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Density, Dynamic Viscosity and Derived Properties of Binary Mixtures of Methanol, Ethanol, *n*-Propanol, and *n*-Butanol with Pyridine at *T* = (293.15, 303.15, 313.15 and 323.15) K

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Densities, viscosities of binary liquid mixtures composed of pyridine and some primary alcohols namely methanol, ethanol, n-propanol and n-butanol were determined at 293.15, 303.15, 313.15 and 323.15 K. From the experimental results obtained, deviation in viscosity ($\Delta\eta$), excess molar volume (V^E), excess Gibbs free energy of activation of viscous flow (ΔG^{*E}), were determined. The deviations in viscosity, excess molar volume and excess Gibbs free energy of activation of viscous flow were correlated with Redlich-Kister polynomial equation. Other parameters like Grunberg-Nissan interaction constant (d') and a modified Kendall-Monroe equation ($E\eta_m$), were used to quantitatively analyze the interactions in the system.

Keywords: Density, Viscosity, Binary mixtures, Pyridine, Alcohols.

1. INTRODUCTION

The mixing of different solvents results in the formation of a solution that is different from ideal [1]. The thermodynamic properties of multicomponent liquid mixtures and their analysis in terms of interpretative models constitute a very interesting subject [2]. The practical need for thermodynamic data for teaching and research as well as for design and set up of industrial processes continue to drive research in the study of multicomponent systems. The characterization of mixtures through their thermodynamic and transport properties is important from the fundamental viewpoint of understand their mixing behavior [3-8]. A thorough knowledge of transport properties of non-aqueous solutions is essential in many chemical and industrial applications [9].

The studies of excess properties such as deviation in viscosity, excess molar volume, excess Gibbs free energy of activation of viscous flow and Grunberg-Nissan interaction constant of binary mixtures are useful in understanding the nature of intermolecular interactions between two liquids [8-12]. Properties such as density and viscosity at several temperatures both for pure chemicals and their binary liquid mixtures over the whole composition range are useful for understanding of the thermodynamic and transport properties associated with heat and fluid flow [6,13]. Binary liquid mixtures due to their unusual behavior have attracted considerable attention due to their importance from both theoretical and practical point of view because these mixtures are used in titration, calorimetry and reaction calorimetry, among other uses [10,14].

Alcohols serve as simple examples of biological and industrially important amphiphilic materials that exist in the liquid state which may be due to hydrogen bonding of their O–H group. They are polar and self-associated liquids. The dipolar association of alcohols decreases when they are mixed with aromatic hydrocarbons due to some specific intermolecular interactions between the alcohol and an aromatic hydrocarbon [13,15]. Primary alcohols have both a proton donor and a proton acceptor group. It is expected that there will be a significant degree of H-bonding leading to self-association in the pure state in addition to mutual association in their binaries [11,16].

In this study, experimental viscosity and density are reported at four temperatures 293.15, 303.15, 313.15 and 323.15 K for binary mixtures of pyridine and some alcohols namely methanol, ethanol, n-propanol and n-butanol. Deviation in viscosity ($\Delta\eta$), excess molar volume (V^E) and excess Gibbs free energy of activation of viscous flow (ΔG^{*E}) have been calculated from the density (ρ), and viscosity (η), data. Modified Kendall-Monroe equation with no parameters has been used in correlating viscosity data of the binary mixtures. Calculated deviation in viscosity and excess functions were fitted to the Redlich-Kister polynomial equation and the results analyzed in terms of molecular interactions.

2. EXPERIMENTAL

2.1. Materials

Reagent grade methanol, ethanol, propanol, butanol and pyridine were purchased from Sigma-Aldrich, South Africa and used without further purification.

2.2 Mixture preparation

Binary mixtures were prepared by weighing appropriate amounts of pyridine and alcohol on an electronic balance. An AE Adam balance (Adam Equipment Inc. USA) model PW124 with a maximum capacity of 120 g, a readability range 0.0001 g and repeatability (S.D.) of 0.00015 g, linearity 0.0002 g, operating temperature +10^{o}C to 40^{o}C was used in all measurements.

2.3 Density measurement

Density measurement of binary mixtures was carried out with an Anton Paar DMA-4500 M digital densitometer thermostatted at different temperatures. Two integrated Pt 100 platinum thermometers were provided for good precision in temperature control internally (T ± 0.01 K). The densimeter protocol includes an automatic correction for the viscosity of the sample. The apparatus is precise to within $1.0x10^{-5}$ g/cm^3, and the uncertainty of the measurements was estimated to be better than ± $1.0x10^{-4}$ g/cm^3. Calibration of the densimeter was performed at atmospheric pressure using doubly distilled and degassed water.

2.4 Viscosity measurement

Viscosity measurements were carried out using Anton Paar SVM 3000 Stabinger Viscometer. The viscometer has a dynamic viscosity range of 0.2 to 20 000 mPa.s, a kinematic viscosity range of 0.2 to 20 000 mm^2/s and a density range of 0.65 to 3 g/cm^3. The instrument is equipped with a maximum temperature range of +105^{o}C and a minimum of 20^{o}C below ambient. Instrument viscosity reproducibility is 0.35% of measured value and density reproducibility of 0.0005 g/cm^3.

3. RESULTS AND DISCUSSION

A comparison of experimentally determined values of density (ρ), and viscosity (η) measured for all pure liquids at 293.15, 303.15, 313.15 and 323.15 K, with literature values are presented in table 1.

Table 1. Comparison of experimental densities (ρ) and viscosities (η) with literature values

Component		T = 293.15		T = 303.15		T = 313.15		T = 323.15	
		ρ (g/cm^3)	η (mPa.s)	ρ (g/cm^3)	η (mPa.s)	ρ (g/cm^3)	η (mPa.s)	ρ (g/cm^3)	η (mPa.s)
Methanol	Experiment	0.7939	0.5990	0.7844	0.5163	0.7748	0.4324	0.7701	0.4055
	Literature	0.7910[17] 0.79112[20]	0.5945[17] 0.5970[18]	0.78180[19,20]	0.510[18]	0.77232[20]	0.456[18]	0.76511[20]	0.403[21]
Ethanol	Experiment	0.8005	0.7893	0.8000	0.7807	0.7916	0.7720	0.7739	0.7630
	Literature	0.78945[18] 0.79008[21]	1.144[18] 1.21[21]	0.7818[19] 0.78100[21]	0.949[18] 1.00[21] 0.995[21]	0.7734[19] 0.77231[21]	0.794[5] 0.83[21] 0.8290[21]	0.76324[21] 0.77220[21]	0.670[18] 0.69[21] 0.6868[21]
n-Propanol	Experiment	0.8045	2.2506	0.7967	1.7153	0.7820	1.4339	0.7730	1.0983
	Literature	0.8041[17] 0.80376[21]	2.256 2.19[21] 2.238[21]	0.79574[21]	1.72 1.73[21] 1.145[21]	0.78746[21]	1.405 1.37[21] 1.381[21]	0.77902[21] 0.77391[21]	1.130 1.10[21] 1.115[21]
n-Butanol	Experiment	0.8115	2.9623	0.8037	2.2662	0.7958	1.7680	0.7876	1.3631
	Literature	0.81 0.8101[17] 0.80979[21]	2.948[18] 2.93[21] 2.963[21]	0.8022[6,8] 0.80209[21] 0.80195[21]	2.2243[9] 2.271[21] 2.26[21]	0.79396[10] 0.79437[21]	1.7734[10] 1.77[21] 1.783[21]	0.78643[21]	1.411[18] 1.41[21] 1.421[21]
Pyridine	Experiment	0.9880	1.4927	0.9780	1.2054	0.9680	1.0098	0.9537	0.7719
	Literature	0.9819[18]		0.9737					

Table 2. Experimental values of density ρ(g/cm^3), viscosity η(mPa.s), deviation in viscosity $\Delta\eta$(mPa.s), excess molar volumes V^E(cm^3/mol), molar volume of mixture V_m(cm^3/mol), excess Gibbs free energy of activation of viscous flow ΔG^{*E}(J/mol), Grunberg-Nissan parameter (d') and modified Kendall and Monroe viscosity correlation $E\eta_m$ (mPa.s) with pyridine (x_1) at 293.15, 303.15, 313.15 and 323.15 K.

(a) Pyridine (1) + Methanol (2) 293.15 K

x_1	ρ(g/cm^3)	η(mPa.s)	$\Delta\eta$(mPa.s)	V^E(cm^3/mol)	V_m(cm^3/mol)	ΔG^{*E}(J/mol)	d'	$E\eta_m$(mPa.s)
1.0000	0.9880	1.4927	0.0000	0.0000	80.0607	0.0000	0.0000	0.0000
0.9003	0.9631	1.0695	-0.3429	1.1566	77.2590	246.3145	4.0691	0.0597
0.8008	0.9529	1.0875	-0.2448	1.0195	73.1754	934.4682	1.9090	0.1174
0.7003	0.9260	0.9154	-0.3359	2.0285	70.1902	1042.7914	0.2595	0.1704
0.6006	0.9182	0.8420	-0.3290	1.4732	65.6766	1183.5755	-0.4439	0.2141
0.4760	0.9038	0.8919	-0.1786	0.9788	60.2352	1603.7511	-0.5838	0.2495
0.4013	0.8712	0.8530	-0.1574	2.1635	58.4541	1642.7594	-1.0329	0.2568
0.2997	0.8537	0.7364	-0.1921	1.7949	54.0516	1302.0910	-2.2584	0.2450
0.2032	0.8283	0.6161	-0.2347	1.8011	50.2265	774.3171	-4.4917	0.2050
0.1028	0.8180	0.6617	-0.1081	0.6436	45.0828	619.5616	-7.9555	0.1268
0.0000	0.7939	0.6870	0.0000	0.0000	40.3577	0.0000	0.0000	0.0000
	303.15 K							
1.0000	0.9780	1.2054	0.0000	0.0000	80.8793	0.0000	0.0000	0.0000
0.9003	0.9532	0.9189	-0.2225	1.1733	78.0614	407.1797	4.6069	0.0510
0.8008	0.9429	0.8804	-0.1972	1.0426	73.9515	965.0874	1.8483	0.0996
0.7003	0.9163	0.7584	-0.2546	2.0517	70.9332	1131.0321	0.3307	0.1434
0.6006	0.9084	0.7169	-0.2320	1.4949	66.3851	1341.7254	-0.2615	0.1789
0.4760	0.8822	0.7234	-0.1456	1.8079	61.7100	1682.0571	-0.5953	0.2069
0.4013	0.8614	0.7170	-0.1040	2.2074	59.1191	1776.9694	-0.8915	0.2120
0.2997	0.8441	0.6291	-0.1266	1.8220	54.6664	1461.2595	-2.0120	0.2010
0.2032	0.8188	0.5222	-0.1716	1.8280	50.8092	891.6245	-4.2118	0.1673
0.1028	0.8084	0.5506	-0.0787	0.6563	45.6182	681.6361	-7.6476	0.1029
0.0000	0.7844	0.5633	0.0000	0.0000	40.8465	0.0000	0.0000	0.0000
	313.15 K							
1.0000	0.9680	1.0098	0.0000	0.0000	81.7149	0.0000	0.0000	0.0000
0.9003	0.9432	0.7937	-0.1585	1.1983	78.8890	523.1829	5.8244	0.0436
0.8008	0.9329	0.7338	-0.1610	1.0653	74.7442	1029.0982	2.2550	0.0848
0.7003	0.9064	0.6492	-0.1876	2.0897	71.7080	1292.6979	0.7252	0.1219
0.6006	0.8985	0.6055	-0.1737	1.5224	67.1166	1497.7828	-0.0085	0.1517
0.4760	0.8723	0.6077	-0.0995	1.8453	62.4104	1864.1643	-0.4174	0.1749
0.4013	0.8513	0.5811	-0.0830	2.2705	59.8205	1886.7004	-0.8833	0.1789
0.2997	0.8344	0.5059	-0.0995	1.8527	55.3019	1562.2187	-2.0820	0.1693
0.2032	0.8093	0.4292	-0.1205	1.8514	51.4056	1052.4287	-4.2199	0.1407
0.1028	0.7988	0.4380	-0.0537	0.6646	46.1665	773.5706	-8.1110	0.0864

