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Densities, viscosities and ultrasonic velocities in ternary liquid mixture of anisole with cyclohexanone and 1- hexanol at 308.15K and 318.15K

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ABSTRACT

Densities , viscosities and ultrasonic velocities has been measured for the ternary mixture involving anisole(1) + cyclohexanone(2) + 1- hexanol(3) at 308.15K and 318.15K over the entire range of mole fraction. Parameters like excess volume, viscosity, adiabatic compressibility, free volume, linear free energy, acoustic impedance, relative association and isothermal compressibility were calculated. From these values the excess properties are also calculated. The deviations of the liquid mixture from ideality have been explained based on the molecular interaction between unlike molecules.

Keywords: density, viscosity, ultrasonic velocity, adiabatic compressibility, acoustic impedance.

INTRODUCTION

The study and understanding of thermodynamic and transport properties of pure liquids, liquid mixtures and solutions have found wide application in chemical, textile, leather, pharmaceutical, nuclear industries and many others. The measurements of ultrasonic velocity in pure liquids and liquid mixtures have been found to be important in predicting the physico- chemical properties of liquid mixtures (1-3). Various studies have been carried out in predicting the nature of interactions between unlike molecules (4-6). The study of nature of interactions in liquid mixtures involving anisole (7-9), cyclohexanone (10-12) and 1- alcohols (13,14) have been studied . However only few work have been reported for ternary liquid mixtures involving anisole (15,16). We report here the nature of interactions in the ternary liquid mixture of anisole with cyclohexanone and 1- hexanol from density, viscosity and ultrasonic velocity measurements at 308.15K and 318.15K. From these values excess volume, adiabatic compressibility, free volume, linear free energy, acoustic impedance, relative association and isothermal compressibility were calculated. Excess values for all these parameters were calculated and fitted to Redlich-Kister polynomial to derive the ternary coefficients and standard deviations between the calculated and fit parameters.

EXPERIMENTAL SECTION

Anisole (Nice Chemicals, Cochin), cyclohexanone (Merck, Mumbai) and 1- hexanol (Loba chemie, Mumbai), all analar grades, were dried using suitable drying agents and distilled based on standard methods (17). Ternary liquid mixtures of various compositions were prepared by mixing measured amount of pure liquids in air tight stoppered bottles of 50ml capacity. Densities of pure liquids and liquid mixtures were measured by relative density method using 10ml relative density bottle with an accuracy of ± 0.001kgm^{-3}. Viscosities of all pure and liquid mixtures were measured using Ostwald viscometer of 10ml capacity with an accuracy of ±0.001cP. Ultrasonic velocities of pure and liquid mixtures were measured by a single crystal variable path interferometer (Mittal Enterprises, New Delhi, Model F-80) at a frequency of 2MHz with an accuracy of ±0.02%. All the measurements were made at both 308.15K and 318.15K with the help of a digital thermostat with a temperature accuracy of ± 0.01K.

RESULTS AND DISCUSSION

The experimental densities (ρ), viscosity (η) and ultrasonic velocity (u) for the pure liquids are presented in Table 1.

Table 1. Densities, viscosities and ultrasonic velocities of pure liquids

	Temperature	Density 10^3Kgm^{-3}	Viscosity cP	Ultrasonic velocity ms^{-1}
Anisole	308.15K	0.9791	1.0941	1372
	318.15K	0.9698	0.9657	1334
Cyclohexanone	308.15K	0.9312	1.6562	1358
	318.15K	0.9234	1.3809	1275
1-Hexanol	308.15K	0.8093	4.4983	1260
	318.15K	0.8017	3.3797	1198

Adiabatic compressibility (Ks) has been calculated from Laplace's equation

$$Ks = 1/\rho u^2 \qquad (1)$$

in which ρ and u are density and ultrasonic velocity in liquid mixture.

Acoustic impedance (Z) has been calculated by the relation (18)

$$Z = u\,\rho \qquad (2)$$

Linear free energy has been calculated by Jacobson's relation (19)

$Lf = K/u\rho^{1/2}$ (3)

K is Jacobson's constant which is temperature dependent constant but independent of the nature of the liquid.

Viscosity has been calculated using the relation

$\eta = (At - B/t)\rho$ (4)

A and B are constants characteristic of viscometer calculated using standard liquids water and nitrobenzene, t time of flow.

