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Density, Viscosity, Refractive Indices, Ultrasonic Velocities and Thermo Acoustical Parameters of Acetophenone + Isoamyl Acetate at 303.15, 313.15 and 323.15 K

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The studies of density, viscosity, refractive indices and ultrasonic velocities are being increasingly used as tools for investigation of the properties of pure components and the nature of intermolecular interactions between the liquid mixture constituents. Density (ρ), viscosity (η), refractive indices (n_D) and ultrasonic velocities (U) have been measured for the binary liquid mixture of acetophenone + isoamyl acetate over the entire composition range at 303.15, 313.15 and 323.15 K. This study involves the evaluation of different thermo acoustical parameters along with the excess properties. The Redlich-Kister model was used to correlate the measured excess properties. The molecular interactions existing between the components were also discussed.

Key Words: Binary mixture, Acetophenone, Isoamyl acetate, Density, Viscosity, Ultrasonic velocity, Refractive index.

INTRODUCTION

Binary liquid mixtures due to their unusual behaviour have attracted considerable attention[1]. Data on some of the properties associated with the liquids and liquid mixtures like density, viscosity, refractive index and ultrasonic velocities find extensive application in chemical engineering process simulation, solution theory and molecular dynamics[2]. These properties are important from practical and theoretical point of view to understand liquid theory. The review of literature on acoustical studies of solutions reveals that ultrasonic measurements are used to estimate the different elastic properties of the molecule from which the type of molecular interactions can be very well understood. Ultrasonic velocity has proved to be useful in understanding the physico-chemical behaviour of the particular system. Ultrasonic velocities have been widely used to study binary liquid mixtures[3]. Acetophenone as an important industrial chemical widely used as an ingredient of flavour and fragrance in soaps, detergents, cosmetics and perfumes. It is also been used as an important intermediate for pharmaceuticals and agrochemicals. Isoamyl acetate used as a fragrance in soaps, perfume, leather polish, varnishes, lacquers and other household products. The thermodynamic properties of system containing acetophenone are helpful for better understanding of the molecular interactions and to design and simulate the different processes of separation. From these experimental results acoustical impedance (Z), isentropic compressibility (K_s), intermolecular free length (L_f), internal pressure (π_i), viscosity deviation ($\Delta\eta$), excess Gibb's energy of activation of viscous flow (G^{E*}) refractive index deviation (Δn_D), intermolecular free length deviation (ΔL_f), internal pressure deviation ($\Delta\pi_i$) and isentropic compressibility deviation (ΔK_s) were derived over the entire mole fraction range. The values have been fitted to Redlich-Kister type[4] equation. Literature survey showed that no measurements have been previously reported for the mixture studied in this paper.

EXPERIMENTAL

The chemicals used were of analytical grade and obtained from Loba chemicals. All the components were dried over anhydrous potassium carbonate and fractionally distilled[5,6]. The purity of the solvents, after purification, was ascertained by comparing their density, viscosity, refractive index and sound of velocity with the corresponding literature values at 303.15 K (Table-1). A thermostatically controlled well-stirred water bath whose temperature was controlled to ± 0.01 K accuracy was used for all the measurements. All the measurements were done by using electronic balance Shimadzu Corporation Japan Type BL 2205 accurate to 0.01 g. The possible uncertainty in the mole fraction was estimated to be less than ± 0.0001.

