IJRAR.ORG E-ISSN: 2348-1269, P-ISSN: 2349-5138

INTERNATIONAL JOURNAL OF RESEARCH AND ANALYTICAL REVIEWS (IJRAR) | IJRAR.ORG

An International Open Access, Peer-reviewed, Refereed Journal

THERMODYNAMICS AND SONOCHEMICAL STUDIES OF BINARY MIXTURES OF (o-, m- AND p-) XYLENES WITH BUTAN-1-OL AT DIFFERENT TEMPERATURES

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***Abstract*:** Density and ultrasonic velocity for binary mixtures of ortho-, meta- and para –xylenes with butan-1-ol have been measured over the entire range of composition at 298.15, 303.15, 308.15 and 313.15K and atmospheric pressure .From the experimental data ,the parameters isentropic compressibility (K_s) ,specific acoustic impedance (Z), intermolecular free length (L_f), relative association (R_A) and molar sound velocity (R)have been calculated. In the present binary liquid mixtures, it has been observed that intermolecular free length (L_f),decreases and ultrasonic velocity increases with increase of mole fractions of xylenes with temperatures. The increase of Z values with increase of x_1 suggests the presence of intermolecular interactions between alcohol and xylene molecules. The decrease of K_s with x_1 (Xylenes), supports the formation of donor-acceptor complexes between xylenes and 1- butanol molecules.

Keywords: Butan-1-ol, Xylenes, Isentropic compressibility (K_s), Specific acoustic impedance (Z), Relative association (R_A).

I. INTRODUCTION:

Ultrasonic velocity measurement through liquid mixtures of non- electrolytes provides an excellent tool to investigate intra and intermolecular interactions between unlike and like molecules. Alcohols exist in the form of aggregates. When they are mixed with other non-electrolyte molecules, the aggregates of alcohols dissociate and form intermolecular complexes with unlike molecules. In order to investigate depolymerisation of these alcohols in the presence of other non-nonelectrolyte molecules, extensive studies of ultrasonic measurements through binary liquid mixtures with alcohols as one of the components have been reported.

The speed of sound was measured [1-3] for primary alcohols at different temperatures. Isentropic compressibilities for binary mixtures of alcohols with methyl cyclohexane were reported by Naidu and Rao[4]. The results have been discussed in terms of depolymerisation of polymerized alcohols by methyl cyclohexane. Many attempts[5-9] were made to measure the ultrasonic behavior of binary mixtures of cycloalkanes with alcohols. Swamy et al.[10] measured Ultrasonic velocities of binary mixtures of toluene with n-propanol, iso-propanol, n-butanol, iso-propanol, n-pentanol, n-hexanol, n-heptanol and cyclohexanol over the whole range of composition at 303.15 K. Isentropic compressibilities and the deviations from linear dependence of isentropic compressibility on volume fraction of toluene have been calculated. Ultrasonic behavior of binary liquid mixtures of benzene, toluene, o-, m-, p-xylenes with triethyl amine was reported by Reddy et al.[11] Ultrasonic velocities in binary liquid mixtures of m-xylene, o-xylene, carbon tetrachloride and tolune in nitrobenzene were obtained by Jajoo et al.[12] More work has been reported about excess, sonochemical and thermodynamic properties of binary liquid mixtures.[13-19]

Experimental Section:

All the chemicals were used from S.D. fine chemicals having purity > 99%. Binary mixtures were prepared by mass in airtight stoppard glass bottles. Densities of pure liquids and their mixtures were determined by using a 15 cm^3 bicapillary pycnometer. The estimated uncertainty of density measurements of solvent and binary mixtures was 0.0005 $g.cm^{-3}$. At least three to four measurements were made, which had an average deviation of $\pm$ 5 x 10^{-5} $g.cm^{-3}$. The Table 1 reveals that literature values and our measured values match very well. An ultrasonic interferometer is simple and direct device to determine the ultrasonic velocity (U) in liquids with high degree of accuracy. From the knowledge of wavelength and frequency U can be obtained by the relation

Velocity = Wave length X Frequency

$$U = \lambda \text{ X } f$$

In present investigation, F-81 ultrasonic interferometer is operating at frequency of 2MHz. The accuracy in velocity measurement was $\pm$ 0.03%.The interferometer cell is a specially designed double walled cell for maintaining the temperature of the liquid constant during the experiment. The maximum capacity of cell is 12 CC.