0.0000	0.7748	0.4324	0.0000	0.0000	41.3526	0.0000	0.0000	0.0000
	323.15 K							
1.0000	0.9581	0.7719	0.0000	0.0000	82.5592	0.0000	0.0000	0.0000
0.9003	0.9332	0.6512	-0.0991	1.3811	79.7344	601.3039	1.4235	0.0391
0.8008	0.9228	0.5974	-0.1312	1.4025	75.5623	974.2366	0.0539	0.0744
0.7003	0.8963	0.5360	-0.1708	2.6000	72.5160	1154.3551	-0.6340	0.1048
0.6006	0.8885	0.5176	-0.1675	2.1619	67.8720	1330.0805	-0.8378	0.1279
0.4760	0.8624	0.5007	-0.1573	2.6732	63.1268	1448.0616	-1.1041	0.1442
0.4013	0.8414	0.4342	-0.2076	3.2220	60.5243	1112.2408	-1.8422	0.1456
0.2997	0.8241	0.3662	-0.2534	2.9769	55.9931	562.4994	-3.0805	0.1355
0.2032	0.7994	1.9850	1.3863	3.0970	52.0423	4893.5986	6.2488	0.1108
0.1028	0.7890	0.3115	-0.2654	2.0302	46.7399	-555.3334	-9.4701	0.0670
0.0000	0.7936	0.5545	0.0000	0.0000	40.3730	0.0000	0.0000	0.0000

(b) Pyridine (1) + Ethanol (2) 293.15 K

x_1	ρ	η	$\Delta\eta$	V^E	V_m	ΔG^{*E}	d'	$E\eta_m$(mPa.s)
1.0000	0.9839	0.9847	0.0000	0.0000	80.3943	0.0000	0.0000	0.0000
0.9003	0.9650	0.9624	-0.0028	0.4941	78.5564	815.8017	1.9634	0.0725
0.8008	0.9459	0.9847	0.0388	0.9325	76.6730	1374.9294	1.1098	0.1318
0.7003	0.9273	0.9100	-0.0161	1.2422	74.6262	1527.1906	0.3621	0.1773
0.6006	0.9159	0.8970	-0.0097	0.9077	71.9596	1692.2294	0.1649	0.2072
0.4760	0.8910	0.8839	0.0016	1.2142	69.3516	1784.8488	-0.0109	0.2215
0.4013	0.8772	0.8650	-0.0027	1.2398	67.6299	1728.7783	-0.1700	0.2169
0.2997	0.8579	0.8524	0.0045	1.2261	65.2396	1591.8659	-0.3716	0.1937
0.2032	0.8406	0.8322	0.0032	1.0342	62.7905	1315.1779	-0.7616	0.1526
0.1028	0.8333	0.8115	0.0021	-0.0469	59.3610	832.0138	-1.8509	0.0888
0.0000	0.8082	0.7893	0.0000	0.0000	57.0032	0.0000	0.0000	0.0000
	303.15 K							
1.0000	0.9739	0.9747	0.0000	0.0000	81.2198	0.0000	0.0000	0.0000
0.9003	0.9550	0.9526	-0.0028	0.5153	79.3790	844.2599	1.9706	0.0717
0.8008	0.9362	0.9747	0.0386	0.9494	77.4675	1422.4482	1.1136	0.1304
0.7003	0.9178	0.9005	-0.0161	1.2615	75.3987	1579.3481	0.3633	0.1754
0.6006	0.9065	0.8874	-0.0098	0.9247	72.7058	1749.5865	0.1645	0.2050
0.4760	0.8820	0.8744	0.0014	1.2228	70.0593	1845.0773	-0.0118	0.2192
0.4013	0.8682	0.8557	-0.0029	1.2598	68.3310	1787.6337	-0.1713	0.2146
0.2997	0.8491	0.8432	0.0044	1.2457	65.9158	1646.1352	-0.3736	0.1917
0.2032	0.8321	0.8232	0.0031	1.0423	63.4319	1359.7686	-0.7648	0.1511
0.1028	0.8247	0.8027	0.0021	-0.0369	59.9800	860.7729	-1.8576	0.0879
0.0000	0.8000	0.7807	0.0000	0.0000	57.5875	0.0000	0.0000	0.0000
		313.15 K						
1.0000	0.8204	0.9645	0.0000	0.0000	96.4164	0.0000	0.0000	0.0000
0.9003	0.9451	0.9426	-0.0027	-12.3956	80.2105	494.5641	1.9771	0.0709

0.8008	0.9265	0.9645	0.0383	-10.5307	78.2785	1133.7723	1.1170	0.1290
0.7003	0.9082	0.8908	-0.0160	-8.7668	76.1957	1337.5954	0.3641	0.1735
0.6006	0.8971	0.8778	-0.0098	-7.6846	73.4676	1555.0493	0.1647	0.2028
0.4760	0.8728	0.8649	0.0013	-5.5925	70.7978	1706.1674	-0.0121	0.2168
0.4013	0.8591	0.8468	-0.0025	-4.4806	69.0548	1679.6202	-0.1698	0.2123
0.2997	0.8402	0.8339	0.0042	-3.0384	66.6140	1574.5593	-0.3753	0.1897
0.2032	0.8233	0.8142	0.0031	-1.8545	64.1099	1319.8600	-0.7669	0.1495
0.1028	0.8160	0.7937	0.0019	-1.5079	60.6195	845.8532	-1.8651	0.0870
0.0000	0.7916	0.7720	0.0000	0.0000	58.1986	0.0000	0.0000	0.0000
	323.15 K							
1.0000	0.9537	0.9544	0.0000	0.0000	82.9401	0.0000	0.0000	0.0000
0.9003	0.9351	0.9325	-0.0028	0.5311	81.0682	899.8397	1.9864	0.0701
0.8008	0.9166	0.9544	0.0381	0.9791	79.1240	1517.5577	1.1231	0.1275
0.7003	0.8985	0.8811	-0.0159	1.3016	77.0183	1683.4651	0.3661	0.1716
0.6006	0.8875	0.8681	-0.0099	0.9487	74.2623	1864.5482	0.1653	0.2006
0.4760	0.8634	0.8552	0.0011	1.2580	71.5685	1966.5755	-0.0129	0.2144
0.4013	0.8498	0.8366	-0.0032	1.3004	69.8105	1904.9068	-0.1745	0.2100
0.2997	0.8311	0.8246	0.0042	1.2821	67.3434	1755.1817	-0.3769	0.1876
0.2032	0.8145	0.8046	0.0027	1.0672	64.8026	1448.6732	-0.7736	0.1479
0.1028	0.8073	0.7843	0.0016	-0.0428	61.2727	916.0792	-1.8788	0.0861
0.0000	0.7830	0.7630	0.0000	0.0000	58.8378	0.0000	0.0000	0.0000

(c) Pyridine (1)+ n-Propanol (2) 293.15 K

x_1	ρ	η	$\Delta\eta$	V^E	V_m	ΔG^{*E}	d'	$E\eta_{m(mPa.s)}$
1.0000	0.9847	1.1771	0.0000	0.0000	80.3290	0.0000	0.0000	0.0000
0.9003	0.9624	1.1804	-0.1237	0.3103	80.2220	629.0965	-7.3237	0.1944
0.8008	0.9847	1.4439	0.0129	-3.0113	76.4915	1264.3551	-2.3991	0.3325
0.7003	0.9100	1.4395	-0.1193	1.5910	80.6656	1492.3448	-1.4879	0.4204
0.6006	0.8970	1.8604	0.1747	1.0656	79.7229	2075.1513	0.0722	0.4618
0.4760	0.8839	1.5432	-0.3012	0.0903	78.2260	1413.7822	-0.3137	0.4565
0.4013	0.8650	1.9658	0.0263	0.4713	78.2945	1837.0767	0.9098	0.4264
0.2997	0.8524	2.0921	0.0232	-0.2107	77.1871	1632.4337	1.6931	0.3571
0.2032	0.8322	2.1453	-0.0465	-0.1364	76.8575	1265.4588	2.7869	0.2645
0.1028	0.8115	2.3774	0.0577	-0.1064	76.4673	913.9462	6.8043	0.1444
0.0000	0.7893	2.4506	0.0000	0.0000	76.1434	0.0000	0.0000	0.0000
	303.15 K							
1.0000	0.9747	0.9691	0.0000	0.0000	81.1532	0.0000	0.0000	0.0000
0.9003	0.9526	0.9840	-0.0732	0.3100	81.0473	703.2704	-6.3315	0.1489
0.8008	0.9747	1.1677	0.0223	-3.0537	77.2763	1304.8547	-2.0841	0.2560
0.7003	0.9005	1.1918	-0.0422	1.6135	81.5166	1621.8207	-1.1772	0.3253
0.6006	0.8874	1.4231	0.1010	1.0980	80.5853	2046.9496	-0.0212	0.3591
0.4760	0.8744	1.2571	-0.1752	0.1084	79.0759	1548.1616	-0.1938	0.3575

