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Speed of Sound, Density, Viscosity and Related Thermodynamic Excess Properties of Binary Liquid Mixtures Containing Butyl Carbitol with 1-Butanol, Toluene and Diethylmalonate at 308.15 and 318.15 K

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In present communication, measurements of experimental density, viscosity and ultrasonic velocity of binary liquid mixtures of butyl carbitol with 1-butanol, toluene and diethylmalonate have been carried out at 308.15 and 318.15 K at atmospheric pressure over the entire composition range of mole fractions. To study the intermolecular interactions present in these liquid mixtures, thermodynamic parameters like V^E, ΔK_S, $\Delta\eta$, ΔL_f, ΔV_f and ΔZ have been calculated. The excess functions and their deviations from ideality have also been correlated using Redlich-Kister type polynomial equation by the method of least-squares. The variations of thermodynamic parameters with composition and temperature have been discussed in terms of molecular interaction in these liquid mixtures. Further, IR spectra of these liquid mixtures have also been recorded and the obtained results have been used to analyze the mixing behaviour of the components.

Keywords: Butyl carbitol, Molecular interaction, Ultrasonic sound, Thermodynamic parameters.

INTRODUCTION

The thermodynamic properties of pure liquid and liquid mixtures are utilized to investigate the intermolecular interactions between the different components of the liquid mixtures and understand engineering applications concerning mass transfer, heat transfer and fluid flow [1]. Glycol ethers [2] are used as scrubbing liquids for absorption of acid gases exhausting from industrial plants, as octane number enhancer, as the polar additive in anionic polymerization and automotive brake fluid as heat absorbents in pumps and chillers and in biomedical processes. Diethylmalonate is used in the synthesis of vitamin B_1 and B_6, pharmaceuticals, agro chemicals and in the industry of flavours and fragrances [3]. Toluene [4] is used in industries as solvent in paint thinner, nail polish removers, glues and correction fluids.

In this work, we report the densities, viscosities and sound velocities for the binary liquid mixtures (butyl carbitol + 1-butanol, toluene and diethylmalonate) over the entire compo-sition range at 308.15 and 318.15 K. These studies are aimed to provide information on the nature of interactions present in these mixtures.

EXPERIMENTAL

Toluene (Merck Chemicals, India, purity > 99%) and 1-butanol (Thermo-Fisher Scientific, India, purity > 99%) was purified by distillation, while butyl carbitol (SRL chemicals, India, purity > 99%) and diethylmalonate (SRL Chemicals, India, purity > 99 %) was used without further purification. The purity was also checked by comparing the measured densities, viscosities and ultrasonic velocities of the pure compounds at various temperatures with those reported in the literature, showing a good agreement and are shown in Table-1.

A series of nine compositions were prepared for each mixture and their physical properties were also measured at the relevant compositions in the mole fraction scale from 0.1 to 0.9 in steps of 0.1. Binary liquid mixtures of different compositions were prepared by mixing fixed quantity of pure liquids in air tight stopper bottles of 50 mL capacity [10] using an analytical balance with an accuracy of ± 0.0001 g. Density, viscosity and ultrasonic velocity of pure and liquid mixtures, where measured using the method available in literature [11, 12]. All the measurements were done at both 308.15 and 318.15 K

TABLE-1
COMPARISON OF EXPERIMENTAL DENSITY (ρ), VISCOSITY (η), ULTRASONIC VELOCITY (U) OF PURE LIQUIDS WITH LITERATURE VALUE AT 308.15 K AND 318.15 K

Liquids	T (K)	ρ (g cm^{-3})		η (mPa s^{-1})		U (m s^{-1})	
		Exp.	Lit.	Exp.	Lit.	Exp.	Lit.
Butyl carbitol	308.15	0.9395	0.9397 [5]	3.6237	3.8721 [5]	1329.5	1326.36 [5]
	318.15	0.9365	–	3.4918	–	1315.5	–
1-Butanol	308.15	0.7981	0.7979 [6]	1.9275	2.008 [6]	1208.5	1209 [6]
	318.15	0.7935	0.7901 [6]	1.8294	1.580 [6]	1196	–
Toluene	308.15	0.8534	0.8378 [7]	0.5037	0.5099 [7]	1273.5	1250 [7]
	318.15	0.8396	–	0.3824	–	1256	–
Diethyl malonate	308.15	1.0440	1.0418 [8,9]	1.6019	1. 6003 [8,9]	1245	1277.0 [8,9]
	318.15	1.0254	1.0283 [8,9]	1.4200	1.4235 [8,9]	1223	1235.5 [8,9]

using a digital thermostat with a temperature precision of ± 0.01 K.

