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Volumetric and Acoustic Properties for Binary Mixtures of *N,N*–Dimethylformamide with 2-Butanol and 2-Pentanol at Temperatures between 298.15 K and 318.15 K

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Abstract The volumetric and acoustic properties for the binary mixtures of *N,N*-dimethylformamide with two polar solvents 2-butanol and 2-pentanol have been measured over the entire range of composition at T= (298.15, 303.15, 308.15, 313.15 and 318.15) K and at atmospheric pressure. From these experimental data, for both the systems, excess molar volume, V^E, versus mole fraction, x_1, of *N,N*-dimethylformamide curves were calculated and found as sigmoid at all temperatures. The partial molar volume of 2- butanol changes almost rapidly but tends to form a hump like in maximum and minimum mole fraction at different temperatures. Excess isentropic compressibility, K_S^E, values were negative for both the systems over the entire composition range. All the excess parameter values were fitted using the Redlich-Kister polynomial smoothing equation. In addition, the results were analyzed in terms of molecular interactions and structural effects.

Keywords 2-Butanol, *N,N*-Dimethylformamide, Excess Molar Volume, Excess Isentropic Compressibility, 2-Pentanol, Molecular interactions

1. Introduction

The volumetric and acoustic properties of binary mixtures would be a great importance in processing the engineering designs and also helpful in getting information about the molecular structure and intermolecular forces in liquid mixtures, which can be very helpful in making the choice of solvent in various applications [1, 2]. The thermodynamic properties of binary liquid mixtures containing protic, aprotic, and associated liquids have been studied previously [3-7].

N,N-dimethylformamide (DMF) is a colorless, non-hydrogen bonded, high-boiling, mobile, highly polar liquid with a faint, characteristic odor. DMF does not decompose at distilling at low pressure and is freely miscible with water, alcohols, ethers, ketones, esters, carbon disulfide, and chlorinated and aromatic hydrocarbons. Molecular interactions of DMF with some solvents reported by various thermodynamic and thermophysical measurements [8]. DMF is aprotic and unassociated [9] in its pure liquid state. It belongs to the so-called super solvents, owing to its miscibility with almost all common polar and no polar solvents [10], probably due to its high polarity with large dipole moment (μ = 3.8D) and moderately high dielectric constant (ε = 36.76) [11]. Several topics and examples of thermodynamic studies are depicted on the basis of the structural behavior of DMF for binary mixtures of non-electrolytes [12].

Alkanols are interesting versatile solvents, used in chemical and technological processes which are inexpensive and easily available at high purity. These polar liquids are self-associated through hydrogen bonding, creating multimers of different degrees [13, 14] whether association is disturbed when they are mixed with another solvent [15]. Alkanols are the -OH functional group containing compounds bonded to a carbon atom. Alkanols of short chain length have a greater proton-donor capacity, so the strength of bonding is expected to decrease with an increase in their chain lengths. Moreover, because of the steric hindrance of alkyl groups, hydrogen bonds are weakened for higher length alkanols [12, 16]. Alkanols are also of interest in their own right and serve as simple examples of biologically and industrially important amphiphilic materials [17].

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Additionally, these solvents (amides and alkanols) and their mixtures are used as reaction solvents and find applications in many chemical and industrial processes [18].

Recently, the substantial research work has been reported on the excess properties of *N, N*-dimethylformamide +2-alkanols [19], formamide+2-alkanols [20], *N,N*-dimethylformamide+1-alkanols [21-25] acetophenone +2-alkanols [26] ethylmethylketone+2-alkanols [27], *N, N*-dimethylaniline+1-alkanols,+2-alkanols,+2-methyl-1-pro panol,+2-methyl-2-propanol [28]. Though a number of studies have been carried out on the physical properties of DMF, the reported results are not so sufficient with the existence of some discrepancies. The study of this work is a part of an ongoing research effort to measure and characterize the binary mixtures containing 2-alkanols [29, 30].

The densities, ultrasonic velocity and excess properties of the binary blends of these compounds have important impact on the injection process. In order to gain a better understanding of the competitive role of H-bonds associations, the nature and dynamics of the molecular structures in these mixtures, we report the consequences of volumetric and acoustic investigations of 2-butanol and 2-pentanol within the temperature range at T= (298.15, 303.15, 308.15, 313.15 and 318.15) and at atmospheric pressure.

Hence, in this paper, we reported the volumetric properties like densities, ρ, excess molar volume, V^E, partial molar volume, V, and acoustic properties like ultrasonic velocity, u, excess ultrasonic velocity, u^E, excess isentropic compressibility, K_S^E, excess acoustic impedance, Z^E, of the binary systems of *N,N*-dimethylformamide with two positional isomeric alkanols, such as, 2-butanol and 2-pentanol. These results have been used to discuss the nature of interaction between unlike molecules in terms of hydrogen bonding, dipole-dipole interaction, proton-acceptor interaction and dispersive forces, and also used to analyze the effect of branching in the alkanol and position of the hydroxyl group in the interaction with the *N,N*-dimethylformamide.

Previously, a large number of researchers have demonstrated that in addition to dipole-dipole and Van der Waals interactions, alcohols are strongly self-associated through H-bonding (O-H•••••O-H) interaction. As the liquid they form clusters or networks with restricted rotations about the H-bonds, and hence show variable degrees of polymeric aggregates. The molecular interactions in the present mixtures are also controlled through the formation of the hydrogen bonds of the type C=O•••••H-O, between unlike molecules [31]. In addition, the effects of difference in shape and steric factors on molecular interactions were observed. The results of the present work were found very much similar with the literatures published previously with a few differences. The findings of this study can be utilized in having a better insight into molecular interactions between the components of the systems. It also helps in understanding the biological systems, synthesis of various compounds, process designing in chemical, petrochemical, pharmaceutical industries, paints, inks as well as in other prospects.

