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Study on Molecular Interactions Using Thermodynamic Excess Properties of Binary Mixture Containing Propiophenone with 1-Propanol, 1-Butanol and 1-Pentanol at Temperatures 303.15, 308.15, 313.15 and 318.15 K

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The current study works out on densities 'ρ', ultrasonic speeds of sound 'u' of binary mixtures of propiophenone with *n*-alcohols *e.g.*, 1-propanol, 1-butanol and 1-pentanol measured over the entire composition range at 303.15 to 318.15 K and at atmospheric pressure 0.1 MPa. The worked out experimental data has been used to calculate various thermodynamic excess parameters like excess molar volume (V^E), excess ultrasonic velocity (U^E), excess acoustic impedance (Z^E), excess intermolecular free-length (L_f^E), deviation in isentropic compressibility (ΔKs), deviation in acoustic impedance (ΔZ) and the computed results were fitted with the Redlich Kister equation to estimate the binary coefficients and standard deviation between experimental and calculated data. Partial molar volume of the binary mixtures was calculated to understand more of the intermolecular reactions in the above binary mixtures.

Keywords: Excess molecular volume, Partial molar volume, Deviation in compressibility, Impedance, Ultrasonic sound.

INTRODUCTION

Study of thermo-physical properties of liquid mixtures has more impact on academic and industrial importance. Thermodynamic properties of binary mixtures are essential to know the behaviour and type of inter molecular interactions between the components of the liquid mixtures at different compositions and temperatures. The excess property study has been used for the design of equipment in heat transfer, fluid mechanics, process calculation and process control. The investigations have been carried out on phenols [1-3] and alcohols [4,5] and more study on phenones [6-12] and on methyl acetate [13] has been reported. Literature survey shows that no work has been reported on these mixtures at such temperatures. Propiophenone with alkanol mixtures are having various applications in chemical industries. Propiophenone is widely used component in perfumes industries. It is used in the synthesis of ketoamphetamines such as cathinone and methcathinone. It is also used as a volatile low toxicity solvent in glues, paints and nail polish removers [13]. The effect of temperature and chain length on 1-alcohols when mixed with propiophenone may induce changes in sign and magnitude of excess thermodynamic and transport properties [14]. Hence, the current study on these binary mixtures has been carried out at 303.15, 308.15, 313.15 and 318.15 K to understand the behaviour of intermolecular interactions.

Alcohols play an important role in many chemical reactions due to the ability to undergo self-association with manifold internal structures and are in wide use in industry and science as reagents, solvents and fuels and attract great attention as useful solvents in the green technology [15]. The effect of molecular size, shape, chain length and degree of molecular association of normal alkanols and associated alkanols on the volumetric, viscosity and acoustic properties of binary mixtures containing acetronitrile, dimethyl sulfoxide, ethyl acetate and benzonitrile have been studied earlier [16-26].

This research paper presents the data on densities, ultrasonic velocity of binary liquid mixtures of propiophenone (PPH) with 1-propanol, 1-butanol and 1-pentanol at 303.15, 308.15, 313.15 and 318.15 K. The experimental values of density, speed of sound are used to calculate various acoustical parameters like acoustic impedance (Z), isentropic compressibility (Ks), Inter molecular free-length (L_f). Various excess properties like excess ultrasonic velocity (U^E), excess acoustic impedance (Z^E), deviation in isentropic compressibility (ΔK_S) and excess inter molecular free-length (L_f^E) were calculated. Variations in excess properties of these binary mixtures at all four temperatures reflect the strong interactions between the

unlike molecules. Calculated excess properties' values were fitted to the Redlich Kister equation to estimate the standard deviations between the experimental and calculated values. A partial molar property is a thermodynamic quantity which indicates how an extensive property of a solution or mixture varies with changes in the molar composition of the mixture at constant temperature and pressure. An extended analysis on partial molar volume is also calculated for the above mixtures at all four temperatures.

EXPERIMENTAL

Propiophenone (purity > 98 %), 1-propanol (purity > 99 %), 1-butanol (purity > 99 %) and 1-pentanol (purity > 99 %) were purchased from S.D. Fine chemicals Ltd, India and used as such.

Estimated purities of the liquid samples are greater than 99 % and water content found less than 0.003 mass %. Mixtures were prepared by mixing weighed amounts of the pure liquids adopting the method of closed system by using Mettler Toledo (ME204) balance with the precision of ± 0.1 mg. Mixtures were allowed to stand for some time before every measurement so as to avoid air bubbles. The purities of the liquids were checked by comparing the values of densities and ultrasonic velocities with literature data (Table-1).

Proper care was taken to avoid any evaporation loss while doing the experiment. The densities of liquids and their mixtures were measured using bi-capillary pycnometer having a capillary diameter of 0.85 mm, which was calibrated using double distilled water. The necessary buoyancy corrections were applied. The density (ρ) values were reproducible within ± 0.2 Kg m^{-3}. The ultrasonic velocity (u) measurements were measured by a single frequency (2 MHz) variable path interferometer with an error of ± 0.0001 % ultrasonic interferometer model F-05 (S. No. 1415342) Mittal Enterprises, New Delhi. The mole fraction of each mixture measured with the masses of the components.

RESULTS AND DISCUSSION

The experimental values of ultrasonic velocity and density of binary mixtures at 303.15, 308.15, 313.15 and 318.15 K with respective mole fractions (x_1) are reported in Table-2.

The density values were used to calculate excess molar volumes V^E using the following equation:

$$V^E = \frac{x_1M_1 + x_2M_2}{\rho_m} - \left\{\frac{x_1M_1}{\rho_1} + \frac{x_2M_2}{\rho_2}\right\} \quad (1)$$

V^E (cm^3 mol^{-1}) is excess molar volume, where ρ_m is the density of the mixture and (x_1, M_1, ρ_1) and (x_2, M_2, ρ_2) are the mole-fraction, the molecular weight and the density of pure components 1 and 2, respectively.

The isentropic compressibility (K_S) was calculated using the Laplace relation:

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

where U is the ultrasonic velocity and ρ the density.

The deviation in isentropic compressibility (ΔK_S) obtained using the relation:

$$\Delta K_S\ (TPa^{-1}) = K_S{}^{12} - \phi_1 K_S{}^{1} - \phi_2 K_S{}^{2} \quad (3)$$

where $K_S{}^{12}$ is the experimental isentropic compressibility of the mixture and ϕ_1, ϕ_2 and $K_S{}^{1}$, $K_S{}^{2}$ are the volume fractions and isentropic compressibility, respectively, of the pure components.