RESULTS AND DISCUSSION

In Tables 2 and 3, the variation of experimental values of density, viscosity and speed of sound and calculated values of V^E, ΔK_S, ΔL_f, $\Delta\eta$, ΔV_f and ΔZ with mole fraction at different temperatures is recorded. The values of excess molar volume (V^E) are of great importance for approaching towards the study of type and extent of intermolecular interactions present in the liquid mixtures [13]. The structural changes of liquid components after mixing are the factors responsible for the values

TABLE-2
PHYSICAL AND THERMODYNAMIC PARAMETERS FOR BINARY LIQUID MIXTURES OF BUTYL CARBITOL WITH 1-BUTANOL, TOLUENE AND DIETHYLMALONATE AT 308.15 K AND 318.15 K

308.15 K						318.15 K					
X_1	ρ (g cm^{-3})	η (mPa s^{-1})	U (m s^{-1})	ΔV^E (cm^3 mol^{-1})	ΔK_S (Tpa^{-1})	X_1	ρ (g cm^{-3})	η (mPa s^{-1})	U (m s^{-1})	ΔV^E (cm^3 mol^{-1})	ΔK_S (Tpa^{-1})
Butyl carbitol + 1-butanol						Butyl carbitol + 1-butanol					
0.0000	0.7981	1.9275	1208	0.0000	0.0000	0.0000	0.7935	1.8294	1196	0.0000	0.0000
0.1043	0.8242	2.2307	1227	-0.1216	-6.8220	0.1043	0.8198	2.0814	1213	-0.0966	-5.6196
0.1654	0.8376	2.4036	1237	-0.1898	-9.8917	0.1654	0.8333	2.2425	1224	-0.1641	-8.8888
0.2681	0.8574	2.6776	1254	-0.2763	-13.8250	0.2681	0.8534	2.5041	1240	-0.2521	-12.4551
0.3494	0.8710	2.8753	1267	-0.3178	-15.6558	0.3494	0.8671	2.6882	1253	-0.2945	-14.4158
0.4705	0.8885	3.1165	1283	-0.3397	-16.2509	0.4705	0.8849	2.9200	1269	-0.3231	-15.0983
0.5742	0.9013	3.2681	1296	-0.3297	-15.5263	0.5742	0.8979	3.0661	1282	-0.3137	-14.3385
0.6476	0.9094	3.3474	1303	-0.3029	-13.9589	0.6476	0.9061	3.1452	1289	-0.2825	-12.6728
0.8028	0.9241	3.4872	1317	-0.1935	-9.3648	0.8028	0.9211	3.2947	1303	-0.1713	-7.8438
0.9023	0.9323	3.5701	1323	-0.1045	-5.2064	0.9023	0.9294	3.3839	1309	-0.0834	-3.4910
1.0000	0.9395	3.6413	1328	0.0000	0.0000	1.0000	0.9368	3.4929	1316	0.0000	0.0000
Butyl carbitol + toluene						Butyl carbitol + toluene					
0.0000	0.8534	0.5037	1273	0.0000	0.0000	0.0000	0.8396	0.3824	1256	0.0000	0.0000
0.0562	0.8611	0.7126	1279	-0.0302	-2.1580	0.0562	0.8481	0.5786	1261	-0.0190	-1.5988
0.1448	0.8723	1.0444	1288	-0.0766	-5.8528	0.1448	0.8605	0.9058	1269	-0.0592	-4.2269
0.2012	0.8789	1.2529	1293	-0.0994	-7.3678	0.2012	0.8678	1.1153	1274	-0.0846	-5.7523
0.3098	0.8903	1.6559	1301	-0.1272	-9.1125	0.3098	0.8806	1.5156	1282	-0.1151	-7.3522
0.3676	0.8958	1.8563	1305	-0.1356	-9.5584	0.3676	0.8868	1.7156	1286	-0.1249	-7.6602
0.4556	0.9036	2.1392	1310	-0.1397	-9.2480	0.4556	0.8956	1.9993	1292	-0.1312	-7.5731
0.6164	0.9162	2.5990	1318	-0.1292	-8.0384	0.6164	0.9099	2.4599	1300	-0.1195	-6.1895
0.7736	0.9268	3.0203	1324	-0.0952	-5.7084	0.7736	0.9219	2.8741	1307	-0.0848	-3.5855
0.8477	0.9312	3.2167	1326	-0.0680	-4.0310	0.8477	0.9269	3.0685	1309	-0.0575	-1.9974
1.0000	0.9395	3.6237	1329	0.0000	0.0000	1.0000	0.9365	3.4918	1315	0.0000	0.0000
Butyl carbitol + diethyl malonate						Butyl carbitol + diethyl malonate					
0.0000	1.0421	1.6069	1246	0.0000	0.0000	0.0000	1.0264	1.4219	1230	0.0000	0.0000
0.1096	1.0298	1.8644	1257	-0.0244	-1.7442	0.1096	1.0158	1.6714	1241	-0.0190	-0.9544
0.1644	1.0238	1.9942	1262	-0.0399	-2.6483	0.1644	1.0106	1.8028	1246	-0.0311	-1.7189
0.3013	1.0091	2.3188	1276	-0.0757	-4.6927	0.3013	0.9979	2.1361	1260	-0.0685	-3.4324
0.4015	0.9986	2.5483	1285	-0.0936	-5.8384	0.4015	0.9887	2.3676	1270	-0.0887	-4.8069
0.5012	0.9883	2.7619	1294	-0.0989	-6.5551	0.5012	0.9798	2.5844	1279	-0.0943	-5.7086
0.6235	0.9759	2.9957	1304	-0.0939	-6.0242	0.6235	0.9689	2.8236	1289	-0.0886	-5.2098
0.7397	0.9644	3.1943	1312	-0.0760	-4.6331	0.7397	0.9588	3.0239	1297	-0.0687	-3.3463
0.7837	0.9601	3.2678	1314	-0.0645	-3.6262	0.7837	0.9550	3.0956	1300	-0.0565	-2.1423
0.8856	0.9503	3.4414	1320	-0.0349	-1.5486	0.8856	0.9463	3.2774	1307	-0.0264	-0.5741
1.0000	0.9395	3.6416	1328	0.0000	0.0000	1.0000	0.9368	3.4930	1316	0.0000	0.0000