2. Experimental Section

2.1. Materials and Method

Table 1. Experimental (bold letter) and Literature Values of Densities, ρ/(Kg.m^{-3}) and ultrasonic velocity, $\boldsymbol{u}$/(ms^{-1}) of the pure *N,N*-dimethylformamide (DMF), 2-Butanol (2-BuOH) and 2-Pentanol (2-PnOH) at different temperatures

	DMF		2-BuOH		2-PnOH	
T/K	ρ	u	ρ	u	ρ	u
298.15	**944.270**	**1458.05**	**802.876**	**1212.46**	**805.395**	**1232.92**
	944.2 [33]	1459.6 [23]	802.56 [26]	1212.11 [40]	805.30 54]	
	944.06 [34]		802.60 [39]	1212.1 [50]	805.24 [26]	
	944.60 [35]		802.690 [40]	1212 [51]	805.40 [39]	
	944.03 [36]		802.634 [41]			
303.15	**939.496**	**1438.45**	**798.683**	**1195.45**	**801.295**	**1214.93**
	939.371 [21]	1434.7 [23]	798.66 [42]	1195 [52]	801.3 [55]	
	939.5 [33]		798.8 [43]	1194 [53]	801.3 [56]	
	939.8 [35]		798.88 [44]		801.3 [57]	
308.15	**934.707**	**1418.88**	**794.386**	**1178.45**	**797.109**	**1197.23**
	934.4 [37]	1420.8 [23]	794.180 [40]	1176.10 [40]	797.1 [58]	
	935.0 [33]		794.18 [45]	1175.8 [50]		
313.15	**929.906**	**1399.66**	**789.992**	**1161.43**	**792.841**	**1179.60**
	929.6 [37]		789.43 [46]		792.8 [58]	
	929.549 [38]		789.41 [47]			
	789.7 [48]					
318.15	**925.096**	**1378.98**	**785.493**	**1144.45**	**788.491**	**1162.00**
	925.49 [35]		785.17 [46]		788.3 [58]	
	925.1 [37]		785.15 [49]			

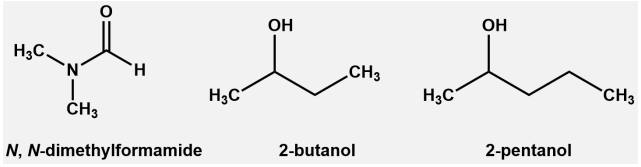

Figure 1. Chemical structures of the compounds used in the experiments

DMF (Aldrich, purity HPLC grade 99.9+ %), 2-BuOH (Aldrich, purity 99+%) and 2-PnOH (Aldrich, purity 98%) were used without further treatment which is shown in Fig. 1. The densities and ultrasonic velocity of pure chemicals were compared with literature values, which show satisfactory agreements as shown in Table 1.

All of the measurements on densities, ρ, and ultrasonic velocity, u, were carried out on an Anton Paar DSA 5000 (Austria-Europe) densimeter. The mixtures samples were prepared by mixing the pure components at different proportions up to ±0.0001g, which was then converted into mole fraction. All molar quantities used in this paper were based on the IUPAC relative atomic mass table [32].

3. Results and Discussion

3.1. Densities and Excess Molar Volumes

The experimental results of the density, ρ, measurements of binary mixtures of DMF with 2-BuOH, and DMF with 2-PnOH as a common component, over the whole composition range expressed as mole fractions, x_1, of DMF ($0 \leq x_1 \leq 1$) at different temperatures are listed in Table 2. The excess molar volumes, V^E, were calculated by using the following relation

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

Where, ρ_1, and ρ_2 represent the densities and M_1 and M_2 the molar volume of component 1 & 2 respectively.

Excess ultrasonic velocities, u^E, from their values in an ideal mixture were calculated from the equation:

$$u^E = u - (\phi_1 u_1 + \phi_2 u_2) \quad (2)$$

Where, ϕ_1 and ϕ_2, u_1 and u_2 are the volume fraction and ultrasonic velocity of component 1 and component 2, respectively.

Excess values of acoustic impedance, (Z^E) were calculated by the following equation:

$$Z^E = Z - (\phi_1 Z_1 + \phi_2 Z_2) \quad (3)$$

Densities, ρ, and ultrasonic velocity, u, were represented by a polynomial equation,

$$Y = \sum_{i=0}^{n} a_i x_1^i \quad (4)$$

Where, x_1 is the mole fraction of DMF, a_i is the regression coefficient and n is the degree of polynomial. The values of ρ and u fitted to equation (4) well for n = 4. The coefficients a_i of Equation (4) and relevant values of r^2 are listed in Table 3.

Table 2. Experimental densities (ρ), excess molar volume (V^E), ultrasonic velocity (u), excess ultrasonic velocity (u^E), excess isentropic compressibility values (K_s^E), acoustical impedance (Z^E) of the systems DMF (x_1) + 2-BuOH (x_2) and + 2-PnOH (x_2) for different molar ratios at different temperatures

x_1	ρ (kgm^{-3})	u (ms^{-1})	V^E (m^{3}mol^{-1})	u^E (ms^{-1})	K_s^Ex10^{-12} (Pa^{-1})	Z^Ex10^{-3} (kgm^{-2}s^{-1})
			T=298.15K			
			DMF+2-BuOH			
0.0000	802.876	1212.46	0.000000	0.0000	0.0000	0.0000
0.0501	808.412	1221.62	0.051275	-1.2418	-5.1712	-4.1991
0.0998	814.391	1230.81	0.057082	-2.5396	-9.5810	-8.1522
0.1494	820.230	1240.03	0.087625	-3.9582	-13.2035	-11.9287
0.1986	826.253	1249.46	0.102792	-5.2549	-16.3315	-15.3106
0.2514	832.892	1259.16	0.111956	-7.2640	-18.2764	-19.1462
0.2994	838.943	1269.01	0.129339	-8.2407	-20.4661	-21.6352
0.3491	845.789	1278.61	0.097950	-10.038	-21.2227	-24.5820
0.3998	853.100	1289.34	0.045545	-11.137	-22.1926	-26.6438
0.4495	860.600	1300.63	-0.025648	-11.644	-23.0998	-27.8472
0.5002	868.514	1312.25	-0.109503	-12.271	-23.2197	-28.8213
0.5498	876.198	1324.71	-0.171279	-12.006	-23.5260	-28.6514
0.5999	884.624	1337.68	-0.282010	-11.573	-23.2564	-27.9612
0.6524	892.730	1351.75	-0.314051	-10.883	-22.3857	-26.6122
0.7001	900.111	1364.88	-0.331903	-10.132	-20.9800	-24.8931
0.7496	907.623	1378.78	-0.324764	-9.3078	-18.8252	-22.6723
0.7999	914.938	1392.87	-0.277351	-8.7504	-15.6421	-20.2480
0.8502	922.106	1407.87	-0.206137	-7.5377	-12.2139	-16.7833
0.9000	928.796	1422.85	-0.090818	-6.4661	-7.9416	-13.0228
0.9501	936.300	1439.38	-0.030371	-4.1948	-3.7221	-7.6586
1.0000	944.270	1458.05	0.000000	0.0000	0.0000	0.0000

