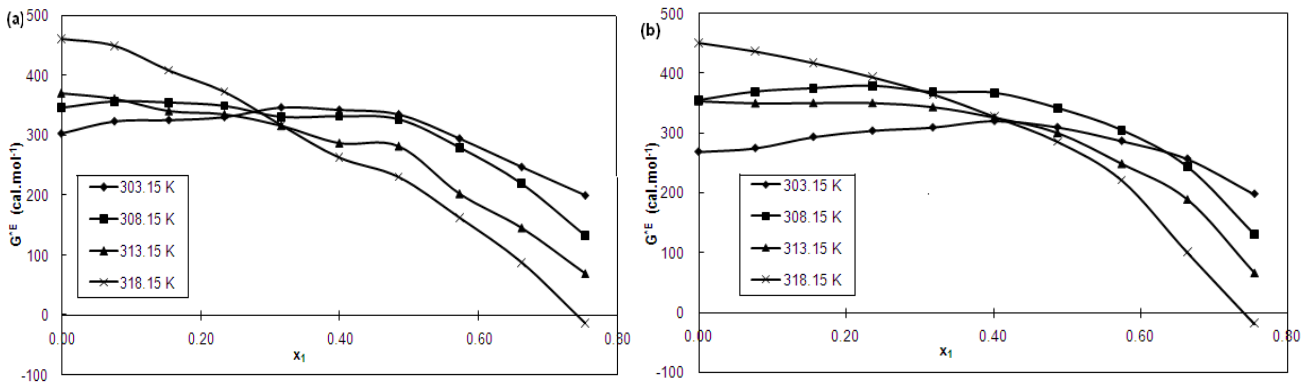


Fig. 3: (a) Variation of excess velocity, u^E, with mole fraction, x_1, for Quinoline+ methanol+o-cresol mixtures at different temperatures:♦,303.15K; ■,308.15K; ▲, 313.15K; and ×, 318.15K. (b) Variation of excess velocity, u^E, with mole fraction, x_1, for Quinoline+ methanol+m-cresol mixtures at different temperatures: ♦,303.15K; ■,308.15K; ▲, 313.15K; and ×, 318.15K. (c) Variation of excess velocity, u^E, with mole fraction, x_1, for Quinoline+ methanol+p-cresol mixtures at different temperatures: ♦,303.15K; ■,308.15K; ▲, 313.15K; and ×, 318.15K

From Figures 3(a-c) it is observed that the excess ultrasonic velocity values for all the mixtures are positive. The positive deviations in u^E from linear dependence suggest the presence of strong interactions between the component molecules [19].



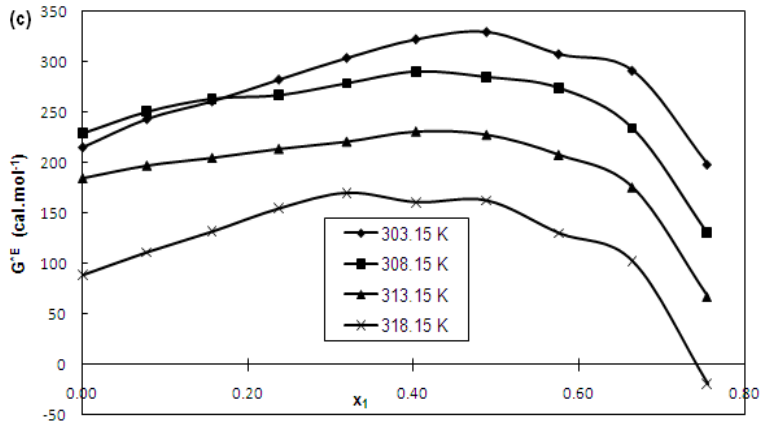


Fig. 4: (a) Variation of excess Gibb's free energy, G^{*E}, with mole fraction, x_1, for Quinoline+ methanol+o-cresol mixtures at different temperatures:♦,303.15K; ■,308.15K; ▲, 313.15K; and ×, 318.15K. (b) Variation of excess Gibb's free energy, G^{*E}, with mole fraction, x_1, for Quinoline+ methanol+m-cresol mixtures at different temperatures: ♦,303.15K; ■,308.15K; ▲, 313.15K; and ×, 318.15K. (c) Variation of excess Gibb's free energy, G^{*E}, with mole fraction, x_1, for Quinoline+ methanol+p-cresol mixtures at different temperatures: ♦,303.15K; ■,308.15K; ▲, 313.15K; and ×, 318.15K

The observed positive values of G^{*E} (Figures 4(a-c)) in all the mixtures indicate the presence of strong interactions between the components of the mixtures [20]. From all the above results we may conclude that there exist stong interactions between the molecules of the mixtures in all the three systems choosen for the study.

CONCLUSIONS

Thermodynamic parameters are sensitive to the molecular interactions present in the liquid mixtures; it supports the results of the present study. The negative values of V_m^E and positive values of $\Delta\eta$, u^E and G^{*E} in all the three mixtures reveal the presence of strong interactions between the component molecules in the mixtures.

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