TABLE-3
THERMODYNAMIC PARAMETERS FOR BINARY LIQUID MIXTURES OF BUTYL CARBITOL WITH 1-BUTANOL, TOLUENE AND DIETHYLMALONATE AT 308.15K AND 318.15 K

308.15 K					318.15 K				
Δη (mPa s)	ΔL_F (10^{-10} m)	ΔV_F (10^{-14} m^3 mol^{-1})	ΔZ (kg m^{-3} s^{-1})	Δπ (Pa)	Δη (mPa s)	ΔL_F (10^{-10} m)	ΔV_F (10^{-14} m^3 mol^{-1})	ΔZ (kg m^{-3} s^{-1})	Δπ (Pa)
Butyl carbitol + 1-butanol					Butyl carbitol + 1-butanol				
0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.1246	-0.9446	-0.0097	17.3258	-644.5605	0.0785	-0.7456	-0.0071	16.1058	-645.90
0.1926	-1.3352	-0.0145	25.2802	-813.1158	0.1378	-1.1612	-0.0123	23.9715	-806.43
0.2907	-1.7722	-0.0210	35.2672	-915.7888	0.2287	-1.6129	-0.0198	33.4419	-901.34
0.3489	-1.9486	-0.0248	40.1655	-903.7941	0.2775	-1.8040	-0.0235	38.2822	-888.01
0.3825	-1.9779	-0.0273	42.6553	-805.1479	0.3078	-1.8435	-0.0260	40.7596	-789.71
0.3566	-1.8342	-0.0260	41.0338	-677.6394	0.2815	-1.6973	-0.0241	39.1007	-664.24
0.3101	-1.6413	-0.0231	37.5213	-573.3188	0.2385	-1.4963	-0.0207	35.5325	-561.83
0.1838	-1.0564	-0.0142	25.2745	-330.5452	0.1299	-0.8868	-0.0113	23.2495	-323.80
0.0963	-0.5656	-0.0076	13.9267	-165.7449	0.0536	-0.3744	-0.0045	11.9033	-162.40
0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
Butyl carbitol + toluene					Butyl carbitol + toluene				
0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.0335	-0.2193	-4.3473	5.4495	106.9350	0.0214	-0.1722	-0.5859	4.9043	88.60
0.0890	-0.5138	-5.6359	13.2458	102.0422	0.0732	-0.3903	-3.9746	11.7003	91.04
0.1215	-0.6460	-5.6281	16.8992	93.7047	0.1073	-0.5449	-4.3350	15.2973	86.14
0.1856	-0.8060	-5.1505	21.6092	74.9683	0.1699	-0.7345	-4.1800	19.8228	71.29
0.2058	-0.8485	-4.7859	23.0236	64.5524	0.1903	-0.7868	-3.9242	21.1078	62.19
0.2140	-0.8521	-4.1710	23.4370	49.7120	0.2002	-0.8118	-3.4486	21.7365	48.65
0.1722	-0.7489	-2.9700	21.1684	27.9746	0.1609	-0.7183	-2.4713	19.3303	27.97
0.1032	-0.5218	-1.7615	15.1437	13.3514	0.0864	-0.4837	-1.4683	13.0478	13.50
0.0682	-0.3714	-1.1865	10.8771	8.1776	0.0504	-0.3314	-0.9892	8.8630	8.30
0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
Butyl carbitol + diethyl malonate					Butyl carbitol + diethyl malonate				
0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.0345	-0.0424	-0.2958	1.5628	0.5434	0.0225	-0.0243	-0.4480	0.8689	0.4841
0.0528	-0.0653	-0.3734	2.4278	0.7781	0.0405	-0.0443	-0.5670	1.6132	0.7266
0.0988	-0.1188	-0.4516	4.4513	1.2460	0.0901	-0.0912	-0.6783	3.3572	1.2441
0.1244	-0.1497	-0.4425	5.6174	1.4400	0.1141	-0.1290	-0.6641	4.7693	1.4385
0.1353	-0.1694	-0.4005	6.3523	1.4767	0.1245	-0.1537	-0.5959	5.6888	1.4775
0.1201	-0.1569	-0.3209	5.8156	1.2952	0.1103	-0.1414	-0.4745	5.1972	1.3007
0.0824	-0.1213	-0.2289	4.4267	0.9416	0.0701	-0.0918	-0.3368	3.3181	0.9300
0.0663	-0.0949	-0.1920	3.4085	0.7904	0.0505	-0.0592	-0.2814	2.1008	0.7581
0.0326	-0.0402	-0.1036	1.3724	0.4306	0.0214	-0.0159	-0.1513	0.5222	0.4029
0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000

