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"SONOCHEMICAL AND THERMODYNAMICS STUDIES OF BINARY LIQUID MIXTURES OF XYLENES (ORTHO-, META- AND PARA) WITH PROPAN-1-OL AT DIFFERENT TEMPERATURES''.

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ABSTRACT:

Density and ultrasonic velocity for binary liquid mixtures of ortho-, meta- and para –xylenes with propan-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 with increase of mole fractions of xylenes, intermolecular free length (L_f) decreases and ultrasonic velocity increases with temperatures. The increase of mole fraction (x_1) with increase of specific acoustic impedance (Z) values suggests the presence of intermolecular interactions between xylene and alcohol molecules.

Keywords:

Xylenes, Propan-1- ol, Isentropic compressibility (K_s), Specific acoustic impedance (Z), Intermolecular free length (L_f), Relative association (R_A), Molar sound velocity (R) .

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 are strongly self associated molecules through hydrogen bonding and for binary solutions rich in alcohols, three dimensional network of hydrogen bond is believed to be present. Xylenes are non-associated and potential electron donors. When they are mixed with other liquids it is expected to give more interactions due to two electron donating CH_3- groups. However on the other hand the increased steric hindrance may arise due to presence of bulky CH_3- groups in xylenes. This hindrance may differ for o-, m- and p-xylenes due to positions of two CH_3- groups.

The speed of sound was measured [1-3] for primary alcohols at different temperatures. Isentropic compressibility's 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 compressibility 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] More work has been reported about excess, sonochemical and thermodynamic properties of binary liquid mixtures.[12-18]

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 operating at frequency of 2MHz. The accuracy in velocity measurement was $\pm$ 0.03%. 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]
propan-1-ol	298.15	0.80015	0.79997[f]	1.948	1.943[a]
	303.15	0.79609	0.79600[a]	1.737	1.725[a]
	308.15	0.79187	0.79170[a]	1.559	1.623[a]
	313.15	0.78758	0.78785[a]	1.403	1.428[a]

[a]Ref 28. [b]Ref 29. [c]Ref 30. [d]Ref 31. [ERef] 32. [f]Ref 33.

Table No 2 : Density and Ultrasonic Velocity Data for the o-Xylene + Propan-1-ol Systems.