The ϕ_i is the ideal state volume fraction and is defined by the relation:

$$\phi_i = \frac{x_iV_i}{\Sigma_{i=1}^{2} x_iV_i} \quad (4)$$

TABLE-1
PHYSICAL PROPERTIES OF PURE COMPONENTS; COMPARISON OF EXPERIMENTAL VALUES OF DENSITIES, ULTRASONIC VELOCITY AND VISCOSITIES OF PURE LIQUIDS WITH THE CORRESPONDING LITERATURE VALUES AT VARIOUS TEMPERATURES

Compound	Temp. (K)	Density (ρ)		Ultrasonic speed (U)	
		Experimental (g/cm^{-3})	Literature (g/cm^{-3})	Experimental ($m\ s^{-1}$)	Literature ($m\ s^{-1}$)
Propiophenone	303.15	1.0045	1.00437(a), 1.00724(f)	1446.41	1458(f); 1440(a)
	308.15	1.0015	1.00896(b) 1.00870(c) 1.00600(a)	1427.038	1438(f); 1432(g)
	313.15	0.9985		1403.95	
	318.15	0.9955		1380	
1-Propanol	303.15	0.7953	0.79559(d), 0.79557(e), 0.7966(h), 0.7963(i), 0.80632(k), 0.806(l)	1197.9	1192(d), 1189(e), 1193(h), 1191(j), 1205.45(k); 202.47(l)
	308.15	0.7914	0.7917(n), 0.7915(m), 0.7915(r)	1166.28	1172.04(n), 1171.41(o)
	313.15	0.7873	0.7872(d), 0.7877(e)	1135.71	1158(d), 1155(e)
	318.15	0.7834		1109.83	
1-Butanol	303.15	0.8022	0.80231(h), 0.80195(q), 0.8023(u), 0.80206(d), 0.80209(e)	1230.114	1226(d), 1222(e), 1227(h), 1224(j)
	308.15	0.7982		1215.873	
	313.15	0.7943	0.79409(d), 0.7943(e)	1201.797	1191(d), 1189(e)
	318.15	0.7904		1188.308	
1-Pentanol	303.15	0.8076	0.80656(d)	1264.786	1262(d), 1265(p)
	308.15	0.804	0.8033(n), 0.8036(s), 0.80345(t)	1243.6	1243.67(n)
	313.15	0.8002	0.79905(d), 0.79905(p)	1220.231	1225(d), 1225(p)
	318.15	0.796		1196.24	

(a): [Ref. 27], (b): [Ref. 28], (c): [Ref. 29], (d): [Ref. 30], (e): [Ref. 31], (f): [Ref. 32], (g): [Ref. 33], (h): [Ref. 34], (i): [Ref. 35], (j): [Ref. 36], (k): [Ref. 37], (l): [Ref. 38], (m): [Ref. 39], (n): [Ref. 40], (o): [Ref. 41], (p): [Ref. 42], (q): [Ref. 43], (r): [Ref. 44], (s): [Ref. 45], (t): [Ref. 46], (u): [Ref. 47].

TABLE-2
EXPERIMENTAL VALUES OF BINARY MIXTURES OF PROPIOPHENONE WITH 1-PROPANOL, 1-BUTANOL AND 1-PENTANOL AT 303.15, 308.15, 313.15 AND 318.15 K

x_1	303.15 K		308.15 K		313.15 K		318.15 K	
	ρ (g cm^{-3})	U (m s^{-1})	ρ (g cm^{-3})	U (m s^{-1})	ρ (g cm^{-3})	U (m s^{-1})	ρ (g cm^{-3})	U (m s^{-1})
Propiophenone + 1-propanol								
0	0.7953	1197.90	0.7914	1166.28	0.7873	1135.71	0.7834	1109.83
0.0588	0.8179	1213.06	0.8146	1182.45	0.8111	1152.50	0.8083	1126.88
0.1233	0.8404	1229.69	0.8372	1199.90	0.8338	1170.51	0.8311	1145.07
0.1943	0.8627	1247.77	0.8596	1218.82	0.8562	1189.99	0.8536	1164.73
0.2728	0.8846	1267.53	0.8815	1239.52	0.8783	1211.27	0.8756	1186.20
0.3601	0.9060	1289.35	0.9030	1262.39	0.8998	1234.77	0.8972	1209.80
0.4577	0.9267	1313.58	0.9237	1287.81	0.9207	1260.89	0.9181	1236.11
0.5676	0.9466	1340.65	0.9437	1316.20	0.9407	1290.10	0.9383	1265.60
0.6924	0.9657	1371.22	0.9629	1348.28	0.9601	1323.16	0.9577	1298.90
0.8351	0.9845	1406.10	0.9819	1384.91	0.9792	1360.80	0.9769	1336.86
1.0000	1.0045	1446.41	1.0015	1427.04	0.9985	1403.95	0.9955	1380.00
Propiophenone + 1-butanol								
0	0.8022	1230.11	0.7982	1215.87	0.7943	1201.80	0.7904	1188.31
0.0710	0.8236	1246.38	0.8199	1232.06	0.8164	1217.62	0.8128	1203.66
0.1468	0.8449	1263.48	0.8415	1248.74	0.8382	1233.87	0.8348	1219.12
0.2278	0.8662	1281.47	0.8629	1266.36	0.8596	1250.57	0.8563	1235.17
0.3145	0.8872	1300.48	0.8839	1284.91	0.8807	1268.45	0.8774	1252.02
0.4077	0.9077	1320.73	0.9045	1304.68	0.9013	1287.34	0.8981	1269.89
0.5080	0.9276	1342.34	0.9245	1325.82	0.9213	1307.57	0.9182	1289.16
0.6163	0.9468	1365.59	0.9438	1348.55	0.9408	1329.37	0.9378	1309.94
0.7336	0.9656	1390.59	0.9628	1373.04	0.9600	1352.86	0.9571	1332.25
0.8610	0.9845	1417.52	0.9817	1399.35	0.9791	1377.91	0.9763	1355.92
1.0000	1.0045	1446.41	1.0015	1427.04	0.9985	1403.95	0.9955	1380.00
Propiophenone + 1-pentanol								
0	0.8076	1264.79	0.8040	1243.60	0.8002	1220.23	0.7960	1196.24
0.0836	0.8279	1280.92	0.8246	1260.49	0.8212	1237.59	0.8174	1214.11
0.1703	0.8484	1297.42	0.8453	1277.15	0.8420	1254.27	0.8384	1230.79
0.2602	0.8689	1314.29	0.8658	1294.18	0.8626	1271.33	0.8591	1247.85
0.3537	0.8891	1331.59	0.8860	1311.65	0.8829	1288.83	0.8794	1265.35
0.4508	0.9090	1349.37	0.9060	1329.60	0.9028	1306.81	0.8994	1283.34
0.5518	0.9285	1367.71	0.9255	1348.12	0.9224	1325.36	0.9191	1301.89
0.6570	0.9474	1386.53	0.9446	1367.13	0.9416	1344.40	0.9385	1320.93
0.7665	0.9662	1405.87	0.9634	1386.67	0.9606	1363.97	0.9577	1340.51
0.8808	0.9850	1425.73	0.9823	1406.74	0.9795	1384.07	0.9767	1360.62
1.0000	1.0045	1446.41	1.0015	1427.04	0.9985	1403.95	0.9955	1380.00

Deviation in ultrasonic speed of sound (ΔU) calculated by using the following relation:

$$\Delta_U = U - (x_1U_1 + x_2U_2) \quad (5)$$

where U is the experimental ultrasonic speed for the mixture, x_1, x_2, U_1, U_2 are the mole fractions and ultrasonic speed of the pure components 1 and 2, respectively.

In an attempt to explore the nature of the interactions, various thermodynamic parameters like intermolecular free length (L_f) [48]; acoustic impedance (Z) [49]; of the binary mixtures have been calculated using the following equations:

$$L_f = K\rho^{\frac{1}{2}} \quad (6)$$

where L_f is the intermolecular free length, K is the isentropic compressibility and ρ is the density.

Acoustic impedance calculated by using the relation:

$$Z = U\rho \quad (7)$$

where Z is the acoustic impedance, U is the ultrasonic speed of sound and ρ is the density.