0.4013	0.8557	1.5283	0.0300	0.4894	79.1454	1884.0999	0.8134	0.3353
0.2997	0.8432	1.6233	0.0352	-0.2029	78.0293	1689.2286	1.5322	0.2824
0.2032	0.8232	1.6496	-0.0238	-0.1320	77.6978	1307.5202	2.4718	0.2104
0.1028	0.8027	1.8072	0.0451	-0.1054	77.3056	936.4934	6.0340	0.1155
0.0000	0.7807	1.8530	0.0000	0.0000	76.9822	0.0000	0.0000	0.0000
	313.15 K							
1.0000	0.9643	0.9638	0.0000	0.0000	82.0284	0.0000	0.0000	0.0000
0.9003	0.9426	0.8368	-0.1738	0.2954	81.9072	383.4332	-5.5588	0.1240
0.8008	0.9645	0.9663	-0.0913	-3.1103	78.0935	998.7747	-1.9771	0.2123
0.7003	0.8908	0.9949	-0.1098	1.6282	82.4042	1415.0639	-1.1742	0.2686
0.6006	0.8778	1.1437	-0.0079	1.1072	81.4666	1820.5142	-0.2812	0.2953
0.4760	0.8649	1.0381	-0.1720	0.1057	79.9445	1457.3345	-0.4604	0.2923
0.4013	0.8463	1.2196	-0.0256	0.4978	80.0245	1764.2923	0.3162	0.2733
0.2997	0.8339	1.2573	-0.0357	-0.2026	78.8995	1551.6114	0.6993	0.2291
0.2032	0.8142	1.3061	-0.0323	-0.1422	78.5566	1277.2762	1.3785	0.1699
0.1028	0.7937	1.3768	-0.0088	-0.0971	78.1822	859.8977	3.4239	0.0928
0.0000	0.7720	1.4339	0.0000	0.0000	77.8497	0.0000	0.0000	0.0000
	323.15 K							
1.0000	0.9544	0.7016	0.0000	0.0000	82.8793	0.0000	0.0000	0.0000
0.9003	0.9325	0.7178	-0.0234	0.3249	82.7943	823.5612	-4.2407	0.0945
0.8008	0.9544	0.8225	0.0418	-3.1482	78.9200	1423.3787	-1.2524	0.1611
0.7003	0.8810	0.8219	0.0014	1.6737	83.3209	1760.1280	-0.7413	0.2029
0.6006	0.8681	0.9078	0.0478	1.1397	82.3769	2057.1076	-0.0479	0.2220
0.4760	0.8552	0.8756	-0.0339	0.1263	80.8513	1828.4744	0.0330	0.2184
0.4013	0.8366	0.9560	0.0169	0.5344	80.9523	1938.6171	0.5392	0.2034
0.2997	0.8246	0.9983	0.0189	-0.2108	79.7894	1738.4873	1.0405	0.1696
0.2032	0.8046	1.0181	0.0004	-0.1095	79.4939	1394.0392	1.7372	0.1251
0.1028	0.7843	1.0804	0.0229	-0.0714	79.1192	967.3196	4.1813	0.0680
0.0000	0.7630	1.0983	0.0000	0.0000	78.7680	0.0000	0.0000	0.0000

(d) Pyridine (1) + n-Butanol (2) 293.15 K

x_1	ρ	η	$\Delta\eta$	V^E	V_m	ΔG^{*E}	d'	$E\eta_m$(mPa.s)
1.0000	0.9853	1.1835	0.0000	0.0000	80.2801	0.0000	0.0000	0.0000
0.9003	0.9630	1.2574	-0.1034	0.2411	81.6236	724.4743	-8.5277	0.2500
0.8008	0.9259	1.6625	0.1244	1.8752	84.3670	1656.9043	-2.4739	0.4173
0.7003	0.9435	1.4495	-0.2671	-1.3390	82.2549	1277.1430	-2.0954	0.5144
0.6006	0.9116	1.7512	-0.1428	-0.1076	84.5886	1703.3661	-0.6637	0.5504
0.4760	0.8934	1.6978	-0.4178	-0.4567	85.6173	1386.4009	-0.3042	0.5261
0.4013	0.8925	2.1925	-0.0560	-1.6131	85.2868	1764.6788	1.0338	0.4812
0.2997	0.8743	2.1732	-0.2560	-1.5398	86.4835	1364.5793	1.5854	0.3913
0.2032	0.8600	2.1685	-0.4323	-1.7275	87.3627	879.9503	2.5886	0.2815
0.1028	0.8434	2.3977	-0.3817	-1.7110	88.4894	476.8354	6.6323	0.1489

0.0000	0.8115	2.9623	0.0000	0.0000	91.3370	0.0000	0.0000	0.0000
	303.15 K							
1.0000	0.9752	0.9765	0.0000	0.0000	81.1116	0.0000	0.0000	0.0000
0.9003	0.9540	1.0421	-0.0630	0.1742	82.3936	777.1638	-7.7198	0.1921
0.8008	0.9166	1.3470	0.1133	1.8887	85.2230	1705.3286	-2.2085	0.3221
0.7003	0.9339	1.1972	-0.1659	-1.3414	83.1004	1380.6199	-1.8382	0.3990
0.6006	0.9024	1.3747	-0.1169	-0.0987	85.4510	1712.2245	-0.6821	0.4291
0.4760	0.8844	1.3657	-0.2866	-0.4456	86.4886	1469.8553	-0.2618	0.4128
0.4013	0.8836	1.7037	-0.0450	-1.6184	86.1459	1788.0016	0.9104	0.3792
0.2997	0.8656	1.6900	-0.1897	-1.5405	87.3527	1395.6712	1.4113	0.3101
0.2032	0.8521	1.7216	-0.2825	-1.7929	88.1727	963.2680	2.4456	0.2244
0.1028	0.8351	1.8864	-0.2472	-1.7123	89.3689	544.5442	6.2007	0.1194
0.0000	0.8037	2.2662	0.0000	0.0000	92.2235	0.0000	0.0000	0.0000
	313.15 K							
1.0000	0.9651	0.8188	0.0000	0.0000	81.9604	0.0000	0.0000	0.0000
0.9003	0.9441	0.8904	-0.0230	0.1827	83.2576	870.9028	-6.7869	0.1507
0.8008	0.9072	1.1079	0.0999	1.9095	86.1060	1749.0260	-1.9675	0.2544
0.7003	0.9243	0.9939	-0.1094	-1.3471	83.9635	1456.7037	-1.6451	0.3171
0.6006	0.8932	1.1123	-0.0856	-0.0940	86.3312	1751.0800	-0.6502	0.3433
0.4760	0.8754	1.1219	-0.1943	-0.4402	87.3777	1563.6854	-0.2064	0.3331
0.4013	0.8747	1.3530	-0.0341	-1.6306	87.0224	1818.0784	0.8047	0.3076
0.2997	0.8569	1.3092	-0.1743	-1.5492	88.2396	1367.2904	1.1370	0.2535
0.2032	0.8435	1.3856	-0.1895	-1.7958	89.0716	1038.4001	2.2830	0.1848
0.1028	0.8268	1.5131	-0.1573	-1.7238	90.2660	615.6071	5.8000	0.0992
0.0000	0.7958	1.7680	0.0000	0.0000	93.1390	0.0000	0.0000	0.0000
	323.15 K							
1.0000	0.9550	0.6958	0.0000	0.0000	82.8272	0.0000	0.0000	0.0000
0.9003	0.9342	0.7687	0.0064	0.1879	84.1399	967.2306	-5.6347	0.1151
0.8008	0.8977	0.9314	0.1025	1.9334	87.0173	1828.1858	-1.5466	0.1924
0.7003	0.9146	0.8450	-0.0508	-1.3543	84.8540	1583.1935	-1.3181	0.2375
0.6006	0.8839	0.9199	-0.0425	-0.0935	87.2395	1838.3713	-0.5197	0.2544
0.4760	0.8663	0.9378	-0.1077	-0.4431	88.2956	1706.3893	-0.0867	0.2436
0.4013	0.8655	1.0881	-0.0072	-1.6340	87.9474	1884.9685	0.7378	0.2230
0.2997	0.8480	1.0579	-0.1052	-1.5619	89.1657	1458.8314	1.0360	0.1816
0.2032	0.8349	1.1189	-0.1086	-1.8271	89.9891	1142.4333	2.0900	0.1308
0.1028	0.8183	1.2040	-0.0905	-1.7453	91.2036	693.1098	5.1957	0.0693
0.0000	0.7876	1.3631	0.0000	0.0000	94.1087	0.0000	0.0000	0.0000

Experimental density (ρ), dynamic viscosity (η), at temperatures of (293.15, 303.15, 313.15 and 323.15 K) are presented in table 2. The table also lists deviation in viscosity, $\Delta\eta$, excess molar volume, V^E and excess Gibbs free energy of activation of viscous flow ΔG^{*E}, for (methanol +

pyridine), (ethanol + pyridine), (*n*-propanol + pyridine) and (*n*-butanol + pyridine) as a function of mole fraction of the alcohol.

To investigate the molecular interaction between pyridine and the alcohols, (methanol, ethanol, *n*-propanol and *n*-butanol), viscosity deviation, $\Delta\eta$, excess molar volumes V^E and excess Gibbs free energy of activation of viscous flow, ΔG^{*E}, have been evaluated from experimental density and viscosity using equations 1 and 2 respectively.

$$V^E = \frac{x_1M_1+x_2M_2}{\rho_m} - \left(\frac{x_1M_1}{\rho_1} + \frac{x_2M_2}{\rho_2}\right) \quad (1)$$

$$\Delta\eta = \eta_m - (x_1\eta_1 + x_2\eta_2) \quad (2)$$

where x_1 and x_2 are the mole fractions calculated from mass fractions. M_1 and M_2 are molar masses, ρ_1 and ρ_2 are densities, η_1 and η_2 are the viscosities of pure components 1 and 2 respectively. ρ_m and η_m are the density and viscosity of the mixture.

