Research & Reviews

ISSN: 2320-2459

Research and Reviews: Journal of Pure and Applied Physics

Studyof Ultrasonic Velocity, Densityand Viscosityin the Binary Mixturesof Benzyl Benzoatewith 1-Octanol andIsophorone

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Research Article

Received: 15/03/2013
Revised: 04/04/2013
Accepted: 09/04/2013

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Keywords: Molecular interactions, binary mixture, benzyl benzoate, isophorone, adiabatic compressibility.

ABSTRACT

Ultrasonic velocity, density and viscosity have been measured using the standard techinques in the binary mixtures of benzyl benzoate (1) and 1-octanol (2) and isophorone (2). In the light of the excess parameters like excess adiabatic compressibility, excess internal pressure, excess enthalpy, excess activation energy etc. computed from the measured data, intermolecular interactions are estimated as strong AB interactions in both the mixtures. It is also observed that all the theories except FLT and VANDAEL have a sharp edge in predicting the velocities theoretically.

INTRODUCTION

Through several binary mixtures of benzyl benzoate[1,2,3,4,5,6,7] have been studied ultrasonically, still the investigations are going on. In view of the importance medicinally, the behaviour of benzyl benzoate in cyclic unsaturated ketone, isophorone and a high alkanol, 1-octanol, has been ultrasonically studied in the present investigation. Benzyl benzoate is an insect repellant and used as a solubilizing agent, in oily injections and medicine for scabbies. 1-octanol, a fattyalcolhol, is used in the manufacture of esters used in perfumes and flavouring, in pharmaceutical industry and also in controlling essential tremors and other types of involuntary neurological tremors. Isophorone, which is an α β – unsaturated cyclic ketone, is used as solvent in some printing inks, paintings, lacquers, adhesives and some pesticides. It is also an ingradient in wood preservations and floor sealants. Ultrasonic velocity, density and viscosity have been measured experimentally at three temperatures in the two binary mixtures of benzoate. From knowledge of excess thermodynamic / acoustic parameters computed, the molecular interactions have been investigated / estimated. Also a theoretical evaluation of velocities has been attempted successfully. Almost all the theories except FLT and VANDAEL agree well with the experiment in both the mixtures. And mostly strong AB interactions are suggested in both the binary mixtures at all temperatures.

EXPERIMENTAL

Ultrasonical velocity has been measured using a single crystal variable path interferometer working at 2 MHz with an accuracy of $\pm$ 0 .05%. Density and viscosity have been measured employing a double stem capillary type pyknometer and Ostwald viscometer with accuracies of 2 parts in 10^5 and $\pm$ 0.1% respectively. All the chemicals used here are of analar / fine grade. Weights are taken using a single pan electronic balance with an accuracy of$\pm$ 0.05 mg. Temperature is maintained to within $\pm$0.01K employing an electronically controlled Thermostatic water bath. For standardizing the measurements, triply distilled water has been chosen as a reference liquid.

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Theoretical Aspects:

Employing the standard theories -- FLT due to JACOBSON, CFT due to SCHAAFFS, NOMOTO, VANDAEL, JUNJIEand JOUYBAN – ACREE, ultrasonic velocity can be evaluated theoretically.

$$U_{FLT} = \frac{K}{L_f \rho_{mix}^{1/2}} \quad (1)$$

$$U_{CFT} = \frac{U_\infty S_{mix} B_{mix}}{V_T^M} \quad (2)$$

$$U_{NOMOTO} = \left(\frac{R}{V}\right)^3 = \left[\frac{(X_A R_A + X_B R_B)}{(X_A V_A + X_B V_B)}\right]^3 \quad (3)$$

$$U_{VANDAEL} = \frac{1}{\left[\left(X_A M_A + X_B M_B\right)\left(\frac{X_A}{M_A U_A^{\,2}} + \frac{X_B}{M_B U_B^{\,2}}\right)\right]^{1/2}} \quad (4)$$

$$U_{JUNJIE} = \frac{\frac{X_A M_A}{\rho_A} + \frac{X_B M_B}{\rho_B}}{\left[\left(X_A M_A + X_B M_B\right)\left(\frac{X_A M_A}{V_A^{\,2}\rho_A} + \frac{X_B M_B}{V_B^{\,2}\rho_B}\right)\right]^{1/2}} \quad (5)$$

Velocity(U) in the mixture due to JOUYBAN – ACREE is given by

$$\ln U_{JA} = X_A \ln U_A + X_B \ln U_B + (X_A X_B/T)\,[A_0 + A_1(X_A - X_B) + A_2(X_A - X_B)^2] \quad (6)$$

The thermodynamic/acoustic and other related parameters can be computed from the following relations.

$$\text{Adiabatic Compressibility} \quad \beta = \frac{1}{U_{exp}^2 \rho_{exp}} \quad (7)$$

$$\text{Internal pressure} \quad \pi = bRT\,[K\eta/U]^{3/2}\rho^{2/3} / M^{7/6} \quad (8)$$

$$\text{Free volume} \quad V_f = [M_{eff}\, U / K\eta]^{3/2} \quad (9)$$

$$\text{Enthalpy} \quad H = \pi V_M \quad (10)$$

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$$\text{Activation Energy} \quad G = RT\,[\ln\eta V_M] \quad (11)$$

All the excess parameters are calculated using the following relation

$$A^E = A^{exp} - A^{ideal} = A^{exp} - (X_1A_1 + X_2A_2) \quad (12)$$

All the quantities used in the above equations have their usual meaning as explained elsewhere[8].

