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Densities, Viscosities, and Speeds of Sound of Binary Liquid Mixtures of Ethylenediamine with Alcohols at T = (293.15 to 313.15) K

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Supporting Information

ABSTRACT: Densities, ρ, and speeds of sound, u, have been measured for the binary liquid mixtures of ethylenediamine, ($NH_2CH_2CH_2NH_2$), with 2-methyl-1-propanol, ($C_4H_{10}O$), 2-propanol, (C_3H_8O), and 1-butanol, ($C_4H_{10}O$), at (293.15, 298.15, 303.15, 308.15, and 313.15) K and viscosities at (298.15, 303.15, and 308.15) K and a pressure of 0.1 MPa as a function of composition. Furthermore, using the experimental results excess molar volumes, V_m^E, isentropic compressibilities, κ_S, deviation in isentropic compressibilities, $\Delta\kappa_S$, deviation in speeds of sound, Δu, and viscosity deviations, $\Delta\eta$, have been calculated. The values of V_m^E, $\Delta\kappa_S$, Δu, and $\Delta\eta$ were fitted to the Redlich−Kister polynomial equation and have been used to discuss the presence of significant interactions between amine and alcohol molecules. The viscosity data have been further used and analyzed in terms of some semiempirical equations.

INTRODUCTION

The increasing use of the alkylamine and alkanol in many industrial processes have greatly stimulated the need for an extensive information on the thermodynamic and transport properties of alkylamine, alkanol and their mixtures. The primary amine and the alkanol 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 pure state in addition to mutual association in their binaries.[1,2] Several workers[1−4] are actively engaged in the investigation of the thermodynamic, acoustic, and transport properties of binary mixtures of alkylamines and alkanols. To the best of our knowledge, no experimental values of densities, ρ, viscosities, η, and speeds of sound, u, have been reported in literature for the binary mixtures of ethylinediamine (EDA) with 2-methyl-1-propanol, or 2-propanol at different temperatures. Very recently these properties for the mixture of EDA and 1-butanol were studied by Shubha and Singh.[3] Therefore, it offers us a good opportunity to study the mixing behavior in such systems.

The objective of the present work is to provide new experimental data on densities, viscosities, speeds of sound, and derived thermodynamic properties of binary mixtures of EDA with 2-methyl-1-propanol, 2-propanol, and 1-butanol at different temperatures and at atmospheric pressure over the entire range of composition.

EXPERIMENTAL SECTION

Materials. Ethylenediamine (EDA, CAS No.: 107-15-3) with a mass fraction purity of 0.99), 2-methyl-1-propanol (with mass fraction purity of 0.99), and 1-butanol (with mass fraction purity of 0.995), were obtained from S. D. Fine Chemicals Ltd., and 2-propanol (with mass fraction purity of 0.995) was obtained from Qualigens Chemicals Limited. All of the chemicals were partially degassed before use. The purification of the liquids was checked by comparing densities, ρ, viscosities, η, and speeds of sound, u, with their corresponding literature values reported in Table 1.

Apparatus and Procedure. The binary mixture (ethylenediamine + alcohol) was prepared by weighing an appropriate amount of ethylenediamine and alcohol on an Afcoset-ER-120A electronic balance, with a precision of ± 0.05 mg, by syringing each component into airtight stoppered bottles to minimize evaporation losses. The pure components were separately degassed by vacuum pump shortly before sample preparation. The accuracy of mole fractions was $\pm 1 \cdot 10^{-4}$.

Densities, ρ, and speeds of sound, u, were measured by using a digital vibrating tube density and speed of sound analyzer (Anton Paar DSA 5000), having two integrated Pt 100 platinum thermometers with a proportional temperature controller that kept the sample at the required temperature. The calibration of the instrument has been reported in our previous paper.[5] The uncertainty in density measurement is $\pm 2 \cdot 10^{-3}$ kg·m^{-3} and for the speed of sound is ± 0.1 m·s^{-1}. The kinematic viscosities ν ($= \eta/\rho$) of pure liquids and liquid mixtures were measured using an Ubbelohde suspended level viscometer. The viscometer was calibrated[5] so as to determine the two constants C_1 and C_2 in the equation $\eta/\rho = C_1 t - C_2/t$, obtained by measuring the flow time t. The uncertainty in the viscosity measurements, based on our work on several pure liquids, was ± 0.003 mPa·s. In viscosity measurements, the temperature of the samples was controlled by using a water bath equipped with a thermostat of ± 0.01 K accuracy. The uncertainties in density, viscosity, and speed of sound are determined by comparing the experimental values of standard liquids with their literature values at that temperature.[5]