Table 1: Comparison of experimental and literature values of densities and ultrasonic velocities for pure liquids

Liquid	Temp. (K)	ρ ($g.cm^{-3}$)		η (mPa.s)	
		Expt.	Lit.	Expt.	Lit.
o-xylene	298.15	0.87596	0.87594[a]	0.748	0.756[a]
	303.15	0.87161	0.87174[a]	0.702	0.709[c]
	308.15	0.86724	0.86754[a]	0.660	0.662[f]
	313.15	0.86275	0.86320[b]	0.622	0.629[d]
m-xylene	298.15	0.86050	0.86009[a]	0.578	0.579[f]
	303.15	0.85608	0.85581[a]	0.546	0.546[a]
	308.15	0.85158	0.85154[a]	0.515	0.517[f]
	313.15	0.84707	0.84726[a]	0.492	0.496[a]
p-xylene	298.15	0.85674	0.85661[a]	0.598	0.605[a]
	303.15	0.85226	0.85225[a]	0.565	0.566[a]
	308.15	0.84773	0.84778[e]	0.528	0.545[e]
	313.15	0.84317	0.84352[a]	0.502	0.514[a]
Butan-1-ol	298.15	0.80560	0.80590[f]	2.586	2.571[a]
	303.15	0.80165	0.80180[a]	2.286	2.285[a]
	308.15	0.79760	0.79815[b]	2.024	2.000[a]
	313.15	0.79349	0.79405[a]	1.797	1.782[a]

[a]Ref 34. [b]Ref 35. [c]Ref 36. [d]Ref 37. [e]Ref 38. [f]Ref 39.