RESULTS AND DISCUSSION

Ultrasonic velocity, density and viscosity have been measured experimentally for the two binary systems 1) benzyl benzoate + 1-octanol and 2) benzyl benzoate + isophorone at three temperatures 30,40 and 50 ^{0}C. The results and discussion have been made systematically and separately for the two systems and a comparison is made at the end. All the pure liquids are highly polar (dipole moment =3.9D, 2.0D and 3.96D for Benzyl benzoate, 1-octanol and isophorone respectively).

Benzyl benzoate + 1 - octanol

Ultrasonic velocity, density and viscosity measured experimentally at three temperatures 30, 40 and 50^{o}C have been presented in Table 1. Variation of experimental velocity with concentration has been shown in Fig.1 along with the theoretically evaluated velocities. Velocity increases from octanol to benzyl benzoate at all the three temperatures. From the maximum percentage deviations i.e., -5.26, 2.25, 1.56, - 3.94, 1.11 and 0.39 in FLT, CFT, NOMOTO, VANDAEL, JUNJIE and JOUYBAN – ACREE respectively, it may be inferred that JOUYBAN – ACREE agrees very well while FLT and VANDAEL show large deviations.

From the measured values of viscosity, density and velocity, thermodynamic and other related parameters like adiabatic compressibility (β), internal pressure (π), enthalpy (H), activation energy (G), free length (L_f) etc., are computed and presented in Table 2. It may be seen that β, π and L_f decrease regularly while H also decreases but not regularly. Molar volume (V_m) and G increase from octanol to benzoate at all temperatures. But to assess the nature of chemical reactions / intermolecular interactions, excess parameters would be of much use and hence the computed excess parameters are shown in Figs. 2-7. $L_f^E$and β^E are both negative upto high concentration (~ 0.8m) and thereafter positive at all temperatures. Π^E and H^E are also negative throughout with a minimum nearly at equimolar concentrations (~0.5m) - less negative after 0.5m. η^E and G^E are also negative throughout. It may be observed that almost all excess parameters are negative and behave similarly with temperature. From the negative excess parameters, it may be indicated that there exists strong interamolecular interactions in the binary mixture and small variation in the parameters at high temperatures may be due to the small relative association between the molecules of 1-octanol also. From $\eta^E$and G^E, negative throughout the composition range at all temperatures, exothermic reactions appear to be predominant.

Benzyl benzoate + isophorone

Isophorone is a cyclic unsaturated ketone which readily reacts with benzoate, an ester. As discussed from Fig.8, velocity increases from isophorone to benzoate monotonously and most linearly at all temperatures. Velocity, density and viscosity in this system are shown in Table 3. Maximum percentage deviations observed in various theories are – 4.38, 0.98, -0.17, -4.26, -0.44 and -0.08 in FLT, CFT, NOMOTO, VANDEAL, JUNJIE and JOUYBAN – ACREE respectively at 30^{o}C. Except FLT and VANDAEL, all the other theories agree reasonably with the experiment (<1% deviation). From the measured data experimentally, thermodynamic / other related parameters are computed and shownin Table 4. β and L_f show decreasing trend from isophorone to benzoate while the others π, V_m, H and G increase. The excess parameters computed for understanding the molecular interactions effectively have been shown in Figs. 9-14. It can be seen that β^E, L_f^E, π^E and H^E are all negative throughout at all temperatures while at 50^{o}C G^E and η^E are negative and positive at 40^{o}C throughout the concentration range. At 30^{0}C,both negative and positive are noticed.

In this system also, it is clearly indicated that strong intermolecular interactions of the type AB besides dipole – dipole interactions are predominant at all temperatures. At 30^{o}C, both endothermic and exothermic type of chemical reactions are suggested which transform into endothermic at 40^{o}C.

At this point of journey, a comparison of our results with those of other researchers in similar systems is desirable and hence is presented hereunder.

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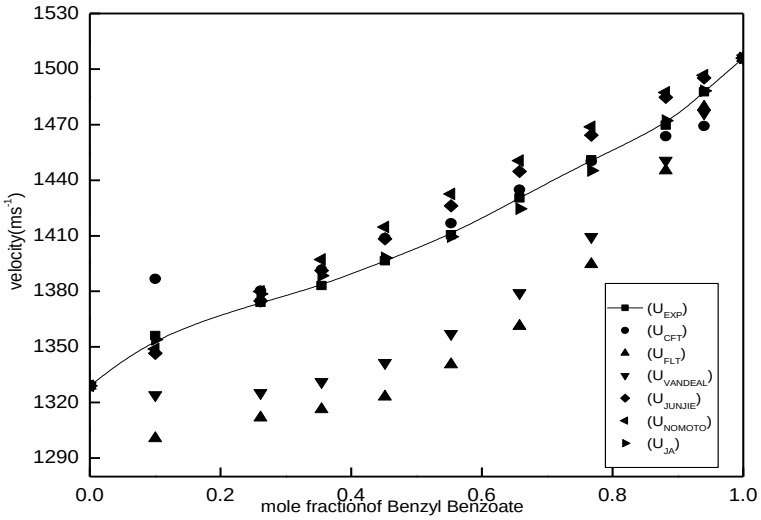

Fig. 1(i). Variation of ultrasonic velocity with the mole fraction of benzyl benzoate at 30°C in the binary mixture: benzyl benzoate + 1-octanol