Excess molar free length (L_f^E) calculated by using the relation:

$$L_f^E = L_f - (x_1L_{f1} + x_2L_{f2}) \quad (8)$$

where L_f^E is excess molar free length, L_f is intermolecular free length of the mixture, x_1, x_2, L_{f1} and L_{f2} are the mole fraction and intermolecular free length for pure components 1 and 2 respectively.

Deviation in acoustic impedance calculated by using the relation:

$$\Delta Z = Z - (x_1Z_1 + x_2Z_2) \quad (9)$$

where ΔZ is the deviation in acoustic impedance, Z is the acoustic impedance of the mixture, x_1, x_2, Z_1, Z_2 are the mole fraction and acoustic impedances of pure components 1 and 2 respectively.

The non-linear variation is a deviation from ideal behaviour which suggests the presence of intermolecular interactions between the component molecules of the mixtures, however the strength of interaction is well reflected in the excess parameters like excess molar volume (V^E), partial molar volume (PMV) and deviation in isentropic compressibility (ΔKs), deviation in ultrasonic speed of sound (ΔU), deviation in

acoustic impedance (ΔZ), excess intermolecular free length (L_f^E), *etc*., as these parameters are found to be more sensitive towards intermolecular interactions in the liquid mixtures [50].

From the thermodynamic point of view – the ultrasonic velocity, isentropic compressibility, deviation in isentropic compressibility, free length along with the other data like excess molar volume, are strongly affected by the changes of concentration and temperature, besides the type of bonding present between the molecules of the constituent liquids.

Hydrogen bonding causes considerable influence on these parameters, because of the fact that specific interactions between the molecules are controlled mainly by hydrogen bond that binds the molecules together. Computed values of the parameters analyzed (V^E, ΔKs, ΔU, ΔZ and L_f^E), are given in Table-3 at 303.15, 308.15, 313.15 and 318.15 K for the binary mixtures of propio-phenone with 1-propanol, 1-butanol and 1-pentanol.

Excess molar volume (V_m^E): Fig. 1 represents the variation of V^E with the mole fraction X_1 of propiophenone with

TABLE-3
VALUES OF EXCESS MOLAR VOLUME (V_m^E), ULTRASONIC VELOCITY (ΔU), EXCESS ISENTROPIC COMPRESSIBILITY (ΔKs), EXCESS ACOUSTIC IMPEDANCE (Δz) AND EXCESS INTERMOLECULAR FREE-LENGTH (L_f^E) FOR THE BINARY LIQUID-MIXTURES OF PROPIOPHENONE WITH 1-PROPANOL, 1-BUTANOL AND 1-PENTANOL AT DIFFERENT TEMPERATURE

x_1	V^E (cm^3 mol^{-1})	ΔKs (pa^{-1})	ΔU ($m\ s^{-1}$)	ΔZ (10^{-3}/ $Kg\ m^{-2}\ s^{-1}$)	L_f^E (10-10 m)	x_1	V^E (cm^3 mol^{-1})	ΔKs (pa^{-1})	ΔU ($m\ s^{-1}$)	ΔZ (10^{-3}/ $Kg\ m^{-2}\ s^{-1}$)	L_f^E (10-10 m)
Propiophenone + 1-propanol at 303.15 K						Propiophenone + 1-propanol at 308.15 K					
0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.0588	-0.1702	-2.1795	0.5366	0.0100	-0.6833	0.0588	-0.2289	-2.5187	0.8284	0.0105	-0.7896
0.1233	-0.3384	-3.9961	1.1423	0.0190	-1.2528	0.1233	-0.3974	-4.5235	1.4707	0.0191	-1.4181
0.1943	-0.4888	-5.3922	1.5846	0.0265	-1.6905	0.1943	-0.5539	-6.0604	1.8840	0.0263	-1.8999
0.2728	-0.6129	-6.3432	1.8337	0.0322	-1.9886	0.2728	-0.6730	-7.0978	2.1138	0.0316	-2.2252
0.3601	-0.6831	-6.8156	1.9667	0.0354	-2.1367	0.3601	-0.7431	-7.6117	2.2235	0.0347	-2.3863
0.4577	-0.6784	-6.7599	1.9337	0.0357	-2.1192	0.4577	-0.7383	-7.5454	2.1867	0.0349	-2.3655
0.5676	-0.5789	-6.1167	1.6812	0.0324	-1.9176	0.5676	-0.6389	-6.8282	1.9024	0.0317	-2.1406
0.6924	-0.3846	-4.8280	1.2605	0.0251	-1.5136	0.6924	-0.4446	-5.3980	1.4619	0.0248	-1.6923
0.8351	-0.1348	-2.8147	0.6655	0.0139	-0.8824	0.8351	-0.1947	-3.1654	0.8740	0.0141	-0.9924
1.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1.0000	0.0000	0.0000	0.0000	0.0000	0.0000
Propiophenone + 1-butanol at 303.15 K						Propiophenone + 1-butanol at 308.15 K					
0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.0710	-0.1375	-1.7424	0.9021	0.0065	-0.5462	0.0710	-0.1774	-1.8616	1.1873	0.0071	-0.5836
0.1468	-0.2800	-3.1309	1.6084	0.0123	-0.9815	0.1468	-0.3401	-3.2954	1.8631	0.0130	-1.0331
0.2278	-0.4141	-4.1526	2.0846	0.0170	-1.3018	0.2278	-0.4769	-4.3469	2.3897	0.0178	-1.3627
0.3145	-0.5105	-4.7884	2.3337	0.0204	-1.5012	0.3145	-0.5696	-4.9892	2.6188	0.0210	-1.5641
0.4077	-0.5520	-5.0351	2.4350	0.0220	-1.5785	0.4077	-0.6039	-5.2327	2.7186	0.0226	-1.6404
0.5080	-0.5198	-4.8746	2.3525	0.0216	-1.5282	0.5080	-0.5695	-5.0656	2.6787	0.0222	-1.5881
0.6163	-0.4020	-4.3012	2.1812	0.0189	-1.3484	0.6163	-0.4529	-4.4750	2.5390	0.0196	-1.4029
0.7336	-0.2209	-3.3008	1.8103	0.0141	-1.0348	0.7336	-0.2800	-3.4538	2.2605	0.0150	-1.0828
0.8610	-0.0409	-1.8703	1.1776	0.0074	-0.5863	0.8610	-0.0917	-1.9780	1.6612	0.0084	-0.6201
1.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1.0000	0.0000	0.0000	0.0000	0.0000	0.0000
Propiophenone +1-pentanol at 303.15 K						Propiophenone +1-pentanol at 308.15 K					
0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.0836	-0.0700	-1.2954	0.9582	0.0030	-0.4061	0.0836	-0.1145	-1.4773	1.5582	0.0037	-0.4631
0.1703	-0.1684	-2.3041	1.7139	0.0058	-0.7223	0.1703	-0.2285	-2.5497	2.3139	0.0066	-0.7993
0.2602	-0.2660	-3.0150	2.2405	0.0082	-0.9452	0.2602	-0.3275	-3.2962	2.8405	0.0089	-1.0334
0.3537	-0.3358	-3.4270	2.5694	0.0099	-1.0744	0.3537	-0.3880	-3.7204	3.1694	0.0105	-1.1663
0.4508	-0.3611	-3.5435	2.7105	0.0106	-1.1109	0.4508	-0.4095	-3.8357	3.3105	0.0112	-1.2025
0.5518	-0.3312	-3.3726	2.6987	0.0103	-1.0573	0.5518	-0.3748	-3.6469	3.2987	0.0109	-1.1433
0.6570	-0.2400	-2.9111	2.4201	0.0087	-0.9126	0.6570	-0.2930	-3.1578	3.0202	0.0095	-0.9900
0.7665	-0.1184	-2.1792	1.8631	0.0061	-0.6832	0.7665	-0.1790	-2.3816	2.4631	0.0070	-0.7466
0.8808	-0.0112	-1.1943	0.9764	0.0028	-0.3744	0.8808	-0.0600	-1.3292	1.5764	0.0038	-0.4167
1.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1.0000	0.0000	0.0000	0.0000	0.0000	0.0000
Propiophenone + 1-propanol at 313.15 K						Propiophenone + 1-propanol at 318.15 K					
0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.0588	-0.2845	-2.8536	1.0076	0.0108	-0.8946	0.0588	-0.3900	-3.2184	1.1568	0.0118	-1.0090
0.1233	-0.4591	-5.0598	1.7139	0.0192	-1.5862	0.1233	-0.5690	-5.5894	1.9230	0.0200	-1.7523
0.1943	-0.6148	-6.7402	2.1623	0.0261	-2.1130	0.1943	-0.7268	-7.3880	2.4097	0.0267	-2.3161
0.2728	-0.7334	-7.8689	2.3897	0.0312	-2.4669	0.2728	-0.8449	-8.5903	2.6682	0.0317	-2.6931
0.3601	-0.8031	-8.4214	2.4696	0.0341	-2.6401	0.3601	-0.9131	-9.1616	2.6882	0.0344	-2.8722
0.4577	-0.7983	-8.3394	2.4002	0.0344	-2.6144	0.4577	-0.9083	-9.0619	2.6188	0.0346	-2.8409
0.5676	-0.6989	-7.5493	2.1236	0.0313	-2.3667	0.5676	-0.8089	-8.2097	2.4097	0.0318	-2.5737
0.6924	-0.5046	-5.9801	1.7234	0.0247	-1.8748	0.6924	-0.6146	-6.5133	2.0124	0.0253	-2.0419
0.8351	-0.2557	-3.5209	1.0825	0.0144	-1.1038	0.8351	-0.3666	-3.8642	1.4154	0.0154	-1.2114
1.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1.0000	0.0000	0.0000	0.0000	0.0000	0.0000