Temp	X_1	ρ	U	K_S	Z x 10^{-3}	L_f	R_A	R	ΔK_S
	0.0000	0.80015	1205.0	860.7	964.2	0.5797	1.000	0.799	0.00
	0.1009	0.81229	1227.8	816.6	997.3	0.5647	0.920	0.854	-8.76
	0.2006	0.82280	1247.0	781.6	1026.0	0.5524	0.866	0.907	-12.77
	0.3002	0.83171	1260.9	756.3	1048.7	0.5434	0.837	0.960	-10.28
	0.4007	0.83948	1274.3	733.6	1069.7	0.5352	0.819	1.015	-7.65
298.15	0.4987	0.84632	1285.5	715.0	1087.9	0.5284	0.816	1.067	-3.84
	0.6016	0.85229	1295.9	698.1	1105.3	0.5221	0.827	1.112	0.66
	0.7004	0.85889	1306.0	682.6	1121.7	0.5163	0.849	1.176	3.88
	0.7987	0.86450	1315.8	668.1	1137.5	0.5108	0.886	1.228	6.53
	0.9008	0.87024	1330.6	649.0	1157.9	0.5034	0.933	1.284	3.83
	1.0000	0.87596	1345.6	630.5	1178.7	0.4962	1.000	1.338	1.00
	0.0000	0.79609	1194.0	881.1	950.5	0.5922	1.000	0.801	0.00
	0.1009	0.80805	1214.2	839.5	981.1	0.5780	0.926	0.855	-5.72
	0.2006	0.81852	1233.3	803.3	1009.4	0.5654	0.871	0.908	-10.35
	0.3002	0.82730	1246.9	777.5	1031.5	0.5583	0.841	0.962	-7.85
	0.4007	0.83505	1259.9	754.4	1052.1	0.5479	0.824	1.016	-5.25
303.15	0.4987	0.84192	1270.7	735.6	1069.8	0.5411	0.821	1.069	-1.23
	0.6016	0.84862	1281.1	718.0	1087.2	0.5346	0.832	1.124	2.90
	0.7004	0.85449	1291.3	701.8	1103.4	0.5285	0.853	1.177	5.81
	0.7987	086004	1301.4	686.6	1119.2	0.5227	0.889	1.230	8.02
	0.9008	0.86581	1315.8	667.2	1139.2	0.5153	0.936	1.286	5.27
	1.0000	0.87161	1331.7	646.9	1160.7	0.5074	1.000	1.340	1.00
	0.0000	0.79187	1178.0	910.0	932.8	0.6057	1.000	0.802	0.00
	0.1009	0.80373	1196.5	869.1	961.7	0.5920	0.929	0.855	-4.36
	0.2006	0.81409	1213.8	833.7	988.2	0.5798	0.877	0.909	-7.53
	0.3002	0.82283	1227.5	806.5	1010.1	0.5703	0.846	0.962	-5.83
	0.4007	0.83058	1239.8	783.2	1029.8	0.5620	0.829	1.016	-2.88
303.15	0.4987	0.83745	1250.0	764.2	1046.8	0.5521	0.827	1.069	1.30
	0.6016	0.84410	1260.5	745.6	1064.0	0.5483	0.836	1.124	4.90
	0.7004	0.85005	1270.3	729.0	1079.8	0.5422	0.858	1.177	7.77
	0.7987	0.85559	1280.6	712.7	1095.6	0.5361	0.892	1.230	9.26
	0.9008	0.86136	1293.8	693.6	1114.4	0.5288	0.940	1.285	7.12
	1.0000	0.86724	1310.7	671.2	1136.7	0.5202	1.000	1.340	1.00
	0.0000	0.78758	1166.0	933.9	918.3	0.6204	1.000	0.803	0.00
	0.1009	0.79932	1182.9	894.1	945.5	0.6070	0.931	0.857	-3.74
	0.2006	0.80958	1198.5	859.9	970.3	0.5953	0.881	0.910	-6.15
	0.3002	0.81825	1210.9	833.5	990.8	0.5861	0.852	0.963	-4.15
	0.4007	0.82595	1221.9	810.9	1009.2	0.5781	0.836	1.017	-0.85
313.15	0.4987	0.83282	1231.4	791.9	1025.5	0.5712	0.833	1.069	2.98
	0.6016	0.83946	1241.1	773.4	1041.9	0.5645	0.842	1.124	6.35
	0.7004	0.84542	1250.1	756.9	1056.9	0.5585	0.863	1.177	9.07
	0.7987	0.85099	1259.4	740.9	1071.7	0.5525	0.897	1.230	10.59
	0.9008	0.85664	1272.1	721.4	1089.7	0.5452	0.943	1.285	7.85
	1.0000	0.86275	1288.2	698.5	1111.4	0.5365	1.000	1.339	1.00