Surianarayana (20) proposed a relation to calculate free volume

$V_f = (M_{eff}\, u/K\, \eta)^{3/2}$ (5)

K is a temperature independent constant which is equal to 4.28×10^9 for all liquids; M_{eff} is effective molecular weight of the mixture calculated using the relation

$M_{eff} = x_1M_1+x_2M_2+x_3M_3$. Where x_1, x_2, x_3, M_1, M_2, M_3 are mole fractions and molar masses of the pure components 1, 2 and 3.

Relative association has been calculated using the relation

$Ra = (\rho/\rho_1)(u_1/u)^{1/3}$ (6)

Isothermal compressibility has been calculated using the relation

$\beta_T = 1.71 \times 10^{-3}/(T^{4/9}u^2\rho^{4/3})$ (7)

T is absolute temperature

Thermal expansion coefficient has been calculated using the relation

$\alpha = (0.0191\beta_T)^{1/4}$ (8)

Excess volume (V^E) has been calculated using the relation

$V^E = ((x_1M_1+x_2M_2+x_3M_3)/\rho) - (x_1M_1/\rho_1) - (x_2M_2/\rho_2) - (x_3M_3/\rho_3)$ (9)

ρ_1, ρ_2 and ρ_3 are densities of pure components 1,2 and 3.

Excess adiabatic compressibility (ΔKs) has been calculated from the relation

$\Delta Ks = Ks - (\varphi_1 Ks_1 + \varphi_2 Ks_2 + \varphi_3 Ks_3)$ (10)

Ks_1, Ks_2, Ks_3 are adiabatic compressibility values of pure liquids and φ_1,φ_2 and φ_3 are volume fraction for pure liquids calculated by the relation

Table 2: Densities, viscosities, ultrasonic velocities and acoustic parameters for the ternary mixture of anisole (1) + cyclohexanone (2) + 1- Hexanol(3) at 308.15K

x_1	x_2	Density $10^3 Kgm^{-3}$	V^E $10^3 m^3 mol^{-1}$	U ms^{-1}	φ_1	φ_2	ΔKs TPa^{-1}	M_{eff}	Ra
0.0472	0.8700	0.9183	0.3870	1362	0.0485	0.8541	-9.2266	*98.9463*	0.9402
0.0493	0.4927	0.8681	0.5537	1214	0.0473	0.4507	104.5433	100.4835	0.9235
0.0525	0.2244	0.8380	0.4860	1196	0.0481	0.1958	106.4875	101.5867	0.8959
0.0459	0.7297	0.8992	0.3478	1244	0.0459	0.6972	90.4112	99.5055	0.9021
0.0885	0.8680	0.9274	0.2226	1366	0.0916	0.8570	-7.5564	99.2011	0.9486
0.1031	0.0646	0.8299	0.2647	1168	0.0923	0.0552	137.8543	102.5335	0.8944
0.1343	0.7318	0.9183	0.0547	1356	0.1363	0.7085	-12.2878	100.024	0.9229
0.1409	0.5866	0.8993	0.1366	1216	0.1392	0.5530	117.2092	100.65	0.9562
0.1429	0.2817	0.8611	0.2138	1208	0.1336	0.2513	99.4232	101.8938	0.8711
0.2000	0.1142	0.8510	0.2150	1200	0.1830	0.0997	101.0287	102.9107	0.9089
0.2242	0.7311	0.9344	0.1635	1368	0.2305	0.7170	-8.6012	100.5628	0.9327
0.2293	0.5401	0.9103	-0.0096	1346	0.2275	0.5111	-15.9487	101.3651	0.9136
0.2415	0.2802	0.8771	0.2492	1212	0.2288	0.2533	102.6359	102.4874	0.9337
0.3234	0.5393	0.9254	0.2567	1350	0.3250	0.5172	-4.9992	101.9285	0.9279
0.3311	0.3326	0.8993	0.2450	1328	0.3205	0.3073	-10.3795	102.8091	0.9071
0.3974	0.0563	0.8731	0.6800	1208	0.3695	0.0499	104.1306	104.3215	0.8841
0.4140	0.5385	0.9415	0.3696	1258	0.4216	0.5232	97.3227	102.472	0.9423
0.4731	0.3895	0.9304	0.5128	1350	0.4722	0.3710	-2.4961	103.4259	0.9312
0.4933	0.1130	0.8993	0.2891	1220	0.4693	0.1025	100.595	104.664	0.9551
0.4785	0.2817	0.9204	0.0725	1242	0.4685	0.2633	89.7331	103.893	0.9284
0.5951	0.0579	0.9103	0.2454	1230	0.5684	0.0527	93.0991	105.493	0.9642
0.6164	0.3317	0.9505	0.3662	1358	0.6212	0.3190	3.1932	104.5137	0.9509
0.6827	0.1731	0.9425	0.1417	1354	0.6741	0.1631	-7.1250	105.5495	0.9446
0.8350	0.1150	0.9615	0.3032	1366	0.8335	0.1096	-1.4509	106.6921	0.9835