of excess molar volume observed. The factors accountable for the sign and values of excess molar volume are (i) disruption of like polar molecules because of addition of dissimilar type of molecules, (ii) chemical association between the dissimilar molecules through the development of H-bonding, (iii) association between the dissimilar molecules due to formation of weak forces like dipole-dipole interactions and (iv) geometrical accommodation of molecules after their mixing.

The first factor is responsible for the weak interaction between the liquid components leading to positive value of excess molar volume. Other three components are responsible for the specific type of interactions among the unlike molecules leading to negative values of excess molar volume [7]. Also V^E becomes a little less negative with increase in the temperature indicating small effect of temperature on the interactions among the components of liquid mixtures.

Excess molar volume was calculated using the following relation [14]:

$$V^E = \frac{X_A M_A + X_B M_B}{\rho_{AB}} - \frac{X_A M_A}{\rho_A} + \frac{X_B M_B}{\rho_B} \quad (1)$$

where X_A and X_B are mole fraction of components A and B; M_A and M_B are the molecular mass of components A and B, ρ_A and ρ_B are the density of components A and B, respectively and ρ_{AB} is the density of mixture.

Excess viscosity can be calculated using the following relation [15]:

$$\Delta\eta = \eta_{mix} - (X_1\eta_1 + X_2\eta_2) \quad (2)$$

where η_{mix} is a viscosity of mixtures, X_1, η_1 and X_2, η_2 are the mole fractions and viscosity of pure components 1 and 2, respectively.

Values of ultrasonic velocity u and density of mixture (ρ_{AB}) were used to calculate isentropic compressibility (K_S), inter molecular free length (L_f), acoustic impedance (Z) and intermolecular free volume (V_f) using the following relation [16]:

$$K_S = \frac{1}{U^2\rho} \quad (3)$$

$$L_f = \frac{K}{u\rho^{1/2}} \quad (4)$$

$$Z = U\rho \quad (5)$$

where U is ultrasonic velocity of pure liquid and liquid mixtures and ρ is the density of pure and mixture, where K is Jacobson's constant, which is temperature-dependant constant but independent of the nature of liquid.

Intermolecular free volume was calculated using the following relation:

$$V_f = \left(\frac{M_{eff}U}{K\eta}\right)^{3/2} \quad (6)$$

where K is a temperature independent constant that is equal to 4.28×10^9 for all the liquids and M_{eff} is the effective molecular weight of the mixture, calculated using the relation no. 7:

$$M_{eff} = X_1M_1 + X_2M_2 + X_3M_3 \quad (7)$$

where, X_1, X_2, X_3 and M_1, M_2, M_3 are the mole fractions and molar masses of the pure components 1, 2 and 3 respectively. The deviation in isentropic compressibility was obtained from the relation [17]:

$$\Delta K_S = K_S - (\phi_1 K_{S1} + \phi_2 K_{S2}) \quad (8)$$

where ϕ_1 and ϕ_2 are the volume fraction of components 1 and 2, K_{S1} and K_{S2} are the isentropic compressibility of components 1 and 2. K_S is the experimental value of isentropic compressibility of the mixture.

The excess properties V^E, ΔK_S, ΔL_f, $\Delta\eta$, ΔV_f and ΔZ were fitted to Redlich-Kister type [18] polynomial equation:

$$\Delta A = X_1X_2 [a + b (X_1 - X_2) + c (X_1 - X_2)] \quad (9)$$

Least squares method was used to derive the adjustable parameters a, b and c.

The standard deviations (σ) presented in this work were computed using relation no. 10:

$$\sigma = \left(\frac{\Sigma(X_{exp} - X_{cal})^2}{N - n}\right)^{1/2} \quad (10)$$

where N is the number of data points and n is the number of co-efficient. Coefficient values of Redlich-Kister type polynomial equation (eqn. 9) and standard deviation (eqn. 10) at different temperatures are presented in Table-4.

V^E and ΔK_S values of butyl carbitol + 1-butanol are large negative (Figs. 1 and 2) over the entire range of composition. The large negative V^E values of (butyl carbitol + 1-butanol) binary liquid mixtures [19,20] shows the presence of appreciable forces between unlike molecules. This could be due to development of hydrogen bonding between the O atom of –OH group of 2-(2-butoxyethoxy) ethanol and H atom of 1-butanol indicating the strong interactions between the components of the studied binary mixtures.