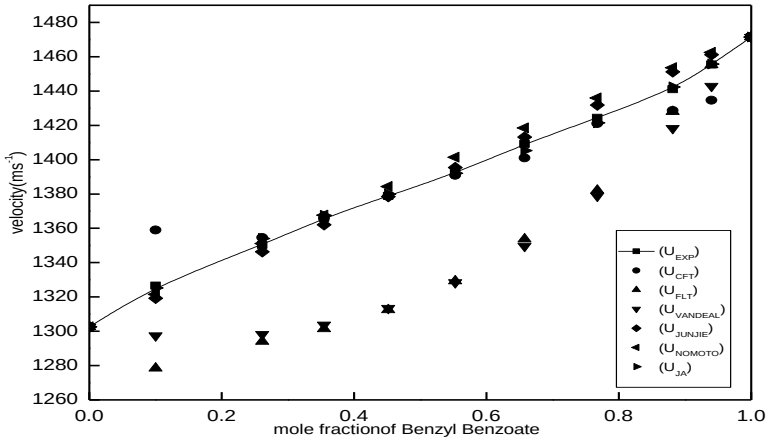

Fig. 1(ii). Variation of ultrasonic velocity with the mole fraction of benzyl benzoate at 40°C in the binary mixture: benzyl benzoate + 1-octanol

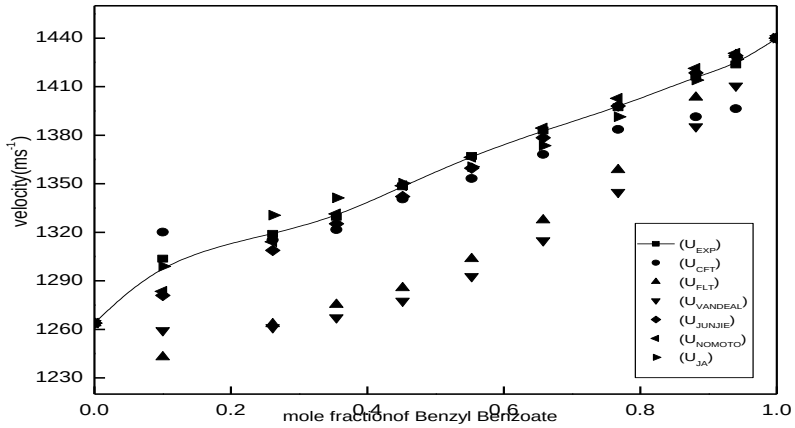

Fig.1(iii). Variation of ultrasonic velocity with the mole fraction of benzyl benzoate at 50°C in the binary mixture: benzyl benzoate + 1-octanol

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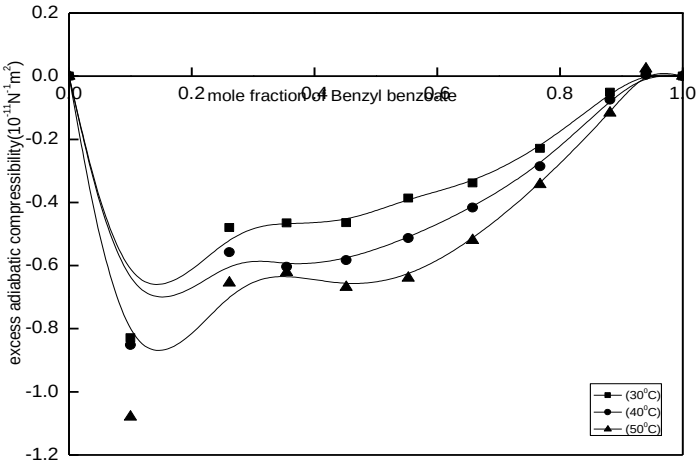

Fig. 2. Variation of excess adiabatic compressibility with the mole fraction of benzyl benzoate at three different temperatures in the binary mixture: Benzyl benzoate + 1-octanol

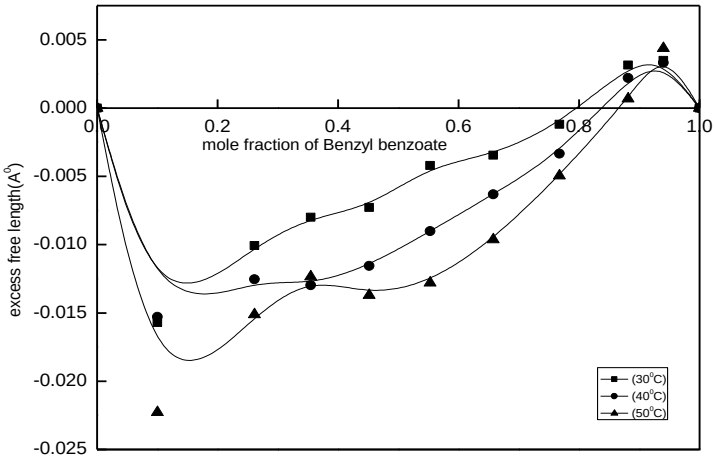

Fig. 3. Variation of excess free length with the mole fraction of benzyl benzoate at three different temperatures in the binary mixture: Benzyl benzoate + 1-octanol

