A Study On Densities And Ultrasound Velocities In The (γ-Butyrolactone + Aliphatic Ester) System At Temperatures (303.15 To 313.15) K

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Abstract

Densities (ρ), and ultrasonic velocities (u), were measured for the binary systems of γ-butyrolactone (GBL) with methyl acetate (MA), ethyl acetate (EA), propyl acetate (PA) and butyl acetate (BA) at 303.15, 308.15, and 313.15 K, over the entire composition range. From the experimental results, excess molar volume (V^E), and deviation in isentropic compressibility ($\Delta\kappa_s$), were calculated. The computed properties have been fitted to a Redlich-Kister type polynomial equation to derive binary coefficients and standard deviations.

Keywords: *γ-butyrolactone, aliphatic nitriles, excess molar volume, deviation in isentropic compressibility.*

1. Introduction

A survey of the literature showed that density, viscosity and speed of sound properties of binary mixtures of ethanenitrile and propanenitrile with MA, EA, BA,[1] densities, viscosities, and refractive indices of MA, EA, and PA with 4-chorotoluene,[2] densities and viscosities of MA, EA, PA, and BA with nitromethane,[3] densities and viscosities of MA, EA, PA, ethyl propionate, and BA with nitroethane,[4] density, viscosity, and speed of sound in binary mixtures of 2-chloroethanol with MA, EA, PA, BA,[5] densities, and speeds of sound properties in binary mixtures of acrylonitrile with ethanenitrile, MA, EA, BA, dimethylformamide, dimethylacetamide, and dimethyl sulfoxide.[6] Earlier we have reported densities, viscosities, and speeds of sound of binary mixtures of sulfolane with EA, PA, and BA.[7] As a continuation of previous work in our laboratory,[7] we here report excess volumes V^E, and deviation in isentropic compressibility $\Delta\kappa_s$, for four binary systems, (γ-butyrolactone + methyl acetate), (γ-butyrolactone + ethyl acetate), (γ-butyrolactone + propyl acetate), and (γ-butyrolactone + butyl acetate) at atmospheric pressure and at the temperature of 303.15, 308.15, and 313.15 K. The experimental values of physical properties were used to calculate excess molar volumes, and isentropic compressibility deviations on mixing over the entire mole fraction range for the binary mixtures. The binary contribution calculated from the Redlich-Kister[8] type polynomial equation, and the adjustable parameters along with the standard deviations between experimental and calculated values are shown.

No experimental data corresponding to density, and speed of sound, have been found in the literature on the binary mixtures studied in this paper.

2. Experimental

2.1. Materials

All the component liquids are of analytical grade and are procured from Sigma-Aldrich, U.S. γ-Butyrolactone was purified by being distilled twice under reduced pressure.[9] Prior to measurements, all the chemicals with stated purities of better than 90 % were kept on molecular sieves (0.3 nm Merck, India) to remove any trace of water and degassed just before use. The densities and speed of sound of pure substances and their comparison with literature values are listed in Table 1.[10-12]

2.2. Apparatus and procedure

Binary mixtures were prepared by mass in air tight bottles. The mass measurements were performed on a Dhona 100 DS, India, single pan analytical balance with a resolution of ± $0.01 \cdot 10^{-6}$ kg. The required properties of the mixture were measured on the same day. The uncertainty in mole fraction was estimated to be less than ± $1 \cdot 10^{-4}$. Density of pure liquids and their mixtures were determined by using a $1 \cdot 10^{-5}$ m^3 double arm pycnometer.[13] The uncertainties in density and excess molar volume values was found to be ± $4 \cdot 10^{-5}$ $g \cdot cm^{-3}$ and $1 \cdot 10^{-3}$ $cm^3 \cdot mol^{-1}$.

Speeds of sound were determined by using an ultrasonic interferometer (Model M-82, Mittal Enterprises, India) operating at a 2 MHz frequency. The working principle used in the measurement of the speed of sound through a medium was based on the accurate determination of the wavelength of ultrasonic waves of known frequency produced by a quartz crystal in the measuring cell.[14,15] The temperature of the solution was controlled by circulating water at a desired temperature through the jacket of the double-walled cell. The speed of sound was measured with relative uncertainty of 0.3%.