TABLE-1
COMPARISON OF EXPERIMENTAL DENSITY (ρ), VISCOSITY (η), REFRACTIVE INDEX (n_D) AND SOUND OF VELOCITY (U) OF PURE LIQUIDS WITH LITERATURE VALUES AT 303.15 K

	Liquid	Experimental	Literature
Density (ρ) g cm^{-3}	Acetophenone	1.0196	1.0194[13-16]
	Isoamyl acetate	0.8633	0.8619[6,17]
Viscosity (η) mpa.s	Acetophenone	1.5412	1.5400[13-16]
	Isoamyl acetate	0.7569	0.7470[6,17]
Refractive index (n_D)	Acetophenone	1.5224	1.5220[12-16]
	Isoamyl acetate	1.3956	1.3957[6,17]
Sound of velocity (U) ms^{-1}	Acetophenone	1460.0	1460[12]
	Isoamyl acetate	1136.8	–

Densities were determined by using a 25 cm^3 bicapillary pycnometer and calibrated with benzene. The precision of the density measurement was estimated to be ± 0.0003 g/cm^3. The pycnometer was thermostated in a transparent walled water bath (maintained constant to ± 0.01 K) for 15 min to attain thermal equilibrium and the liquid level in the two arms was obtained with a traveling microscope which read to 0.01 mm. The precision of the measurements was estimated to be ± 0.0003 g cm^{-3}.

The kinematic viscosities were measured with an Oswald viscometer previously calibrated with water. The viscometer is filled with liquid or liquid mixtures and its limbs were closed with Teflon caps, taking due precautions to minimize the evaporation losses. The flow time measurements were made by using an electronic stopwatch with a precision of ± 0.01 s. An average of four or five sets of flow times for each liquid or liquid mixture was taken for the purpose of calculations of viscosity. The uncertainty in the viscosity was estimated to be less than 0.0003 m pa s.

Refractive indices were measured using thermostatically controlled Abbe refractometer with accuracy ± 0.0001 units. Water was circulated in to the prism of the refractometer by a circulation pump connected to an external thermostated water bath. Calibration was performed by measuring the refractive indices of doubly distilled water and propyl alcohol at defined temperatures. The sample mixture was directly injected in to the prism assembly of the instrument using a syringe. The solutions were pre thermostated at the temperature of the experiment to achieve a quick thermal equilibrium.

TABLE-2
EXPERIMENTAL DENSITY (ρ), VISCOSITY (η) REFRACTIVE INDICES (n_D), ULTRASONIC VELOCITIES (u) AND CALCULATED ACOUSTICAL IMPEDANCE (z), ISENTROPIC COMPRESSIBILITY (Ks), FREE LENGTH (L_f) AND INTERNAL PRESSURE (π_i) OF ACETOPHENONE AND ISOAMYL ACETATE AT 303.15, 313.15 AND 323.15 K