Table 2: Ultrasonic velocity data for the o-Xylene + Butan-1-ol systems

Temp	X_1	ρ	U	K_S	Z x 10^{-3}	L_f	R_A	R	ΔK_S
298.15	0.0000	0.80560	1236.2	812.3	995.9	0.5632	1.000	0.987	0.00
	0.1002	0.81486	1252.1	782.8	1020.3	0.5528	0.962	1.023	-6.23
	0.2004	0.82344	1265.3	758.5	1041.9	0.5442	0.937	1.058	-8.61
	0.3012	0.83118	1276.3	738.6	1060.8	0.5370	0.922	1.093	-7.86
	0.4017	0.83821	1285.7	721.7	1077.7	0.5308	0.916	1.129	-5.23
	0.5070	0.84523	1294.4	706.1	1094.1	0.5251	0.918	1.165	-1.55
	0.6010	0.85126	1300.3	694.8	1106.9	0.5208	0.930	1.197	3.37
	0.6921	0.85688	1307.0	683.2	1119.9	0.5165	0.944	1.229	6.78
	0.8026	0.86351	1316.7	668.0	1137.0	0.5107	0.965	1.267	8.88
	0.9049	0.86974	1330.5	649.5	1157.2	0.5036	0.981	1.304	5.57
	1.0000	0.87596	1345.6	630.5	1178.7	0.4962	1.000	1.338	1.00
303.15	0.0000	0.80165	1225.5	830.6	982.4	0.5750	1.000	0.989	0.00
	0.1002	0.81080	1239.6	802.6	1005.1	0.5652	0.965	1.025	-4.44
	0.2004	0.81930	1252.5	778.0	1026.2	0.5565	0.940	1.060	-6.97
	0.3012	0.82685	1263.2	757.9	1044.5	0.5492	0.925	1.095	-6.15
	0.4017	0.83384	1271.8	741.4	1060.5	0.5432	0.921	1.130	-2.94
	0.5070	0.84084	1279.9	726.0	1076.2	0.5375	0.924	1.167	1.07
	0.6010	0.84687	1286.9	713.0	1089.8	0.5327	0.932	1.199	4.53
	0.6921	0.85250	1292.9	701.7	1102.2	0.5285	0.947	1.231	8.43
	0.8026	0.85912	1302.9	685.7	1119.3	0.5224	0.966	1.269	9.85
	0.9049	0.86531	1315.5	667.8	1138.3	0.5155	0.985	1.306	7.29
	1.0000	0.87161	1331.7	646.9	1160.7	0.5074	1.000	1.340	1.00
308.15	0.0000	0.79760	1212.2	853.2	966.9	0.5865	1.000	0.991	0.00
	0.1002	0.80664	1224.9	826.3	988.1	0.5772	0.967	1.026	-3.67
	0.2004	0.81507	1236.6	802.3	1007.9	0.5688	0.943	1.061	-5.73
	0.3012	0.82256	1246.1	782.9	1025.0	0.5619	0.930	1.096	-4.37
	0.4017	0.82952	1254.2	766.4	1040.4	0.5559	0.925	1.131	-1.42
	0.5070	0.83650	1261.1	751.7	1054.9	0.5505	0.929	1.167	3.19
	0.6010	0.84249	1267.0	739.4	1067.4	0.5460	0.939	1.199	7.21
	0.6921	0.84810	1273.3	727.3	1079.9	0.5415	0.951	1.231	10.10
	0.8026	0.85470	1282.7	711.1	1096.3	0.5355	0.970	1.269	11.27
	0.9049	0.86080	1294.4	693.4	1114.2	0.5287	0.988	1.306	8.71
	1.0000	0.86724	1310.7	671.2	1136.7	0.5202	1.000	1.340	1.00
313.15	0.0000	0.79349	1198.5	877.4	951.0	0.6013	1.000	0.992	0.00
	0.1002	0.80242	1209.6	851.8	970.6	0.5925	0.970	1.027	-2.72
	0.2004	0.81079	1219.9	828.8	989.1	0.5844	0.948	1.061	-4.18
	0.3012	0.81821	1228.6	809.7	1005.3	0.5776	0.935	1.097	-2.89
	0.4017	0.82514	1235.4	794.1	1019.4	0.5720	0.931	1.131	0.67
	0.5070	0.83208	1241.5	779.7	1033.0	0.5668	0.935	1.167	5.29
	0.6010	0.83802	1246.9	767.5	1044.9	0.5624	0.944	1.199	9.09
	0.6921	0.84360	1252.8	755.3	1056.9	0.5579	0.955	1.231	11.63
	0.8026	0.85015	1261.7	738.9	1072.6	0.5518	0.973	1.269	12.30
	0.9049	0.85617	1272.5	721.3	1089.5	0.5452	0.990	1.305	9.63
	1.0000	0.86275	1288.2	698.5	1111.4	0.5365	1.000	1.339	1.00