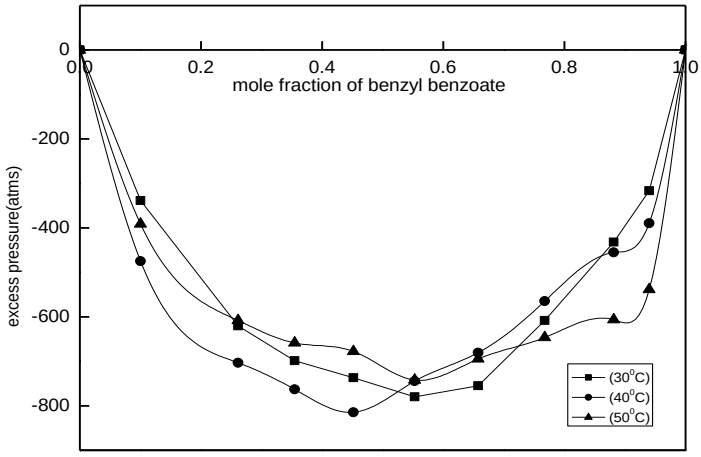

Fig. 4. Variation of excess internal pressure with the mole fraction of benzyl benzoate at three different temperatures in the binary mixture: Benzyl benzoate + 1-octanol

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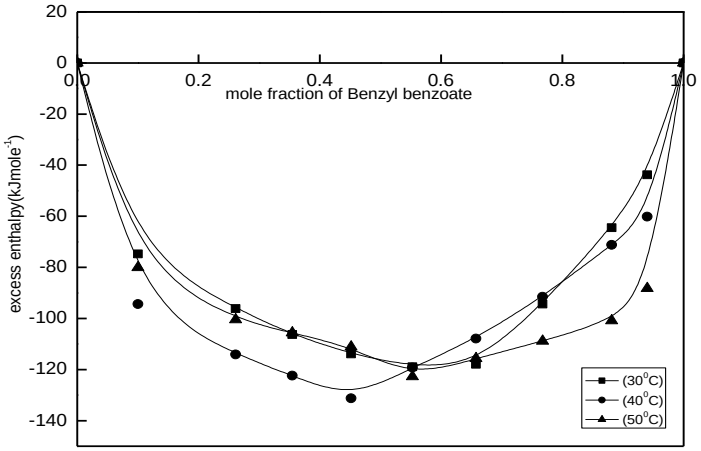

Fig. 5. Variation of excess enthalpy with the mole fraction of benzyl benzoate at three different temperatures in the binary mixture: benzyl benzoate + 1-octanol

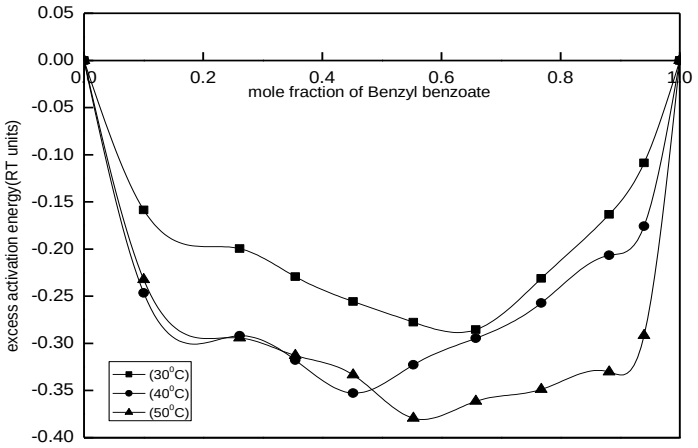

Fig. 6.Variation of excess activation energy with the mole fraction of benzyl benzoate at three different temperatures in the binary mixture: benzyl benzoate + 1-octanol

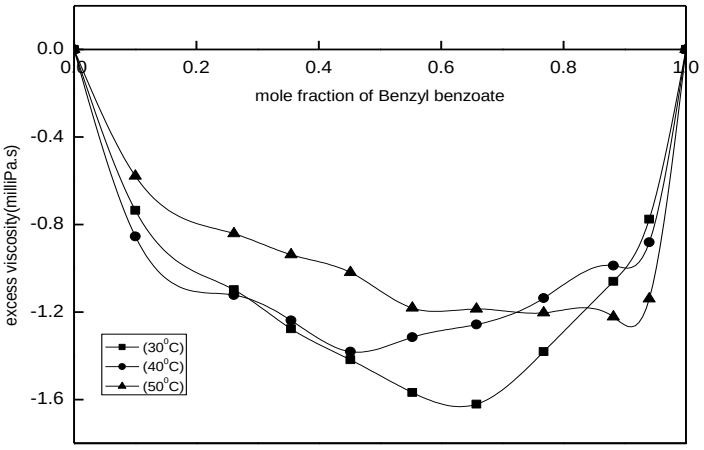

Fig. 7. Variation of excess viscosity with the mole fraction of benzyl benzoate at three different tempeartures in the binary mixture: benzyl benzoate + 1-octanol

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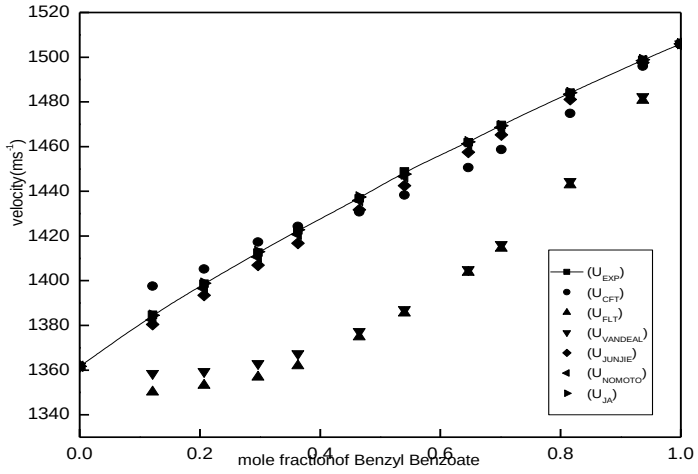