Table No 3: Density and Ultrasonic Velocity Data for the m-Xylene + Propan-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	0.80015	1205.0	860.7	964.2	0.5797	1.000	0.799	0.00
	0.1010	0.80992	1222.4	826.3	990.0	0.5680	0.930	0.855	-4.20
	0.2007	0.81799	1237.6	798.2	1012.3	0.5583	0.881	0.910	-5.91
	0.2996	0.82497	1249.1	776.9	1030.5	0.5508	0.853	0.965	-3.76
	0.4002	0.83125	1258.4	759.7	1046.0	0.5446	0.839	1.020	0.39
	0.5003	0.83688	1267.7	743.5	1060.9	0.5388	0.836	1.075	3.45
	0.5999	0.84198	1276.1	729.3	1074.5	0.5336	0.846	1.130	6.57
	0.7004	0.84679	1283.6	716.7	1086.9	0.5290	0.869	1.185	9.91
	0.8004	0.85123	1291.6	704.2	1099.4	0.5244	0.904	1.241	11.87
	0.9009	0.85574	1304.2	687.0	1116.1	0.5179	0.946	1.297	8.08
	1.0000	0.86050	1320.2	666.8	1136.0	0.5102	1.000	1.354	1.00
303.15	0.0000	0.79609	1194.0	881.1	950.5	0.5922	1.000	0.801	0.00
	0.1010	0.80568	1209.4	848.6	974.4	0.5812	0.933	0.856	-3.14
	0.2007	0.81369	1223.4	821.1	995.5	0.5717	0.885	0.911	-4.93
	0.2996	0.82062	1233.6	800.8	1012.3	0.5645	0.858	0.966	-2.52
	0.4002	0.82690	1242.0	784.0	1027.0	0.5586	0.845	1.021	1.46
	0.5003	0.83249	1250.0	768.0	1040.6	0.5532	0.843	1.076	4.92
	0.5999	0.83759	1257.3	755.2	1053.1	0.5483	0.852	1.131	8.23
	0.7004	0.84236	1264.5	742.4	1065.2	0.5436	0.874	1.186	10.92
	0.8004	0.84680	1271.8	730.1	1077.0	0.5391	0.908	1.241	12.67
	0.9009	0.85127	1283.2	713.4	1092.3	0.5329	0.949	1.297	9.00
	1.0000	0.85608	1298.7	692.6	1111.8	0.5250	1.000	1.353	1.00
303.15	0.0000	0.79187	1178.0	910.0	932.8	0.6057	1.000	0.802	0.00
	0.1010	0.80132	1192.6	877.4	955.7	0.5948	0.935	0.857	-2.41
	0.2007	0.80925	1206.2	849.3	976.1	0.5852	0.887	0.912	-4.08
	0.2996	0.81614	1215.9	828.8	992.3	0.5781	0.860	0.966	-1.24
	0.4002	0.82240	1224.2	811.4	1006.8	0.5720	0.847	1.022	2.70
	0.5003	0.82800	1232.0	795.7	1020.1	0.5664	0.845	1.077	6.23
	0.5999	0.83309	1240.0	780.7	1033.0	0.5610	0.853	1.131	8.51
	0.7004	0.83784	1246.3	768.4	1044.2	0.5566	0.876	1.186	12.19
	0.8004	0.84227	1253.8	755.3	1056.0	0.5518	0.909	1.241	13.53
	0.9009	0.84672	1265.1	737.9	1071.2	0.5455	0.950	1.298	9.58
	1.0000	0.85158	1280.5	716.2	1090.4	0.5374	1.000	1.354	1.00
313.15	0.0000	0.78758	1166.0	933.9	918.3	0.6204	1.000	0.803	0.00
	0.1010	0.79693	1180.0	900.9	940.5	0.6093	0.935	0.859	-1.45
	0.2007	0.80477	1193.7	872.0	960.7	0.5995	0.888	0.914	-2.69
	0.2996	0.81162	1203.5	850.7	976.8	0.5921	0.861	0.969	0.38
	0.4002	0.81786	1212.0	832.4	991.2	0.5857	0.848	1.024	4.43
	0.5003	0.82347	1220.1	815.8	1004.7	0.5798	0.846	1.079	7.88
	0.5999	0.82854	1227.8	800.6	1017.3	0.5744	0.854	1.134	10.85
	0.7004	0.83329	1235.2	786.6	1029.3	0.5693	0.876	1.189	13.43
	0.8004	0.83771	1243.0	772.6	1041.3	0.5643	0.909	1.245	14.65
	0.9009	0.84211	1254.3	754.8	1056.3	0.5577	0.951	1.301	10.82
	1.0000	0.84707	1270.6	731.2	1076.3	0.5489	1.000	1.358	1.00

Table No 4: Density and Ultrasonic Velocity Data for the p-Xylene + Propan-1-ol System.