In all the property measurements the temperature was controlled within ± 0.01 K using a constant temperature bath (INSREF model IRI-016 C, India), and the temperature was monitored with a platinum resistance thermometer with an accuracy of ± 0.001 K and an uncertainty of ± 0.004 K.

3. Results and discussion

Experimental values of density (ρ) and speed of sound (u) for the binary mixtures of γ-butyrolactone with methyl acetate, ethyl acetate, propyl acetate, and butyl acetate at T = (303.15, 308.15 and 313.15) K as a function of mole fraction are listed in Table 2.

The excess molar volumes (V^E) have been evaluated from density using

$$V^E = (x_1M_1 + x_2M_2)/\rho_m - (x_1M_1/\rho_1 + x_2M_2/\rho_2) \quad (1)$$

where ρ_m is the density of the mixture; x_1, M_1, ρ_1 and x_2, M_2 and ρ_2 are the mole fraction, molar mass and density of pure components respectively.

The deviations in isentropic compressibility ($\Delta\kappa_s$) have been evaluated using the equation

$$\Delta\kappa_s = \kappa_s - (\Phi_1\kappa_{s1} + \Phi_2\kappa_{s2}) \quad (2)$$

where κ_{s1}, κ_{s2} and κ_s are the isentropic compressibility of the pure components and observed isentropic compressibility of liquid mixture respectively.

Φ_i is the volume fraction and is calculated from the individual pure molar volumes, V_i, using the relation

$$\Phi_1 = x_iV_i/(\Sigma x_iV_i) \quad (3)$$

The excess or deviation properties ΔY were fitted by the method of non linear least squares to a Redlich-Kister type polynomial equation[8]

$$\Delta Y = x_1x_2\,\Sigma A_i\,(x_1 - x_2)^i \quad (4)$$

where A_0, A_1, A_2, A_3, and A_4 are adjustable binary coefficients. The coefficients A_i were estimated using multiparametric regression analysis based on a nonlinear least-squares method. The number of A_i parameters was optimized using F-test and is found to be (m = 5). In each case, the optimum number of coefficients A_i is determined from an examination of the variation of standard deviation (σ) as calculated by

$$\sigma(\Delta Y) = [\Sigma\,(\Delta Y_{obs} - \Delta Y_{cal})^2/(n - m)]^{1/2} \quad (5)$$

where 'n' represents the number of experimental points and 'm' is the number of coefficients used in fitting the data. The coefficients A_i and the estimated standard deviations (σ)V^E and (σ)$\Delta\kappa_s$ of the fit are shown in Table 3.

3.1. *Excess Molar Volume (V^E).*

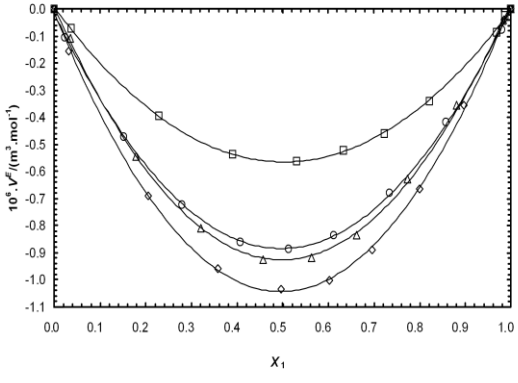
Dependences of excess molar volumes (V^E) for the mixtures investigated on the mole fraction of component 1, (x_1) at T = 303.15, 308.15 and 313.15 K are depicted graphically in figures 1, 2, 3, & 4. It can be seen that values of excess molar volume (V^E) are all negative for the four binary mixtures over the entire composition range, indicating negative deviations from ideal behaviors. The largest deviations are all located at $x_1 \approx$ 0.5-0.6. The negative V^E values for aliphatic esters at 303.15 K fall in the sequence:

BA < MA < EA < PA < 0

The above observation seen at all the temperatures (i.e., 308.15 and 313.15 K) and are more negative with increasing chain length of the ester molecules except in the case of BA.

The negative V^E values suggest specific interactions acting between the mixing components. The interactions may be classified as dipole-dipole forces resulting from the polarizability of ester molecules by the dipoles of γ-butyrolactone molecules (because of its large dipole moment μ = 4.12D).