RESULTS AND DISCUSSION

The density, ρ, speed of sound, u, excess molar volume, V_m^E, and isentropic compressibility, κ_S are reported in Table 2, and the viscosity, η, is reported in Table 3. Meanwhile, deviations in

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Table 1. Experimental and Literature Values of Densities (ρ^*), Viscosities (η^*), and Speeds of Sound (u^*) of Pure Liquid Components at T = 298.15 K

components	$\rho^* \cdot 10^{-3}$/kg·m^{-3}		η^*/mPa·s		u^*/m·s^{-1}	
	exptl	lit.	exptl	lit.	exptl	lit.
ethylenediamine	0.892389	-	1.405	1.540[17]	1671.6	-
2-methyl-1-propanol	0.798104	0.7982[18]	3.307	3.333[19]	1188.8	1189.0[20]
2-propanol	0.781051	0.7811[21]	2.063	2.061[21]	1140.4	1139.3[21]
1-butanol	0.805953	0.805907[22]	2.565	2.573[23]	1241.1	1240.6[24]

isentropic compressibility, $\Delta\kappa_S$, deviations in speed of sound, Δu, and deviations in viscosity, $\Delta\eta$, for binary liquid mixtures of ($NH_2CH_2CH_2NH_2$) + 2-methyl-1-propanol ($C_4H_{10}O$), + 2-propanol (C_3H_8O), and + 1-butanol ($C_4H_{10}O$) are reported in the Supporting Information.

Excess molar volumes, deviation in speeds of sound, and deviation in isentropic compressibilities were derived respectively from the expressions:

$$V_m^E = \sum_{i=1}^{2} x_i M_i (\rho^{-1} - \rho_i^{-1}) \quad (1)$$

where ρ_i is the density, of the pure components, ρ is the density of mixtures, and x_i is the mole fraction. The isentropic compressibility, κ_S, was calculated using the Newton−Laplace equation:

$$\kappa_S = -V_m^{-1}(\delta V_m/\delta P)_S = (\rho u^2)^{-1} = V_m(Mu^2)^{-1} \quad (2)$$

V_m is the molar volume and M the molar mass of the mixture. The deviations in isentropic compressibility $\Delta\kappa_S$ from the ideal mixing values were estimated from the following equation:

$$\Delta\kappa_S = \kappa_S - (\varphi_1\kappa_{S1} + \varphi_2\kappa_{S2}) \quad (3)$$

The deviations of the speed of sound from their values in an ideal mixture were calculated from equation:

$$\Delta u = u - (\varphi_1 u_1 + \varphi_2 u_2) \quad (4)$$

where φ_1, φ_2, u_1, and u_2 are the volume fraction and speed of sound of component 1 (amine) and component 2 (alcohol), respectively.

The deviations of the viscosities from the linear dependence were calculated from the relationship:

$$\Delta\eta = \eta - \sum_{i=1}^{2} x_i\eta_i \quad (5)$$

where η and η_i are viscosities of the mixture and the pure component i, respectively.

The experimental results were fitted in the Redlich−Kister polynomial equation[6]

$$Y(x) = x_1x_2\sum_{i=1}^{p} A_i(x_1 - x_2)^i \quad (6)$$

where p is the number of estimated parameters of A_i. The standard deviation was calculated using the relation:

$$\sigma = \left[\sum_{i=1}^{n}\{Y(x)_{exptl} - Y(x)_{cal}\}^2/(n-p)\right]^{1/2} \quad (7)$$

where $Y(x)_{exptl}$, $Y(x)_{cal}$ are the values of the experimental and calculated properties (ρ, η, V_m^E), respectively, and n is the number

Table 2. Densities (ρ), Excess Molar Volumes (V_m^E), Speeds of Sound (u), and Isentropic Compressibilities (κ_S) for Binary Mixtures at (293.15 to 313.15) K