Table 3: Ultrasonic velocity data for the m-Xylene + Butan-1-ol systems

Temp	X_1	ρ	U	K_S	$Z \times 10^{-3}$	L_f	R_A	R	ΔK_S
298.15	0.0000	0.80560	1236.2	812.3	995.9	0.5632	1.000	0.987	0.00
	0.1006	0.81297	1247.8	790.0	1014.4	0.5554	0.968	1.024	-3.27
	0.2012	0.81951	1257.8	771.3	1030.8	0.5488	0.946	1.061	-4.24
	0.3014	0.82541	1265.6	756.4	1044.6	0.5434	0.934	1.098	-2.56
	0.4017	0.83093	1270.5	745.6	1055.7	0.5395	0.935	1.134	2.23
	0.5072	0.83639	1276.7	733.5	1067.8	0.5352	0.937	1.172	5.63
	0.6010	0.84104	1281.5	724.0	1077.8	0.5317	0.945	1.206	9.06
	0.7013	0.84588	1286.7	714.1	1088.4	0.5280	0.960	1.242	12.22
	0.8032	0.85063	1293.0	703.2	1099.9	0.5240	0.978	1.279	13.94
	0.9021	0.85533	1303.9	687.7	1115.3	0.5182	0.992	1.316	10.01
	1.0000	0.86050	1320.2	666.8	1136.0	0.5102	1.000	1.354	1.00
303.15	0.0000	0.80165	1225.5	830.6	982.4	0.5750	1.000	0.989	0.00
	0.1006	0.80883	1234.9	810.7	998.8	0.5680	0.972	1.026	-1.85
	0.2012	0.81527	1244.0	792.6	1014.2	0.5617	0.950	1.063	-3.14
	0.3014	0.82113	1250.2	779.2	1026.6	0.5569	0.940	1.099	-0.83
	0.4017	0.82652	1254.6	768.7	1037.0	0.5531	0.939	1.135	3.47
	0.5072	0.83205	1259.2	758.0	1047.7	0.5493	0.943	1.173	7.44
	0.6010	0.83668	1263.1	749.1	1056.8	0.5460	0.951	1.206	10.87
	0.7013	0.84148	1267.6	739.6	1066.7	0.5425	0.965	1.242	13.75
	0.8032	0.84618	1273.4	728.8	1077.5	0.5386	0.981	1.279	14.91
	0.9021	0.85079	1282.9	714.2	1091.5	0.5331	0.995	1.316	11.25
	1.0000	0.85608	1298.7	692.6	1111.8	0.5250	1.000	1.353	1.00
308.15	0.0000	0.79760	1212.2	853.2	966.9	0.5865	1.000	0.991	0.00
	0.1006	0.80455	1220.7	834.1	982.1	0.5799	0.973	1.027	-1.22
	0.2012	0.81089	1228.7	816.9	996.3	0.5739	0.953	1.064	-1.76
	0.3014	0.81671	1234.3	803.7	1008.1	0.5693	0.943	1.100	0.72
	0.4017	0.82217	1238.4	793.1	1018.2	0.5655	0.942	1.136	4.80
	0.5072	0.82761	1242.8	782.3	1028.6	0.5616	0.945	1.174	8.56
	0.6010	0.83219	1246.2	773.8	1037.1	0.5586	0.954	1.208	12.21
	0.7013	0.83697	1250.5	764.1	1046.6	0.5550	0.967	1.243	14.85
	0.8032	0.84164	1255.8	753.4	1056.9	0.5512	0.983	1.280	16.08
	0.9021	0.84621	1264.6	739.0	1070.1	0.5458	0.997	1.317	12.53
	1.0000	0.85158	1280.5	716.2	1090.4	0.5374	1.000	1.354	1.00
313.15	0.0000	0.79349	1198.5	877.4	951.0	0.6013	1.000	0.992	0.00
	0.1006	0.80026	1207.1	857.6	966.0	0.5945	0.973	1.029	-0.70
	0.2012	0.80652	1214.7	840.3	979.7	0.5885	0.955	1.066	-0.14
	0.3014	0.81229	1220.5	826.4	991.4	0.5836	0.945	1.102	2.66
	0.4017	0.81779	1225.3	814.5	1002.0	0.5793	0.943	1.138	6.35
	0.5072	0.82314	1229.9	803.1	1012.4	0.5753	0.947	1.176	10.52
	0.6010	0.82773	1234.0	793.4	1021.4	0.5718	0.955	1.210	13.76
	0.7013	0.83248	1238.8	782.8	1031.3	0.5679	0.967	1.246	16.30
	0.8032	0.83708	1244.7	771.1	1041.9	0.5637	0.984	1.283	17.30
	0.9021	0.84158	1253.3	756.5	1054.8	0.5583	0.999	1.320	14.28
	1.0000	0.84707	1270.6	731.2	1076.3	0.5489	1.000	1.358	1.00