The excess Gibbs free energy of activation of viscous flow was obtained from equation 3.

$$\Delta G^{*E} = RT[ln\eta_m V_m - (x_1 ln\eta_1 V_1 + x_2 ln\eta_2 V_2)] \quad (3)$$

where R is the universal constant of gases, T is the absolute temperature, V_1 and V_2 are the molar volumes of component 1 and 2, x_1 and x_2 represents the mole fraction of component 1 and 2. V_m is obtained from equation 4 below. η_1, η_2 and η_m are the viscosity of component 1 and 2 and mixture respectively.

$$V_m = \frac{x_1M_1+x_2M_2}{\rho_m} \quad (4)$$

The values of V^E, $\Delta\eta$ and ΔG^{*E} were correlated by a Redlich-Kister [22] type polynomial, equation 5 and 6.

$$\Delta Y = x_1x_2\sum_{k=1}^{n} A_k(2x_1-1)^{k-1} \quad (5)$$

$$\Delta Y = x_1x_2[A_o + A_1(2x_1-1)^1 + A_2(2x_1-1)^2 + A_3(2x_1-1)^3 + A_4(2x_1-1)^4] \quad (6)$$

The values of the parameters A_k, are obtained by fitting the equation to the experimental values with the least-squares method. The correlated results for excess molar volume, viscosity deviation and excess Gibbs free energy of activation of viscous flow are presented in table 3. The standard deviation $\sigma(\Delta Y)$ is calculated from equation 7.

$$\sigma(\Delta Y) = \left[\frac{\sum(Y_{expt}-Y_{calc})^2}{N-n}\right]^{1/2} \quad (7)$$

where ΔY is the excess volume, V^E, deviation in viscosity $\Delta\eta$, and excess Gibbs free energy of activation of viscous flow, ΔG^{*E}. The subscript *expt* and *calc* represents the experimental and calculated values respectively. *N* and *n* are the number of experimental data points and the number of coefficients in the Redlich-Kister polynomial equation.

Table 3. Adjustable parameters A_i, with standard deviations $\sigma(\Delta Y)$, for deviation in viscosity ($\Delta\eta$), Excess volume (V^E), and Excess Gibbs free energy (ΔG^{*E}), for binary mixtures at various temperatures.

Parameter/Function	*T/K*	A_o	A_1	A_2	A_3	A_4	σ
$\Delta\eta$ (mPa.s)				Pyridine (1) +Methanol (2)			
	293.15	-0.2585	0.1306	3.0×10^{-15}			0.58
	303.15	-0.1866	0.0943	9.0×10^{-16}			0.42
	313.15	-0.1403	0.0737	1.0×10^{-15}			0.31
	323.15	-0.1711	0.3293	9.0×10^{-16}			0.07
V^E (cm^3/mol)	293.15	1.1668	0.0408	-3.0×10^{-13}	1.0×10^{-13}		3.7
	303.15	1.2504	0.0598	-3.0×10^{-13}	2.0×10^{-13}		4.0
	313.15	1.2774	0.0561	-2.0×10^{-13}	1.0×10^{-13}		4.1
	323.15	1.5192	0.08764	-7.0×10^{-13}	5.0×10^{-13}		6.2
ΔG^{*E} (kJ/mol)	293.15	0.75	0.198				2.7
	303.15	0.84	0.196	-7.0×10^{-12}			3.0
	313.15	0.94	0.200	-3.0×10^{-10}	1.0×10^{-10}		3.3
	323.15	0.78	0.524	-7.0×10^{-12}			3.4
				Pyridine (1) + Ethanol (2)			
$\Delta\eta$ (mPa.s)	293.15	0.0035	-0.0036				0.0024
	303.15	0.0034	-0.0036	-1.0×10^{-17}			0.0023
	313.15	0.0034	-0.0036	-1.0×10^{-17}			0.0022
	323.15	0.0034	-0.0038	-2.0×10^{-17}			0.0014
V^E (cm^3/mol)	293.15	0.8153	-0.1314	-7.0×10^{-15}			2.36
	303.15	0.8313	-0.1377	-2.0×10^{-14}			2.40
	313.15	-8.9159	7.6559	-2.0×10^{-12}	7.0×10^{-13}		10.79
	323.15	0.8572	-0.1458	-1.0×10^{-13}			2.46
ΔG^{*E} (kJ/mol)	293.15	1.14	27.614	0.2719	0.0021		4.09
	303.15	1.18	27.879	0.2719	0.0021		4.23
	313.15	0.94	256.35	0.2719	0.0021		3.98
	323.15	1.26	28.095	0.2719	0.0021		4.51
				Pyridine (1)+ *n*-Propanol (2)			
$\Delta\eta$ (mPa.s)	293.15	-0.0647	0.0005	6.0×10^{-5}	-8.0×10^{-15}	3.0×10^{-7}	0.08
	303.15	-0.0246	0.0343	-7.0×10^{-15}	3.0×10^{-14}	-7.0×10^{-15}	0.007
	313.15	-0.1026	0.0855	-1.0×10^{-14}	9.0×10^{-15}		0.14
	323.15	0.006	0.005	-9.0×10^{-14}	1.0×10^{-13}	-6.0×10^{-14}	0.04
V^E (cm^3/mol)	293.15	-0.1217	0.2543	-4.0×10^{-16}			0.24
	303.15	-0.1209	0.2639	4.0×10^{-16}			0.27
	313.15	-0.1341	0.2823	4.0×10^{-16}			0.27
	323.15	-0.1222	0.2908	4.0×10^{-16}			0.33
ΔG^{*E} (kJ/mol)	293.15	1.08	0.113				3.64
	303.15	1.14	0.090				4.9
	313.15	0.91	0.277				3.5
	323.15	1.25	0.0411				4.0
				Pyridine (1) + *n*-Butanol (2)			
$\Delta\eta$ (mPa.s)	293.15	-0.0513	-0.248	9×10^{-13}	-2×10^{-12}	7×10^{-13}	0.92
	303.15	-0.0283	-0.1763	7×10^{-13}	-1×10^{-12}	2×10^{-13}	0.63
	313.15	-0.01	-0.1374	9×10^{-13}	-1×10^{-12}	5×10^{-13}	0.44
	323.15	0.014	-0.1011	3×10^{-13}	-6×10^{-13}	2×10^{-13}	0.24
V^E (cm^3/mol)	293.15	0.354	-1.8625	5×10^{-12}	-1×10^{-11}	4×10^{-12}	4.09
	303.15	0.3431	-1.8606	7×10^{-12}	-4×10^{-12}	4×10^{-12}	4.13
	313.15	0.3509	-1.8763	5×10^{-12}	-1×10^{-11}	4×10^{-12}	4.14
	323.15	0.3594	-1.9024	4×10^{-12}	-7×10^{-12}	5×10^{-12}	4.19
ΔG^{*E} (kJ/mol)	293.15	1.16	0.273	1×10^{-10}	-1×10^{-10}		3.69
	303.15	1.20	-0.296	-1×10^{-10}	1×10^{-10}		3.52
	313.15	1.26	-0.288	-7×10^{-12}			3.42
	323.15	1.34	-0.295	1×10^{-10}	-1×10^{-10}		3.93

Kendall and Monroe [23] derived equation 8 for analyzing the viscosity of binary mixtures based on zero adjustable parameter.

$$\eta_m = \left(x_1\eta_1^{1/3} + x_2\eta_2^{1/3}\right)^3 \quad (8)$$

$$E\eta_m = x_1x_2\left(x_1\eta_1^{1/3} + x_2\eta_2^{1/3}\right)^3 \quad (9)$$

where $E\eta_m$ is a modified Kendall-Monroe equation, 9.

The predictive ability of some selected viscosity models such as the one parameter model of Frenkel [24] equation 10 and Hind [25] equation 11, apply to the studied binary mixtures.

$$\ln\eta = x_1^2 ln\eta_1 + x_2^2 ln\eta_2 + 2x_1x_2 ln\eta_{12} \quad (10)$$

$$\eta = x_1^2\eta_1 + x_2^2\eta_2 + 2x_1x_2\eta_{12} \quad (11)$$

where η_{12} is a constant attributed to unlike pair interactions. Its value is obtained from equation 12.

$$\eta_{12} = 0.5\eta_1 + 0.5\eta_2 \quad (12)$$

Grunberg and Nissan [26] formulated equation 13 to determine the molecular interactions leading to viscosity changes with one parameter to estimate the dynamic viscosity of binary liquid mixtures.

$$ln\eta_m = x_1 ln\eta_1 + x_2 ln\eta_2 + x_1x_2d' \quad (13)$$

where d' is an interaction parameter which is a function of the composition and temperature of binary liquid mixture.

McAllister's three-body interaction model derived for the viscosity of a mixture based on Eyring's rate theory enables correlation of the kinematic viscosity of binary liquid mixtures with mole fraction [15]. The three-body model is presented in equation 14.

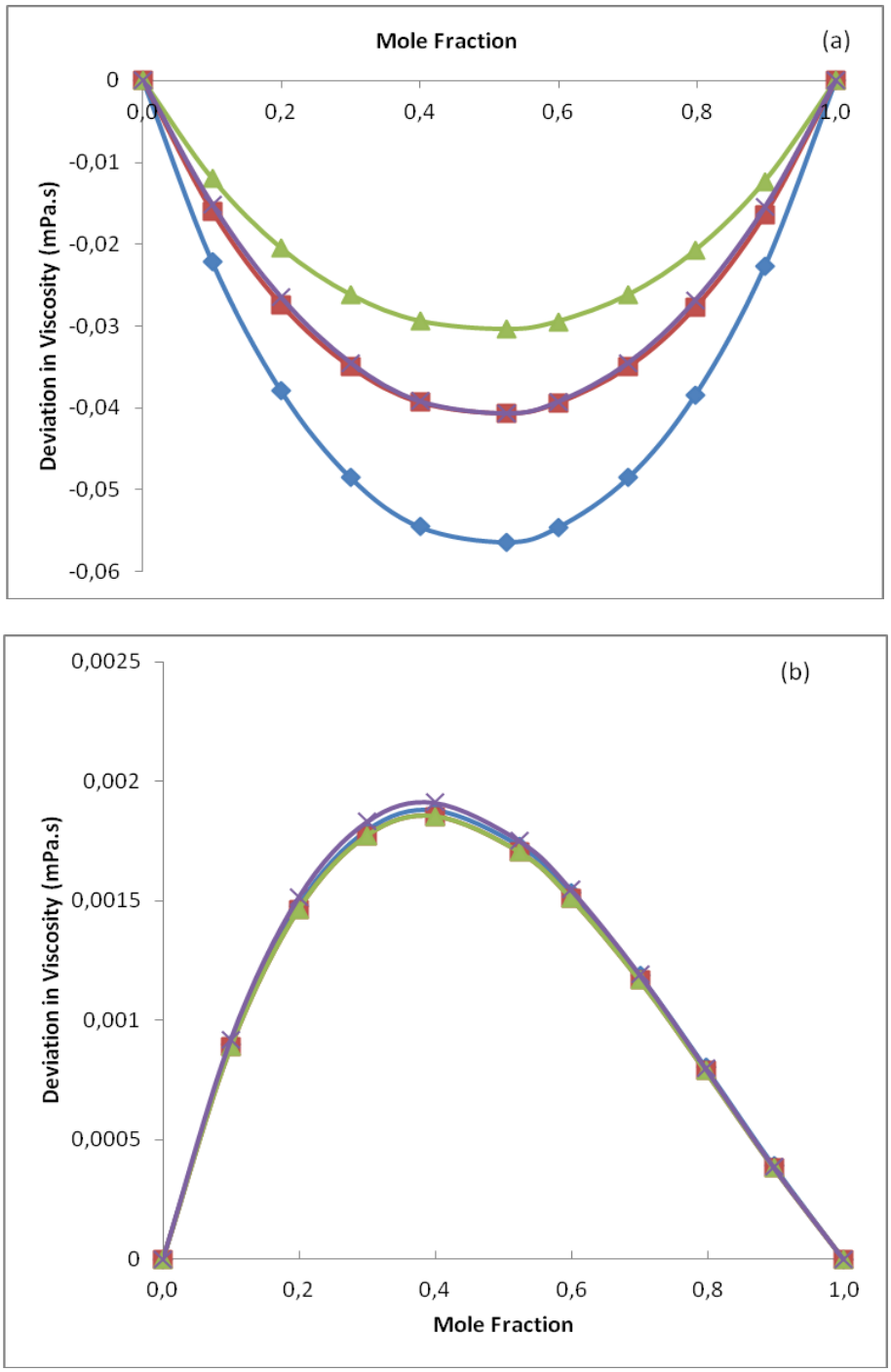
$$lnv = x_1^3 lnv_1 + x_2^3 lnv_2 + 3x_1^2x_2 lnv_{12} + 3x_1x_2^2 lnv_{21} - ln\left[x_1 + \left(\frac{x_2M_2}{M_1}\right)\right] + 3x_1^2x_2 ln\left[\left(\frac{2}{3}\right) + \left(\frac{M_2}{3M_1}\right)\right] + 3x_1x_2^2 ln\left[\left(\frac{1}{3}\right) + \left(\frac{2M_2}{3M_1}\right)\right] + x_2^3 ln\left(\frac{M_2}{M_1}\right) \quad (14)$$