x_1	ρ (kgm^{-3})	u (ms^{-1})	V^E (m^{3}mol^{-1})	u^E (ms^{-1})	K_s^Ex10^{-12} (Pa^{-1})	Z^Ex10^{-3} (kgm^{-2}s^{-1})
			T=303.15K			
			DMF+2-BuOH			
0.0000	798.683	1195.45	0.000000	0.0000	0.0000	0.0000
0.0501	804.205	1204.05	0.050489	-1.6905	-4.7153	-4.5084
0.0998	810.160	1213.28	0.056186	-2.8361	-9.5071	-8.2930
0.1494	815.793	1222.38	0.107076	-4.2611	-13.2556	-12.0326
0.1986	821.967	1232.09	0.102739	-5.1634	-17.0121	-15.0515
0.2514	828.575	1242.07	0.112298	-6.7679	-19.5129	-18.5090
0.2994	834.597	1251.07	0.130175	-8.4795	-20.8673	-21.5846
0.3491	841.410	1261.13	0.099046	-9.6960	-22.3289	-24.0125
0.3998	848.374	1271.33	0.078713	-11.198	-22.8548	-26.3975
0.4495	855.730	1282.56	0.017839	-11.642	-23.8699	-27.5328
0.5002	863.058	1294.26	-0.012715	-12.059	-24.2150	-28.3169
0.5498	871.791	1306.23	-0.181361	-12.155	-24.1624	-28.4634
0.5999	880.089	1318.99	-0.283729	-11.800	-23.8197	-27.8505
0.6524	888.162	1332.79	-0.315971	-11.239	-22.8122	-26.6333
0.7001	895.575	1345.88	-0.339602	-10.399	-21.4570	-24.8553
0.7496	902.987	1359.92	-0.326055	-9.2973	-19.4825	-22.4121
0.7999	910.279	1373.88	-0.278927	-8.7283	-16.1945	-20.0085
0.8502	917.420	1388.77	-0.207420	-7.4816	-12.6736	-16.5530
0.9000	924.083	1403.50	-0.091358	-6.5149	-8.1639	-12.9352
0.9501	931.500	1419.60	-0.025713	-4.5252	-3.5903	-7.8925
1.0000	939.496	1438.45	0.000000	0.0000	0.0000	0.0000

x_1	ρ (kgm^{-3})	u (ms^{-1})	V^E (m^{3}mol^{-1})	u^E (ms^{-1})	K_s^Ex10^{-12} (Pa^{-1})	Z^Ex10^{-3} (kgm^{-2}s^{-1})
			T=308.15K			
			DMF+2-BuOH			
0.0000	794.386	1178.45	0.000000	0.0000	0.0000	0.0000
0.0501	799.909	1187.21	0.048321	-1.4189	-5.2843	-4.2386
0.0998	805.852	1196.13	0.052924	-2.7622	-9.9534	-8.1359
0.1494	811.647	1205.16	0.083677	-4.1436	-13.8914	-11.7969
0.1986	817.619	1214.21	0.099496	-5.5916	-17.0415	-15.2279
0.2514	824.200	1224.04	0.109487	-7.2216	-19.6102	-18.6732
0.2994	830.197	1233.22	0.127871	-8.6383	-21.3950	-21.4751
0.3491	836.983	1243.08	0.096824	-9.9342	-22.8131	-23.9467
0.3998	843.914	1253.43	0.077225	-11.162	-23.6858	-26.0782
0.4495	851.244	1264.12	0.015938	-12.021	-24.2601	-27.5582
0.5002	858.582	1275.48	-0.018379	-12.650	-24.3995	-28.5211
0.5498	867.345	1287.80	-0.194019	-12.268	-24.8854	-28.2551
0.5999	875.517	1300.48	-0.287560	-11.862	-24.5944	-27.6053
0.6524	883.548	1314.28	-0.318651	-11.162	-23.6976	-26.2788
0.7001	890.938	1326.99	-0.342646	-10.573	-22.0559	-24.7441
0.7496	898.324	1340.68	-0.328901	-9.6856	-19.8240	-22.5215
0.7999	905.581	1354.86	-0.280483	-8.7568	-16.7463	-19.8244
0.8502	912.702	1369.23	-0.208802	-7.8881	-12.7768	-16.7536
0.9000	919.312	1384.32	-0.089231	-6.4186	-8.5232	-12.7191
0.9501	926.800	1399.72	-0.031276	-4.9829	-3.3449	-8.2442
1.0000	934.707	1418.88	0.000000	0.0000	0.0000	0.0000

x_1	ρ (kgm^{-3})	u (ms^{-1})	V^E (m^{3}mol^{-1})	u^E (ms^{-1})	K_s^Ex10^{-12} (Pa^{-1})	Z^Ex10^{-3} (kgm^{-2}s^{-1})
			T=313.15K			
			DMF+2-BuOH			
0.0000	789.992	1161.43	0.000000	0.0000	0.0000	0.0000
0.0501	795.526	1170.01	0.045291	-1.5019	-5.3627	-4.2605
0.0998	801.469	1178.72	0.047758	-2.9578	-10.0492	-8.2085
0.1494	807.356	1187.62	0.066279	-4.3707	-14.1039	-11.8595
0.1986	813.213	1196.81	0.093225	-5.5797	-17.7261	-15.0631
0.2514	819.776	1206.62	0.103114	-7.1221	-20.5108	-18.4038
0.2994	825.751	1216.04	0.122113	-8.1997	-22.8104	-20.8974
0.3491	832.513	1225.18	0.091270	-10.112	-23.4640	-23.8609
0.3998	839.423	1235.42	0.071621	-11.342	-24.3717	-25.9780
0.4495	846.723	1246.27	0.010791	-11.934	-25.2984	-27.2198
0.5002	854.077	1257.18	-0.027503	-12.903	-25.0425	-28.4673
0.5498	862.573	1269.35	-0.180460	-12.562	-25.5214	-28.2404
0.5999	870.920	1281.79	-0.294255	-12.283	-25.0983	-27.7110
0.6524	878.920	1295.33	-0.324707	-11.725	-24.0432	-26.5248
0.7001	886.292	1308.84	-0.349062	-10.226	-23.2749	-24.2021
0.7496	893.641	1321.96	-0.333599	-9.7932	-20.5040	-22.4063
0.7999	900.874	1336.23	-0.284362	-8.6557	-17.5023	-19.5493
0.8502	907.973	1350.64	-0.211815	-7.6266	-13.5118	-16.3642
0.9000	914.536	1365.40	-0.088684	-6.3663	-8.8990	-12.5593
0.9501	922.000	1380.80	-0.029888	-4.8074	-3.6099	-8.0152
1.0000	929.906	1399.66	0.000000	0.0000	0.0000	0.0000