Temp	X_1	ρ	U	K_S	Z x 10^{-3}	L_f	R_A	R	ΔK_S
	0.0000	0.80015	1205.0	860.7	964.2	0.5797	1.000	0.799	0.00
	0.1009	0.80960	1222.9	825.9	990.1	0.5679	0.927	0.855	-7.18
	0.2011	0.81765	1236.7	799.7	1011.2	0.5588	0.880	0.911	-9.23
	0.3006	0.82421	1247.5	779.6	1028.2	0.5517	0.851	0.966	-7.82
298.15	0.4009	0.82992	1256.5	763.2	1042.8	0.5459	0.837	1.022	-4.85
	0.5005	0.83494	1264.3	749.3	1055.6	0.5409	0.835	1.077	-1.39
	0.6009	0.83965	1271.2	737.0	1067.4	0.5364	0.844	1.133	2.21
	0.7006	0.84403	1277.8	725.6	1078.5	0.5323	0.866	1.188	5.18
	0.8006	0.84821	1284.7	714.3	1089.7	0.5281	0.899	1.243	7.04
	0.9011	0.85243	1295.0	699.5	1103.9	0.5226	0.942	1.299	4.39
	1.0000	0.85674	1306.2	684.1	1119.1	0.5168	1.000	1.355	1.00
	0.0000	0.79609	1194.0	881.1	950.5	0.5922	1.000	0.801	0.00
	0.1009	0.80532	1209.5	848.8	974.0	0.5812	0.932	0.857	-5.09
	0.2011	0.81331	1222.8	822.3	994.5	0.5721	0.884	0.912	-7.73
	0.3006	0.81975	1232.8	802.7	1010.6	0.5652	0.856	0.967	-6.23
	0.4009	0.82541	1241.1	786.5	1024.4	0.5595	0.842	1.023	-3.25
303.15	0.5005	0.83045	1248.1	773.0	1036.5	0.5547	0.840	1.078	0.36
	0.6009	0.83513	1254.5	760.9	1047.7	0.5503	0.850	1.134	3.85
	0.7006	0.83951	1260.9	749.2	1058.5	0.5461	0.870	1.189	6.35
	0.8006	0.84367	1267.2	738.1	1069.1	0.5420	0.903	1.244	8.24
	0.9011	0.84782	1276.9	723.4	1082.6	0.5366	0.945	1.300	5.49
	1.0000	0.85226	1288.2	707.1	1097.9	0.5305	1.000	1.355	1.00
	0.0000	0.79187	1178.0	910.0	932.8	0.6057	1.000	0.802	0.00
	0.1009	0.80094	1192.3	878.3	955.0	0.5951	0.934	0.857	-3.66
	0.2011	0.80884	1205.3	851.0	974.9	0.5858	0.887	0.913	-6.23
	0.3006	0.81525	1215.0	830.9	990.5	0.5788	0.859	0.968	-4.50
	0.4009	0.82090	1223.3	814.0	1004.2	0.5729	0.844	1.024	-1.64
303.15	0.5005	0.82591	1230.3	799.9	1016.0	0.5679	0.842	1.079	1.94
	0.6009	0.83058	1236.9	787.0	1027.3	0.5633	0.851	1.135	5.14
	0.7006	0.83496	1243.1	775.0	1037.9	0.5590	0.872	1.190	7.83
	0.8006	0.83911	1249.5	763.3	1048.5	0.5548	0.905	1.245	9.52
	0.9011	0.84323	1258.9	748.3	1061.5	0.5493	0.947	1.301	6.87
	1.0000	0.84773	1271.0	730.2	1077.5	0.5426	1.000	1.357	1.00
	0.0000	0.78758	1166.0	933.9	918.3	0.6204	1.000	0.803	0.00
	0.1009	0.79651	1178.3	904.3	938.5	0.6104	0.937	0.859	-2.86
	0.2011	0.80429	1189.7	878.4	956.9	0.6017	0.892	0.914	-5.16
	0.3006	0.81080	1197.8	859.6	971.2	0.5952	0.865	0.969	-3.13
	0.4009	0.81632	1204.5	844.4	983.3	0.5899	0.852	1.024	0.41
313.15	0.5005	0.82135	1210.8	830.5	994.5	0.5850	0.850	1.079	3.41
	0.6009	0.82602	1216.3	818.3	1004.7	0.5807	0.859	1.134	6.67
	0.7006	0.83037	1221.6	807.0	1014.4	0.5767	0.879	1.189	9.27
	0.8006	0.83450	1227.4	795.4	1024.3	0.5725	0.910	1.244	10.52
	0.9011	0.83858	1236.2	780.3	1036.7	0.5671	0.949	1.300	7.15
	1.0000	0.84317	1247.2	762.5	1051.6	0.5605	1.000	1.355	1.00

RESULTS AND DISCUSSIONS:

Ultrasonic velocity measurement through liquid mixtures of non-electrolytes provide an excellent tool to investigate inter and intermolecular interactions between like and unlike 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. Thermodynamic functions are highly useful in understanding the interactions in binary liquid mixtures. 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) and excess isentropic compressibility(ΔK_s) are calculated from ultrasonic velocity with use of following relation.