V^E and ΔK_S values of butyl carbitol + toluene are negative (Figs. 1 and 2) over the entire range of composition. In toluene [21-23], the methyl (electron-donating) group releases electron toward the benzene ring mainly due to hyper conjugation and partly due to inductive effect. Thus, hyperconjugation overcomes on inductive effect in toluene. Due to this reason, the

TABLE-4
COEFFICIENT VALUES OF REDLICH-KISTER TYPE POLYNOMIAL EQUATION AND STANDARD DEVIATION AT DIFFERENT TEMPERATURES

Temp. (K)	Butyl carbitol + 1-butanol				Butyl carbitol + toluene				Butyl carbitol + diethyl malonate			
	a	b	c	σ	a	b	c	σ	a	b	c	σ
	V^E (cm^3 mol^{-1})				V^E (cm^3 mol^{-1})				V^E (cm^3 mol^{-1})			
308.15	-1.3647	0.0139	0.0182	0.0016	-0.5712	0.0082	0.0038	0.0001	-0.3943	-0.0071	0.0173	0.0017
318.15	-1.2880	-1.2880	0.0456	0.0043	-0.5300	-0.5300	0.0170	0.0015	-0.3723	-0.3723	0.0271	0.0026
	η (mPa s)				η (mPa s)				η (mPa s)			
308.15	12.6138	0.8327	2.7362	0.2572	9.0783	1.5293	2.0094	0.1259	11.0099	1.0132	2.5873	0.2494
318.15	11.8209	0.8089	2.5872	0.2441	8.5139	1.5221	1.8664	0.1133	10.3011	1.0303	3.4062	0.2165
	ΔK_s (Tpa^{-1})				ΔK_s (Tpa^{-1})				ΔK_s (Tpa^{-1})			
308.15	-64.4650	-4.3615	1.4466	0.1191	-38.0013	-0.9190	0.0988	0.0675	-25.1341	-0.5186	1.5431	0.1474
318.15	-59.8680	-4.1561	2.9801	0.0580	-30.5313	-0.2020	1.2928	0.0761	-20.9446	-0.2830	2.5551	0.2546
	Δη (mPa s)				Δη (mPa s)				Δη (mPa s)			
308.15	12.6138	0.8328	2.7363	0.0044	9.0784	1.5294	2.0095	0.0040	11.0099	1.0133	2.5873	0.0036
318.15	11.8209	0.8090	2.5872	0.0069	8.5140	1.5222	1.8664	0.0057	10.3011	1.0303	3.4063	0.0049
	ΔZ (kg m^{-3} s^{-1})				ΔZ (kg m^{-3} s^{-1})				ΔZ (kg m^{-3} s^{-1})			
308.15	170.4490	-2.3739	0.2899	0.0676	95.5373	-2.1823	-0.9472	0.0267	24.3264	0.0282	-1.7506	0.1689
318.15	162.5890	-2.8225	-1.1215	0.0648	87.5372	-2.6676	-2.1164	0.0704	20.8677	-0.1269	-2.7021	0.2606
	ΔL_F (10^{-10} m)				ΔL_F (10^{-10} m)				ΔL_F (10^{-10} m)			
308.15	-7.8471	0.3006	-0.0828	0.0131	-3.4722	0.1213	0.0403	0.0028	-0.6513	-0.0033	0.0428	0.0041
318.15	0.2613	0.2169	-0.4294	0.0461	0.1036	0.0291	-0.1081	0.0133	0.0079	0.0071	-0.0151	0.0015
	ΔV_F (10^{-14} m^3 mol^{-1})				ΔV_F (10^{-14} m^3 mol^{-1})				ΔV_F (10^{-14} m^3 mol^{-1})			
308.15	-0.1065	0.0015	0.0021	0.0002	-16.7175	3.0377	-1.8190	0.3578	-1.6253	0.1648	-0.0635	0.0054
318.15	-0.1004	0.0021	0.0059	0.0073	-15.4174	1.7947	0.5459	0.1104	-2.4237	0.2551	-0.0984	0.1808
	Δπ (10^{-05} Nm^{-2})				Δπ (10^{-05} Nm^{-2})				Δπ (10^{-05} Nm^{-2})			
308.15	-31.5636	3.7569	-2.1696	0.2763	1.8588	-0.6563	0.5670	0.0997	5.7671	-0.1358	-0.1949	0.0187
318.15	-30.9358	3.7561	-2.2333	0.2825	1.8731	-0.5812	0.4377	0.0813	5.7835	-0.1233	-0.2779	0.0268