x_1	ρ (kgm^{-3})	u (ms^{-1})	V^E (m^{3}mol^{-1})	u^E (ms^{-1})	K_s^Ex10^{-12} (Pa^{-1})	Z^Ex10^{-3} (kgm^{-2}s^{-1})
			T=318.15K			
			DMF+2-BuOH			
0.0000	785.493	1144.45	0.000000	0.0000	0.0000	0.0000
0.0501	791.051	1153.18	0.041059	-1.1903	-6.0408	-3.9514
0.0998	797.007	1162.05	0.040246	-2.3238	-11.3693	-7.5830
0.1494	802.800	1170.06	0.068053	-4.4623	-14.3935	-11.7636
0.1986	808.747	1179.26	0.083327	-5.4960	-18.3960	-14.7807
0.2514	815.297	1189.19	0.093066	-6.7385	-21.6887	-17.8313
0.2994	821.258	1198.05	0.112293	-8.2103	-23.4871	-20.6159
0.3491	828.010	1207.39	0.080656	-9.7482	-24.6455	-23.2354
0.3998	834.906	1217.38	0.060756	-11.049	-25.4637	-25.3852
0.4495	842.169	1227.66	0.001664	-12.032	-25.9114	-26.9423
0.5002	849.532	1238.48	-0.039438	-12.905	-25.7594	-28.0988
0.5498	857.973	1250.18	-0.189891	-12.849	-25.9308	-28.1208
0.5999	866.297	1262.61	-0.304044	-12.393	-25.7211	-27.4465
0.6524	874.271	1276.14	-0.333749	-11.644	-24.8573	-26.1110
0.7001	881.621	1288.89	-0.357560	-10.719	-23.4775	-24.3220
0.7496	888.933	1302.11	-0.339798	-9.9928	-20.9345	-22.2959
0.7999	896.151	1316.15	-0.289991	-8.8846	-17.8208	-19.5059
0.8502	903.226	1329.94	-0.215809	-8.2718	-13.3164	-16.7448
0.9000	909.776	1344.78	-0.091491	-6.7268	-8.8314	-12.7387
0.9501	917.100	1360.39	-0.021133	-4.7488	-3.7415	-7.8754
1.0000	925.096	1378.98	0.000000	0.0000	0.0000	0.0000

x_1	ρ (kgm^{-3})	u (ms^{-1})	V^E (m^{3}mol^{-1})	u^E (ms^{-1})	K_s^Ex10^{-12} (Pa^{-1})	Z^Ex10^{-3} (kgm^{-2}s^{-1})
			T=298.15K			
			DMF+2-PnOH			
0.0000	805.395	1232.92	0.000000	0.0000	0.0000	0.0000
0.0506	809.787	1243.72	0.086953	2.6215	-6.9014	-5.2599
0.0997	814.631	1253.19	0.111081	3.9170	-12.0262	-10.3656
0.1515	820.338	1264.92	0.080191	6.7564	-19.2089	-13.4722
0.2000	825.973	1275.99	0.036050	9.2426	-25.3288	-15.8154
0.2497	832.099	1286.87	-0.029593	11.0537	-30.3950	-18.0205
0.2909	837.416	1296.11	-0.094141	12.5572	-34.2966	-19.2533
0.3510	845.299	1308.59	-0.171780	13.3787	-37.7918	-21.5534
0.3999	851.924	1318.91	-0.231484	13.8700	-39.9375	-22.8602
0.4494	858.917	1329.61	-0.298001	14.2909	-41.6106	-23.4451
0.5003	866.108	1339.99	-0.338600	13.7371	-41.6812	-24.4293
0.5491	873.099	1350.49	-0.361805	13.3899	-41.3566	-24.6236
0.6002	880.493	1361.89	-0.366601	13.0286	-40.3641	-24.2129
0.6500	887.881	1372.39	-0.363067	11.6412	-37.8895	-23.9535
0.7000	895.201	1383.21	-0.323630	10.1075	-34.5356	-23.3926
0.7499	902.527	1394.11	-0.260953	8.1884	-30.1502	-22.5966
0.7999	909.972	1405.16	-0.182933	5.9188	-24.8192	-21.3525
0.8500	917.745	1415.08	-0.107975	1.9834	-17.6316	-20.5395
0.9000	925.898	1426.03	-0.041979	-1.4556	-10.3073	-18.0540
0.9500	933.999	1437.92	0.055230	-4.5380	-2.3921	-14.5868
1.0000	944.270	1458.05	0.000000	0.0000	0.0000	0.0000

x_1	ρ (kgm^{-3})	u (ms^{-1})	V^E (m^{3}mol^{-1})	u^E (ms^{-1})	K_s^Ex10^{-12} (Pa^{-1})	Z^Ex10^{-3} (kgm^{-2}s^{-1})
			T=303.15K			
			DMF+2-PnOH			
0.0000	801.295	1214.93	0.000000	0.0000	0.0000	0.0000
0.0506	805.702	1225.76	0.082507	2.7103	-7.3910	-5.0418
0.0997	810.540	1236.45	0.104533	5.2844	-14.4381	-9.0018
0.1515	816.272	1247.80	0.066828	7.8077	-21.5428	-12.2251
0.2000	821.868	1257.94	0.024205	9.4255	-26.8252	-15.2368
0.2497	828.337	1268.91	-0.087389	11.3917	-32.6281	-16.7940
0.2909	833.318	1278.26	-0.113738	13.0606	-36.5962	-18.2516
0.3510	841.237	1290.49	-0.199946	13.7157	-40.0897	-20.5525
0.3999	847.995	1300.59	-0.278183	14.0575	-42.2800	-21.7460
0.4494	854.694	1310.71	-0.314954	13.9720	-43.2681	-23.0899
0.5003	861.886	1321.72	-0.359273	14.1265	-44.0772	-23.4091
0.5491	868.917	1331.99	-0.389957	13.6269	-43.6197	-23.6338
0.6002	876.136	1342.91	-0.379755	12.8697	-42.0887	-23.7615
0.6500	883.479	1353.81	-0.374990	11.9673	-39.9339	-23.1014
0.7000	890.808	1364.31	-0.339436	10.2018	-36.2394	-22.7020
0.7499	898.243	1374.43	-0.289828	7.5942	-31.1553	-22.3480
0.7999	905.546	1385.90	-0.200843	5.8398	-25.9569	-20.8190
0.8500	913.278	1395.95	-0.124727	2.1332	-18.6279	-19.8425
0.9000	921.494	1406.99	-0.066932	-1.1133	-11.1824	-17.0951
0.9500	929.400	1417.92	0.044794	-5.0490	-2.1743	-14.7081
1.0000	939.496	1438.45	0.000000	0.0000	0.0000	0.0000