X_1	ρ (g cm^{-3})	η (m pa.s)	n_D	U (m s^{-1})	Z* (kg m^{-2} s^{-1})	Ks_*10^{-10} (m^2 N^{-1})	L_f*10^{-11} (m)	$\pi_i \times 10^6$ (pa)
303.15 K								
0	0.8633	0.7569	1.3956	1136.8	981.40	8.9633	6.2123	2.6323
0.1238	0.8789	0.8353	1.4083	1169.1	1027.57	8.3239	5.9866	2.7906
0.2412	0.8945	0.9138	1.4210	1201.4	1074.72	7.7449	5.7747	2.9446
0.3527	0.9101	0.9922	1.4336	1233.8	1122.99	7.2174	5.5745	3.0946
0.4587	0.9258	1.0706	1.4463	1266.0	1172.09	6.7392	5.3867	3.2414
0.5597	0.9414	1.1491	1.4590	1298.4	1222.38	6.3007	5.2085	3.3849
0.6560	0.9570	1.2275	1.4717	1330.7	1273.59	5.9005	5.0404	3.5258
0.7479	0.9727	1.3059	1.4844	1363.0	1325.80	5.5338	4.8812	3.6642
0.8357	0.9883	1.3843	1.4970	1395.4	1379.13	5.1963	4.7301	3.8002
0.9196	1.0039	1.4628	1.5097	1427.7	1433.37	4.8866	4.5869	3.9343
1.0000	1.0196	1.5412	1.5224	1460.0	1488.62	4.6011	4.4509	4.0664
313.15								
0	0.8528	0.6413	1.3941	1109.5	946.18	9.5257	6.5215	2.5129
0.1238	0.8684	0.7093	1.4064	1142.5	992.19	8.8216	6.2759	2.6657
0.2412	0.8841	0.7767	1.4186	1175.6	1039.32	8.1844	6.0450	2.8127
0.3527	0.8997	0.8441	1.4309	1208.7	1087.49	7.6077	5.8281	2.9560
0.4587	0.9154	0.9115	1.4431	1242.0	1136.88	7.0821	5.6232	3.0956
0.5597	0.9310	0.9789	1.4554	1274.8	1186.85	6.6094	5.4323	3.2328
0.6560	0.9466	1.0462	1.4677	1307.8	1238.02	6.1764	5.2513	3.3671
0.7479	0.9623	1.1136	1.4799	1340.9	1290.32	5.7797	5.0799	3.4988
0.8357	0.9779	1.1810	1.4922	1373.9	1343.56	5.4173	4.9180	3.6284
0.9196	0.9936	1.2484	1.5044	1406.9	1397.84	5.0849	4.7647	3.7559
1.0000	1.0092	1.3152	1.5167	1440.0	1453.25	4.7786	4.6190	3.8806
323.15								
0	0.8432	0.6025	1.3867	1083.0	913.17	10.111	6.8367	2.5249
0.1238	0.8590	0.6721	1.3987	1117.5	959.95	9.3218	6.5643	2.6879
0.2412	0.8749	0.7411	1.4106	1152.6	1008.35	8.6042	6.3066	2.8435
0.3527	0.8907	0.8095	1.4225	1187.0	1057.24	7.9685	6.0691	2.9942
0.4587	0.9065	0.8779	1.4345	1221.0	1106.85	7.3994	5.8484	3.1415
0.5597	0.9223	0.9459	1.4464	1256.0	1158.46	6.8727	5.6364	3.2833
0.6560	0.9382	1.0138	1.4584	1290.0	1210.24	6.4053	5.4414	3.4233
0.7479	0.9540	1.0824	1.4703	1324.0	1263.11	5.9796	5.2574	3.5616
0.8357	0.9698	1.1505	1.4822	1358.4	1317.42	5.5879	5.0823	3.6960
0.9196	0.9857	1.2192	1.4942	1393.5	1373.52	5.2247	4.9144	3.8282
1.0000	1.0015	1.2855	1.5061	1428.0	1430.13	4.8966	4.7576	3.9553

The velocity of the sound was measured by using a variable path, single crystal interferometer (Mittal Enterprises, New Delhi). The interferometer was calibrated using toluene. The interferometer cell was filled with the test liquid and water was circulated around the measuring cell from a thermostat. The uncertainty was estimated to be ± 0.1 m s^{-1}.

The acoustical impedance (Z) was calculated by the equation:

$$Z = \rho U \quad (1)$$

where ρ is the density mixture and U is the ultrasonic velocity of the mixture, The isentropic compressibility (K_s) was calculated by the equation:

$$K_s = 1/\rho U^2 \quad (2)$$

where ρ is the density of mixture and U is the ultrasonic velocity of the mixture.

Eqn. 1 is based on one dimensional analysis of the situation when an ultrasonic wave passes through a liquid medium.

The internal pressure of pure liquids and their binary liquid mixtures are calculated by the relation:

$$\pi_i = bRT\,(K\eta/U)^{1/2}\,(\rho^{2/3}/M_{eff}^{7/6}) \quad (3)$$

where η is the viscosity of the mixture which is to be found (m Pa-s), 'b' is the packing factor (1.78), T is the temperature of the mixture (K), ρ is the density of the mixture (g/cm^3), M_{eff} is effective mass of the mixture ($M_1X_1 + M_2X_2$), U is the ultrasonic velocity of the mixture (m/s), R is gas constant given as 8.314 J/mol K and K is the constant of value 4.28×10^9 independent of temperature.