x_1	$\rho \cdot 10^{-3}$ / kg·m^{-3}	$V_m^E \cdot 10^6$ / m^3·mol^{-1}	u / m·s^{-1}	κ_S / TPa^{-1}
293.15 K				
x_1 $NH_2CH_2CH_2NH_2$ (EDA) + x_2 $C_4H_{10}O$ (2-Methyl-1-propanol)				
0.0664	0.809959	−0.3717	1236.6	809.56
0.0967	0.813410	−0.5069	1250.3	786.43
0.1652	0.821052	−0.7675	1280.8	742.49
0.2002	0.824886	−0.8789	1296.8	720.87
0.2617	0.831170	−1.0061	1324.1	686.26
0.3019	0.835134	−1.0611	1342.3	664.55
0.4093	0.844755	−1.0657	1389.6	612.98
0.5014	0.852563	−0.9798	1431.0	572.78
0.6011	0.861131	−0.8508	1478.0	531.56
0.6911	0.868086	−0.6271	1520.5	498.26
0.7887	0.876941	−0.4552	1571.6	461.66
0.8471	0.882565	−0.3562	1603.7	440.54
0.8998	0.887801	−0.2643	1634.3	421.74
0.9391	0.892054	−0.2129	1658.5	407.56
x_1 $NH_2CH_2CH_2NH_2$ (EDA) + x_2 C_3H_8O (2-Propanol)				
0.0557	0.793696	−0.2429	1187.2	893.96
0.1036	0.800705	−0.4506	1213.8	847.65
0.1560	0.807902	−0.6215	1241.5	803.06
0.2011	0.813922	−0.7432	1265.8	766.77
0.2548	0.820862	−0.8574	1295.1	726.36
0.3018	0.826514	−0.9116	1320.2	694.19
0.3892	0.836546	−0.9533	1366.4	640.23
0.5109	0.849474	−0.8906	1430.5	575.29
0.6005	0.858745	−0.8032	1478.9	532.38
0.6988	0.868400	−0.6475	1530.6	491.54
0.7969	0.878278	−0.4941	1584.3	453.60
0.8369	0.881835	−0.3901	1605.3	440.07
0.8931	0.887269	−0.2736	1635.6	421.27
0.9409	0.892340	−0.2045	1662.9	405.27
x_1 $NH_2CH_2CH_2NH_2$ (EDA) + x_2 $C_4H_{10}O$ (1-Butanol)				
0.0528	0.815585	−0.2638	1279.7	748.65
0.1135	0.822116	−0.5243	1302.1	717.38
0.1488	0.825769	−0.6477	1315.9	699.29
0.2140	0.832273	−0.8279	1341.7	667.41
0.2602	0.836674	−0.9180	1360.5	645.66
0.3286	0.842718	−0.9817	1387.1	616.76
0.3985	0.848216	−0.9566	1414.5	589.23
0.5036	0.856280	−0.8595	1456.5	550.53
0.5965	0.863365	−0.7313	1494.7	518.40
0.7081	0.872153	−0.5543	1543.9	480.97
0.7931	0.879359	−0.4102	1584.7	452.82
0.8504	0.884315	−0.3309	1613.6	434.30
0.8979	0.888772	−0.2674	1639.1	418.79
0.9360	0.892567	−0.2254	1660.5	406.31
298.15 K				
x_1 $NH_2CH_2CH_2NH_2$ (EDA) + x_2 $C_4H_{10}O$ (2-Methyl-1-propanol)				
0.0664	0.806006	−0.3707	1219.4	834.33
0.0967	0.809413	−0.5045	1233.1	812.52
0.1652	0.816964	−0.7626	1263.3	766.91
0.2002	0.820750	−0.8726	1279.2	744.51
0.2617	0.826969	−0.9990	1306.3	708.61
0.3019	0.830903	−1.0547	1324.5	685.97
0.4093	0.840455	−1.0611	1371.4	632.66