Propiophenone + 1-butanol at 313.15 K						Propiophenone + 1-butanol at 318.15 K					
0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.0710	-0.2139	-1.9638	1.4624	0.0077	-0.6157	0.0710	-0.2409	-2.0534	1.7375	0.0082	-0.6437
0.1468	-0.3943	-3.4645	2.3982	0.0140	-1.0861	0.1468	-0.4399	-3.5844	2.6733	0.0146	-1.1237
0.2278	-0.5320	-4.5035	2.7286	0.0186	-1.4119	0.2278	-0.5819	-4.6572	3.1971	0.0195	-1.4600
0.3145	-0.6200	-5.1596	3.0688	0.0218	-1.6175	0.3145	-0.6705	-5.3006	3.4169	0.0226	-1.6617
0.4077	-0.6500	-5.3936	3.1274	0.0233	-1.6909	0.4077	-0.6960	-5.5233	3.4314	0.0240	-1.7316
0.5080	-0.6085	-5.2137	3.0866	0.0229	-1.6345	0.5080	-0.6545	-5.3441	3.4722	0.0237	-1.6754
0.6163	-0.4999	-4.6159	2.9890	0.0205	-1.4471	0.6163	-0.5500	-4.7478	3.4998	0.0215	-1.4884
0.7336	-0.3395	-3.5829	2.7694	0.0161	-1.1232	0.7336	-0.3900	-3.7005	3.3209	0.0171	-1.1601
0.8610	-0.1499	-2.0669	2.0534	0.0094	-0.6480	0.8610	-0.1959	-2.1555	2.5640	0.0104	-0.6757
1.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1.0000	0.0000	0.0000	0.0000	0.0000	0.0000
Propiophenone +1-pentanol at 313.15 K						Propiophenone +1-pentanol at 318.15 K					
0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.0836	-0.1570	-1.6571	2.0082	0.0043	-0.5195	0.0836	-0.1970	-1.8632	2.5082	0.0049	-0.5841
0.1703	-0.2880	-2.8007	2.7639	0.0073	-0.8780	0.1703	-0.3455	-3.0864	3.2639	0.0079	-0.9676
0.2602	-0.3819	-3.5833	3.2905	0.0095	-1.1234	0.2602	-0.4449	-3.9160	3.7905	0.0101	-1.2277
0.3537	-0.4420	-4.0272	3.6194	0.0110	-1.2625	0.3537	-0.4950	-4.3755	4.1194	0.0114	-1.3717
0.4508	-0.4520	-4.1376	3.7605	0.0116	-1.2971	0.4508	-0.5010	-4.4847	4.2605	0.0120	-1.4060
0.5518	-0.4217	-3.9349	3.7487	0.0113	-1.2336	0.5518	-0.4667	-4.2612	4.2487	0.0117	-1.3359
0.6570	-0.3450	-3.4142	3.4702	0.0100	-1.0704	0.6570	-0.3960	-3.7049	3.9702	0.0106	-1.1615
0.7665	-0.2350	-2.5884	2.9131	0.0078	-0.8115	0.7665	-0.2875	-2.8218	3.4131	0.0084	-0.8846
0.8808	-0.1099	-1.4655	2.0264	0.0046	-0.4594	0.8808	-0.1569	-1.6199	2.5264	0.0054	-0.5078
1.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1.0000	0.0000	0.0000	0.0000	0.0000	0.0000

1-propanol, 1-butanol and 1-pentanol at 303.15, 308.15, 313.15 and 318.15 K. In present study, V^E is negative for the binary mixtures over the entire composition range for all four temperatures. It shows the predominance of specific interactions and it results in decrease in volume which includes depolymerization of self-associated 1-alcohols by the addition of propiophenone or formation of new bonds (hydrogen bond) between propiophenone and alcohols and other complex forming interactions [51].

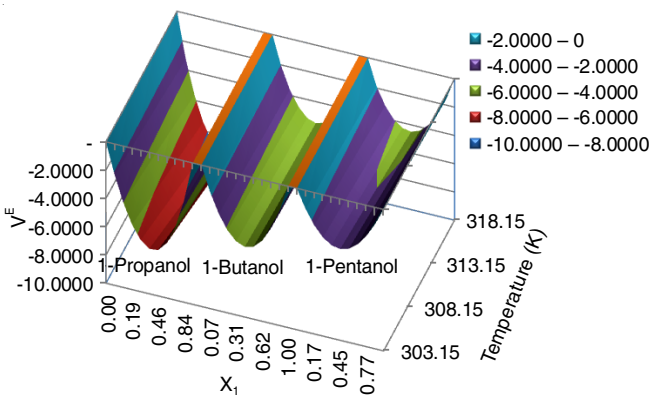

Fig. 1. Combined 3D details of V^E with mole fraction at temperatures 303.15-318.15 K for propiophenone + (1-propanol, 1-butanol and 1-pentanol)

As suggested by Sah *et al*. [52] excess molar volume is the resultant effect of physical contribution which is non-specific interaction (dispersion type) between the real species causing positive V^E, chemical which is specific interaction may due to charge transfer type forces and other complex forming interaction resulting negative V^E and structural contribution arising from interstitial accommodation and changes in free volume also causes negative V^E. However, the positive values of V^E are due to physical interaction mainly consisting of dispersion force or weak dipolar-dipolar interactions.