The intermolecular free length (L_f) was calculated by the equation:

$$L_f = K\,(K_s)^{1/2} \quad (4)$$

RESULTS AND DISCUSSION

Measured values of densities, viscosities, refractive indices and ultrasonic velocities and derived parameters of acoustical impedance (Z), isentropic compressibility (K_s), intermolecular free length (L_f) and internal pressure (π_i) of (acetophenone + isoamyl acetate) mixture at temperatures of 303.15, 313.15 and 323.15 K are listed in Table-2. On the basis of the theory of absolute reaction rates[7] excess Gibb's free energy of activation of viscous flow for binary liquid mixtures are obtained by using following expression:

TABLE-3
DEVIATIONS OF VISCOSITY (Δη), REFRACTIVE INDICES (Δn_D), EXCESS GIBBS ENERGY (ΔG^E), ISENTROPIC COMPRESSIBILITY (ΔKs), INTERMOLECULAR FREE LENGTH (ΔL_f) AND INTERNAL PRESSURE ($\Delta\pi_i$) OF ACETOPHENONE + ISOAMYL ACETATE AT 303.15, 313.15 AND 323.15 K

X_1	Δη (m pa.s)	Δn_D	ΔG^E (J mol^{-1})	ΔKs $\times 10^{-11}$ (m^2 N^{-1})	$\Delta L_f \times 10^{-11}$ (m)	$\Delta\pi_i \times 10^6$ (Pa)
			303.15 K			
0	0	0	0	0	0	0
0.1238	-0.01866	-0.00302	34.1892	-0.09941	-0.00762	-0.01917
0.2412	-0.03232	-0.00523	55.3535	-0.16623	-0.01277	-0.03358
0.3527	-0.04136	-0.00669	66.5177	-0.20728	-0.01648	-0.04355
0.4587	-0.04611	-0.00745	69.9275	-0.22283	-0.01750	-0.04884
0.5597	-0.04688	-0.00758	67.2800	-0.22082	-0.01783	-0.05015
0.6560	-0.04396	-0.00711	59.8775	-0.20100	-0.01635	-0.04731
0.7479	-0.03758	-0.00608	48.7320	-0.16693	-0.01367	-0.04065
0.8357	-0.02800	-0.00453	34.6384	-0.12153	-0.01025	-0.03053
0.9196	-0.01540	-0.00249	18.2266	-0.06509	-0.00553	-0.01687
1.0000	0	0	0	0	0	0
			313.15 K			
0	0	0	0	0	0	0
0.1238	-0.01544	-0.00292	38.7019	-0.11644	-0.01012	-0.01652
0.2412	-0.02717	-0.00505	61.0660	-0.19620	-0.01762	-0.03004
0.3527	-0.03494	-0.00647	72.7767	-0.24350	-0.02232	-0.03933
0.4587	-0.03902	-0.00721	76.2399	-0.26563	-0.02547	-0.04477
0.5597	-0.03968	-0.00733	73.2426	-0.25899	-0.02427	-0.04564
0.6560	-0.03717	-0.00687	65.2586	-0.23502	-0.02209	-0.04306
0.7479	-0.03169	-0.00588	53.2824	-0.19552	-0.01871	-0.03701
0.8357	-0.02346	-0.00438	38.1880	-0.14120	-0.01354	-0.02746
0.9196	-0.01264	-0.00241	20.6449	-0.07518	-0.00714	-0.01473
1.0000	0	0	0	0	0	0
			323.15 K			
0	0	0	0	0	0	0
0.1238	-0.01495	-0.00284	49.8275	-0.14408	-0.01500	-0.01408
0.2412	-0.02615	-0.00493	79.1856	-0.24943	-0.02862	-0.02643
0.3527	-0.03392	-0.00630	93.0792	-0.30353	-0.03419	-0.03523
0.4587	-0.03795	-0.00703	96.9556	-0.31957	-0.03443	-0.03965
0.5597	-0.03893	-0.00715	91.9505	-0.31960	-0.03645	-0.04226
0.6560	-0.03678	-0.00671	80.8086	-0.28501	-0.03135	-0.04006
0.7479	-0.03093	-0.00574	66.8322	-0.23160	-0.02426	-0.03312
0.8357	-0.02278	-0.00428	47.9954	-0.16554	-0.01687	-0.02425
0.9196	-0.01141	-0.00236	27.6565	-0.09102	-0.01030	-0.01218
1.0000	0	0	0	0	0	0