x_1	ρ (kgm^{-3})	u (ms^{-1})	V^E (m^{3}mol^{-1})	u^E (ms^{-1})	K_s^Ex10^{-12} (Pa^{-1})	Z^Ex10^{-3} (kgm^{-2}s^{-1})
			T=308.15K			
			DMF+2-PnOH			
0.0000	797.109	1197.23	0.000000	0.0000	0.0000	0.0000
0.0506	801.536	1208.13	0.077592	2.8492	-7.9807	-4.7820
0.0997	806.370	1218.43	0.097506	5.1022	-14.9265	-8.8973
0.1515	812.122	1229.81	0.053811	7.7302	-22.5459	-11.9121
0.2000	817.685	1239.98	0.012255	9.4501	-28.1865	-14.7926
0.2497	824.126	1250.96	-0.099811	11.5023	-34.3471	-16.2411
0.2909	829.112	1259.43	-0.129392	12.3561	-37.5274	-18.3041
0.3510	836.942	1271.97	-0.209293	13.4187	-41.6171	-20.2996
0.3999	843.557	1281.90	-0.274735	13.6727	-43.7063	-21.6956
0.4494	850.438	1292.22	-0.335653	13.8729	-45.2291	-22.5081
0.5003	857.445	1302.69	-0.362927	13.5785	-45.4675	-23.4065
0.5491	864.399	1313.62	-0.388811	13.8293	-45.7278	-23.0491
0.6002	871.761	1324.51	-0.396805	13.1398	-44.3135	-22.8896
0.6500	878.964	1334.85	-0.380667	11.7760	-41.5249	-22.7931
0.7000	886.310	1345.68	-0.349628	10.4429	-38.1006	-21.9657
0.7499	893.744	1355.53	-0.302505	7.6712	-32.6514	-21.7394
0.7999	901.109	1366.02	-0.221465	5.0467	-26.5252	-20.8990
0.8500	908.803	1377.11	-0.143936	2.4943	-19.8571	-18.9282
0.9000	916.977	1387.69	-0.084678	-1.0938	-11.7865	-16.5658
0.9500	924.896	1398.52	0.024152	-5.0067	-2.4282	-14.1558
1.0000	934.707	1418.88	0.000000	0.0000	0.0000	0.0000

x_1	ρ (kgm^{-3})	u (ms^{-1})	V^E (m^{3}mol^{-1})	u^E (ms^{-1})	K_s^Ex10^{-12} (Pa^{-1})	Z^Ex10^{-3} (kgm^{-2}s^{-1})
			T=313.15K			
			DMF+2-PnOH			
0.0000	792.841	1179.60	0.000000	0.0000	0.0000	0.0000
0.0506	797.301	1190.40	0.071086	2.8087	-8.3689	-4.6638
0.0997	802.145	1200.00	0.087278	4.4209	-14.7562	-9.1830
0.1515	807.889	1210.89	0.041528	6.6232	-22.1885	-12.4676
0.2000	813.304	1221.96	0.016129	9.3051	-29.3060	-14.6737
0.2497	819.826	1232.94	-0.110194	11.4227	-35.9432	-15.9083
0.2909	824.859	1242.04	-0.148027	12.9621	-40.1992	-17.2890
0.3510	832.691	1253.97	-0.231902	13.4983	-43.8218	-19.6430
0.3999	839.281	1263.67	-0.297398	13.5925	-45.8007	-21.1512
0.4494	845.990	1273.99	-0.341722	13.8660	-47.3398	-22.0753
0.5003	853.098	1284.59	-0.383305	13.7794	-47.9155	-22.6225
0.5491	859.934	1295.21	-0.399088	13.7970	-47.8461	-22.5848
0.6002	867.282	1306.19	-0.408455	13.2806	-46.5483	-22.2636
0.6500	874.535	1316.59	-0.399939	12.0606	-43.8210	-21.9482
0.7000	881.790	1327.01	-0.361977	10.4042	-39.8625	-21.5057
0.7499	889.234	1337.29	-0.317994	8.1523	-34.6740	-20.7873
0.7999	896.660	1347.92	-0.244472	5.7606	-28.5234	-19.6409
0.8500	904.314	1357.53	-0.164827	1.8245	-20.2973	-18.9713
0.9000	912.550	1368.39	-0.112906	-1.3841	-12.2571	-16.1962
0.9500	920.623	1379.91	-0.018580	-4.5037	-3.2498	-12.8595
1.0000	929.906	1399.66	0.000000	0.0000	0.0000	0.0000

x_1	ρ (kgm^{-3})	u (ms^{-1})	V^E (m^{3}mol^{-1})	u^E (ms^{-1})	K_s^Ex10^{-12} (Pa^{-1})	Z^Ex10^{-3} (kgm^{-2}s^{-1})
			T=318.15K			
			DMF+2-PnOH			
0.0000	788.491	1162.00	0.000000	0.0000	0.0000	0.0000
0.0506	792.992	1172.42	0.063641	2.5429	-8.3967	-4.6957
0.0997	797.854	1182.75	0.075266	4.9992	-16.3891	-8.4031
0.1515	803.584	1193.40	0.028572	7.0853	-23.9488	-11.6879
0.2000	809.325	1203.95	-0.041647	9.3665	-31.1538	-13.7322
0.2497	815.449	1214.93	-0.119938	11.6099	-37.8479	-15.2708
0.2909	820.427	1223.41	-0.153244	12.6365	-41.5590	-17.0755
0.3510	828.400	1235.61	-0.257961	13.6040	-46.0487	-18.8185
0.3999	834.982	1245.27	-0.325098	13.7938	-48.2203	-20.1975
0.4494	841.643	1255.77	-0.366254	14.3889	-50.1649	-20.8589
0.5003	848.706	1266.24	-0.405250	14.3226	-50.7338	-21.4001
0.5491	855.509	1276.79	-0.419593	14.4190	-50.7105	-21.3020
0.6002	862.702	1287.61	-0.414704	13.9036	-49.2169	-21.1521
0.6500	869.909	1297.73	-0.403766	12.5659	-46.2000	-20.9808
0.7000	877.203	1307.79	-0.371836	10.7178	-41.8991	-20.6457
0.7499	884.709	1317.31	-0.335802	7.8802	-35.9527	-20.3657
0.7999	892.198	1327.81	-0.269640	5.5391	-29.6498	-19.1054
0.8500	899.814	1337.92	-0.187512	2.2902	-21.6909	-17.8904
0.9000	908.012	1348.71	-0.133515	-0.7943	-13.3897	-15.0978
0.9500	916.149	1359.95	-0.045789	-3.9927	-3.9702	-11.7989
1.0000	925.096	1378.98	0.000000	0.00000	0.00000	0.00000

Table 3. Fitting coefficients a_i of polynomial Equation (4) and the value of r^2 for the systems DMF (x_1) + 2-BuOH (x_2) and + 2-PnOH (x_2) at different temperatures