Figs. 1 & 2 and Table-3 illustrates that the excess molar volumes for all the three binary systems over the whole composition range are negative and the value decreases with increase in temperatures at 303.15, 308.15, 313.15 and 318.15 K. The maximum deviation in excess molar volume is obtained at 0.3601, 0.4077 and 0.4508 mole fractions of propiophenone for (propiophenone + 1-propanol), propiophenone + 1-butanol) and (propiophenone + 1-pentanol) respectively at 303.15 K.

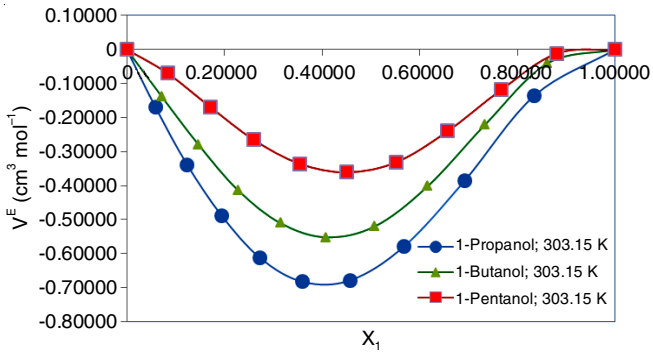

Fig. 2. V^E with mole fraction for propiophenone + (1-propanol, 1-butanol and 1-pentanol) at 303.15 K

The order of V^E values under the current study is 1-propanol < 1-butanol < 1-pentanol. V^E negative values are increasing with increase in temperatures. It shows that the extent of hydrogen bond and self-association decreases with increasing chain length of the alkanols [53].

Deviations in isentropic compressibility (ΔK_S): Figs. 3 and 4 shows that ΔKs values are negative over the whole composition range at the four temperatures. In the current study, the ΔKs values are negative because of strong molecular interactions between priopiophenone and 1-alkanols. Kiyohara and Benson [54] suggested that ΔKs is the result of several opposing effects. Strong molecular interactions occur through charge transfer, dipole induced-dipole and dipole-dipole interactions

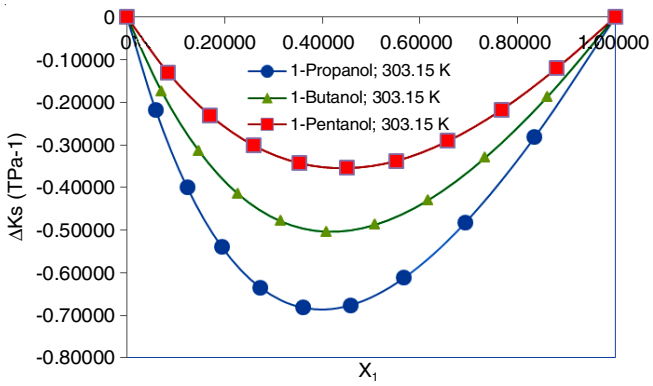

Fig. 3. Deviation in isentropic compressibility for propiophenone + (1-propanol, 1-butanol and 1-pentanol) at 303.15 K

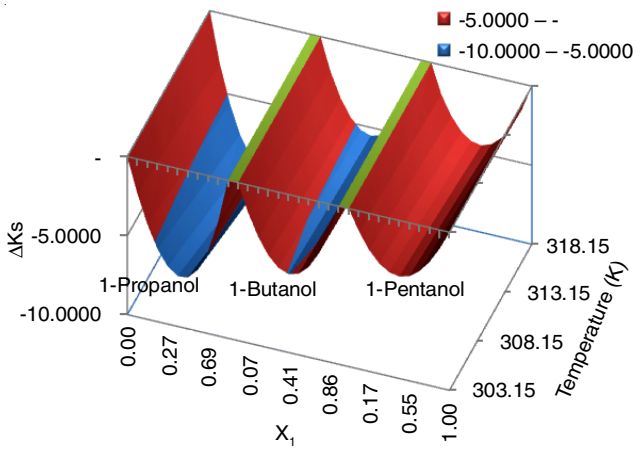

Fig. 4. Combined 3D details of deviation in isentropic compressibility for propiophenone + (1-propanol, 1-butanol and 1-pentanol) at 303.15-318.15 K

[55], closer approach of unlike molecules tends to contraction in the volume and reduction in the size leads to a more compact structure making ΔKs negative, whereas de-clustering of the 1-alkanol structures tends to make ΔKs positive. Fort and Moore [56] notice that the negative excess free volume tends to decrease as the strength of the interaction between the unlike molecules increases although they do not parallel with the excess compressibility. The trend is negative, order of ΔKs values is propanol < butanol < pentanol.

Excess intermolecular free length (L_f^E): In Figs. 5 and 6, L_f^E values are negative for the whole composition range at all four temperatures. According to Ramamoorthy and Alwan [57]

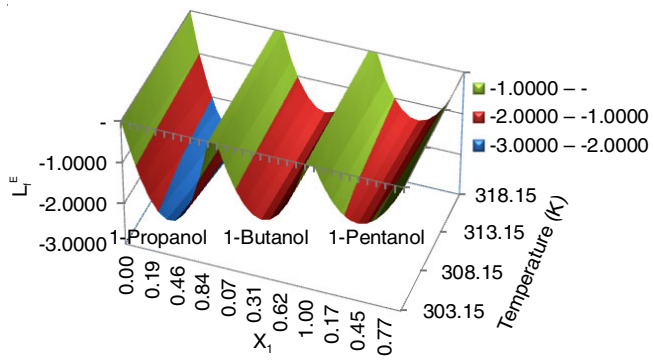

Fig. 5. Combined 3D details for variation of excess intermolecular free length with mole fraction of propiophenone + (1-propanol, 1-butanol and 1-pentanol) at 303.15-318.15 K

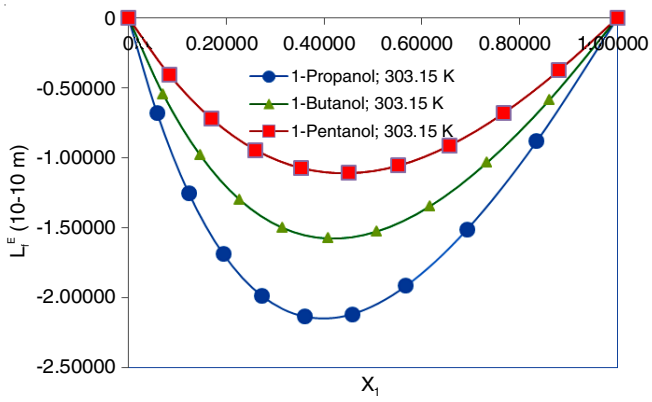

Fig. 6. Variation of excess intermolecular free length with mole fraction of propiophenone with 1-propanol, 1-butanol and 1-pentanol at 303.15 K

negative values of excess intermolecular free length L_f^E indicate that sound waves cover longer distances due to decrease in intermolecular free length ascribing the dominant nature of hydrogen bond interaction between unlike molecules.

In this study, it is observed that the values of excess intermolecular free length and ΔKs are negative. It shows strong intermolecular interactions between the unlike molecules in the binary system. The trend of (L_f^E) is negative and the order of values are propanol < butanol < pentanol.