$$G^E = RT [\ln \eta_{mix} V_{mix} - X_1 \ln \eta_1 V_1 - X_2 \ln \eta_2 V_2] \quad (5)$$

The excess properties or deviation were calculated by using

$$\Delta A = A_1 - (X_1 A_1 + X_2 A_2) \quad (6)$$

where A, A_1 and A_2 refer to the viscosity, refractive index, isentropic compressibility, internal pressure and free length of the mixture and pure components 1 and 2, respectively. The excess values or deviation of binary mixture presented in Table-3.

The excess Gibb's free energy of activation of viscous flow and deviations of thermo physical and acoustical properties were fitted to a Redlich-Kister equation of the type:

$$Y = X_1 X_2 \Sigma A_i (X_1 - X_2)^i \quad (7)$$

where Y is either G^E, $\Delta\eta$, Δn_D, $\Delta\pi_i$, ΔK_s and ΔL_f and n is the degree of polynomial. Coefficients Ai were obtained by fitting eqn. 7 to experimental results using a least square regression method. In each case, the optimum number of coefficients ascertained from an examination of the variation and standard deviation was calculated using the relation:

$$S(Y) = [\Sigma (A_{exp} - A_{cal})^2 / (N-n)]^{1/2} \quad (8)$$

where N is the number of data points and n is the number of coefficients. The calculated values of coefficients (Ai) along with the average per cent deviation of acetophenone with isoamyl acetate are given in Table-4.

Fig. 1 depicts the variation $\Delta\eta$ with the mole fraction X_i of acetophenone. $\Delta\eta$ values are always negative for all the studied temperatures and for any composition. The increase in temperature and the viscosity deviation values become less negative. The excess Gibbs free energy of activation of viscous flow (ΔG^{*E}) is positive over the whole mole fraction range for binary mixtures at different temperatures (Fig. 2). The sign values of values of (ΔG^{*E}) can be considered as a reliable criterion for detecting or excluding the presence of interaction between unlike molecules. The positive ΔG^{*E} values are also indicative of the strong molecular interaction between acetophenone and isoamyl acetate.

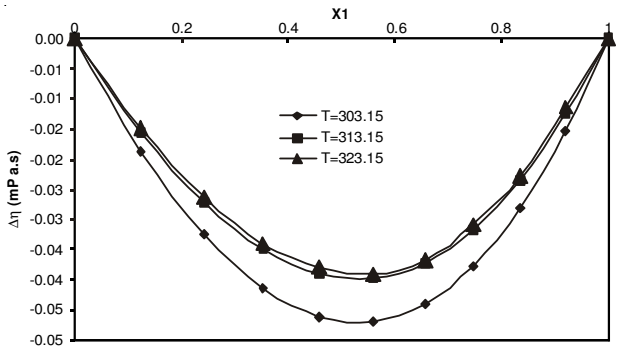

Fig. 1. Deviations in viscosity ($\Delta\eta$) for acetophenone and isoamyl acetate at 303.15, 313.15 and 323.15 K