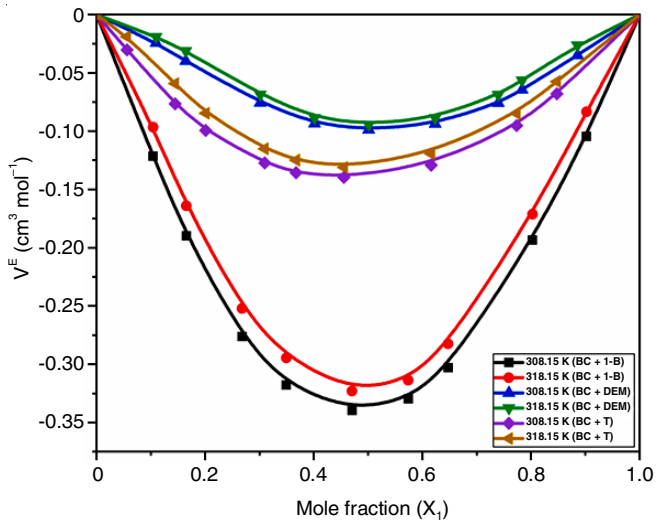

Fig. 1. V^E against the mole fraction of BC at 308.15 K and 318.15 K for the binary mixtures of butyl carbitol (BC) with 1-butanol (1-B), toluene (T) and diethyl malonate (DEM)

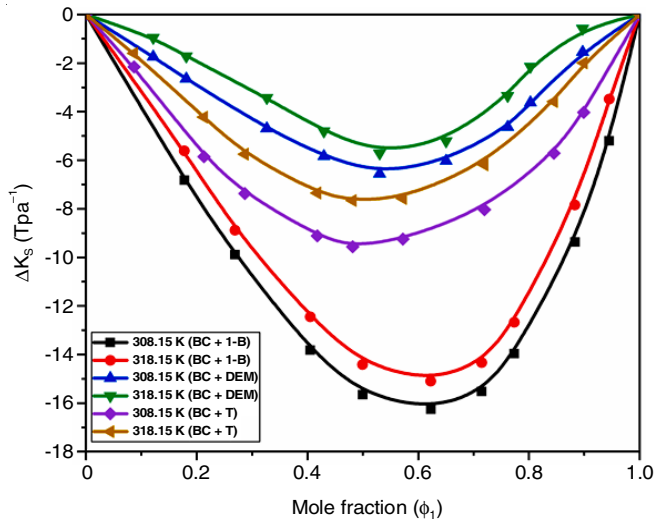

Fig. 2. ΔK_S against the volume fraction of BC at 308.15 K and 318.15 K for the binary mixtures of butyl carbitol (BC) with 1-butanol (1-B), toluene (T) and diethyl malonate (DEM)

negative charge on the methylene group of toluene molecule is stabilized so that hydrogen atom has a positive charge. Thus in all the mixtures, there may be an interaction between the positive charge on hydrogen atom of toluene molecule and with a negative charge on the O atom of –OH group of 2-(2-butoxyethoxy) ethanol, leads to a partial H-bond due to hyperconjugation nature of the toluene molecule.

The V^E and ΔK_S values of butyl carbitol + diethylmalonate are negative (Figs. 1 and 2) over the entire range of composition. The hydrogen atom of the ester group can be attracted by O atom of –OH group of 2-(2-butoxyethoxy) ethanol, hence it involves hydrogen bonding. It is well known that the alkyl group is an electron-donating group and this ability increase with the chain length of the ester molecule [24,25]. Moreover, the negative values in the present investigation is an indication of the effective packing effect caused by interstitial accommodation as the chain length of ester molecules increases, which in turn increases the intermolecular interaction.

Deviations in viscosity of all the three liquid mixtures (Fig. 3) are positive over the entire composition range. Fig. 3 shows the decrease in positive values with increasing temperature. The lowest positive value of butyl carbitol + diethylmalonate indicates that the intermolecular force is larger than that of other mixtures. This supports the earlier suggestion that diethylmalonate shows more interaction than the other liquids with butyl carbitol. The highest positive value of $\Delta\eta$ shows that there is dispersion force between butyl carbitol with 1-butanol and deviates more from ideality. The positive values of $\Delta\eta$ for toluene, diethylmalonate and 1-butanol are consecutively higher than diethylmalonate and lower than 1-butanol, so it reveals that the interaction is less than diethyl malonate and higher than 1-butanol. The order of the $\Delta\eta$ values in the graph matches with V^E and ΔK_S values.

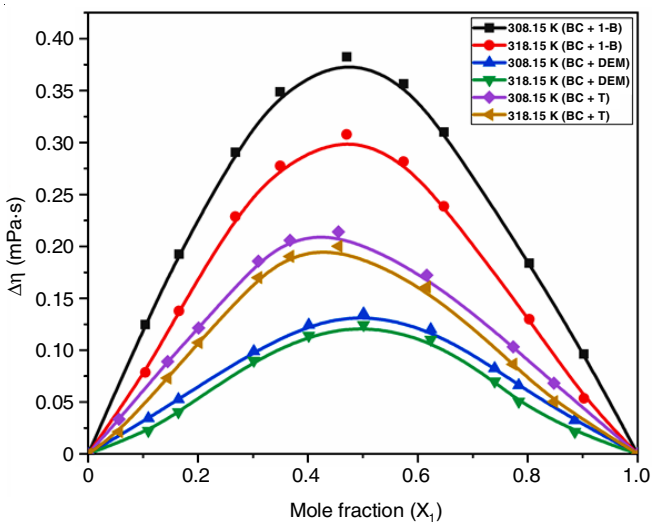

Fig. 3. $\Delta\eta$ against the mole fraction of BC at 308.15 K and 318.15 K for the binary mixtures of butyl carbitol (BC) with 1-butanol (1-B), toluene (T) and diethyl malonate (DEM)