where v, v_1 and v_2 are the kinematic viscosities of the mixture, viscosity of component 1 and 2 respectively, v_{12} and v_{21} are interaction parameters. The correlating ability of equations 9, 10, 11 and

13 were tested by calculating the average percentage deviations (*APD*) between the experimental and the calculated viscosity as shown in equation 15.

$$APD = \frac{100}{N}\sum_{i=1}^{N}\left[\frac{(\eta_{expt}-\eta_{calc})}{\eta_{expt}}\right] \quad (15)$$

where η_{expt} and η_{calc} represent the viscosity of experimental and calculated data, N is the number of experimental data points.



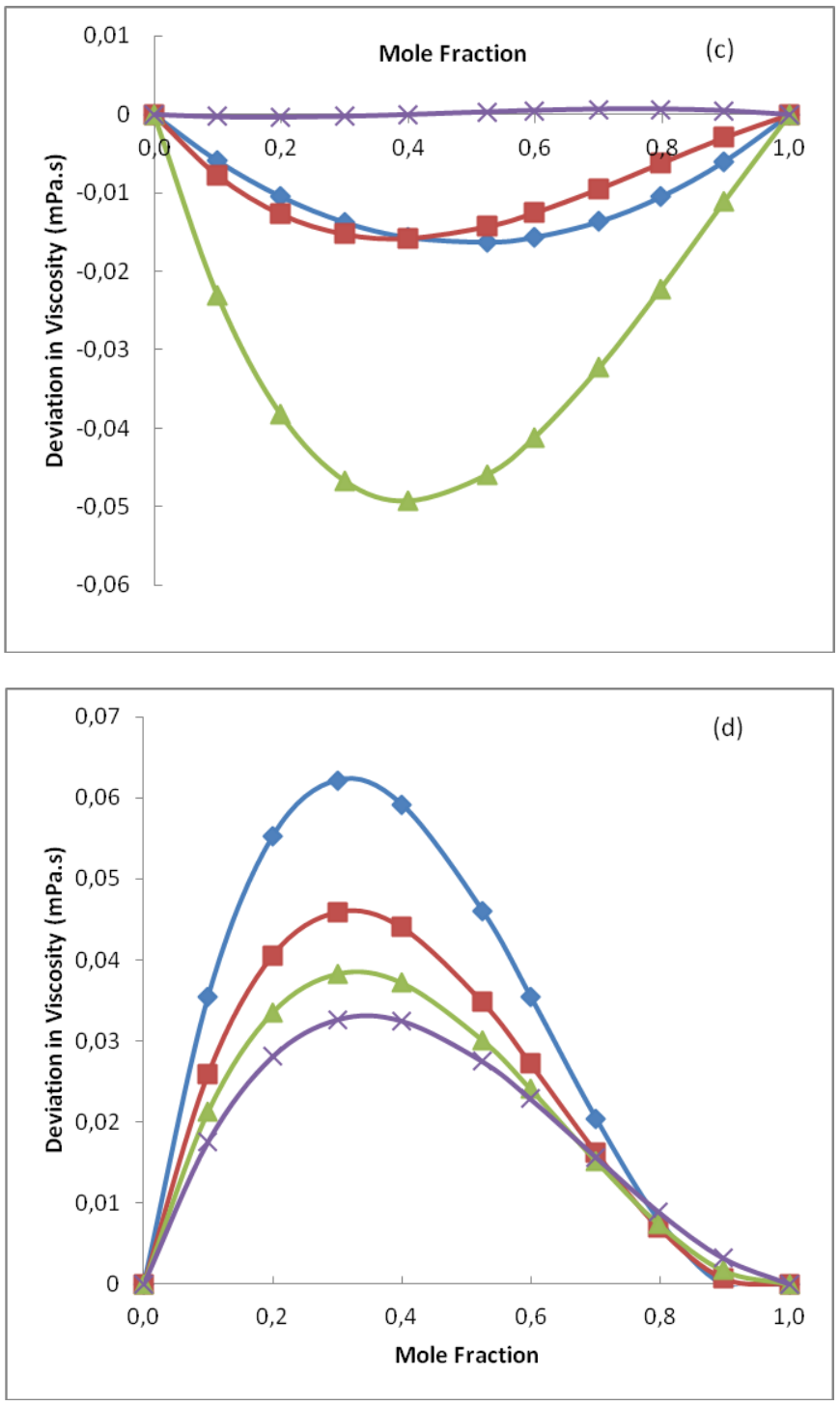
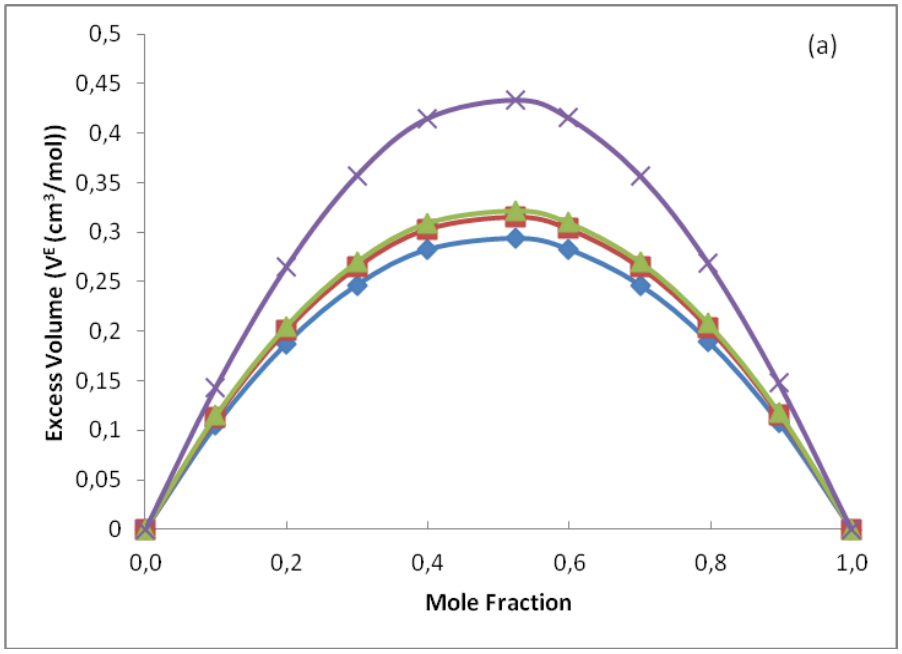
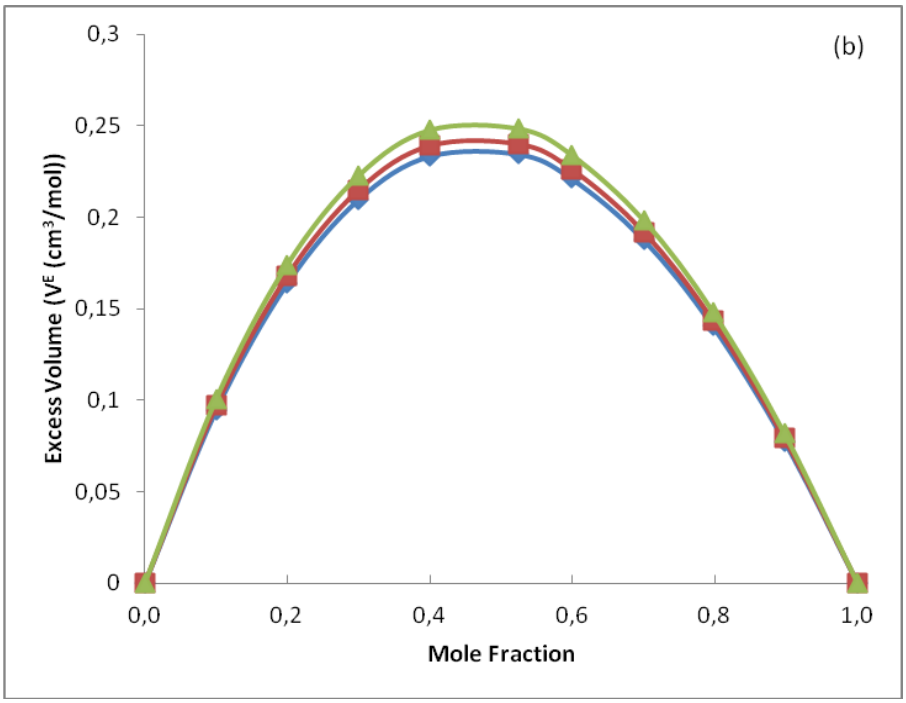


Figure 1. Plots of deviation in viscosity (Δη) against mole fraction for the system (a) Pyridine (1) + Methanol (2) (b) Pyridine (1) + Ethanol (2) (c) Pyridine (1) + n-Propanol (2) (d) Pyridine (1) + *n*-Butanol (2) at different temperatures: ♦, 293.15 K; ■, 303.15 K;▲, 313.15 K; x, 323.15 K. The solid line represents the corresponding correlation by the Redlich-Kister equation.