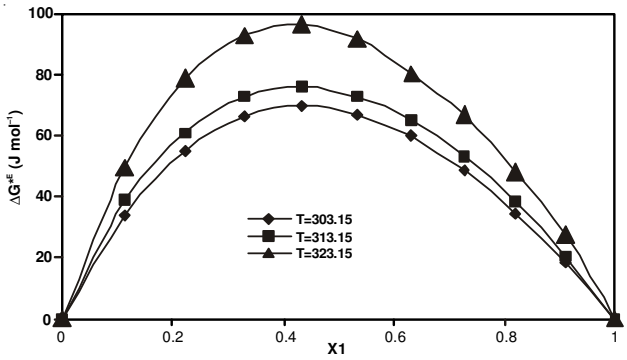

Fig. 2. Deviations in excess Gibb's free energy of activation of viscous flow for acetophenone and isoamyl acetate at 303.15, 313.15 and 323.15 K

TABLE-4
REDLICH-KISTER CONSTANTS FOR ACETOPHENONE-ISOAMYL ACATATE AT 303.15, 313.15 AND 323.15 K

Temperature (K)	a_0	a_1	a_2	a_3	a_4	a_5	a_6	S
Internal pressure deviations								
303.15	-0.2059	-0.0086	0.1936	-0.1385	-0.8319	0.1454	0.8431	0.0019
313.15	-0.1873	-0.0103	0.1635	-0.1270	-0.6706	0.1359	0.6934	0.0015
323.15	-0.1693	-0.0251	0.1280	-0.0613	-0.4813	0.0853	0.5220	0.0014
Viscosity deviations								
303.15	-0.1709	-0.0364	-0.2553	0.2590	0.4159	–	–	0.0207
313.15	-0.1454	-0.0311	-0.2027	0.0227	0.3140	–	–	0.0028
323.15	-0.1436	-0.0312	-0.1801	0.0251	0.3170	–	–	0.0024
Excess isentropic compressibility								
303.15	-8.228	-0.6412	-11.659	0.308	-8.228	–	–	0.1918
313.15	-9.729	0.5101	-13.211	-0.8902	22.439	–	–	0.4956
323.15	-11.93	1.5293	-15.935	-2.0122	27.249	–	–	1.4890
Excess free length								
303.15	-0.0654	-0.0046	-0.0965	0.0013	0.1581	–	–	0.0016
313.15	-0.0919	-0.0038	-0.1122	-0.0004	0.1994	–	–	0.0021
323.15	-0.1337	0.0240	-0.1532	-0.0315	0.2796	–	–	0.0031
Excess Gibbs energy for viscous flow								
303.15	255.36	-66.521	367.63	72.252	-610.68	–	–	4.4750
313.15	276.14	-78.011	438.4	84.55	-699.94	–	–	5.3180
323.15	346.10	-113.43	594.8	125.55	-920.13	–	–	7.1690
Refractive index deviations								
303.15	-0.0276	-0.0059	-0.0413	0.0042	0.0672	–	–	0.0005
313.15	-0.0267	-0.0057	-0.0399	0.0040	0.0650	–	–	0.0005
323.15	-0.0260	-0.0057	-0.0392	0.0040	0.0636	–	–	0.0005

In the present investigation, negative ΔK_s values[8] obtained over the range of temperatures studied for all compositions (Fig. 3) are attributed to chemical forces operating between unlike molecules of the binary mixtures[9,10]. The existence of the specific interactions is also supported by values of excess intermolecular free length (ΔL_f). The variation of excess intermolecular free length ΔL_f is shown in Fig. 4. Excess inter molecular free length is found to be negative[11] indicating specific interaction. The effect of an increase in the temperature appears to increase the excess properties, suggesting the presence of specific molecular interactions. As the temperature increases the values of ΔK_s and ΔL_f become more negative which may be due to thermal dissociation of hetero-aggregates in liquid mixtures and more interstitial accommodation of one component into another. The internal pressure deviations ($\Delta\pi_i$) for acetophenone and isoamyl acetate system are negative over the entire range of composition (Fig. 5). The refractive index deviations Δn_D for acetophenone-isoamyl acetate system are negative over the entire range of composition (Fig. 6). This reveals that the strength of specific interaction is not only the factor influencing the refractive index deviations of liquid mixtures. The molecular size and shape of the components also play an important role.