Deviation in ultrasonic speed (ΔU) and deviation in acoustic impedance (ΔZ): Table-3 shows that the values of ΔU are positive for all the binary mixtures of propiophenone with 1-propanol, 1-butanol and 1-pentanol. Generally, negative values of indicate dispersion forces due to weak interactions whereas positive values of ΔU indicate strong interactions [58]. The sign and magnitude of ΔU play important roles in describing molecular rearrangements among the component molecules in the mixtures which reflect intermolecular interactions between the molecules.

Figs. 7 and 8 show that the ΔU values are positive for the whole composition range at four temperatures for the current binary mixtures. The trend is positive in ΔU and order of values is 1-propanol < 1-butanol < 1-pentanol. In the current study, ΔZ values are also positive for the whole composition range at all four temperatures (Fig. 9). The trend is positive in ΔZ and the order of values is 1-propanol < 1-butanol < 1-pentanol.

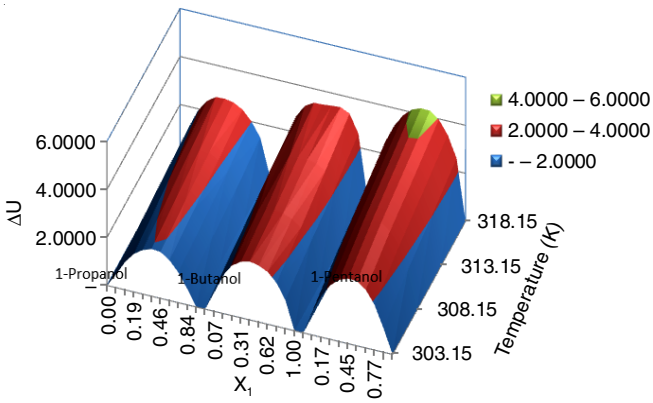

Fig. 7. Deviation in ultrasonic speed velocity with mole fraction of propiophenone with 1-propanol, 1-butanol and 1-pentanol at 303.15-318.15 K

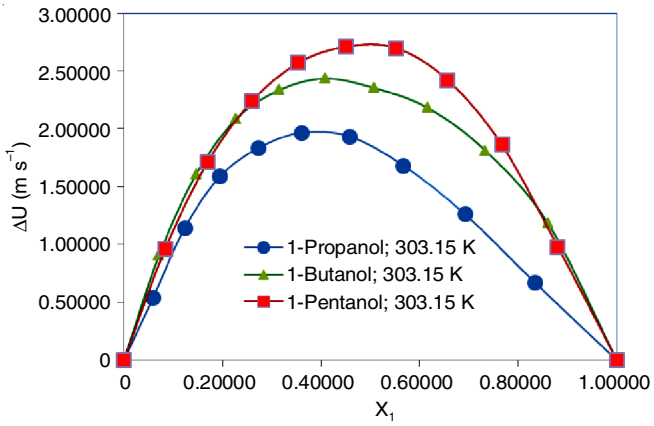

Fig. 8. Deviation in ultrasonic speed velocity with mole fraction of propiophenone with 1-propanol, 1-butanol and 1-pentanol at 303.15 K

Partial molar volume (PMV): Behaviour of the mixtures is further analyzed by calculating the partial excess molar volumes of the components. V^E_{m1} and V^E_{m2} for each 1-alkanols are calculated with respect to their mole fractions at all four temperatures 303.15, 308.15, 313.15 and 318.15 K.

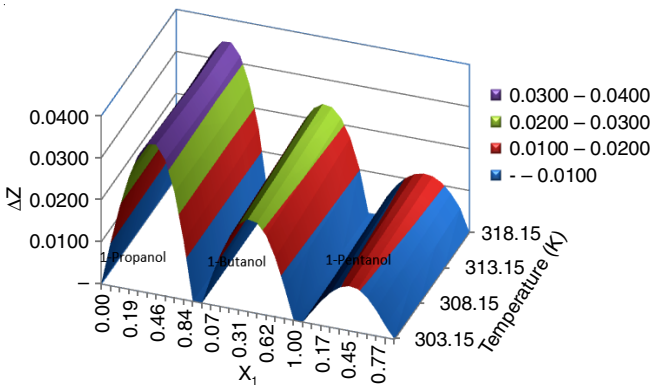

Fig. 9. Combined 3D details for deviation in acoustic impedance with mole fraction of propiophenone + (1-propanol, 1-butanol and 1-pentanol) at 303.15-318.15 K

The partial molar volumes, $\bar{V}^{\circ}_{m,1}$ of component 1 (propiophenone) and $\bar{V}^{\circ}_{m,2}$ of component 2 (1-propanol, 1-butanol, 1-pentanol) in the mixture over entire composition range are calculated using the following relations:

$$\bar{V}^{\circ}_{m,1} = V^E_m + V^*_{m,1} + x_2\left(\frac{\partial V^E_m}{\partial x_1}\right)T,P \qquad (10)$$