The effect of temperature on V^E is noteworthy. There is a gradual decrease, followed by an increase in V^E with a rise in temperature for all mixtures except for EA, as shown in Fig. 4. Also the curves do not show

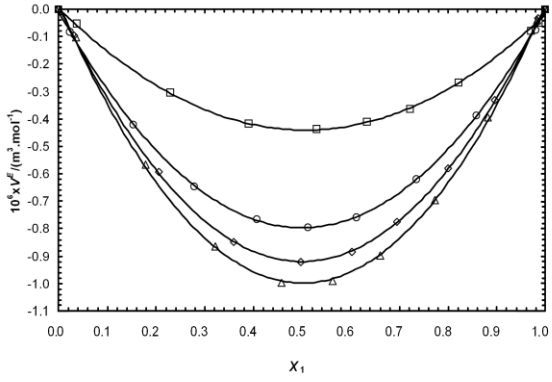


any systematic variation with the size of the esters.

Figure 1. Plots of excess molar volumes, V^E, as a function of mole fraction x at T = 303.15 K; ○, {GBL (1) + MA (2)}; Δ; {GBL (1) + EA (2)}; □, {GBL (1) + PA (2)}; ◊, {GBL (1) + BA (2)}; the symbols represent experimental values and lines represent the smoothed data of this work.

The negative deviations at all the temperatures and at equimolar composition follow the same trend but show different values except for EA and PA at 308.15 K which interchanges.

Figure 2. Plots of excess molar volumes, V^E, as a function of mole fraction x at T = 308.15 K; ○, {GBL (1) + MA (2)}; Δ; {GBL (1) + EA (2)}; □, {GBL (1) + PA (2)}; ◊, {GBL (1) + BA (2)}; the symbols represent



experimental values and lines represent the smoothed data of this work.

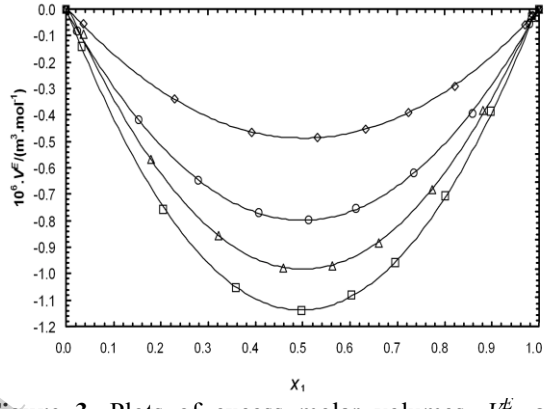


Figure 3. Plots of excess molar volumes, V^E, as a function of mole fraction x at T = 313.15 K; ○, {GBL (1) + MA (2)}; Δ; {GBL (1) + EA (2)}; □, {GBL (1) + PA (2)}; ◊, {GBL (1) + BA (2)}; the symbols represent experimental values and lines represent the smoothed data of this work.

3.2. *Deviation in Isentropic Compressibility ($\Delta\kappa_s$).*

The variation of $\Delta\kappa_s$ with volume fraction, Φ_1, of γ-butyrolactone with esters is presented in Table 2. All the systems of GBL + esters show negative deviations through $\Delta\kappa_s$ isotherms over the entire range of volume fractions and become more negative at higher temperatures. The negative $\Delta\kappa_s$ values for aliphatic esters at 313.15 K fall in the sequence:

PA < BA < EA < MA < 0

From the above observation $\Delta\kappa_s$ values become more negative with increasing chain length of the ester except for PA at 313.15 K. The $\Delta\kappa_s$ vs Φ_1 *curve at* 303.15 and 313.15 K show the same trends, but these are not shown to avoid overcrowding. However, the effect of temperature on $\Delta\kappa_s$ values is significant.

In general the dependence of V^E and $\Delta\kappa_s$ on composition is unsymmetrical and the magnitude varies with the type of solvent used.