The plots of deviation in viscosity against mole fraction at 293.15, 303.15, 313.15 and 323.15 K for pyridine + methanol, pyridine + ethanol, pyridine + *n*-propanol and pyridine + *n*-butanol are presented in figure 1 (a-d). Deviations in viscosity were found to be both negative and positive.

Negative deviations are observed for pyridine +methanol and pyridine + *n*-propanol mixtures while positive deviations were observed for pyridine + ethanol and pyridine + *n*-butanol mixtures. The negative values of the deviation in viscosity ($\Delta\eta$) suggest the existence of weak intermolecular interactions upon mixing in methanol and n-propanol while the positive values of deviation observed in ethanol and n-butanol relate to strong intermolecular interaction between pyridine, ethanol and n-butanol. This shows that the strength of the specific forces is not the factor influencing the viscosity deviation in the liquid mixture. This leads to suggestions that combinations of an interactive and non-interactive force are responsible in these positive and negative interactions [9,27]. The figures also clearly show a general deviation in viscosity to decrease with increase in temperature.





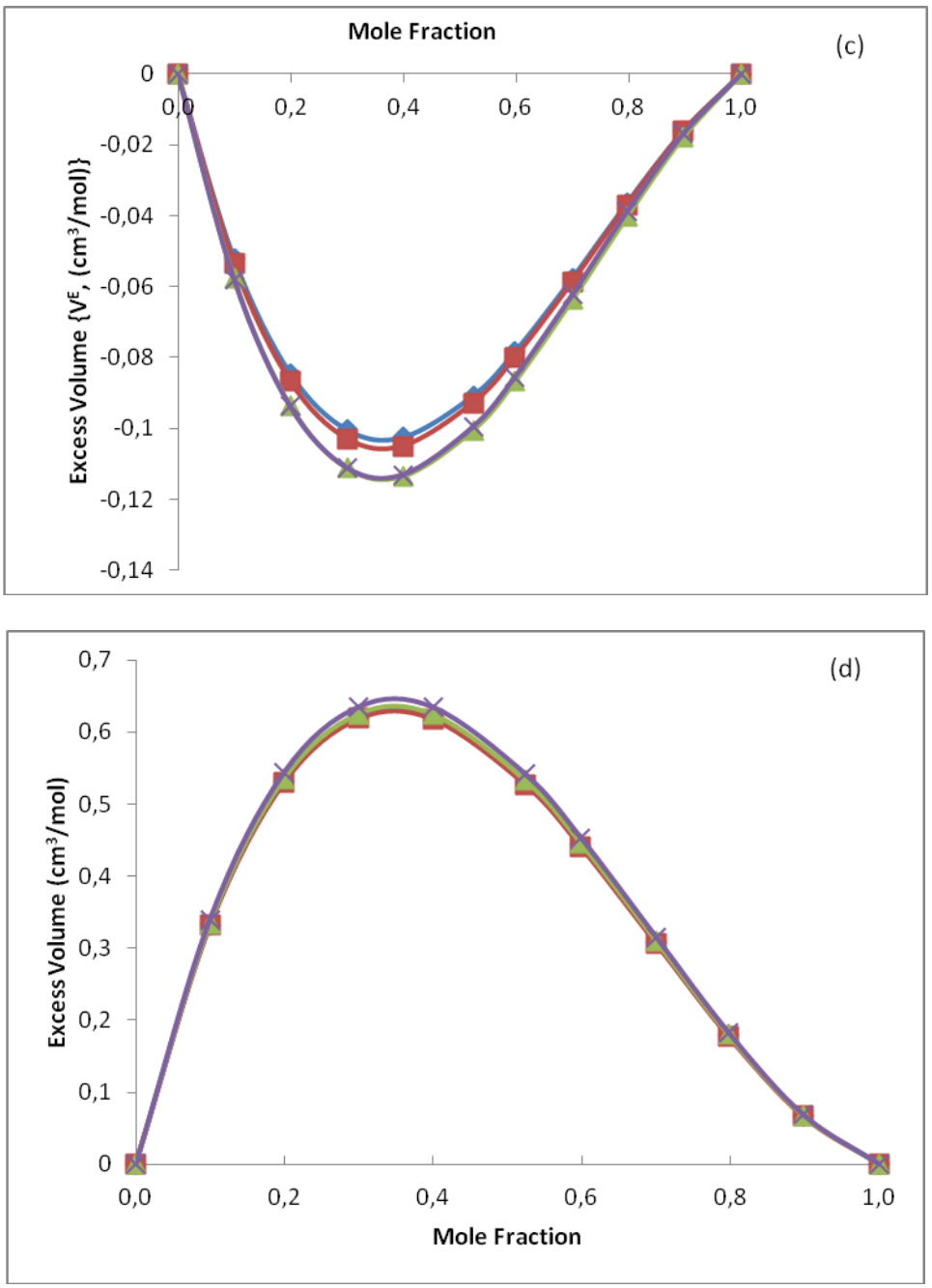
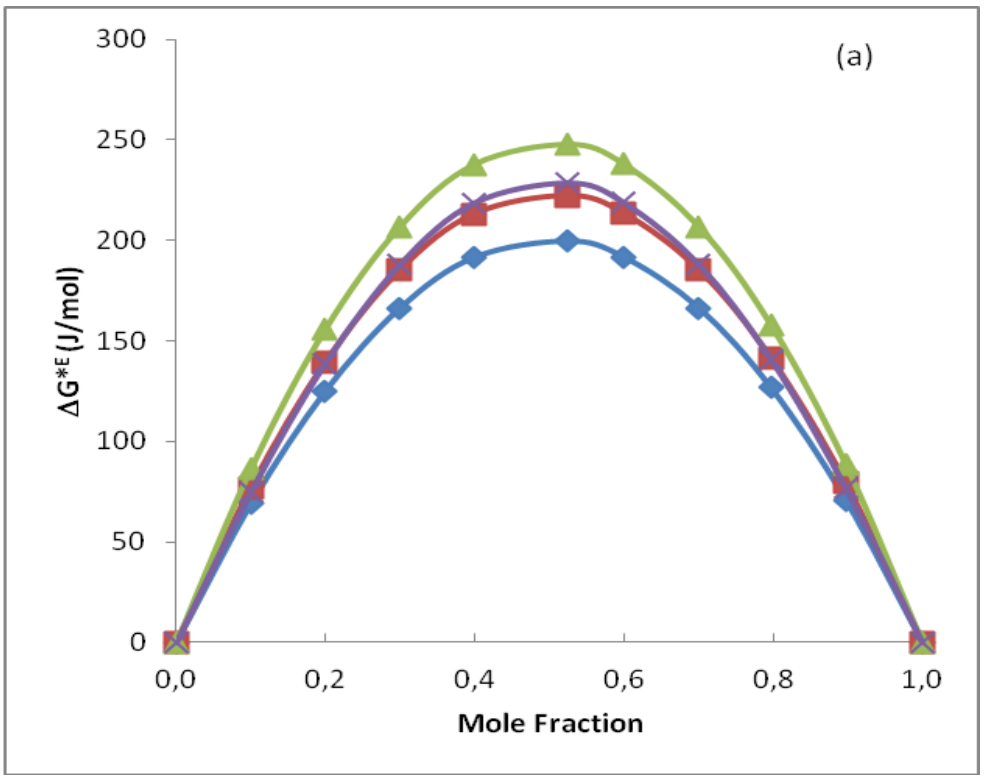
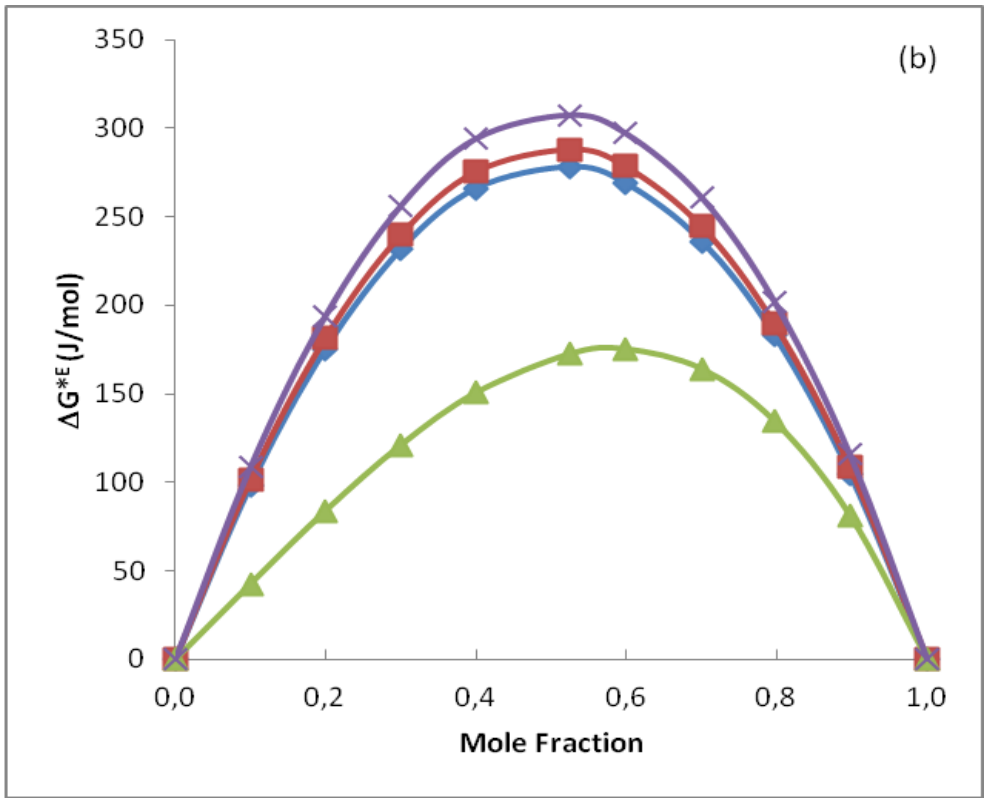


Figure 2. Plots of Excess molar volume (V^E) against mole fraction for the system (a) Pyridine (1) + Methanol (2) (b) Pyridine (1) + Ethanol (2) (c) Pyridine (1) + n-Propanol (2) (d) Pyridine (1) + *n*-Butanol (2) at different temperatures: ♦, 293.15 K; ■, 303.15 K;▲, 313.15 K; x, 323.15 K. The solid line represents the corresponding correlation by the Redlich-Kister equation.