TABLE-4
EXCESS PARTIAL MOLAR VOLUME AT 303.15, 308.15, 313.15 AND 318.15 K

x_1	303.15 K		308.15 K		313.15 K		318.15 K	
	$\bar{V}^E_{m,1}$	$\bar{V}^E_{m,2}$	$\bar{V}^E_{m,1}$	$\bar{V}^E_{m,2}$	$\bar{V}^E_{m,1}$	$\bar{V}^E_{m,2}$	$\bar{V}^E_{m,1}$	$\bar{V}^E_{m,2}$
Propiophenone + 1-propanol								
0.0000	0.2232	0.0000	0.0853	0.0000	-0.0439	0.0000	-0.1660	0.0000
0.0588	-0.4890	0.0167	-0.5796	0.0157	-0.6675	0.0148	-0.7517	0.0140
0.1233	-1.0177	0.0567	-1.0722	0.0533	-1.1277	0.0501	-1.1821	0.0472
0.1943	-1.3504	0.0992	-1.3800	0.0930	-1.4122	0.0870	-1.4450	0.0813
0.2728	-1.4818	0.1134	-1.4970	0.1053	-1.5147	0.0967	-1.5337	0.0885
0.3601	-1.4184	0.0567	-1.4281	0.0485	-1.4389	0.0387	-1.4509	0.0290
0.4577	-1.1844	-0.1262	-1.1949	-0.1322	-1.2042	-0.1416	-1.2143	-0.1513
0.5676	-0.8300	-0.4998	-0.8432	-0.5033	-0.8534	-0.5120	-0.8637	-0.5217
0.6924	-0.4375	-1.1226	-0.4505	-1.1316	-0.4603	-1.1454	-0.4696	-1.1605
0.8351	-0.1213	-2.0047	-0.1281	-2.0495	-0.1332	-2.0906	-0.1379	-2.1316
1.0000	0.0000	-2.9957	0.0000	-3.1608	0.0000	-3.2926	0.0000	-3.4167
Propiophenone + 1-butanol								
0.0000	0.5768	0.0000	0.1054	0.0000	-0.6599	0.0000	-1.2043	0.0000
0.0710	-0.3411	0.0247	-0.5877	0.0192	-1.0509	0.0113	-1.4032	0.0061
0.1468	-0.9419	0.0760	-1.0387	0.0592	-1.2890	0.0332	-1.5025	0.0158
0.2278	-1.2504	0.1152	-1.2631	0.0892	-1.3796	0.0437	-1.5020	0.0122
0.3145	-1.3046	0.0992	-1.2862	0.0731	-1.3344	0.0151	-1.4052	-0.0280
0.4077	-1.1572	-0.0161	-1.1446	-0.0291	-1.1724	-0.0874	-1.2208	-0.1364
0.5080	-0.8762	-0.2684	-0.8874	-0.2572	-0.9220	-0.3052	-0.9649	-0.3561
0.6163	-0.5427	-0.6755	-0.5762	-0.6440	-0.6224	-0.6860	-0.6638	-0.7442
0.7336	-0.2435	-1.2107	-0.2811	-1.2010	-0.3251	-1.2815	-0.3582	-1.3772
0.8610	-0.0534	-1.7607	-0.0725	-1.8936	-0.0936	-2.1412	-0.1081	-2.3562
1.0000	0.0000	-2.0520	0.0000	-2.5970	0.0000	-3.3008	0.0000	-3.8144
Propiophenone + 1-pentanol								
0.0000	0.6595	0.0000	-0.0365	0.0000	-0.7312	0.0000	-1.3571	0.0000
0.0836	-0.1635	0.0242	-0.5540	0.0150	-0.9421	0.0058	-1.3014	-0.0022
0.1703	-0.6259	0.0651	-0.8246	0.0376	-1.0198	0.0097	-1.2107	-0.0145
0.2602	-0.8023	0.0789	-0.8973	0.0355	-0.9880	-0.0087	-1.0865	-0.0474
0.3537	-0.7708	0.0304	-0.8242	-0.0190	-0.8726	-0.0695	-0.9322	-0.1148
0.4508	-0.6106	-0.1016	-0.6587	-0.1460	-0.7017	-0.1917	-0.7537	-0.2346
0.5518	-0.3973	-0.3169	-0.4532	-0.3536	-0.5045	-0.3912	-0.5603	-0.4305
0.6570	-0.1974	-0.5833	-0.2557	-0.6316	-0.3103	-0.6789	-0.3654	-0.7324
0.7665	-0.0595	-0.8237	-0.1043	-0.9433	-0.1469	-1.0579	-0.1880	-1.1790
0.8808	-0.0026	-0.8978	-0.0207	-1.2150	-0.0381	-1.5196	-0.0544	-1.8195
1.0000	0.0000	-0.5786	0.0000	-1.3212	0.0000	-2.0381	0.0000	-2.7165

$$\bar{V}^{o}_{m,2} = V^{E}_{m} + V^{*}_{m,2} - x_1\left(\frac{\partial V^{E}_{m}}{\partial x_1}\right)T,P \quad (11)$$

where, $V^{*}_{m,1}$, $V^{*}_{m,2}$ are molar volumes of pure components propiophenone with 1-propanol, 1-butanol and 1-pentanol. The derivative of $\left(\frac{\partial V^{E}_{m}}{\partial x_1}\right)$ obtained by using below equations.

$$\bar{V}^{o}_{m,2} = V^{*}_{m,2} + x_1^2\Sigma^{n}_{i=0}A_i(1-2x_1)^{i} + 2x_1x_2^2\Sigma^{n}_{i=1}A_i(1-2x_1)^{i-1} \quad (12)$$

$$\bar{V}^{o}_{m,1} = V^{*}_{m,1} + x_2^2\Sigma^{n}_{i=0}A_i(1-2x_1)^{i} + 2x_1x_2^2\Sigma^{n}_{i=1}A_i(1-2x_1)^{i-1} \quad (13)$$

The excess partial molar volumes $\bar{V}^{o,E}_{m,1}$, $\bar{V}^{o,E}_{m,2}$ over the whole composition range are calculated by using the below relation [59]:

$$\bar{V}^{o,E}_{m,1} = \bar{V}^{o}_{m,1} - V^{*}_{m,1} \quad (14)$$

$$\bar{V}^{o,E}_{m,2} = \bar{V}^{o}_{m,2} - V^{*}_{m,2} \quad (15)$$

The values of excess partial molar volumes are given in Table-4. The negative excess partial molar volumes may be indicative of solute-solvent [60,61] interactions between unlike molecules, whereas positive values indicate the presence of strong self-association between like molecules. The excess partial molar volume of propiophenone with alkanols at infinite dilution are calculated by using the relation:

$$\bar{V}^{o,E,\infty}_{m,2} = \bar{V}^{o,\infty}_{m,2} - V^{*}_{m,2} \quad (16)$$

$$\bar{V}^{o,E,\infty}_{m,1} = \bar{V}^{o,\infty}_{m,1} - V^{*}_{m,1} \quad (17)$$

The values of excess partial molar volume of at infinite dilution $\bar{V}^{o,E,\infty}_{m,1}$, $\bar{V}^{o,E,\infty}_{m,2}$ for the selected binary systems at different temperatures are presented in Table-5. Negative values of $\bar{V}^{o,E,\infty}_{m,1}$, $\bar{V}^{o,E,\infty}_{m,2}$ of propiophenone with 1-alkanols shows stronger solute–solvent interactions at infinite dilution. Fig. 10 shows the behaviour of excess partial molar volume at 303.15 K.

Redlich-Kister equation: The variation of V^{E}, K_s^{E}, ΔZ, ΔU and L_f^{E} with mole fraction were fitted to the Redlich-Kister equation of the type:

$$Y^{E} = x_ix_2\{a0 + a1(x_1-x_2) + a2(x_1-x_2)^2\} \quad (18)$$

where Y^{E} is for V^{E}, ΔU, ΔKs, ΔZ and L_f^{E} in the polynomial degree. The values of a0, a1 and a2 are the coefficients of the polynomial equation and were obtained by the method of least-squares and are given in Table-6 along with standard deviation values. The standard deviations are calculated by using the equation:

TABLE-5
EXCESS PARTIAL MOLAR VOLUME AT INFINITE DILUTION

Temp. (K)	$\bar{V}^{o}_{m,1}$ (cm^3 mol^{-1})	$V^{*}_{m,1}$ (cm^3 mol^{-1})	$\bar{V}^{o,E}_{m,1}$ (cm^3 mol^{-1})	$\bar{V}^{o}_{m,2}$ (cm^3 mol^{-1})	$V^{*}_{m,2}$ (cm^3 mol^{-1})	$\bar{V}^{o,E}_{m,2}$ (cm^3 mol^{-1})
Propiophenone + 1-propanol						
303.15	133.8021	133.5789	0.2232	72.5733	75.5690	-2.9957
308.15	134.0644	133.9790	0.0853	72.7806	75.9414	-3.1608
313.15	134.3376	134.3816	-0.0439	73.0443	76.3368	-3.2926
318.15	134.6206	134.7865	-0.1660	73.3001	76.7169	-3.4167
Propiophenone + 1-butanol						
303.15	134.1557	133.5789	0.5768	90.3418	92.3938	-2.0520
308.15	134.0844	133.9790	0.1054	90.2633	92.8603	-2.5970
313.15	133.7217	134.3816	-0.6599	90.0166	93.3174	-3.3008
318.15	133.5822	134.7865	-1.2043	89.9611	93.7755	-3.8144
Propiophenone + 1-pentanol						
303.15	134.2384	133.5789	0.6595	108.5720	109.1506	-0.5786
308.15	133.9425	133.9790	-0.0365	108.3181	109.6393	-1.3212
313.15	133.6504	134.3816	-0.7312	108.1219	110.1600	-2.0381
318.15	133.4295	134.7865	-1.3571	108.0247	110.7412	-2.7165