Table 2 continued

$\Delta\eta$ cP	ΔL_f $/10^6$ m	$\Delta Z/$ $10^3 Kgm^{-2}s^{-1}$	$\Delta\beta_T$ $/10^{-12} K^{-1} Kg^{-1}sm^2$	$\Delta\alpha$ $/10^{-5} K^{-1}Kg^{-1}sm^2$	ΔV_f $/10^{-8} m^3mol^{-1}$
0.0935	-0.1092	2.77	-0.1389	-0.3168	-1.6309
-0.9644	1.3291	-102.44	1.6406	4.9291	-0.1673
-1.0556	1.2861	-89.46	1.6748	4.6823	-0.4626
-0.3504	-1.1846	88.88	-1.3868	-4.3585	0.1994
0.0842	-0.1130	5.91	-0.1370	-0.3616	-1.4396
-0.8623	1.5752	-99.53	2.1179	5.6076	-1.0942
-0.2053	-0.9976	80.04	-1.1491	-3.7157	-0.2020
-0.5526	1.4792	-115.45	1.7633	5.4613	-1.4283
-1.0356	-1.2688	80.89	-1.6156	-4.5744	0.2333
-1.1388	1.2527	-91.20	1.5682	4.5607	-1.3928
-0.0150	-1.1530	102.30	-1.2607	-4.3715	-0.3813
-0.5028	-1.1710	90.13	-1.3826	-4.3518	0.3132
-1.0477	1.3204	-103.38	1.5864	4.8689	-1.1135
-0.3430	-1.0593	85.3756	-1.2086	-3.9539	0.1089
-0.8265	-1.0570	72.23	-1.2993	-3.8556	0.6927
-1.1233	-1.1300	67.25	-1.4485	-4.0396	-0.5792

-0.0950	-0.9992	87.08	-1.0904	-3.7705	-0.2055
-0.3556	-1.0966	88.33	-1.2379	-4.0918	0.4413
-0.9385	1.3233	-109.94	1.5307	4.9099	-2.0309
-0.5963	-0.9219	67.00	-1.1137	-3.4020	0.1330
-0.8521	1.2449	-106.79	1.4089	4.6396	-2.1833
-0.0875	-0.9894	87.31	-1.0733	-3.7471	-0.5078
-0.4102	-1.0358	85.41	-1.1780	-3.8983	1.0999
-0.0973	-0.0073	-4.64	-0.0266	0.0180	-1.3748

Table 3: Densities, viscosities, ultrasonic velocities and acoustic parameters for the ternary mixture of Anisole (1) + Cyclohexanone (2) + 1-Hexanol (3) at 318.15K