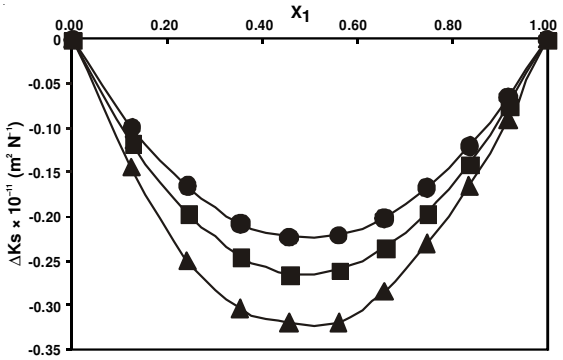

Fig. 3. Deviations in internal pressure (ΔKs) for acetophenone and isoamyl acetate at 303.15, 313.15 and 323.15 K

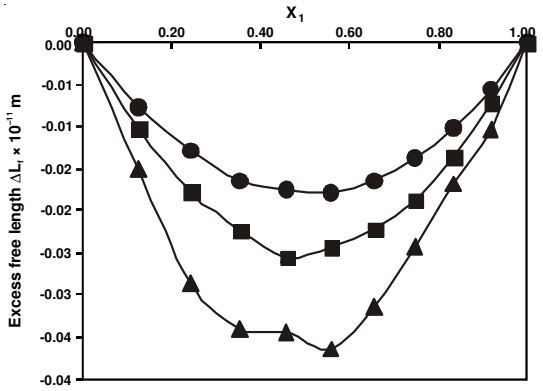

Fig. 4. Deviations in intermolecular free length for acetophenone and isoamyl acetate at 303.15, 313.15 and 323.15 K

Conclusion

Densities, viscosities, refractive indices and ultrasonic velocities for a two binary mixtures have been measured. Deviations of viscosity, refractive index, free length, isentropic compressibility, internal pressure and excess Gibbs free energy for mixtures of acetophenone and isoamyl acetate were obtained from the experimental results and fitted by the Redlich Kister equations.

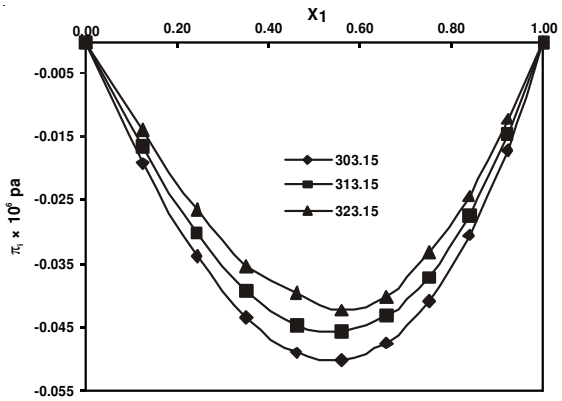

Fig. 5. Deviations in internal pressure ($\Delta\pi_i$) for acetophenone and isoamyl acetate at 303.15, 313.15 and 323.15 K

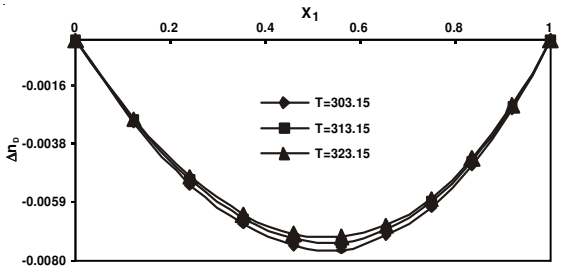

Fig. 6. Deviations in refractive index (Δn_D) for acetophenone and isoamyl acetate at 303.15, 313.15 and 323.15 K

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