Fig.8(i). Variation of velocity with the mole fraction of benzyl benzoate at 30^{0}C in the binary mixture: benzyl benzoate + isophorone

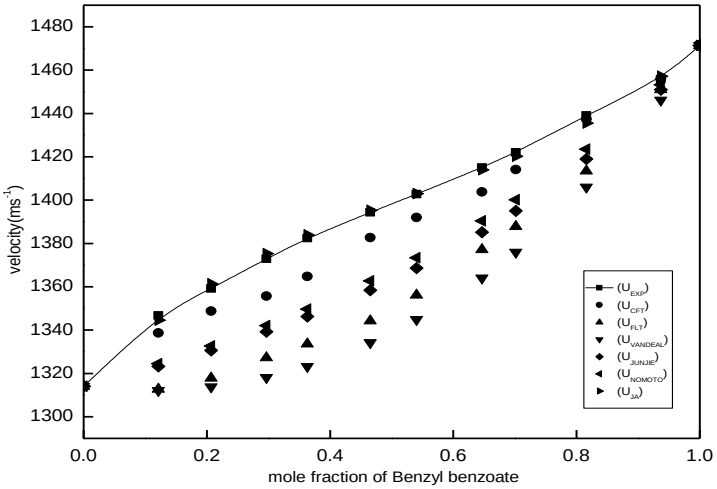

Fig. 8(ii). Variation of velocity with the mole fraction of benzyle benzoate at 40^{0}C in the binary mixture; benzyl benzoate + isophorone

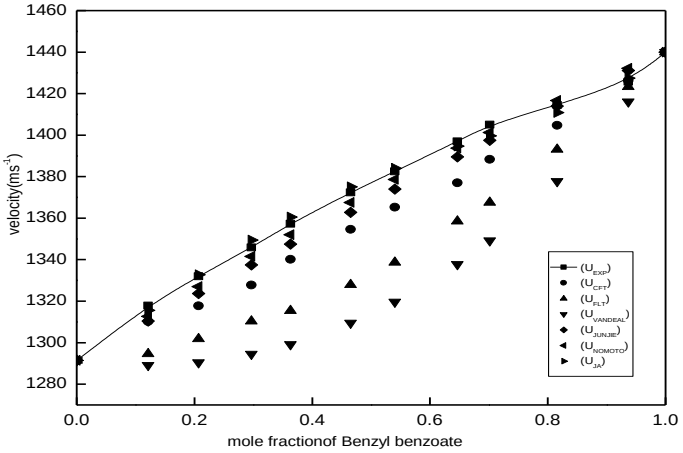

Fig. 8(iii). Variation of velocity with the mole fraction of benzyl benzoate at 50^{0}C in the binary mixture: benzyl benzoate + isophorone

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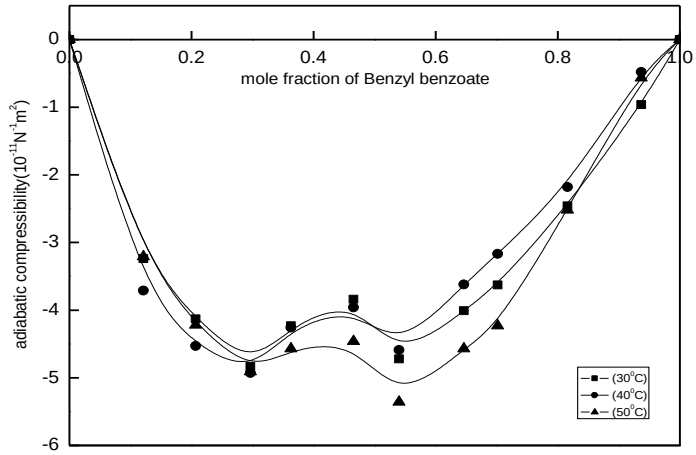

Fig. 9. Variation of excess adiabatic compressibility with the mole fraction of benzyl benzoate at three different temperatures in the binary mixture: Benzyl benzoate + isophorone

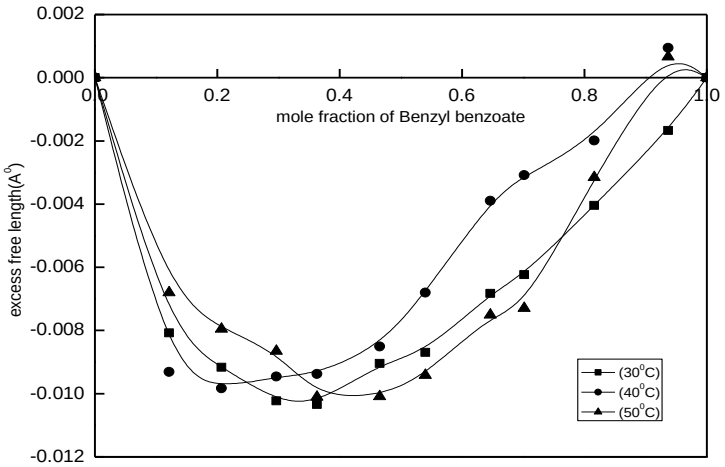

Fig. 10. Variation excess free length with the mole fraction of benzyl benzoate at three different temperatures in the binary mixture: benzyl benzoate + isophorone