The plots of excess molar volume against mole fraction at 293.15, 303.15, 313.15 and 323.15 K for pyridine + methanol, pyridine + ethanol, pyridine + *n*-propanol and pyridine + *n*-butanol are presented in figure 2 (a-d). Excess parameters associated with a liquid mixture are a quantitative measure of deviation in the behavior of the liquid mixture from ideality [3,4]. These functions are found to be sensitive towards the intermolecular forces and also on the difference in size and shape of

the molecules. Excess volumes of liquid mixtures reflect the result of different contributions arising from structural changes undergone by the pure cosolvent.





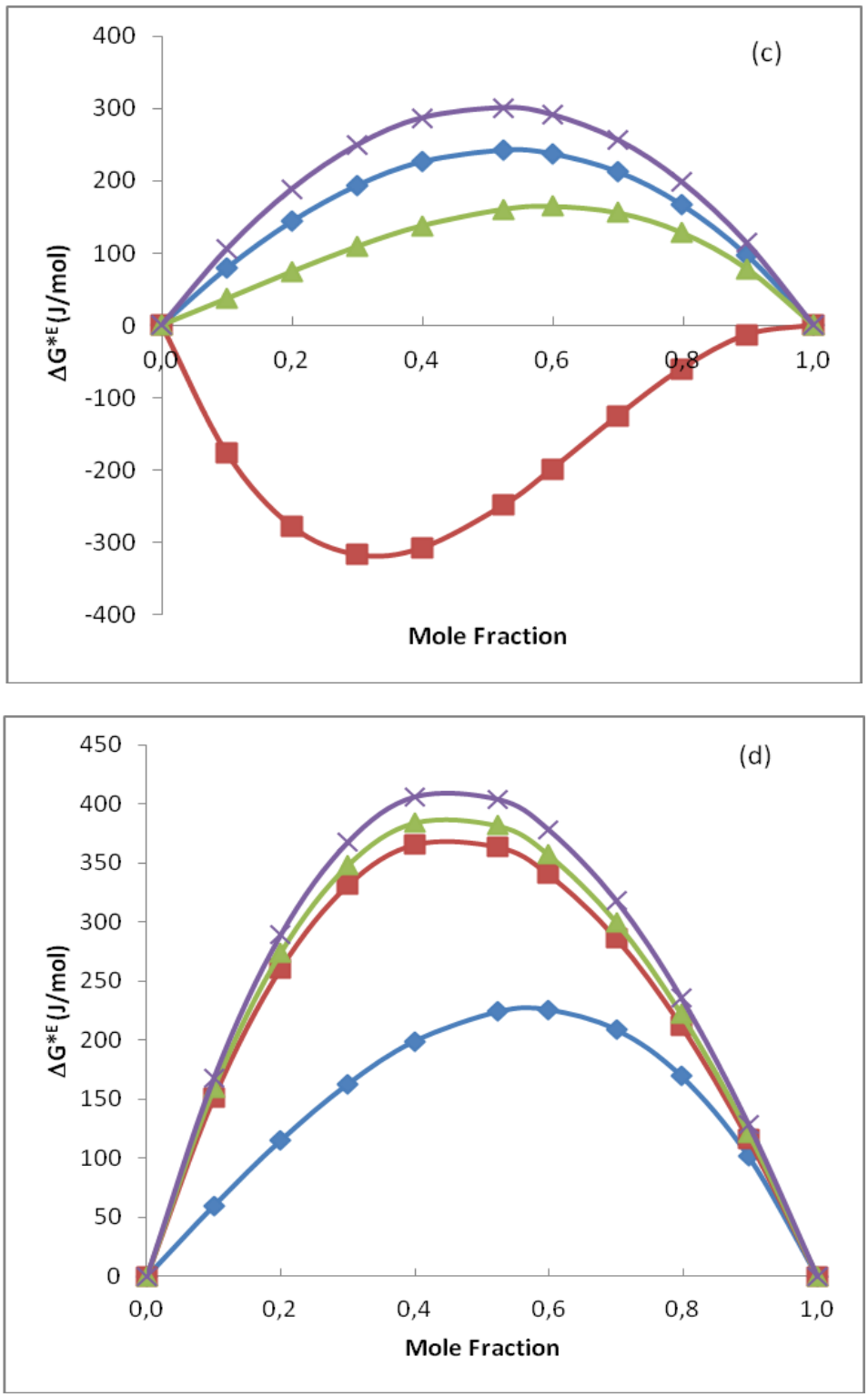
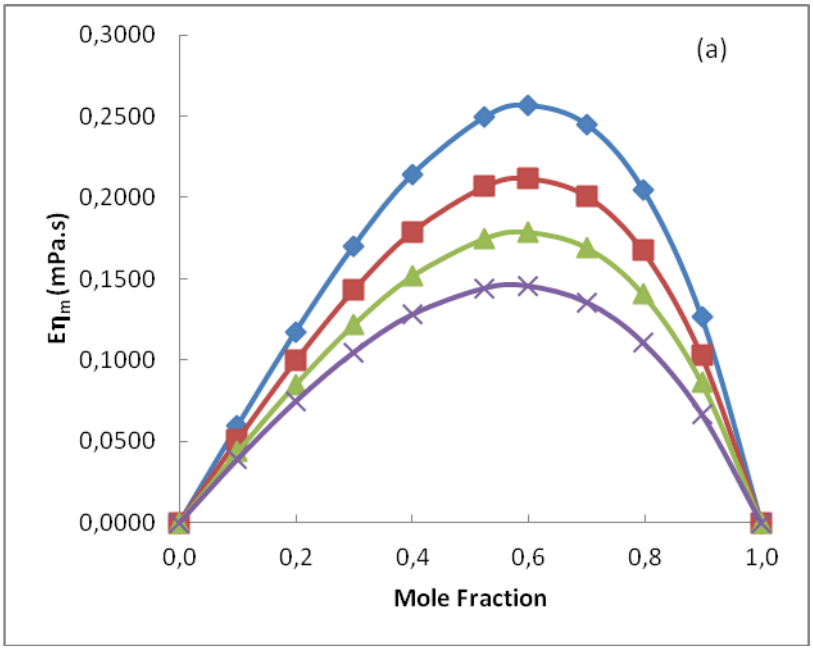
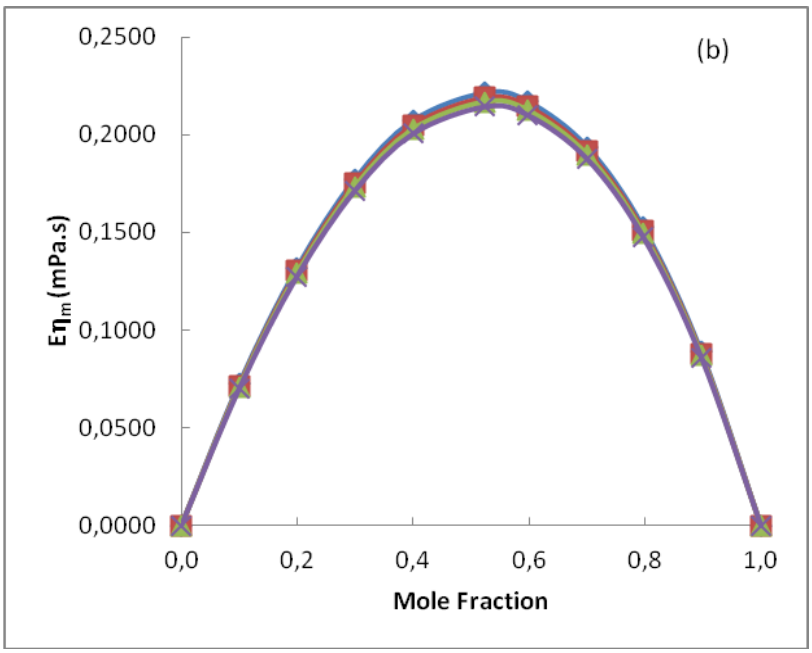


Figure 3. Plots of Excess Gibbs free energy of activation of viscous flow (ΔG^{*E}) against mole fraction for the system system (a) Pyridine (1) + Methanol (2) (b) Pyridine (1) + Ethanol (2) (c) Pyridine (1) + n-Propanol (2) (d) Pyridine (1) + *n*-Butanol (2) at different temperatures: ♦, 293.15 K; ■, 303.15 K;▲, 313.15 K; x, 323.15 K. The solid line represents the corresponding correlation by the Redlich-Kister equation.

Positive contributions arise from breakup of interactions between molecules namely, the rupture of hydrogen bonded chains and the loosening of dipole interactions [28]. The values of V^E for the mixtures of pyridine + methanol, pyridine + ethanol and pyridine + n-butanol are positive while for the mixture pyridine + n-propanol is negative. In all plots, V^E increases with increase in temperature. The values of V^E are the result of contributions from several opposing effects. Negative excess molar volume can be attributed to strong interactions between unlike molecules through hydrogen bonding as observed in the system pyridine + n-propanol.