The observed value of ΔL_f, ΔV_f, $\Delta\pi_i$ reflect the same idea as obtained above. The $\Delta\pi_i$ values of the liquid mixtures decreases and ΔV_f values increases with the increase in temperature for the three liquid mixtures. The nature of interaction for the three liquid mixtures decreases as the temperature is increased, due to an increase in the thermal motion of interacting molecules. The negative values [26] of $\Delta\eta$ arise due to dispersion forces between the mixing liquids and positive values arise due to attractive forces like dipole-dipole interaction. The ΔZ behaves in an opposite manner to ΔL_f, positive and negative deviation of the mixtures indicates the extent of association or dissociation between the mixing components. The observed values of $\Delta\eta$ and ΔZ are positive over the entire composition range (Figs. 3 and 5), which strongly supports the above idea. The order of the interaction among the systems is butyl carbitol + diethylmalonate < butyl carbitol + toluene < butyl carbitol + 1-butanol.

FT-IR studies: FTIR results obtained for 1-butanol, toluene and diethylmalonate binary liquid mixtures with butyl carbitol in a molar fraction of 0.5 are shown in Figs. 6-8. According to FT-IR analysis, pure butyl carbitol molecule shows a peak at

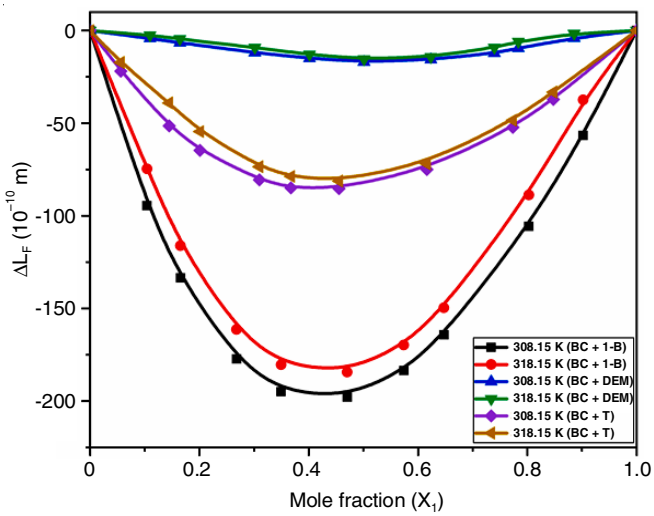

Fig. 4. ΔL_F against the mole fraction of BC at 308.15 K and 318.15 K for the binary mixtures of butyl carbitol (BC) with 1-butanol (1-B), toluene (T) and diethyl malonate (DEM)

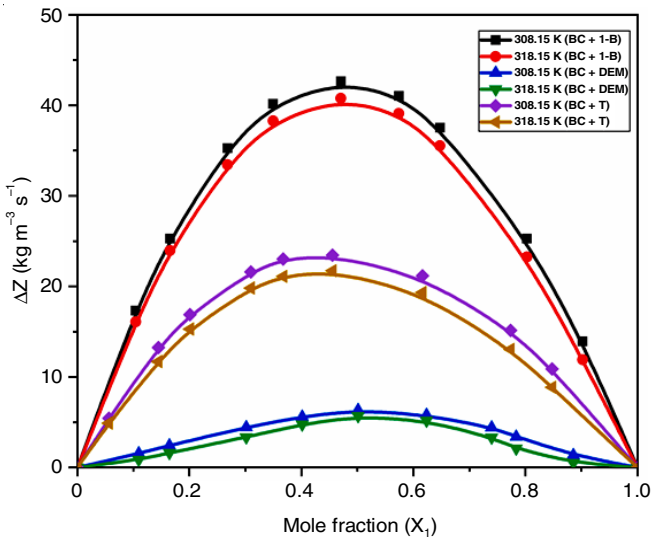

Fig. 5. ΔZ against the mole fraction of BC at 308.15 K and 318.15 K for the binary mixtures of butyl carbitol (BC) with 1-butanol (1-B), toluene (T) and diethyl malonate (DEM)

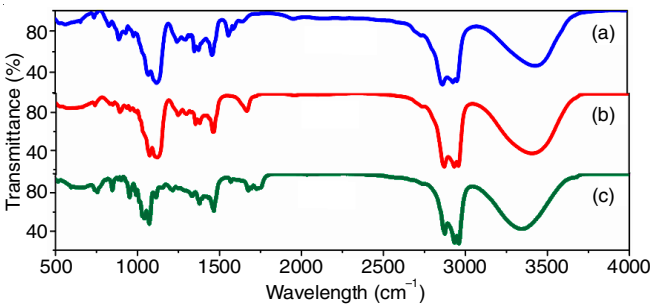

Fig. 6. FT-IR spectrum (a) pure butyl carbitol liquid, (b) equimolar mixture of butyl carbitol + 1-butanol, (c) pure 1-butanol liquid