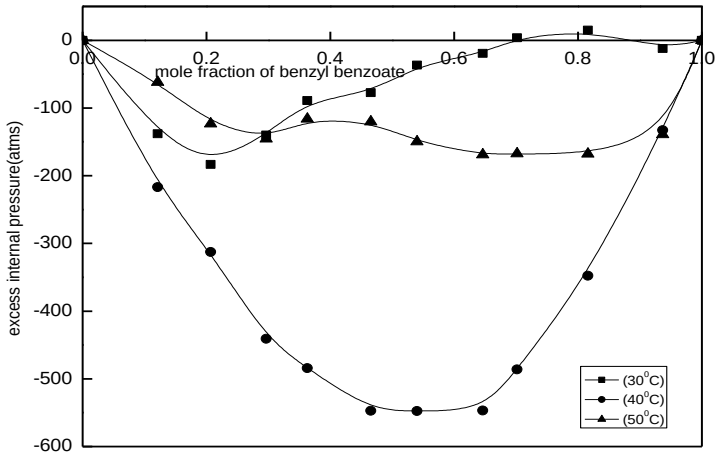

Fig.11. Variation of excess internal pressure with the mole fraction of benzyl benzoate at three different temperatures in the binary mixture: Benzyl benzoate + isophorone

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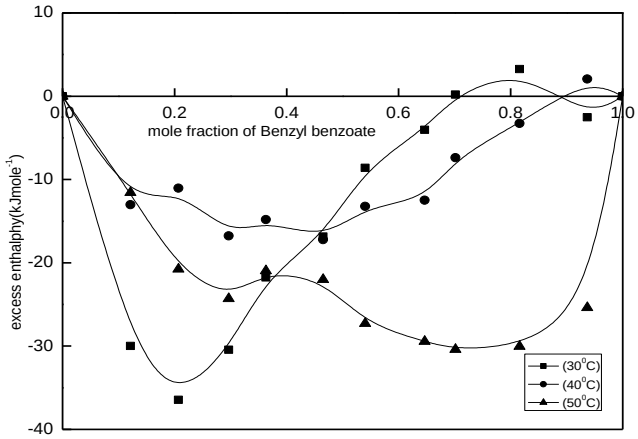

Fig. 12. Variation of excess enthalpy with the mole fraction of benzyl benzoate at three different temperatures in the binary mixture : Benzyl benzoate + isophorone

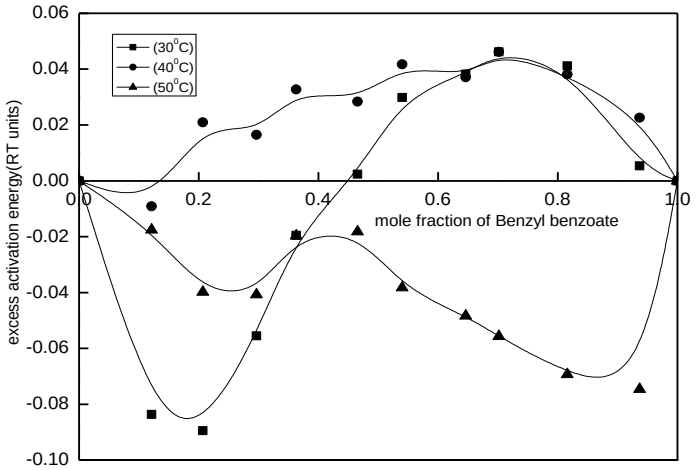

Fig. 13.Variation of excess activation energy with the mole fraction of benzyl benzoate at three different temperatures in the binary mixture: benzyl benzoate + isophorone

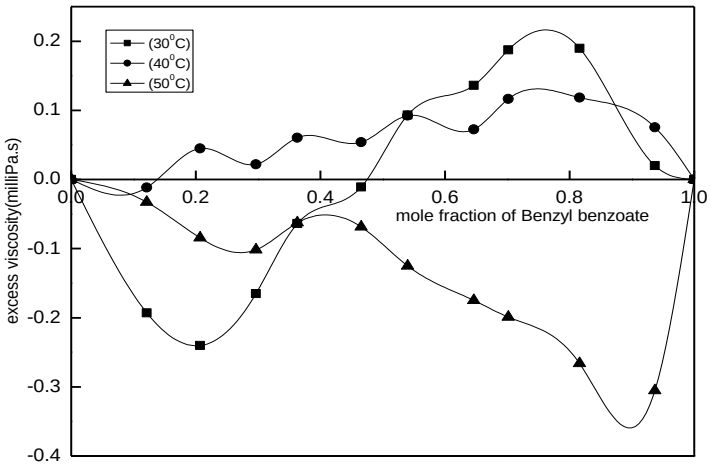

Fig. 14. Variation of excess viscosity with the mole fraction of benzyl benzoate at three different tempeartures in the binary mixture: benzyl benzoate and isophorone

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Table 1: Experimental data of Velocity (U), density (ρ) and viscosity (η) as a function mole fraction of benzyl benzoate at three different temperatures in the binary mixture: benzyl benzoate + 1-octanol.