x_1	x_2	Density $10^3 Kgm^{-3}$	V^E $10^3 m^3 mol^{-1}$	U ms^{-1}	φ_1	φ_2	ΔKs TPa^{-1}	M_{eff}	Ra
0.0472	0.8700	0.9124	0.1622	1334	0.0485	0.8541	-62.7572	98.9463	0.9408
0.0493	0.4927	0.8632	0.1744	1186	0.0473	0.4507	61.3079	100.4835	0.9256
0.0525	0.2244	0.8320	0.2289	1166	0.0481	0.1958	69.3036	101.5867	0.8973
0.0459	0.7297	0.8933	0.1144	1412	0.0459	0.6972	-150.3430	99.5055	0.9039
0.0885	0.8680	0.9205	0.1086	1132	0.0916	0.8570	182.3281	99.2011	1.0025
0.1031	0.0646	0.8300	-0.9130	1150	0.0923	0.0552	80.1227	102.5335	0.8993
0.1343	0.7318	0.9114	-0.0719	1424	0.1363	0.7085	-142.0980	100.0240	0.9196
0.1409	0.5866	0.8933	-0.1191	1162	0.1392	0.5530	114.5951	100.6500	0.9645
0.1429	0.2817	0.8511	0.5057	1174	0.1336	0.2513	74.0827	101.8938	0.9158
0.2000	0.1142	0.8461	-0.2159	1170	0.1830	0.0997	68.0998	102.9107	0.9114
0.2242	0.7311	0.9285	-0.0925	1424	0.2305	0.7170	-122.8670	100.5628	0.9368
0.2293	0.5401	0.9024	-0.0308	1408	0.2275	0.5111	-138.4370	101.3651	0.9139
0.2415	0.2802	0.8712	-0.0341	1170	0.2288	0.2533	88.2003	102.4874	0.9385
0.3234	0.5393	0.9195	-0.0213	1218	0.3250	0.5172	65.3449	101.9285	0.9773
0.3311	0.3326	0.8933	-0.0464	1198	0.3205	0.3073	67.5116	102.8091	0.9548
0.3974	0.0563	0.8682	0.2322	1162	0.3695	0.0499	101.6860	104.3215	0.9374
0.4140	0.5385	0.9365	-0.0344	1444	0.4216	0.5232	-126.3700	102.4720	0.9405
0.4731	0.3895	0.9255	0.0905	1420	0.4722	0.3710	-119.2940	103.4259	0.9346
0.4933	0.1130	0.8933	-0.0337	1196	0.4693	0.1025	71.1225	104.6640	0.9553
0.4785	0.2817	0.9114	0.1344	1212	0.4685	0.2633	68.4340	103.8930	0.9703
0.5951	0.0579	0.9024	0.1666	1200	0.5684	0.0527	76.6503	105.4930	0.9639
0.6164	0.3317	0.9446	0.0445	1238	0.6212	0.3190	68.1936	104.5137	0.9985
0.6827	0.1731	0.9365	-0.2006	1226	0.6741	0.1631	71.0070	105.5495	0.9933
0.8350	0.1150	0.9556	-0.0568	1236	0.8335	0.1096	80.8800	106.6921	1.0108

Table 3 continued

Δη cP	ΔL_f $/10^6$ m	ΔZ/ $10^3 Kgm^{-}s^{-1}$	$\Delta\beta_T$ $/10^{-12} K^{-1} Kg^{-1} sm^2$	Δα $/10^{-5} K^{-1} Kg^{-1} sm^2$	ΔVf $/10^{-8} m^3 mol^{-1}$
0.0513	-0.7720	52.31	-0.9227	-2.7587	-1.0135
-0.6450	0.7841	-60.01	0.9977	2.8809	0.1109
-0.7057	0.8240	-56.47	1.1183	2.9693	-0.2607
-0.1939	-1.7916	127.38	-2.1626	-6.5281	0.5443
0.1242	2.0158	-136.25	2.6177	7.2282	-3.459
-0.7124	0.8902	-54.26	1.2167	3.1357	-0.6291
-0.1625	-1.7919	133.92	-2.0693	-6.4854	0.9959
-0.3756	1.3300	-96.58	1.7250	4.9041	-1.4486
-0.7959	0.8920	-69.95	1.1985	3.4008	-0.2960
-0.7512	0.7971	-61.95	1.0746	3.0700	-1.3053
-0.0325	-1.6584	128.44	-1.8198	-5.9172	0.5567

-0.3487	1.7079	116.53	-1.9922	-5.9782	1.0936
-0.6949	1.0341	-82.37	1.3760	4.0111	-1.1406
-0.2455	0.7018	-65.29	0.9497	2.9849	-1.4165
-0.5498	0.7826	-72.71	1.0436	3.2847	-1.2240
-0.8061	1.1449	-96.24	1.5815	4.6371	-1.8975
-0.0617	1.7889	137.14	-1.8620	-6.1684	0.7830
-0.2693	1.6295	111.60	-1.7250	-5.4035	1.6012
-0.6512	0.7958	-80.93	1.1043	3.5713	-1.7145
-0.3968	0.7362	-76.40	1.0349	3.3569	-1.9793
-0.5939	0.8294	-88.46	1.1834	3.8621	-1.9484
-0.0918	0.6114	-68.43	0.9566	3.0867	-2.3567
-0.3164	0.6783	-77.31	1.0330	3.4179	-1.0822
-0.1423	0.7026	-82.52	1.1472	3.7295	-1.1447

$\Phi 1 = (x_1 M_1/\rho_1)/(x_1 M_1/\rho_1 + x_2 M_2/\rho_2 + x_3 M_3/\rho_3)$ (11)

Excess values of other parameters are calculated using the relation

$A^E = A_{exp} - A_{id}$ (12)

$A_{id} = \sum x_i A_i$, x_i and A_i are mole fraction and parameters of the i^{th} component liquid.

Experimental densities, viscosities, ultrasonic velocities and excess parameters for the ternary system are presented in Table 2 and Table 3 for 308.15K and 318.15K respectively.