3441.86 cm^{-1}, which is due to -OH group. Pure 1-butanol molecule has a peak at 3350.14 cm^{-1}, which is characteristic of the –OH group. The equimolar mixture of butyl carbitol + 1-butanol (Fig. 6) shows a broadband at 3417.35 cm^{-1}. The change in frequency from 3441.86 to 3417.52 cm^{-1} and changes in

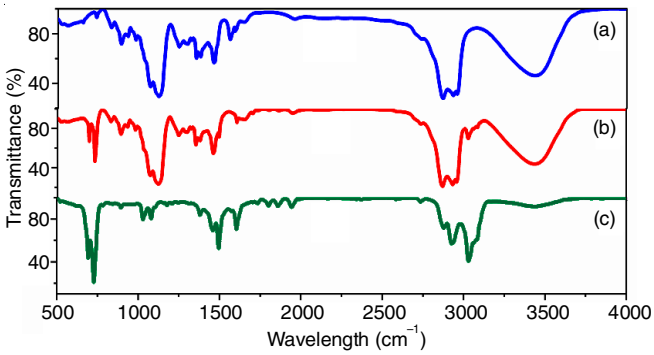

Fig. 7. FT-IR spectrum: (a) pure butyl carbitol liquid, (b) equimolar mixture of butyl carbitol + toluene, (c) pure toluene liquid

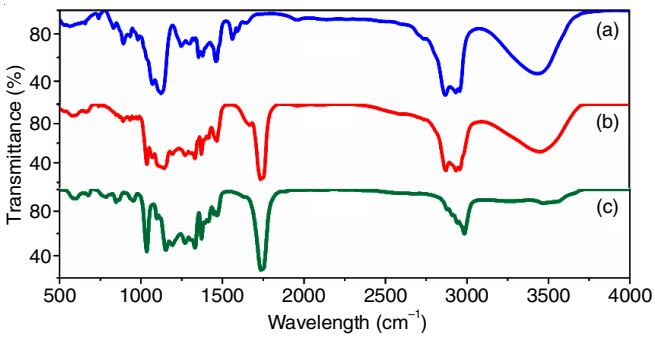

Fig. 8. FT-IR spectrum: (a) pure butyl carbitol liquid, (b) equimolar mixture of butyl carbitol + diethyl malonate, (c) pure diethyl malonate liquid

intensity confirms the existence of an intermolecular hydrogen bond between the hydrogen atom of 1-butanol molecule and O atom of –OH group of 2-(2-butoxyethoxy) ethanol.

Pure toluene molecule shows a peak at 3036.33 cm^{-1} which is due to –C-H bond. The equimolar mixture of butyl carbitol + diethyl malonate (Fig. 7) shows a broad peak at 3447.78 cm^{-1}. The change in frequency from 3341.86 to 3453.78 cm^{-1} and changes in intensity confirmed the existence of an intermolecular hydrogen bond between the hydrogen atom of the toluene molecule and –O atom of –OH group of 2-(2-butoxyethoxy)-ethanol.

Pure diethyl malonate molecule shows a peak at 1745.13 cm^{-1} which is due to C=O bond. The equimolar mixture of butyl carbitol + diethyl malonate (Fig. 8) shows a broad peak at 3453.9.35 cm^{-1}. The change in frequency from 3441.86 to 3453.35 cm^{-1} and changes in intensity confirmed the existence of an intermolecular hydrogen bond between the hydrogen atom of the ester molecule and –O atom of –OH group of 2-(2-butoxyethoxy)ethanol.

Conclusion

In present work, study of the mixing properties of binary liquid mixtures of butyl carbitol with diethylmalonate, toluene and 1-butanol have been carried out. Experimental density data utilized to calculate the excess molar volumes, which display negative deviations as the function of mole fractions at different temperature. The negative values of excess molar volumes suggest the stronger hydrogen bond formations between unlike molecules than those between like molecules. The negative value of excess properties for butyl carbitol + diethyl malonate,

butyl carbitol + toluene and butyl carbitol + 1-butanol shows the existence of H-bonding between them and stronger hydrogen bond formations between butyl carbitol + 1-butanol. The positive and negative deviations in intermolecular free length ΔL_f and acoustic impedance ΔZ respectively suggest the intermolecular interaction among the components of the binary mixtures. The intermolecular interaction decreases with increasing temperature, because of thermal motion. The corresponding thermodynamic excess parameters were calculated with the formulas reported earlier and fitted to a Redlich-Kister type polynomial equation to determine the variable coefficients. The behaviour of the liquid mixtures and the deviation from ideality has been discussed based on experimental and calculated values. The formation of hydrogen bonds between the unlike molecules has been confirmed by the investigation of FT-IR spectroscopy.

CONFLICT OF INTEREST

The authors declare that there is no conflict of interests regarding the publication of this article.

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