4. *Conclusion*

In this paper we present the new experimental values on density and speeds of sound are measured between T = 303.15, 308.15 and 313.15 K and at

different compositions for four binary liquid mixtures of (□-butyrolactone +methyl acetate), (□-butyrolactone +ethyl acetate), (□-butyrolactone +propyl acetate), and (□-butyrolactone +butyl acetate). We have calculated the excess molar volume (V^E) and deviation in isentropic compressibility ($\square\square_s$) using the Redlich-Kister type equation. Excess molar volume and deviation in isentropic compressibility are negative. The sign of these quantities have been discussed in terms of the dipole-dipole interactions.

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TABLE 1 – Experimental Densities (ρ) and Speeds of Sound (u) of Pure Liquids at T = 303.15 K.

Component	$10^{-3}\cdot\rho$ /(kg·m^{-3})		u/(m·s^{-1})	
	exptl.	lit.	exptl.	lit.
γ-Butyrolactone	1.12032	1.11883[10]	1498.8	1500.0[11]
Methyl Acetate	0.92124	0.92180[12]	1134.6	1134.0[1]
Ethyl Acetate	0.8907	0.88890[3]	1128.0	
Propyl Acetate	0.86992		1164.3	
Butyl Acetate	0.87135	0.87136[6]	1179.4	1176.0[1]

TABLE 2 – The Values of Density (ρ), Speed of Sound (u), Excess Molar Volume (V^E) and Deviation in Isentropic Compressibility ($\Delta\kappa_s$) for the Binary Liquid Mixtures at Various Temperatures.

x_1	$10^{-3}\cdot\rho/(kg\cdot m^{-3})$	$u/(m\cdot s^{-1})$	$10^{6}\cdot V^E/(m^3\cdot mol^{-1})$	$10^{11}\cdot\Delta\kappa_s/(m^2\cdot N^{-1})$
		γ-Butyrolactone (1) + Methyl Acetate (2)		
		T = 303.15 K		
0.0000	0.9219	1134.6	0.0000	0.0000
0.0233	0.9275	1142.8	-0.1051	-0.7131
0.1531	0.9568	1190.5	-0.4735	-3.9539
0.2795	0.9845	1240.4	-0.7219	-6.1949
0.4079	1.0117	1293.2	-0.8592	-7.4714
0.5127	1.0331	1337.4	-0.8863	-7.8083
0.6124	1.0525	1379.1	-0.8340	-7.5105
0.7347	1.0753	1426.2	-0.6792	-6.2182
0.8586	1.0971	1467.7	-0.4192	-3.9619
0.9794	1.1172	1497.8	-0.0771	-0.7922
1.0000	1.1203	1498.8	0.0000	0.0000
		T = 308.15 K		
0.0000	0.9143	1112.2	0.0000	0.0000
0.0233	0.9197	1120.1	-0.0841	-0.6982
0.1531	0.9486	1167.2	-0.4205	-4.0587
0.2795	0.9762	1216.0	-0.6467	-6.3121
0.4079	1.0032	1269.1	-0.7669	-7.6981
0.5127	1.0247	1313.1	-0.7935	-8.0308
0.6124	1.0445	1354.5	-0.7599	-7.6873
0.7347	1.0677	1403.1	-0.6182	-6.3939
0.8586	1.0901	1446.4	-0.3883	-4.0723
0.9794	1.1109	1477.8	-0.0774	-0.7330
1.0000	1.1141	1480.9	0.0000	0.0000
		T = 313.15 K		
0.0000	0.9081	1089.6	0.0000	0.0000
0.0233	0.9136	1098.0	-0.0848	-0.8244
0.1531	0.9426	1148.3	-0.4194	-4.8111
0.2795	0.9703	1200.5	-0.6487	-7.4946
0.4079	0.9975	1257.4	-0.7703	-9.1593
0.5127	1.0191	1304.3	-0.7956	-9.5675
0.6124	1.0390	1348.1	-0.7551	-9.1778
0.7347	1.0626	1398.0	-0.6216	-7.6403
0.8586	1.0854	1440.0	-0.3956	-4.8566