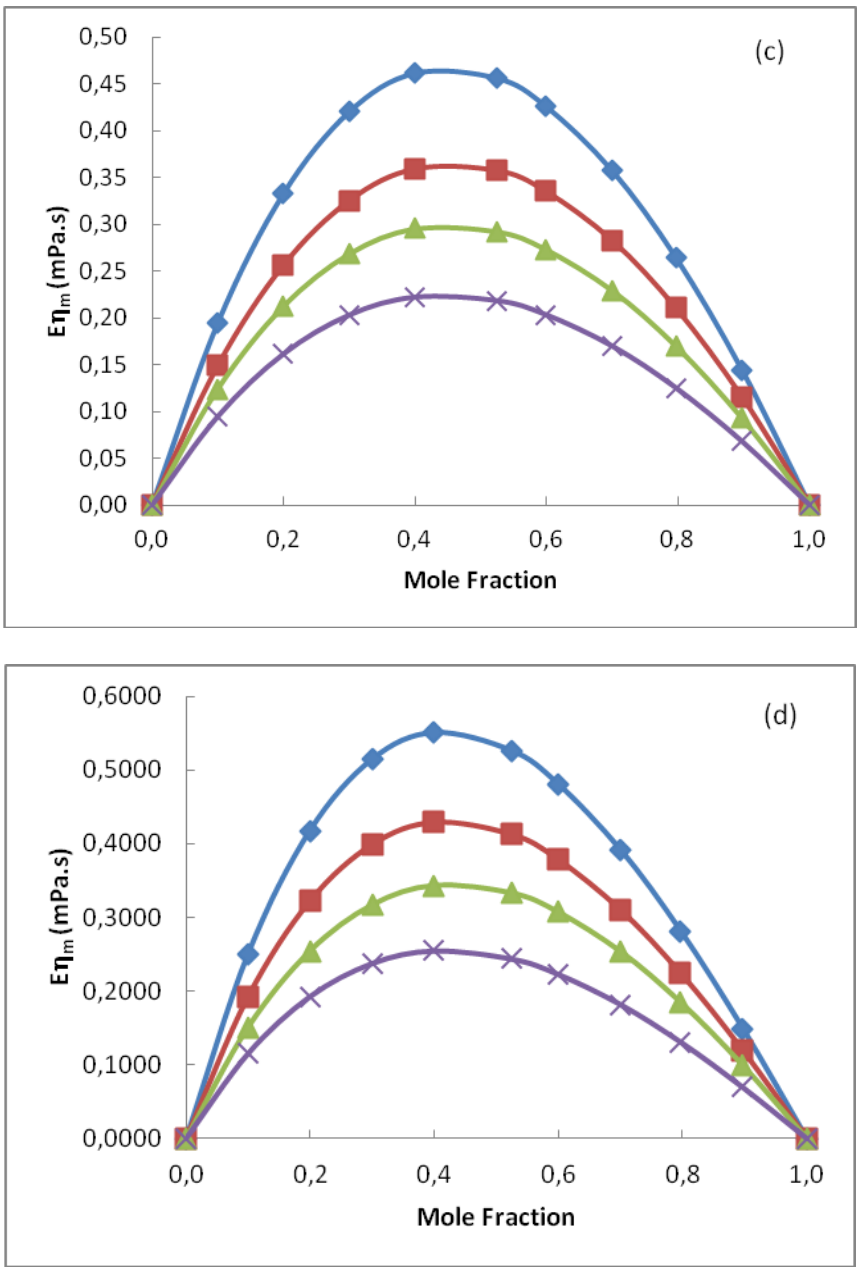


Figure 4. Plots of modified Kendall and Monroe viscosity correlation $E\eta_m$ (mPa.s) against mole fraction for the system (a) Pyridine (1) + Methanol (2) (b) Pyridine (1) + Ethanol (2) (c) Pyridine (1) + n-Propanol (2) (d) Pyridine (1) + n-Butanol (2) at different temperatures: ♦, 293.15 K; ■, 303.15 K;▲, 313.15 K; x, 323.15 K.

The plots of excess Gibbs free energy of activation of viscous flow against mole fraction at 293.15, 303.15, 313.15 and 323.15 K for pyridine + methanol, pyridine + ethanol, pyridine + *n*-propanol and pyridine + *n*-butanol are presented in figure 3(a-d). Excess properties provide information about the molecular interactions and macroscopic behavior of fluid mixtures which can be used to test and improve thermodynamic models for calculating and predicting fluid phase equilibria [4]. The magnitude of ΔG^{*E} represents the strength of interaction between unlike molecules [29,30]. Excess Gibbs free energy of activation of viscous flow were found to be positive for all plots. In all

plots, ΔG^{*E} increased with increase in temperature. The positive values of excess Gibbs free energy of activation of viscous flow indicate the presence of specific and strong interactions in the systems under investigation [31,32]. The excess Gibbs free energy of activation of viscous flow attains a maximum between 0.44 – 0.6 of the mole fraction of pyridine.

A comparison of experimental thermodynamic data of multicomponent mixtures with that calculated by means of various predictive methods is very useful from different points of view: (i) it suggests which model is more appropriate to the characteristics of the system, (ii) it may indicate which parameters should be improved when the model involves group contributions and (iii) it may allow the identification of some model as a convenient reference for the interpretation of the deviations observed [4]. The viscosity data have been correlated with semi-empirical equations of modified Kendall and Monroe, Frenkel, Hind, and Grunberg-Nissan. The values of the Grunberg and Nissan constant (d') and modified Kendall-Monroe ($E\eta_m$) for all systems under study are presented in table 2. Grunberg-Nissan interaction parameters are both positive and negative while the modified Kendall-Monroe viscosity correlation data are all positive. Plots for the modified Kendall-Monroe viscosity correlation are presented in figure 4(a-d). Plots of modified Kendall-Monroe viscosity correlation at different temperatures show decrease in viscosity with increase in temperature. The values of Frenkel and Hind are presented in table 4.

Table 4. Fitting parameters with Average Percentage Deviations (APD) for binary mixtures at various temperatures.

Temperature	Frenkel		Hind	
K	η_{12}	APD	η_{12}	APD
Pyridine (1) + Methanol (2)				
293.15	1.04585	-0.617	1.04585	0.363
303.15	0.86085	-0.699	0.86085	0.147
313.15	0.7261	-0.619	0.7261	0.177
323.15	0.5887	-0.472	0.5887	0.005
Pyridine (1) + Ethanol (2)				
293.15	0.8870	0.073	0.8870	0.129
303.15	0.8777	0.075	0.8777	0.132
313.15	0.8683	0.076	0.8683	0.133
323.15	0.8587	0.078	0.8587	0.135
Pyridine (1) + *n*-Propanol (2)				
293.15	1.8739	0.622	1.8739	0.843
303.15	1.4604	0.743	1.4604	0.941
313.15	1.1989	0.913	1.1989	0.887
323.15	0.89995	0.508	0.89995	0.050
Pyridine (1) + *n*-Butanol (2)				
293.15	2.2275	-1.858	2.2275	-1.095
303.15	1.7358	-1.604	1.7358	-0.977
313.15	1.3889	-1.247	1.3889	-0.786
323.15	1.0295	0.051	1.0295	0.581

Positive and negative Grunberg-Nissan parameters indicate the presence of both strong and weak interactions between unlike molecules [9].

4. CONCLUSION

The deviation in viscosity, excess molar volume and excess Gibbs free energy of activation of viscous flow for the systems pyridine + methanol, pyridine + ethanol, pyridine + *n*-propanol and pyridine + *n*-butanol at T = 293.15, 303.15, 313.15 and 323.15 K has been reported. There is intermolecular interaction among the components of the binary mixtures leading to possible hydrogen bond formation of the type N···H─O between unlike molecules confirming hydrogen bonding formation between pyridine and the alcohol mixtures.

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References

1. V. Serheyev. *Chem. Chem. Tech*. 5(3) (2011) 241.
2. G. Conti, P. Gianni, L. Lepori, E. Matteoli. *J. Pure & Appl. Chem.* 67(1) (1995) 1849.
3. B. R. Kumar, B. Satyanarayana, S. A. Banu, K. A. Jyoti, T. S. Jyostna, N. Satyanarayana. *Ind. J. Pure & Appl. Phys.* 47 (2009) 511.
4. S. Parveen, M. Yasmin, M. Gupta, J. P. Shukla. *Int. J. Thermodyn.* 13(2) (2010) 59.
5. S. Singh, B. P. S. Sethi, R. C. Katyal, V. K. Rattan. *J. Chem. Eng. Data* 49 (2004) 1373.
6. R. A. Clara, A. C. G. Marigliano, V. V. Campos, H. N. Solimo. *Fluid Phase Equilib*. 293 (2010) 151.
7. B. Gonzalez, N. Calvar, E. Gomez, A. Dominguez. *J. Chem. Thermodyn.* 39 (2007) 1578.
8. S. C. Bhatia, J. Sangwan, R. Bhatia. *J. Mol. Liq.* 161 (2011) 95.
9. B. Sathyanarayana, B. Ranjithkumar, T. S. Jyostna, N. Satyanarayana. *J. Chem. Thermodyn.* 39 (2007) 16 .
10. R. Baskaran, T. R. Kubendran. *Int. J. Appl. Sci. Eng.* 8(2) (2010) 149.
11. Fedeles, O. Ciocirlan, O. Iulian. *U. P. B. Sci. Bull. B* 71(4) (2009) 99.
12. M. L. J. Kijevcanin, V. Z. Kostic, I. R. Radovic, B. D. Djordjevic, S. P. Serbanovic. *Chem. Ind. Chem Eng.* 14(4) (2008) 223.
13. S. S. Patil, S. R. Mirgane. *Rasayan J. Chem.* 4(2) (2011) 445.
14. E. M. Zivkovic, M. L. Kijevcanin, I R. Radovic, S. P. Serbanovic, B. D. Djordjevic. *Fluid Phase Equilib.* 299 (2010) 191.
15. S. C. Bhatia, R. Rani, R. Bhatia. *J. Mol. Liq*. 159 (2011) 132.
16. S. L. Oswal, H. S. Desai. *Fluid Phase Equilib.* 161 (1999) 191.
17. V. E. Vittal Prasad, S. B. Agrawal, A. B. Bajaj, D. H. L. Prasad. *Phys. Chem. Liq.: Int. J.* 38(4) (2000) 433.
18. Lange's Handbook of Chemistry 10^{th} edition, 1525 – 1528
19. N. Deenadayalu, I. Bahadur, T. Hofman, *J. Chem. Thermodyn.* 42 (2010) 726.
20. M. M. Taib, A. K. Ziyada, C. D. Wilfred, T. Murugesan. *J. Mol. Liq.* 158 (2011) 101.
21. J. Liu,C. Zhu, Y. Ma. *J. Chem. Eng, Data.* 56 (2011) 2095.
22. O. Redlich, A. T. Kister. *Ind. Eng. Chem.* 40(2) (1948) 345.
23. J. Kendall, K. P. Monroe. *J. Am. Chem. Soc.* 39(9) (1917) 1787.
24. Frenkel Ya. I.. *Petroleum (London)* 9 (1946) 27.
25. R. K. Hind, E. McLaughlin, A. R. Ubbelohde. *Trans Faraday Soc.* 56 (1960) 328.

26. L. Grunberg, A. H. Nissan. *Nature* 164 (1949) 799.
27. B. Mokhtarani, A. Sharifi, H. R. Mortaheb, M. Mirzaei, M. Mafi, F. Sadeghian. *J. Chem. Thermodyn.* 41 (2009) 323.
28. M. M. El-Banna. *Can. J. Chem.* 75 (1997) 1890.
29. P. N. Tshibangu, S. N. Ndwandwe, E. D. Dikio *Int. J. Electrochem. Sci.* 6(3) (2011) 2201.
30. M. Shafiq, S. Asif, M. Farooqui. *A. J. Biochem. Pharm. Res.* 1(2) (2011) 419.
31. K. Sreekanth, D. S. Kumar, M. Kondaiah, D. K. Rao. *J. Phys B.* 406 (2011) 854.
32. A. G. Peshwe, B. R. Arbad, S. P. Pachaling. *Int. J. Chem. Sci.* 7(3) (2009) 1505.