Mole fraction of Benzyl Benzoate	Velocity (ms^{-1})	Density (kgm^{-3})	Viscosity (milliPa.s)
	30°C		
0.0000	1329.0	0817.08	5.94246
0.1001	1356.2	0877.75	5.26443
0.2611	1374.0	0909.92	4.99247
0.3547	1383.1	0936.52	4.86827
0.4516	1396.5	0966.48	4.78386
0.5527	1410.5	0989.99	4.69386
0.6575	1430.4	1019.75	4.70313
0.7673	1451.0	1047.48	5.00869
0.8816	1469.7	1073.02	5.39983
0.9402	1487.7	1084.73	5.72010
1.0000	1506.0	1119.35	6.53251
	40°C		
0.0000	1302.5	0810.4	4.29108
0.1001	1326.6	0870.76	3.50401
0.2611	1350.3	0904.35	3.34844
0.3547	1366.1	0931.12	3.29877
0.4516	1378.9	0958.53	3.22506
0.5527	1392.1	0985.11	3.36526
0.6575	1409.1	1009.63	3.50132
0.7673	1424.3	1040.85	3.70438
0.8816	1441.1	1062.43	3.94012
0.9402	1455.4	1074.63	4.09262
1.0000	1471.4	1109.77	5.02109
	50°C		
0.0000	1263.8	0803.03	3.02700
0.1001	1303.7	0863.68	2.55662
0.2611	1319.0	0896.33	2.47525
0.3547	1329.6	0919.54	2.49005
0.4516	1348.3	0950.97	2.52673
0.5527	1367.1	0977.54	2.49015
0.6575	1382.9	1005.62	2.62573
0.7673	1397.2	1033.51	2.75749
0.8816	1416.2	1054.99	2.90154
0.9402	1423.6	1066.30	3.06896
1.0000	1440.0	1107.09	4.29833

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Table 2. Various thermodynamic parameters – enthalpy, internal pressure, free length, free volume, molar volume, adiabatic compressibility, activation energy as a function of mole fraction benzyl benzoate at three temperatures in the binary mixture : benzyl benzoate + 1-octanol.

Mole fraction of Benzyl Benzoate	Adiabatic compressibility (10^{-10} N^{-1} m^2)	Internal pressure (atms)	Molar volume (10^{-3}L.mole^{-1})	Free length (A^0)	Enthalpy (kJ.mole^{-1})	Activation energy (RT units)
			30^{0}C			
0.0000	6.93	5774	159.38	0.52526	920	6.85
0.1001	6.19	5254	157.72	0.49661	829	6.72
0.2611	5.82	4682	166.66	0.48144	780	6.72
0.3547	5.58	4434	170.12	0.47143	754	6.72
0.4516	5.30	4221	173.07	0.45961	730	6.72
0.5527	5.08	3995	177.34	0.44961	708	6.72
0.6575	4.79	3831	180.59	0.43684	692	6.74
0.7673	4.53	3779	184.41	0.42490	697	6.83
0.8816	4.31	3749	188.75	0.41447	708	6.93
0.9402	4.16	3759	191.15	0.40724	718	7.00
1.0000	3.94	3967	189.62	0.39602	752	7.12
			40^{0}C			
0.0000	7.27	4929	160.70	0.53815	792	6.54
0.1001	6.52	4311	158.99	0.50973	685	6.32
0.2611	6.06	3852	167.68	0.49139	646	6.33
0.3547	5.75	3659	171.11	0.47868	626	6.33
0.4516	5.49	3468	174.51	0.46740	605	6.33
0.5527	5.24	3394	178.22	0.45668	605	6.40
0.6575	4.99	3308	182.40	0.44566	603	6.46
0.7673	4.73	3267	185.58	0.43424	606	6.53
0.8816	4.53	3213	190.64	0.42480	612	6.62
0.9402	4.39	3195	192.94	0.41823	616	6.67
1.0000	4.16	3499	191.25	0.40708	669	6.87
			50^{0}C			
0.0000	7.80	4177	162.17	0.55717	677	6.20
0.1001	6.81	3694	160.29	0.52080	592	6.01
0.2611	6.41	3331	169.18	0.50530	563	6.04
0.3547	6.15	3195	173.26	0.49491	554	6.07
0.4516	5.78	3088	175.89	0.47991	543	6.10
0.5527	5.47	2933	179.46	0.46683	526	6.10
0.6575	5.20	2884	183.13	0.45501	528	6.17
0.7673	4.96	2832	186.90	0.44423	529	6.24
0.8816	4.73	2768	191.98	0.43379	531	6.32
0.9402	4.63	2783	194.45	0.42924	541	6.39
1.0000	4.36	3267	191.72	0.41646	626	6.71

ISSN: 2320-2459

Table 3: Experimental data of Velocity (U), density (ρ) and viscosity (η) as a function of mole fraction of benzyl benzoate at three different temperatures in the binary mixture : benzyl benzoate + isophorone.

Mole fraction of Benzyl Benzoate	Velocity (ms^{-1})	Density (kgm^{-3})	Viscosity (milliPa.s)
	30°C		
0.0000	1361.7	0917.44	2.41697
0.1214	1384.8	0959.98	2.53400
0.2070	1398.7	0976.81	2.72920
0.2967	1412.5	0996.43	3.08110
0.3629	1422.5	1009.02	3.40344
0.4653	1436.8	1024.39	3.82782
0.5402	1448.9	1036.88	4.22890
0.6464	1461.9	1054.97	4.73233
0.7015	1469.7	1065.21	5.04246
0.8161	1484.1	1085.29	5.63072
0.9370	1498.8	1108.42	6.15597
1.0000	1506.0	1119.35	6.53251
	40°C		
0.0000	1314.0	0911.86	1.74391
0.1214	1346.7	0945.20	1.97122
0.2070	1359.2	0962.42	2.21543
0.2967	1372.9	0976.96	2.40865
0.3629	1382.4	0990.03	2.6202
0.4653	1394.4	1012.13	2.90645
0.5402	1402.8	1024.84	3.17999
0.6464	1415.0	1041.00	3.52703
0.7015	1422.0	1052.20	3.77825
0.8161	1439.1	1075.94	4.25204
0.9370	1455.9	1095.61	4.77298
1.0000	1471.4	1109.77	5.02109
	50°C		
0.0000	1291.4	0903.23	1.45190
0.1214	1317.9	0933.82	1.62368
0.2070	1332.0	0950.31	1.73328
0.2967	1345.9	0968.10	1.90183
0.3629	1357.3	0984.33	2.09006
0.4653	1372.4	1005.18	2.33728
0.5402	1382.5	1019.86	2.48436
0.6464	1397.0	1037.30	2.75354
0.7015	1405.0	1049.84	2.90987
0.8161	1414.5	1069.95	3.25440
0.9370	1425.7	1093.35	3.70871
1.0000	1440.0	1107.09	4.29833