All the calculated excess parameters were fitted to Redlich – Kister (21) type polynomial equation

$A^E = x_1 x_2 x_3[a+bx_1(x_2-x_3) + cx_1^2(x_2-x_3)^2]$ (13)

by the method of least squares to derive the adjustable parameters a, b and c. From these a, b and c values theoretical values for all excess parameters were calculated and the standard deviation values were calculated using the relation

$\sigma = [(A^E{}_{exp} - A^E{}_{cal})^2/(n-m)]^{1/2}$ (14)

here n is the number of measurements and m the number of adjustable parameters. The values of a, b, c and σ are given in the table 4a and 4b.

Excess volume values are positive over entire range of mole fraction at 308.15K predicting breaking of liquid order on mixing and presence of non-specific physical interaction and unfavorable interaction between unlike molecules (22).This leads to the expansion of volume during mixing. This nature is also supported by the negative excess value of viscosity. Negative excess values of viscosity predict the easier flow of liquid mixture as compared with the pure liquids. This may be due to difference in size and shape of mixing components and loss of dipolar association in pure liquid component (23).

Table 4a : Adjustable parameters a, b, c and standard deviation values for the excess acoustical parameters at 308.15K.

parameters	a	b	c	σ
V^E (10^3 m^3mol^{-1})	6.6008	-15.5156	688.5430	0.0161
ΔKs (TPa^{-1})	1713	-19107	96939	2.8026
Δ η (cP)	-28.5425	178.507	-400.342	0.0138
ΔL_f (m)	$5.3143x10^{10}$	$-2.1476x10^{12}$	$-1.9159x10^{13}$	0.9762
Δz ($10^3Kgm^{-2}s^{-1}$)	-2293	18134	-85739	2.1177
$\Delta\beta_T$ ($K^{-1}Kg^{-1}sm^2$)	$3.2232x10^{-10}$	$-3.0916x10^{-9}$	$1.1438x10^{-8}$	0.4119
Δα ($K^{-1}Kg^{-1}sm^2$)	$0.1026x10^{-2}$	$-0.9053x10^{-2}$	0.0373	0.1082
ΔV_f (m^3mol^{-1})	$-2.9461x10^{-7}$	$1.0137x10^{-6}$	$-3.4996x10^{-5}$	0.0013

Table 4a : Adjustable parameters a, b, c and standard deviation values for the excess acoustical parameters at 308.15K.

Parameters	a	b	c	σ
V^E (10^3 m^3mol^{-1})	0.6592	-11.0380	48.8179	0.0026
ΔKs (TPa^{-1})	2508	-8549	63320	3.5018
Δ η (cP)	-19.6353	122.347	-313.276	0.0104
ΔL_f (m)	$3.5508x10^7$	$-1.1719x10^8$	$7.3498x10^8$	0.0421
Δz ($10^3Kgm^{-2}s^{-1}$)	-2607	7956	-61324	2.8085
$\Delta\beta_T$ ($K^{-1}Kg^{-1}sm^2$)	$4.1001x10^{-10}$	$-1.4260x10^{-9}$	$7.6541x10^{-9}$	0.5099
Δα ($K^{-1}Kg^{-1}sm^2$)	$0.1228x10^{-2}$	$-0.4009x10^{-2}$	0.0260	0.1432
ΔV_f (m^3mol^{-1})	$-3.6719x10^{-7}$	$1.2762x10^{-7}$	$-2.8825x10^{-5}$	0.0667

The above nature of interaction is also predicted by the positive and low negative values of excess adiabatic compressibility and excess linear free energy. As temperature is raised to 318.15K the excess values decreases, may be due to opening up of pure component aggregates and thus increasing the nature of interaction. The excess values of Z and V_f are negative and the negative values decreases with increase in temperature. The excess values of β_T and α are positive and their values decreases with temperature. Though anisole and 1- hexanol are dipolar and self aggregated molecules, may cause expansion of volume by mixing with cyclohexanone due to difference in size and shape. This may lead to non-specific interaction between the three mixing components. At 318.15K the excess properties of Z, β_T, α and V_f shows similar trend but their values decreases predicting increase in nature of interaction between the mixing components at higher temperatures.

CONCLUSION

From density, viscosity and ultrasonic velocity, related acoustical parameters and their excess values for the ternary liquid mixtures of anisole, cyclohexanone and 1- hexanol for various mole fractions at 308.15K and 318.15K has been studied. It is found that bond breaking or breaking of liquid order predominates over other type of molecular interactions between the mixing components.

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