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Table 2. Continued

x_1	$\rho \cdot 10^{-3}$ / kg·m^{-3}	$V_m^E \cdot 10^6$ / m^3·mol^{-1}	u / m·s^{-1}	κ_S / TPa^{-1}
0.5014	0.848217	−0.9773	1412.2	591.11
0.6011	0.856736	−0.8507	1458.7	548.54
0.6911	0.863637	−0.6273	1500.6	514.19
0.7887	0.872420	−0.4558	1550.9	476.56
0.8471	0.878003	−0.3574	1582.6	454.69
0.8998	0.883197	−0.2659	1612.7	435.32
0.9391	0.887411	−0.2144	1636.5	420.76
x_1 $NH_2CH_2CH_2NH_2$ (EDA) + x_2 C_3H_8O (2-Propanol)				
0.0557	0.789425	−0.2413	1169.4	926.29
0.1036	0.796388	−0.4491	1195.9	877.88
0.1560	0.803523	−0.6184	1223.4	831.46
0.2011	0.809506	−0.7401	1247.6	793.56
0.2548	0.816397	−0.8535	1276.6	751.58
0.3018	0.822019	−0.9078	1301.6	717.86
0.3892	0.832023	−0.9515	1347.5	661.96
0.5109	0.844925	−0.8913	1410.9	594.53
0.6005	0.854173	−0.8050	1458.9	549.98
0.6988	0.863796	−0.6494	1510.0	507.74
0.7969	0.873662	−0.4975	1563.2	468.43
0.8369	0.877194	−0.3922	1583.7	454.49
0.8931	0.882613	−0.2757	1613.9	435.00
0.9409	0.887669	−0.2067	1640.7	418.47
x_1 $NH_2CH_2CH_2NH_2$ (EDA) + x_2 $C_4H_{10}O$ (1-Butanol)				
0.0528	0.811698	−0.2630	1262.9	772.38
0.1135	0.818144	−0.5212	1285.0	740.21
0.1488	0.821746	−0.6430	1298.6	721.61
0.2140	0.828164	−0.8210	1324.3	688.55
0.2602	0.832509	−0.9099	1342.9	666.03
0.3286	0.838245	−0.9493	1368.8	636.74
0.3985	0.843943	−0.9456	1396.3	607.76
0.5036	0.851945	−0.8545	1437.7	567.83
0.5965	0.858974	−0.7278	1475.5	534.74
0.7081	0.867696	−0.5532	1523.9	496.24
0.7931	0.874839	−0.4268	1564.1	467.26
0.8504	0.879750	−0.3313	1592.5	448.23
0.8979	0.884168	−0.2683	1617.5	432.26
0.9360	0.887932	−0.2270	1638.7	419.41
303.15 K				
x_1 $NH_2CH_2CH_2NH_2$ (EDA) + x_2 $C_4H_{10}O$ (2-Methyl-1-propanol)				
0.0664	0.802020	−0.3697	1202.4	862.42
0.0967	0.805385	−0.5022	1216.0	839.74
0.1652	0.812854	−0.7587	1246.1	792.28
0.2002	0.816592	−0.8672	1261.8	769.14
0.2617	0.822547	−0.9928	1288.6	731.92
0.3019	0.826644	−1.0483	1306.7	708.48
0.4093	0.836134	−1.0569	1353.1	653.21
0.5014	0.843853	−0.9755	1393.5	610.26
0.6011	0.852325	−0.8512	1439.5	566.22
0.6911	0.859166	−0.6276	1480.6	530.66
0.7887	0.867883	−0.4567	1530.2	492.07
0.8471	0.873420	−0.3585	1561.5	469.54
0.8998	0.878575	−0.2673	1591.2	449.53
0.9391	0.882760	−0.2164	1614.6	434.51
x_1 $NH_2CH_2CH_2NH_2$ (EDA) + x_2 C_3H_8O (2-Propanol)				
0.0557	0.785100	−0.2446	1151.7	960.23
0.1036	0.792016	−0.4492	1178.0	909.85
0.1560	0.799108	−0.6183	1205.3	861.37
0.2011	0.805044	−0.7388	1229.4	821.88
0.2548	0.811904	−0.8528	1258.2	778.04