Table 4: Ultrasonic velocity data for the p-Xylene + Butan-1-ol system

Temp	X_1	ρ	U	K_S	$Z \times 10^{-3}$	L_f	R_A	R	ΔK_S
298.15	0.0000	0.80560	1236.2	812.3	995.9	0.5632	1.000	0.987	0.00
	0.1020	0.81268	1247.7	790.4	1014.0	0.5555	0.967	1.025	-4.85
	0.2009	0.81901	1256.5	773.4	1029.1	0.5495	0.945	1.061	-6.49
	0.3012	0.82465	1263.2	760.0	1041.7	0.5447	0.933	1.098	-5.25
	0.4016	0.82963	1268.4	749.2	1052.3	0.5409	0.930	1.135	-2.21
	0.5071	0.83461	1273.3	739.0	1062.7	0.5372	0.932	1.174	1.18
	0.6002	0.83884	1277.0	731.0	1071.2	0.5343	0.940	1.207	4.51
	0.7011	0.84324	1281.5	722.1	1080.6	0.5310	0.953	1.244	7.20
	0.8033	0.84765	1285.9	713.5	1090.0	0.5278	0.972	1.281	9.62
	0.9043	0.85214	1295.6	699.1	1104.0	0.5225	0.985	1.319	5.65
	1.0000	0.85674	1306.2	684.1	1119.1	0.5168	1.000	1.355	1.00
303.15	0.0000	0.80165	1225.5	830.6	982.4	0.5750	1.000	0.989	0.00
	0.1020	0.80861	1235.0	810.8	998.6	0.5681	0.970	1.027	-3.38
	0.2009	0.81482	1243.2	794.1	1013.0	0.5622	0.949	1.063	-5.28
	0.3012	0.82037	1248.8	781.6	1024.5	0.5578	0.938	1.100	-3.57
	0.4016	0.82527	1253.2	771.5	1034.2	0.5541	0.935	1.136	-0.38
	0.5071	0.83019	1255.6	764.0	1042.4	0.5514	0.942	1.174	5.21
	0.6002	0.83435	1260.7	754.1	1051.9	0.5478	0.945	1.209	6.16
	0.7011	0.83877	1264.3	745.9	1060.5	0.5448	0.958	1.245	9.10
	0.8033	0.84313	1268.9	736.6	1069.8	0.5415	0.975	1.282	10.57
	0.9043	0.84756	1276.9	723.6	1082.2	0.5367	0.989	1.320	7.56
	1.0000	0.85226	1288.2	707.1	1097.9	0.5305	1.000	1.355	1.00
308.15	0.0000	0.79760	1212.2	853.2	966.9	0.5865	1.000	0.991	0.00
	0.1020	0.80443	1220.7	834.2	982.0	0.5800	0.972	1.028	-2.66
	0.2009	0.81055	1228.2	817.9	995.5	0.5743	0.951	1.064	-4.24
	0.3012	0.81596	1233.5	805.5	1006.5	0.5699	0.941	1.101	-2.55
	0.4016	0.82087	1237.4	795.6	1015.7	0.5664	0.938	1.138	0.82
	0.5071	0.82576	1241.3	785.9	1025.0	0.5629	0.940	1.176	4.18
	0.6002	0.82989	1244.1	778.5	1032.5	0.5603	0.948	1.210	7.61
	0.7011	0.83427	1247.5	770.2	1040.8	0.5573	0.961	1.246	10.44
	0.8033	0.83860	1251.9	760.9	1049.8	0.5539	0.977	1.283	11.73
	0.9043	0.84295	1259.7	747.6	1061.9	0.5490	0.990	1.321	8.41
	1.0000	0.84773	1271.0	730.2	1077.5	0.5426	1.000	1.357	1.00
313.15	0.0000	0.79349	1198.5	877.4	951.0	0.6013	1.000	0.992	0.00
	0.1020	0.80021	1205.5	859.9	964.7	0.5953	0.974	1.029	-2.18
	0.2009	0.80624	1211.4	845.2	976.7	0.5902	0.955	1.065	-3.08
	0.3012	0.81152	1215.5	834.0	986.4	0.5863	0.945	1.102	-1.09
	0.4016	0.81642	1218.3	825.2	994.6	0.5832	0.943	1.138	2.45
	0.5071	0.82131	1221.9	815.5	1003.6	0.5797	0.944	1.176	4.89
	0.6002	0.82541	1223.3	809.6	1009.7	0.5776	0.953	1.210	9.13
	0.7011	0.82973	1226.1	801.7	1017.3	0.5748	0.965	1.246	11.64
	0.8033	0.83401	1229.7	792.9	1025.6	0.5714	0.980	1.283	12.85
	0.9043	0.83828	1236.5	780.2	1036.5	0.5670	0.993	1.320	9.39
	1.0000	0.84317	1247.2	762.5	1051.6	0.5605	1.000	1.355	1.00