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

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

$$Z = \rho U$$

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

$R_A = (Lf/L_f ')^3$

$$\Delta Ks = Ks_{12} - Ks_1 \emptyset_1 - Ks_2 \emptyset_2$$

Where M is the average molecular weight, K is the temperature dependent constant whose values are 1.976×10^{-6} , 1.995×10^{-6} , 2.008×10^{-6} , 2.030×10^{-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 [19]. Acoustic impedance (Z), for alcohols + xylenes increases with increase of mole fraction of xylene at all temperatures. As expected K_s, decreases with increase of xylene concentrations in all binary mixtures at all temperatures. Rajendran[20] 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).[21-22] The slight increase in R with increase of X_1 may caused due to specific interactions between xylene and alkanol molecules forming donor- acceptor complexes through π ----HO bond formation.[23-27]

Graphical representation of ΔK_s and Z as a function of $\emptyset$ (xylene) at 298.15 K for the binary mixtures of xylenes (o-,m-and p-) with propan-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.

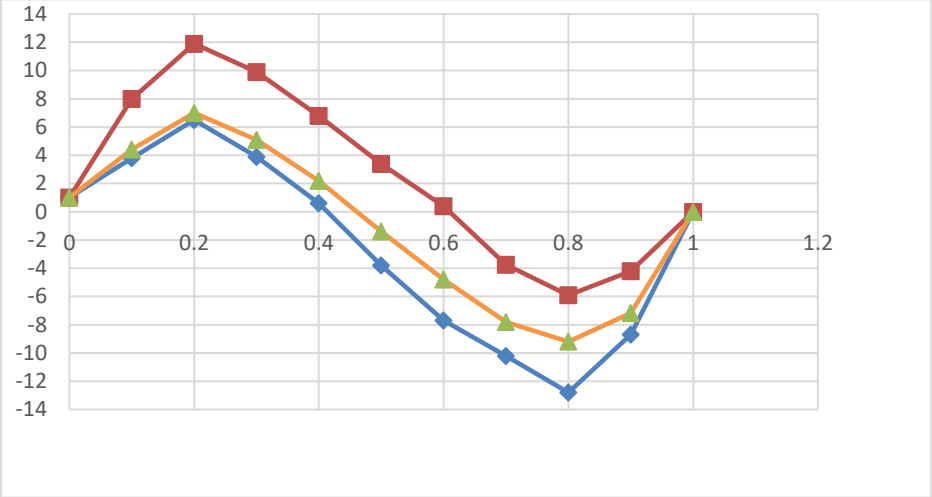

Fig. 1. ΔK_S values at 298.15K for X_1 propan-1-ol + (1-X_1) o-xylene(♦), m-xylene(■), p-xylene(▲).

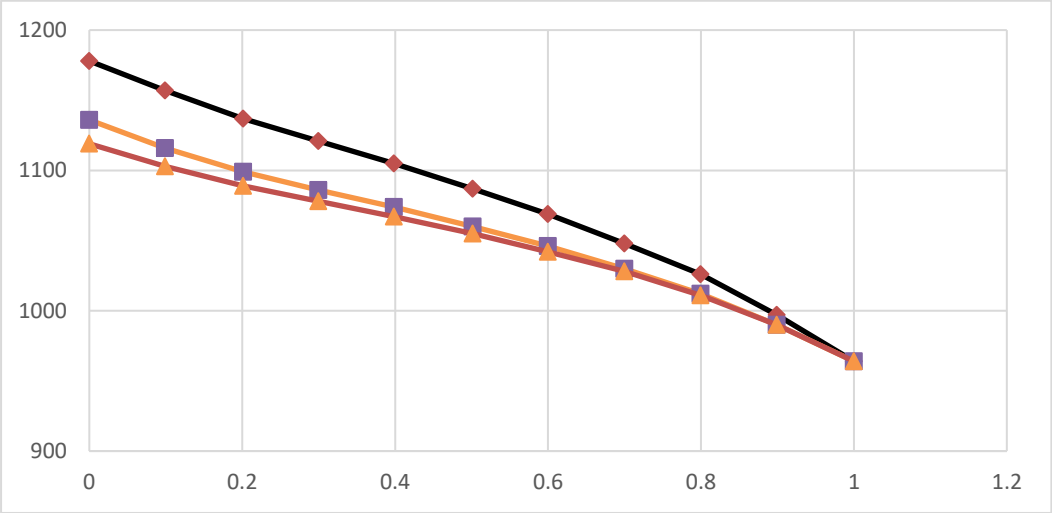

Fig. 2. Z values at 298.15K for X_1 propan-1-ol + (1-X_1) o-xylene(♦), m-xylene(■), p-xylene(▲).

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