Table 2. Continued

x_1	$\rho \cdot 10^{-3}$ / kg·m^{-3}	$V_m^E \cdot 10^6$ / m^3·mol^{-1}	u / m·s^{-1}	κ_S / TPa^{-1}
0.3018	0.817496	−0.9069	1283.0	743.13
0.3892	0.827475	−0.9524	1328.5	684.73
0.5109	0.840354	−0.8943	1391.5	614.60
0.6050	0.849588	−0.8094	1439.0	568.40
0.6988	0.859186	−0.6537	1489.5	524.63
0.7969	0.869028	−0.5018	1542.1	483.90
0.8369	0.872546	−0.3957	1562.4	469.51
0.8931	0.877945	−0.2785	1592.1	449.35
0.9409	0.882985	−0.2093	1618.7	432.21
x_1 $NH_2CH_2CH_2NH_2$ (EDA) + x_2 $C_4H_{10}O$ (1-Butanol)				
0.0528	0.807785	−0.2620	1245.9	797.51
0.1135	0.814153	−0.5184	1267.9	764.04
0.1488	0.817703	−0.6385	1281.4	744.84
0.2140	0.824038	−0.8145	1306.8	710.56
0.2602	0.828329	−0.9023	1325.3	687.29
0.3286	0.833896	−0.9307	1350.9	657.12
0.3985	0.839656	−0.9430	1378.1	627.08
0.5036	0.847599	−0.8502	1419.1	585.86
0.5965	0.854570	−0.7249	1456.3	551.78
0.7081	0.863224	−0.5521	1504.0	512.12
0.7931	0.870306	−0.4268	1543.5	482.29
0.8504	0.875173	−0.3319	1571.4	462.73
0.8979	0.879552	−0.2695	1596.1	446.31
0.9360	0.883288	−0.2290	1616.8	433.08
308.15 K				
x_1 $NH_2CH_2CH_2NH_2$ (EDA) + x_2 $C_4H_{10}O$ (2-Methyl-1-propanol)				
0.0664	0.797994	−0.3692	1185.4	891.82
0.0967	0.801327	−0.5013	1198.8	868.28
0.1652	0.808708	−0.7554	1228.7	819.02
0.2002	0.812410	−0.8636	1244.4	794.82
0.2617	0.818501	−0.9881	1271.1	756.16
0.3019	0.822366	−1.0438	1288.9	731.96
0.4093	0.831795	−1.0545	1334.9	674.65
0.5014	0.839474	−0.9753	1374.9	630.17
0.6011	0.847896	−0.8528	1420.2	584.74
0.6911	0.854681	−0.6288	1460.9	548.23
0.7887	0.863336	−0.4578	1509.8	508.14
0.8471	0.868829	−0.3604	1540.7	484.84
0.8998	0.873941	−0.2692	1570.1	464.17
0.9391	0.878094	−0.2184	1593.1	448.71
x_1 $NH_2CH_2CH_2NH_2$ (EDA) + x_2 C_3H_8O (2-Propanol)				
0.0557	0.780720	−0.2446	1134.0	995.95
0.1036	0.787599	−0.4524	1160.1	943.34
0.1560	0.794649	−0.6212	1187.3	892.68
0.2011	0.800552	−0.7416	1211.2	851.50
0.2548	0.807374	−0.8551	1239.8	805.76
0.3018	0.812943	−0.9093	1264.5	769.35
0.3892	0.822898	−0.9561	1309.7	708.48
0.5109	0.835760	−0.8999	1372.1	635.55
0.6005	0.844983	−0.8160	1419.1	587.63
0.6988	0.854563	−0.6601	1469.1	542.21
0.7969	0.864380	−0.5076	1521.1	500.01
0.8369	0.867885	−0.4005	1541.2	485.11
0.8931	0.873264	−0.2821	1570.6	464.24
0.9409	0.878288	−0.2123	1596.8	446.52
x_1 $NH_2CH_2CH_2NH_2$ (EDA) + x_2 $C_4H_{10}O$ (1-Butanol)				
0.0528	0.803849	−0.2614	1228.5	824.19
0.1135	0.810137	−0.5158	1250.7	789.05