T/K	a_0	a_1	a_2	a_3	a_4	r^2
			DMF (x_1) + 2-BuOH (x_2)			
298.15	803.2	100.1	74.79	-19.7	-14.99	0.999
303.15	799.0	101.3	61.15	4.927	-27.93	0.999
308.15	794.7	101.6	58.75	7.474	-28.81	0.999
313.15	790.3	102.4	53.73	14.14	31.74	0.999
318.15	785.8	102.5	53.66	13.02	-30.97	0.999
			DMF (x_1) +2-PnOH (x_2)			
298.15	805.5	75.89	163.5	-190.1	88.90	1.000
303.15	801.3	78.27	153.9	-177.7	83.21	1.000
308.15	797.1	80.07	143.0	-160.6	74.65	1.000
313.15	792.8	81.44	135.3	-148.5	68.68	1.000
318.15	788.5	84.12	123.0	-131.1	60.51	1.000

The excess values were fitted by the Redlich-Kister type [59] polynomial equation,

$$Y^E = x_1 x_2 \sum_{i=0}^{n} A_i (2x_1 - 1)^I \qquad (5)$$

Where, A_i is the i^{th} fitting coefficient of the Redlich-Kister polynomial equation and all the other terms has their usual significance.

The relevant standard deviations, σ, were calculated by using the relation.

$$\sigma = \left[\sum \frac{\left(Y^E_{\exp} - Y^E_{cal}\right)^2}{n-p-2} \right]^{1/2} \qquad (6)$$

Where, n and p are the number of experimental points and number of parameters retained respectively.

All the coefficients, A_i, of Equation (5) and their relevant σ values by using Equation (6) are listed in Table 4 and Table 5.

Table 4. Fitting coefficients, A_i, of Redlich-Kister polynomial Equation (5) and the values of standard deviation, σ (V^E) for the systems DMF (x_1) + 2-BuOH (x_2), + 2-PnOH (x_2) at different temperatures

T/K	A_0	A_1	A_2	A_3	A_4	σ
			DMF (x_1) + 2-BuOH (x_2)			
298.15	-0.478	3.240	0.096	-3.297	0.000	0.019
303.15	-0.381	3.381	-0.156	-3.536	0.000	0.033
308.15	-0.396	3.404	-0.201	-3.667	0.000	0.032
313.15	-0.408	3.403	-0.273	-3.744	0.000	0.032
318.15	-0.453	3.406	-0.249	-3.799	0.000	0.032
			DMF (x_1) +2-PnOH (x_2)			
298.15	-1.393	1.455	2.696	-0.868	0.000	0.016
303.15	-1.491	1.290	2.591	-0.465	0.000	0.020
308.15	-1.514	1.257	2.382	-0.286	0.000	0.018
313.15	-1.565	1.179	2.193	0.121	0.000	0.014
318.15	-1.626	1.048	1.967	0.445	0.000	0.009

Table 5. Fitting coefficients, A_i, of Redlich-Kister polynomial Equation (5) and the values of standard deviation, σ (κ_S^E) for the systems DMF (x_1) + 2-BuOH (x_2), + 2-PnOH (x_2) at different temperatures

T/K	A_0	A_1	A_2	A_3	A_4	σ
DMF (x_1) + 2-BuOH (x_2)						
298.15	-93.5153	-11.4287	-34.0103	42.2295	0.0000	0.1603
303.15	-96.1862	-8.1695	-43.8001	35.1468	0.0000	0.1869
308.15	-98.6827	-9.1456	-41.2808	26.8524	0.0000	0.1609
313.15	-101.318	-5.1141	-53.2063	9.3358	0.0000	0.2519
318.15	-104.452	-0.7432	-51.069	-3.3151	0.0000	0.3156
DMF (x_1) +2-PnOH (x_2)						
298.15	-166.954	4.467	-10.6169	-47.4952	0.0000	0.3838
303.15	-175.488	6.2921	-13.3810	-38.6877	0.0000	0.4852
308.15	-182.787	0.7014	-12.9916	-11.3229	0.0000	0.5459
313.15	-192.133	3.8177	-16.4858	-32.7977	0.0000	0.5214
318.15	-203.334	-2.0622	-0.41980	11.8165	0.0000	0.4570

The densities of the systems DMF + 2-BuOH and DMF + 2-PnOH in the whole range of composition at 5 different temperatures (between 298.15 and 318.15 K) are displayed as in Table 2. The values of V^E, *(calc.)* were obtained from Equation (5) by using the best-fit values of Ai coefficients. The variations of V^E with mole fraction x_1, of DMF at various temperatures, along with the smoothed V^E values by using Equation (5), are presented graphically in Fig. 2 and Fig. 3.

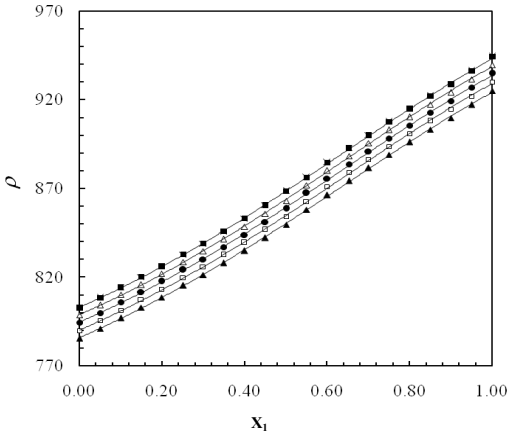

Figure 2. Density (ρ) of DMF(x_1) + 2-BuOH(x_2) system for different molar ratios at different temperatures

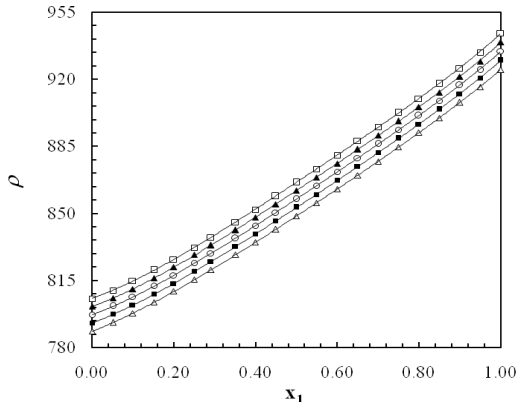

Figure 3. Density (ρ) of DMF(x_1) + 2-PnOH(x_2) system for different molar ratios at different temperatures

The calculated values of V^E are presented in Table 2 and Figure 4 and Figure 5 also.