0.9794	1.1061	1467.6	-0.0568	-0.8902
1.0000	1.1096	1469.0	0.0000	0.0000
		γ-Butyrolactone (1) + Ethyl Acetate (2)		
		T = 303.15 K		
0.0000	0.8907	1128.0	0.0000	0.0000
0.0362	0.8982	1135.7	-0.1093	-0.5452
0.1797	0.9294	1180.2	-0.5455	-3.9380
0.3221	0.9611	1230.1	-0.8092	-6.3965
0.4587	0.9922	1284.0	-0.9242	-7.8490
0.5634	1.0164	1328.7	-0.9173	-8.2284
0.6616	1.0394	1371.4	-0.8344	-7.8375
0.7741	1.0658	1418.5	-0.6264	-6.3478
0.8822	1.0915	1460.1	-0.3550	-3.8745
0.9836	1.1161	1495.1	-0.0439	-0.6716
1.0000	1.1203	1498.8	0.0000	0.0000
		T = 308.15 K		
0.0000	0.8836	1097.2	0.0000	0.0000
0.0362	0.8910	1107.0	-0.1011	-0.9280
0.1797	0.9225	1149.3	-0.5652	-4.2417
0.3221	0.9546	1198.4	-0.8642	-6.7813
0.4587	0.9860	1252.3	-0.9962	-8.2971
0.5634	1.0103	1297.3	-0.9895	-8.6600
0.6616	1.0334	1342.3	-0.8992	-8.3279
0.7741	1.0600	1393.6	-0.6957	-6.8747
0.8822	1.0856	1441.2	-0.3946	-4.3814
0.9836	1.1098	1476.0	-0.0402	-0.6870
1.0000	1.1141	1480.9	0.0000	0.0000
		T = 313.15 K		
0.0000	0.8781	1089.8	0.0000	0.0000
0.0362	0.8855	1099.0	-0.0940	-0.8560
0.1797	0.9170	1141.0	-0.5665	-4.2861
0.3221	0.9490	1189.4	-0.8566	-6.8622
0.4587	0.9804	1242.0	-0.9784	-8.3400
0.5634	1.0049	1286.2	-0.9710	-8.7006
0.6616	1.0281	1330.7	-0.8833	-8.3820
0.7741	1.0549	1381.4	-0.6824	-6.9161
0.8822	1.0807	1426.8	-0.3833	-4.2794
0.9836	1.1050	1463.4	-0.0230	-0.6497
1.0000	1.1096	1469.0	0.0000	0.0000

		γ-Butyrolactone (1) + Propyl Acetate (2)		
		T = 303.15 K		
0.0000	0.8699	1164.3	0.0000	0.0000
0.0331	0.8766	1170.6	-0.1579	-0.5600
0.2060	0.9120	1209.8	-0.6900	-3.3423
0.3598	0.9461	1253.4	-0.9596	-5.3981
0.4979	0.9789	1298.5	-1.0342	-6.4733
0.6031	1.0056	1338.5	-1.0037	-6.8234
0.6963	1.0304	1376.1	-0.8881	-6.5071
0.8010	1.0597	1421.5	-0.6638	-5.4323
0.8973	1.0878	1462.4	-0.3551	-3.4568
0.9853	1.1153	1497.0	-0.0389	-0.7287
1.0000	1.1203	1498.8	0.0000	0.0000
		T = 308.15 K		
0.0000	0.8685	1155.8	0.0000	0.0000
0.0331	0.8746	1161.5	-0.0966	-0.4511
0.2060	0.9092	1196.1	-0.5923	-2.7242
0.3598	0.9425	1234.4	-0.8488	-4.3554
0.4979	0.9746	1275.8	-0.9208	-5.2972
0.6031	1.0007	1312.4	-0.8847	-5.5604
0.6963	1.0250	1348.4	-0.7757	-5.3286
0.8010	1.0539	1392.4	-0.5811	-4.4056
0.8973	1.0821	1436.2	-0.3312	-2.8383
0.9853	1.1091	1473.8	-0.0332	-0.4227
1.0000	1.1141	1480.9	0.0000	0.0000
		T = 313.15 K		
0.0000	0.8589	1143.6	0.0000	0.0000
0.0331	0.8655	1149.2	-0.1416	-0.4971
0.2060	0.9014	1183.1	-0.7565	-2.9221
0.3598	0.9357	1221.2	-1.0541	-4.6770
0.4979	0.9687	1262.3	-1.1413	-5.6663
0.6031	0.9952	1299.1	-1.0831	-5.9543
0.6963	1.0200	1335.7	-0.9585	-5.7466
0.8010	1.0491	1381.2	-0.7077	-4.8315
0.8973	1.0773	1424.9	-0.3872	-3.0972
0.9853	1.1045	1465.6	-0.0321	-0.6679
1.0000	1.1096	1469.0	0.0000	0.0000