II. RESULTS AND DISCUSSIONS:

Thermodynamic functions are highly useful in understanding the interactions in binary liquid mixtures. Ultrasonic velocity measurement through liquid mixtures of non-electrolytes provide an excellent tool to investigate inter and intramolecular interactions between like and unlike molecules. Ultrasonic velocity (U) is a thermodynamic function .Many other thermodynamic functions such as isentropic compressibility[K_s], specific acoustic impedance [Z], relative association[R_A], intermolecular free length [L_f], Rao's constant[R] are calculated from ultrasonic velocity with use of following relation.

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

$Z = \rho U$

$R_A = (Lf/L_f\text{'})^3$

$L_f = K (K_s)^{1/2}$

$R = (M/ \rho) U^{1/3}$

Where M is the average molecular weight, K is the temperature dependent constant whose values are 1.976 x 10^{-6}, 1.995 x 10^{-6}, 2.008 x 10^{-6}, 2.030 x 10^{-6} at 298.15, 303.15, 308.15, 313.15 K . L_F' is the free length of ideal mixture.

Tables 2, 3 and 4 contains values of U and all the calculated parameters Viz. K_s, Z, L_f, R_A, R for the all binary mixtures as a function of X (xylenes) at 298.15, 303.15, 308.15, 313.15 K temperature. It is observed from the tables that ultrasonic velocity increases with increases of xylene concentrations in binary mixtures at all temperature.

In the present binary mixtures it has been observed that intermolecular free length decreases and ultrasonic velocity increases with increase of mole fractions of xylenes at all temperatures. The nature of variation of L_f strongly depends on the size of molecules of both the components in the mixture [20]. Acoustic impedance [Z], for alcohols + xylenes increases with increase of mole fraction of xylene at all temperatures. The increase of Z values with increase of X_1suggests the presence of intermolecular interactions between alcohol-xylene molecules forming π---HO donor-acceptor complexes. As expected K_s, decreases with increase of xylene concentrations in all binary mixtures at all temperatures. Rajendran[21] suggested that K_s should decreases with increase of ultrasonic velocity which gives insight into structure making and structure breaking capacity of in binary mixture as observed in the present investigation. The molar sound velocity, also known as Rao's constant [R].[22-23] The slight increase in R with increase of X_1 may cause due to specific interactions between xylene and alkanol molecules forming donor- acceptor complexes through π----HO bond formation.[24-29] **Alco**hols are associated through H- bonding.[30-33] When a small quantity of xylene is added to alcohol, the breaking of H-bonding of alcohols takes place leading to close packing of monomers, which is responsible for the continuous decrease in internal pressure in all binary mixtures. It is futher seen that internal pressure in all binary mixtures decreases with increase of temperature due to dissociation of alcohol-xylene and alcohol-alcohol aggregates.

Graphical representation of ΔK_s and Z as a function of ∅ (xylene) at 298.15 K for the binary mixtures of xylenes (o-,m- and p-) with butan-1-ol illustrated in figure 1 and 2. The values of ΔK_s for the mixtures of alcohols with xylenes are negative in mixtures rich in alcohols and positive in mixtures rich in xylenes. ΔK_s values for all binary systems of xylenes with n-alkanol are negative in lower concentration region of xylene molecules. This implies that there are strong specific interactions between xylene and alkanol molecules in this region. The benzene ring in xylene has high π electron cloud density due to presence of two electron donating -CH_3groups**.** This makes xylene molecules proton acceptor and alkanol proton donars, thus forming π ----HO donar- acceptor complexes. The ΔK_s values increase with increase of temperatures.