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Table 4:Various thermodynamic parameters – enthalpy, internal pressure, free length, free volume, molar volume, adiabatic compressibility, activation energy as a function of mole fraction benzyl benzoate at three temperatures in the binary mixture : benzyl benzoate + isophorone.

Mole fraction of Benzyl Benzoate	Adiabatic compressibility (10^{-10} N^{-1} m^2)	Internal pressure (atms)	Molar volume (10^{-3}L.mole^{-1})	Free length (A^0)	Enthalpy (kJ.mole^{-1})	Activation energy (RT units)
			30^{0}C			
0.0000	5.88	3666	150.65	0.48379	552	5.90
0.1214	5.43	3565	153.33	0.46506	547	5.96
0.2070	5.23	3545	157.18	0.45645	557	6.06
0.2967	5.03	3615	160.75	0.44752	581	6.20
0.3629	4.90	3686	163.60	0.44159	603	6.32
0.4653	4.73	3729	168.55	0.43391	628	6.47
0.5402	4.59	3792	171.87	0.42769	652	6.59
0.6464	4.43	3841	176.37	0.42023	677	6.73
0.7015	4.35	3881	178.51	0.41599	693	6.80
0.8161	4.18	3927	183.02	0.40812	719	6.94
0.9370	4.02	3936	187.28	0.39988	737	7.05
1.0000	3.94	3967	189.62	0.39602	752	7.12
			40^{0}C			
0.0000	6.35	1914	232.77	0.50288	445	6.01
0.1214	5.83	2165	215.05	0.48194	466	6.05
0.2070	5.62	2399	204.61	0.47322	491	6.12
0.2967	5.43	2618	194.77	0.46499	510	6.15
0.3629	5.28	2830	187.25	0.45874	530	6.14
0.4653	5.08	3162	175.67	0.44980	555	6.23
0.5402	4.96	3450	168.08	0.44433	580	6.28
0.6464	4.80	3860	157.92	0.43707	610	6.32
0.7015	4.70	4134	152.36	0.43259	630	6.35
0.8161	4.49	4714	141.11	0.42271	665	6.40
0.9370	4.31	5395	130.41	0.41407	704	6.43
1.0000	4.16	5771	124.54	0.40708	719	6.44
			50^{0}C			
0.0000	6.64	2888	153.07	0.51413	442	5.40
0.1214	6.16	2872	157.63	0.49547	453	5.54
0.2070	5.93	2843	161.56	0.48595	459	5.63
0.2967	5.70	2854	165.45	0.47649	472	5.75
0.3629	5.51	2909	167.71	0.46858	488	5.86
0.4653	5.28	2944	171.77	0.45859	506	5.99
0.5402	5.13	2943	174.74	0.45195	514	6.07
0.6464	4.94	2964	179.38	0.44349	532	6.20
0.7015	4.82	2986	181.12	0.43832	541	6.27
0.8161	4.67	3029	185.65	0.431265	562	6.40
0.9370	4.50	3104	189.86	0.42327	589	6.56
1.0000	4.36	3267	191.72	0.41646	626	6.71

In the binary mixtures of benzyl benzoate with alcohols (methanol, ethanol, 1-propoanol, 1-butanol), strong AB interactions besides dipole – dipole interactions are estimated and the strength of the interaction decreased with the chain length. In the ketone systems (MEK & BMK) and acetophenone system also, mostly strong intermolecular interactions are suggested through weak interactions at some concentrations are not totally ruled out. The nature of chemical reaction is endothermic in the systems with alcohols and the two ketones (MEK & BMK) at all temperatures. In acetophenone system, endothermic type of reactions is also predominant at some concentrations and temperature. Other studies in benzyl benzoate and methyl benzoate[9]and other systems[10, 11]also reveal similar type of interactions from the variation of excess thermodynamic parameters.

ISSN: 2320-2459

Referring to the above literature on benzyl benzoate systems, our results and discussion are in conformity. In the mixture with the unsaturated cyclic ketone, exothermic at 30°C and endothermic at high temperatures and totally exothermic at all temperatures in the higher alcohol system are noticed. In both the systems, strong AB interactions besides dipole – dipole interactions are suggested. The highly polar nature of the constituents – the benzoate and isophorone may be the reason for (nearly equal) stronger interactions in the binary mixture compared to the other mixture.

ACKNOWLEDGEMENTS

The authors thank SVU and VSU authorities for providing the necessary facilities for carrying out this work. One of the authors (JG) acknowledges the sanction of RGNF (Senior) by the UGC.

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