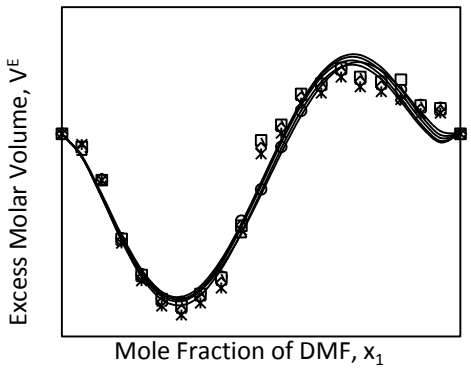

Figure 4. Excess Molar Volume (V^E) of DMF (x_1) + 2-BuOH (x_2) system for different molar ratios at different temperatures

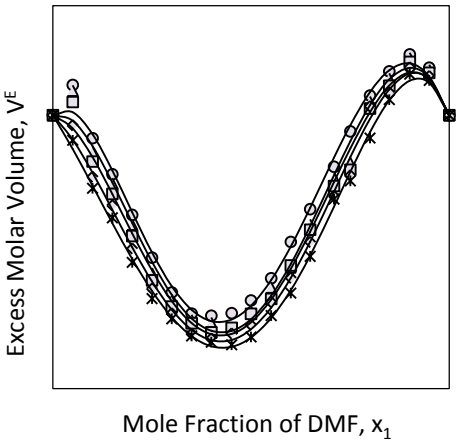

Figure 5. Excess Molar Volume (V^E) of DMF (x_1) + 2-PnOH (x_2) system for different molar ratios at different temperatures

For both systems excess molar volume, V^E, versus mole fraction, x_1, curves were found as sigmoid at all temperatures. In the system of 2-BuOH + DMF, the V^E values initially increases forming a small maximum in the alcohol-rich region nearly at $x_1 = 0.05$ - 0.30 and V^E then decreases and made a deep negative lobe with minimum nearly at $x_1 = 0.70$ at all temperatures. Likewise, V^E versus x_1 curves showed maxima at $x_2 = 0.05$ - 0.10 and minima nearly at $x = 0.55$ for the system 2-PnOH + DMF.

The order of positive excess molar volumes, V^E, follows as 2-BuOH + DMF > 2-PnOH + DMF. For both systems, as T rises, the magnitude of the positive V^E increases but that of negative V^E decreases, that means, dV^E/dT is positive. The sign of V^E of solutions depends upon the relative magnitude of expansion and contraction on mixing up of the components. If the factors causing expansion outweigh the factors causing contraction, the values of V^E becomes positive. But when the contractive factors are dominant over the expansive factors, the overall V^E becomes negative.

3.2. Partial Molar Volumes

In addition to other volumetric properties, partial molar

volumes ($\overline{V}_1$ and $\overline{V}_2$) and excess partial molar volumes ($\overline{V}_1^E$ and $\overline{V}_2^E$), of DMF, alkanols over the entire composition range were determined using Equation (7) and Equation (8)

$$\overline{V}_1 = V^E + V_1^* + x_2 (\delta V^E/x_1)_{T,P} \quad (7)$$

$$\overline{V}_2 = V^E + V_2^* - x_1 (\delta V^E/x_1)_{T,P} \quad (8)$$

Here, V_1^* and V_2^* are the molar volumes of pure components 1 and 2. Following the procedures of Maham *et al.* [60] the partial derivatives $(\partial V^E/\partial x_1)_{T,P}$ were obtained.

Accordingly, by differentiating Equation (5) for V^E with respect to x_1 and x_2 subsequently, substituting of its value in Equation (7) and Equation (8) lead to the following Equation (9) and Equation (10):

$$\overline{V}_1 = V^E + V_1^* + x_2^2 \sum_{i=0}^{n} A_i (1-2x_1)^i - 2x_1 x_2^2 \sum_{i=0}^{n} i\, A_i (1-2x_1)^{i-1} \quad (9)$$

$$\overline{V}_2 = V^E + V_2^* + x_1^2 \sum_{i=0}^{n} A_i (1-2x_1)^i + 2x_1^2 x_2 \sum_{i=1}^{n} i\, A_i (1-2x_1)^{i-1} \quad (10)$$

Again, partial molar volumes at infinite dilution V^{1o} and V^{2o} for component 1 and 2 were obtained from Equation (4) and Equation (5) at the limit of $x_2 \to 0$ and $x_1 \to 0$, respectively. These were further used to calculate excess partial molar volumes at infinite dilution V_1^{oE} and V_2^{oE} by using the following relations

$$\overline{V}_1^{oE} = \overline{V}_1^{o} - V_1^* \quad (11)$$

$$\overline{V}_2^{oE} = \overline{V}_2^{o} - V_2^* \quad (12)$$

It is concluded from the Fig. 6 and Fig. 7 that, the partial molar volumes, $\overline{V}_1$ of 2-PnOH was slightly risen in the DMF-poor region, but rapidly decreased in the DMF-rich region at lower temperatures. Considering, the significances of $\overline{V}_i$, the falling $\overline{V}_1$ of 2-PnOH and also to form the minimum at $x_2 \approx 0.85$ reveals that in the overall volume expansion, the alkanol contributes slightly but in contraction 2-PnOH contributes quite significantly making the corresponding V^E values negative in the DMF-rich region.

This is just in accordance to the result that, in the stated region above ($x_2 = 0.30$) the value of $(V_1 - V_1)$ falls to be negative with minimum near $x_2 = 0.80$ for the system of 2-PnOH + DMF. Likewise, the partial molar volume of 2-BuOH ($\overline{V}_1$) though apparently changes very small in the whole range of concentration, ($\overline{V}_1 - V_1$) values are all quite large, being positive in the region $0 < x_2 < 0.65$ and sharp negative above $x_2 > 0.65$. This clearly signifies that, contribution of 2-BuOH towards V^E is positive in the former region, while it is negative above $x_2 > 0.65$. However, the magnitudes of V^E, whether positive or negative are greater for 2-BuOH + DMF than that for 2-PnOH + DMF.