		γ-Butyrolactone (1) + Butyl Acetate (2)		
		T = 303.15 K		
0.0000	0.8714	1179.4	0.0000	0.0000
0.0373	0.8773	1185.5	-0.0721	-0.4682
0.2303	0.9110	1222.9	-0.3941	-2.8135
0.3922	0.9434	1263.7	-0.5356	-4.5352
0.5311	0.9750	1306.5	-0.5627	-5.5265
0.6337	1.0010	1343.7	-0.5220	-5.8220
0.7232	1.0261	1379.1	-0.4628	-5.5616
0.8221	1.0565	1422.3	-0.3423	-4.6206
0.9702	1.1089	1484.8	-0.0857	-0.9947
0.9867	1.1150	1492.3	-0.0241	-0.4377
1.0000	1.1203	1498.8	0.0000	0.0000
		T = 308.15 K		
0.0000	0.8672	1158.2	0.0000	0.0000
0.0373	0.8730	1165.0	-0.0532	-0.5727
0.2303	0.9058	1203.5	-0.3064	-3.1137
0.3922	0.9377	1244.6	-0.4174	-4.8971
0.5311	0.9688	1287.8	-0.4360	-5.9265
0.6337	0.9947	1325.0	-0.4112	-6.2031
0.7232	1.0196	1361.0	-0.3651	-5.9418
0.8221	1.0500	1404.4	-0.2680	-4.9239
0.9702	1.1027	1469.7	-0.0789	-1.2214
0.9867	1.1092	1476.3	-0.0505	-0.5900
1.0000	1.1141	1480.9	0.0000	0.0000
		T = 313.15 K		
0.0000	0.8624	1136.8	0.0000	0.0000
0.0373	0.8682	1143.7	-0.0555	-0.6225
0.2303	0.9013	1183.5	-0.3410	-3.4624
0.3922	0.9332	1226.1	-0.4639	-5.4539
0.5311	0.9645	1270.7	-0.4856	-6.5790
0.6337	0.9904	1309.0	-0.4541	-6.8589
0.7232	1.0152	1346.6	-0.3922	-6.5894
0.8221	1.0456	1391.9	-0.2918	-5.4962
0.9702	1.0979	1457.4	-0.0581	-1.3093
0.9867	1.1043	1464.0	-0.0250	-0.6103
1.0000	1.1096	1469.0	0.0000	0.0000

TABLE 3. The Binary Coefficients (A_i) and Standard Errors (σ) of γ-Butyrolactone (1) + Aliphatic Esters (2)

Function	T/K	A_0	A_1	A_2	A_3	A_4	σ
	γ-Butyrolactone (1) + Methyl Acetate (2)						
	303.15						
	308.15						
$10^6 \cdot V^E/(m^3 \cdot mol^{-1})$	313.15						
	303.15						
$10^{11} \cdot \Delta\kappa_s/(m^2 \cdot N^{-1})$	308.15						
	313.15						
	γ-Butyrolactone (1) + Ethyl Acetate (2)						
	303.15						
	308.15						
$10^6 \cdot V^E/(m^3 \cdot mol^{-1})$	313.15						
	303.15						
$10^{11} \cdot \Delta\kappa_s/(m^2 \cdot N^{-1})$	308.15						
	313.15						
	γ-Butyrolactone (1) + Propyl Acetate (2)						
	303.15						
	308.15						
$10^6 \cdot V^E/(m^3 \cdot mol^{-1})$	313.15						
	303.15						
$10^{11} \cdot \Delta\kappa_s/(m^2 \cdot N^{-1})$	308.15						
	313.15						
	γ-Butyrolactone (1) + Butyl Acetate (2)						
	303.15						
	308.15						
$10^6 \cdot V^E/(m^3 \cdot mol^{-1})$	313.15						
	303.15						
$10^{11} \cdot \Delta\kappa_s/(m^2 \cdot N^{-1})$	308.15						
	313.15						