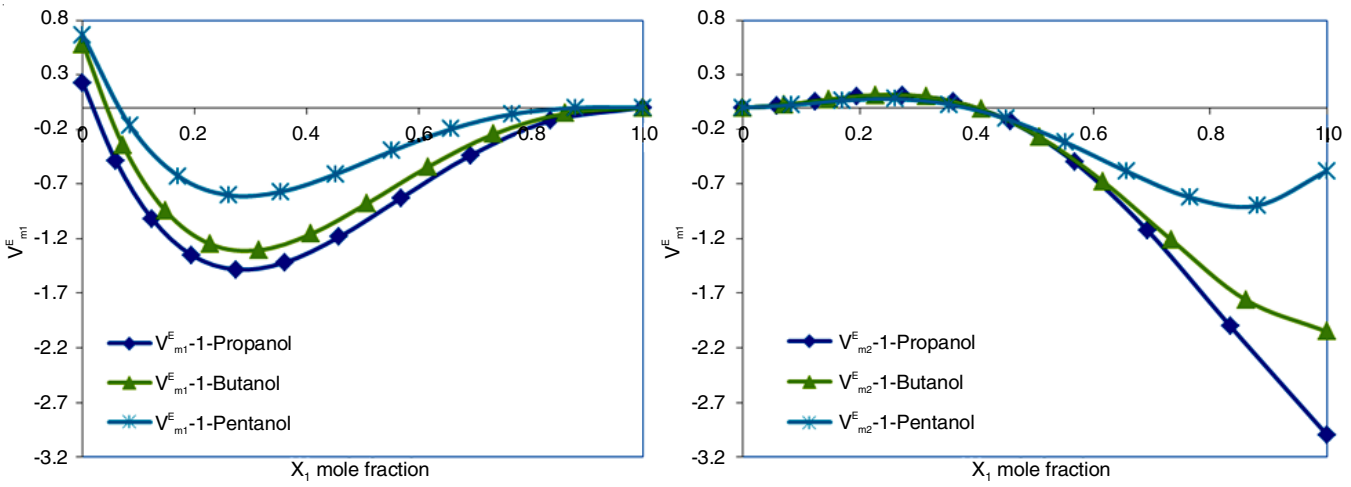

Fig. 10. Excess partial molar volume $V^{E}_{m,1}$ and $V^{E}_{m,2}$ for the components 1-propanol, 1-butanol and 1-pentanol at 303.15 K

TABLE-6
COEFFICIENTS OF REDLICH-KISTER POLYNOMIAL EQUATION

Property	T (K)	Propiophenone + 1-propanol				Propiophenone + 1-butanol				Propiophenone + 1-pentanol			
		a0	a1	a2	σ	a0	a1	a2	σ	a0	a1	a2	σ
V^E	303.15	-2.6048	1.6094	1.2186	0.0011	-2.2887	1.3144	1.5512	0.0015	-1.4122	0.6191	1.4526	0.0014
	308.15	-2.6373	1.6231	1.0995	0.0015	-2.2887	1.3512	1.0430	0.0014	-1.5954	0.6423	0.9165	0.0013
	313.15	-2.6745	1.6243	1.0063	0.0015	-2.4538	1.3205	0.4735	0.0014	-1.7710	0.6534	0.3863	0.0014
	318.15	-2.7138	1.6254	0.9224	0.0015	-2.6412	1.3050	0.1318	0.0013	-1.9590	0.6797	-0.0778	0.0013
ΔU	303.15	7.6054	-3.8571	-0.8097	0.0503	9.56188	-2.12061	3.34692	0.01813	10.9893	-1.69421	-0.07332	0.05273
	308.15	8.1616	-4.6902	3.2094	0.0410	10.5797	-1.26539	8.0081	0.04257	12.9332	-2.00349	6.80894	0.1093
	313.15	8.9705	-5.0215	5.6171	0.0578	12.1714	-1.3096	11.6869	0.06822	14.391	-2.2355	11.9707	0.17767
	318.15	9.8931	-4.5909	8.2391	0.0610	13.6505	-0.37613	15.9034	0.06361	16.0108	-2.49329	17.706	0.25726
ΔKs	303.15	-26.2147	11.1536	-4.0939	0.0266	-19.5945	6.5697	-1.5542	0.0062	-13.9875	3.4616	-0.06289	0.00287
	308.15	-29.0989	12.6885	-5.9914	0.0612	-20.3121	6.9089	-2.4688	0.0169	-15.0709	3.9440	-1.1306	0.0159
	313.15	-32.0535	14.2321	-7.7879	0.0921	-20.8888	7.2481	-3.3633	0.0273	-15.9217	4.2523	-2.6685	0.1217
	318.15	-34.6373	15.7964	-10.4098	0.1425	-21.3913	7.4528	-4.1751	0.0336	-17.5066	4.9867	-3.2258	0.0453
ΔZ	303.15	0.1392	-0.0529	-0.0059	0.0000	0.0868	-0.0252	-0.0133	0.0000	0.0424	-0.0083	-0.0153	0.0001
	308.15	0.1353	-0.0522	0.0069	0.0002	0.0889	-0.0240	-0.0031	0.0001	0.0444	-0.0073	-0.0073	0.0001
	313.15	0.1323	-0.0510	0.0174	0.0004	0.0917	-0.0231	0.0068	0.0001	0.0457	-0.0066	0.0056	0.0001
	318.15	0.1321	-0.0504	0.0356	0.0007	0.0949	-0.0215	0.0142	0.0001	0.0470	-0.0060	0.0152	0.0002
L_f^E	303.15	-8.21831	3.49666	-1.28344	0.00833	-6.14289	2.0596	-0.48726	0.00193	-4.38508	1.08518	-0.01959	0.00088
	308.15	-9.12252	3.97783	-1.87829	0.01919	-6.36786	2.16593	-0.77397	0.00529	-4.72481	1.23633	-0.35402	0.00497
	313.15	-10.0488	4.46176	-2.4415	0.02886	-6.54863	2.27227	-1.05439	0.00855	-5.08162	1.38679	-0.6613	0.00935
	318.15	-10.8588	4.95217	-3.26348	0.04467	-6.70624	2.33646	-1.30807	0.01051	-5.48838	1.5636	-1.01081	0.0142

$$\sigma(Y^E) = \frac{\left\{\Sigma_{i=1}^{n}(Y_{obs}^E - Y_{cal}^E)^2\right\}^{1/2}}{n-m} \quad (19)$$

where n is the total number of experimental points and m is the number of coefficients.

Conclusion

In this paper, the excess thermodynamic properties show that molecular interactions are predominant in the binary mixtures. The negative values of V^E are due to the strong intermolecular attractions between unlike molecules. The current study shows that the strong dipole-dipole interactions are observed in 1-propanol as compared to that of 1-butanol and 1-pentanol. From the measured thermodynamic properties data, the values of excess molar volumes, deviation in acoustic impedance, excess free length, deviation in speed of sound and excess isentropic compressibility have been correlated by Redlich-Kister type polynomial equation to derive the coefficients and standard deviation. Excess partial molar volume values at infinite dilution indicate stronger solute-solvent interactions. Finally, the strength of interactions is more in 1-propanol than that of 1-butanol and 1-pentanol.

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