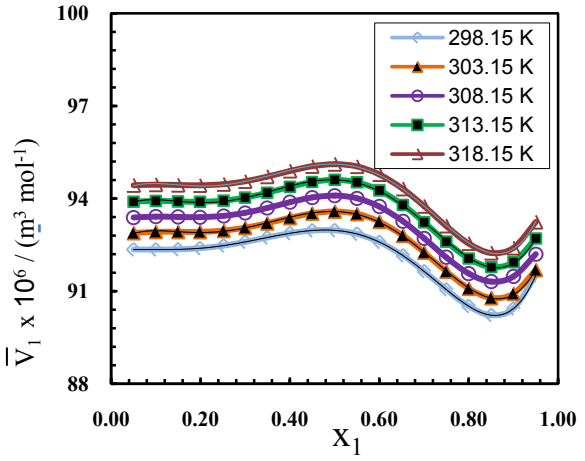

Figure 6. Partial molar volume ($\overline{V}_1$) of 2-BuOH in 2-BuOH(x2) + DMF(x1) system for different molar ratios at different temperatures

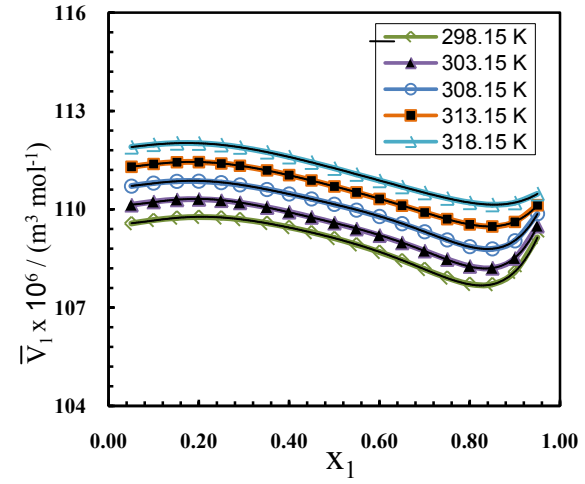

Figure 7. Partial molar volume ($\overline{V}_1$) of 2-PnOH in 2-PnOH (x_2) + DMF (x_1) system for different molar ratios at different temperatures

3.3. Excess Isentropic Compressibility

The isentropic compressibility, K_S, was calculated from density, ρ, and ultrasonic velocity, u, assuming that ultrasonic absorption is negligible, using the Newton-Laplace equation, $K_S = 1/\rho u^2$.

Then, the excess isentropic compressibility, K_S^E, ($\pm 10^{-1}$ TPa^{-1}) can be determined according to the following equation:

$$K_S^E = K_S - (\phi_1 K_{S1} + \phi_2 K_{S2}) \quad (13)$$

where, ϕ_1 and ϕ_2, are the volume fraction of component 1 and 2.

The excess isentropic compressibility, K_S^E, for both the binary systems is plotted as a function of the mole fraction in Fig. 8 and Fig. 9. The K_S^E values are negative at all temperatures, and they are more negative for the mixture containing 2-pentanol mixture. The K_S^E minimum value occurs at $x_1 \approx 0.55$ for DMF + 2-butanol system and at $x_1 \approx 0.50$ for DMF + 2-pentanol system. On the other hand, when temperature increases the K_S^E values decrease, in other words, they are more negative.

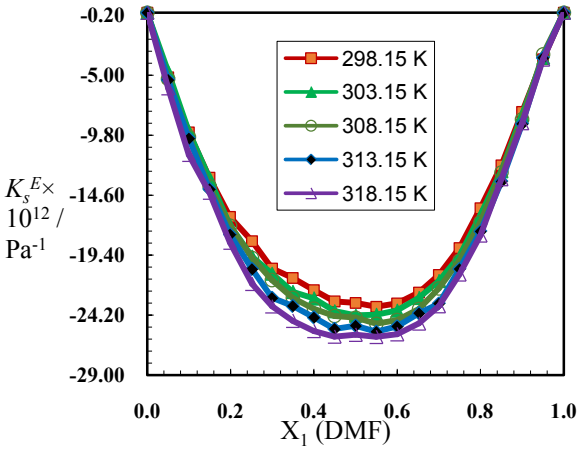

Figure 8. Excess isentropic compressibilities (K_s^E), for *N,N*-dimethylformamide (X_2) + 2-BuOH (X_1) system at different molar ratios at different temperatures

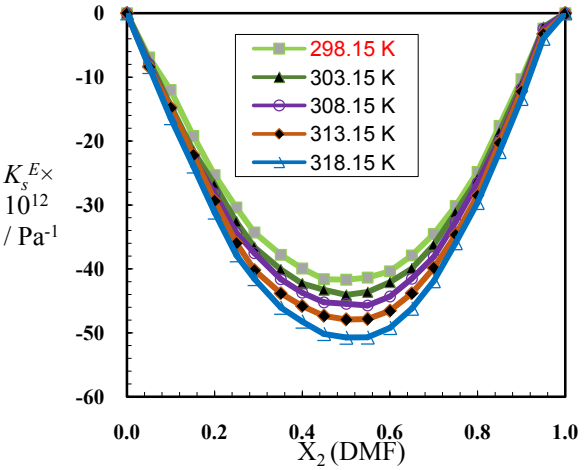

Figure 9. Excess isentropic compressibilities, (K_s^E), for *N,N*-dimethylformamide (X_2) + 2-PnOH (X_1) system at different molar ratios at different temperatures

However, values of K_S showed an inverse behavior as compared to ultrasonic velocity u; K_S was decreased with increasing concentration of DMF. It is primarily the compressibility that decreases due to structural changes of molecules in the mixture leading to an increase in ultrasonic velocity. Again, the negative K_S^E and V^E values were found to follow the order: 2-PnOH + DMF > 2-BuOH + DMF, i.e. K_S^E and V^E values became more negative as the chain length of alcohol molecules increases. This may be explained by considering the electron-repelling tendency (+ *I* effect) of -CH_3 group(s). The presence of three methyl groups at the α- and γ-carbons in 2-PnOH and at the α- and β-positions in 2-BuOH increase the electron density at the O of -OH group of alcohol molecules that overall sequence follows: 2-BuOH < 2-PnOH. So that, the proton-accepting ability of oxygen atom of -OH group also should be in the same sequence. As a result, the extent of negative deviations in K_S^E and V^E clearly indicates that the strength of interaction between DMF and alcohol molecules should follow the order: 2-PnOH + DMF > 2-BuOH + DMF.

4. Conclusions

The density, ultrasonic velocity and some excess volumetric and acoustic properties of binary mixtures of *N,N* dimethylformamide with 2-butanol and 2-pentanol have been measured over the entire range of composition at T= (298.15, 303.15, 308.15, 313.15 and 318.15) K and at atmospheric pressure. The Redlich-Kister polynomial equation was used to correlate the results. From the experimental data, the positive V^E suggests that, DMF-alkanol interaction (structure making effect) is weaker than that of DMF-DMF and alkanol-alkanol interactions (structure breaking effect) and vice-versa for the negative V^E. Dissociation of H-bonded structures of 2-alkanols results into positive V^E, whereas, dissociation/formation of weak H-bonds between DMF and 2-alkanols leads to negative V^E. In applications, this study would be helpful in understanding the physical properties as well as chemical properties of mixtures between DMF and other solvents.

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