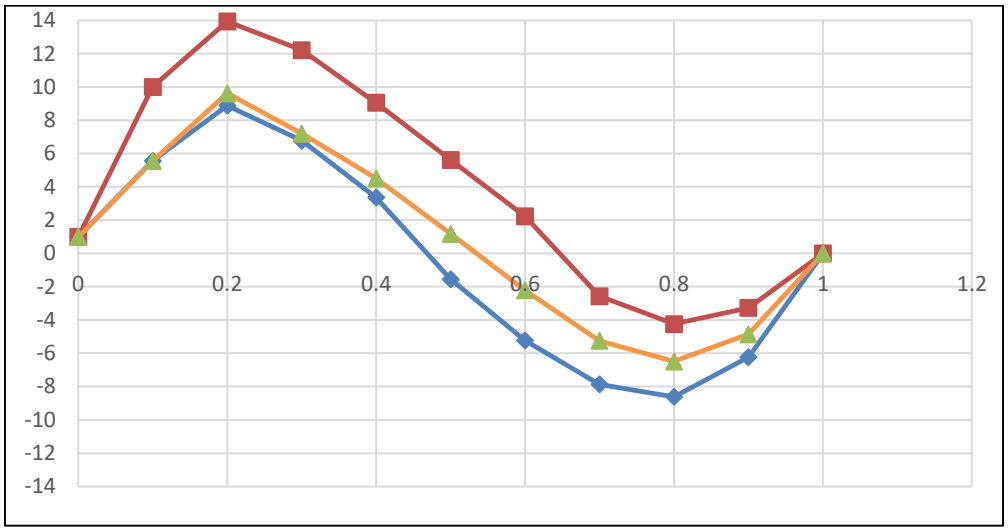

Figure 1: ΔK_S values at 298.15K for X_1 butan-1-ol + (1-X_1) o-xylene(♦), m-xylene(■), p-xylene(▲).

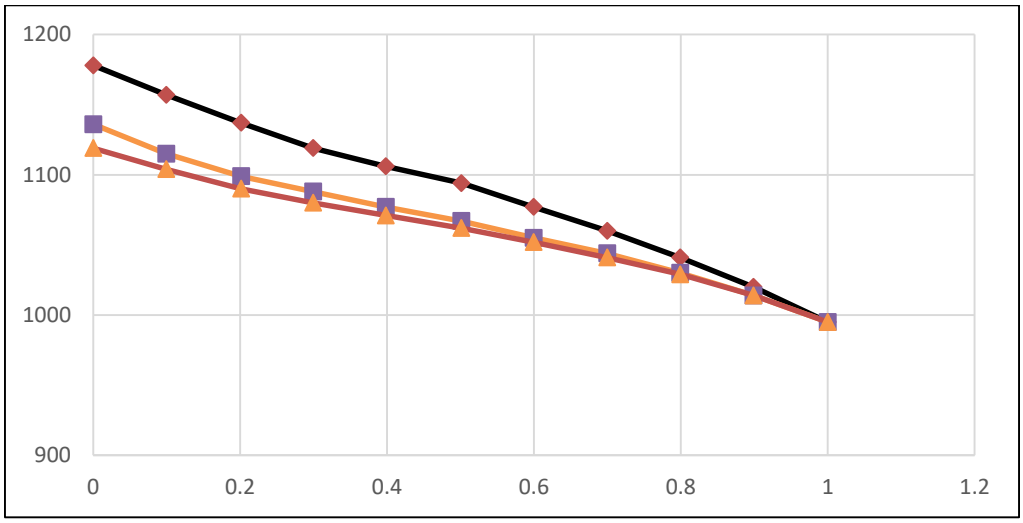

Figure 2: Z values at 298.15K for X_1 butan-1-ol + (1-X_1) o-xylene(♦), m-xylene(■), p-xylene(▲).

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