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Table 2. Continued

x_1	$\rho \cdot 10^{-3}$ / kg·m^{-3}	$V_m^E \cdot 10^6$ / m^3·mol^{-1}	u / m·s^{-1}	κ_S / TPa^{-1}
0.1488	0.813640	−0.6345	1264.3	768.94
0.2140	0.819892	−0.8083	1289.5	733.45
0.2602	0.824133	−0.8955	1307.8	709.42
0.3286	0.829828	−0.9424	1333.6	677.53
0.3985	0.835352	−0.9369	1360.0	647.21
0.5036	0.843235	−0.8461	1400.4	604.72
0.5965	0.850157	−0.7228	1437.1	569.52
0.7081	0.858739	−0.5515	1484.2	528.65
0.7931	0.865762	−0.4273	1523.0	497.94
0.8504	0.870584	−0.3328	1550.5	477.78
0.8979	0.874924	−0.2709	1574.8	460.85
0.9360	0.878628	−0.2309	1595.3	447.23
		313.15 K		
x_1 $NH_2CH_2CH_2NH_2$ (EDA) + x_2 $C_4H_{10}O$ (2-Methyl-1-propanol)				
0.0664	0.793931	−0.3706	1168.4	922.67
0.0967	0.797223	−0.5013	1181.6	898.39
0.1652	0.804528	−0.7539	1211.5	846.87
0.2002	0.808187	−0.8609	1227.1	821.78
0.2617	0.814225	−0.9852	1253.6	781.46
0.3019	0.818055	−1.0407	1271.2	756.45
0.4093	0.827425	−1.0531	1316.9	696.91
0.5014	0.835064	−0.9758	1356.4	650.90
0.6011	0.843444	−0.8554	1400.9	604.08
0.6911	0.850175	−0.6309	1441.2	566.28
0.7887	0.858761	−0.4603	1489.6	524.80
0.8471	0.864211	−0.3619	1520.1	500.75
0.8998	0.869284	−0.2706	1548.9	479.51
0.9391	0.873407	−0.2201	1571.7	463.49
x_1 $NH_2CH_2CH_2NH_2$ (EDA) + x_2 C_3H_8O (2-Propanol)				
0.0557	0.776266	−0.2487	1116.4	1033.65
0.1036	0.783114	−0.4572	1142.2	978.74
0.1560	0.790129	−0.6261	1169.4	925.56
0.2011	0.796005	−0.7465	1193.2	882.43
0.2548	0.802798	−0.8601	1221.6	834.75
0.3018	0.808346	−0.9143	1246.1	796.72
0.3892	0.818281	−0.9621	1291.2	732.96
0.5109	0.831131	−0.9074	1353.4	656.87
0.6005	0.840349	−0.8243	1399.4	607.61
0.6988	0.849909	−0.6674	1449.1	560.34
0.7969	0.859706	−0.5138	1500.5	516.60
0.8369	0.863196	−0.4053	1520.3	501.17
0.8931	0.868560	−0.2856	1549.4	479.58
0.9409	0.873569	−0.2149	1575.4	461.23
x_1 $NH_2CH_2CH_2NH_2$ (EDA) + x_2 C_4H_8O (1-Butanol)				
0.0528	0.799876	−0.2610	1210.7	852.85
0.1135	0.806088	−0.5136	1233.6	815.18
0.1488	0.809548	−0.6313	1247.2	794.12
0.2140	0.815723	−0.8034	1272.3	757.29
0.2602	0.819910	−0.8893	1290.4	732.48
0.3286	0.825879	−0.9700	1315.8	699.32
0.3985	0.831025	−0.9315	1341.9	668.18
0.5036	0.838849	−0.8425	1381.7	624.41
0.5965	0.845718	−0.7207	1418.1	588.00
0.7081	0.854232	−0.5508	1464.4	545.87
0.7931	0.861194	−0.4274	1502.7	514.20
0.8504	0.865971	−0.3331	1529.7	493.52
0.8979	0.870277	−0.2720	1553.1	476.07
0.9360	0.873950	−0.2325	1573.7	462.03

Table 3. Viscosities (η) for Binary Mixtures at (298.15, 303.15, and 308.15) K

x_1	η/(mPa·s) 298.15 K	303.15 K	308.15 K
x_1 $NH_2CH_2CH_2NH_2$ (EDA) + x_2 $C_4H_{10}O$ (2-Methyl-1-propanol)			
0.0664	3.231	2.711	2.180
0.0967	3.209	2.699	2.169
0.1652	3.134	2.648	2.147
0.2002	3.094	2.625	2.135
0.2617	3.006	2.553	2.097
0.3019	2.940	2.502	2.059
0.4093	2.724	2.325	1.924
0.5014	2.495	2.135	1.776
0.6011	2.216	1.895	1.583
0.6911	1.958	1.698	1.446
0.7887	1.716	1.502	1.314
0.8471	1.591	1.404	1.248
0.8998	1.501	1.335	1.191
0.9391	1.456	1.294	1.154
x_1 $NH_2CH_2CH_2NH_2$ (EDA) + x_2 C_3H_8O (2-Propanol)			
0.0557	2.034	1.761	1.555
0.1036	2.017	1.746	1.543
0.1560	1.997	1.733	1.528
0.2011	1.978	1.718	1.516
0.2548	1.962	1.698	1.497
0.3018	1.940	1.675	1.480
0.3892	1.896	1.629	1.440
0.5109	1.809	1.558	1.375
0.6005	1.725	1.496	1.322
0.6988	1.637	1.429	1.266
0.7969	1.543	1.357	1.205
0.8369	1.511	1.331	1.183
0.8931	1.469	1.298	1.152
0.9409	1.438	1.271	1.129
x_1 $NH_2CH_2CH_2NH_2$ (EDA) + x_2 C_4H_8O (1-Butanol)			
0.0528	2.609	2.298	2.001
0.1135	2.591	2.300	2.009
0.1488	2.584	2.292	1.995
0.2140	2.530	2.245	1.954
0.2602	2.484	2.213	1.923
0.3286	2.411	2.145	1.864
0.3985	2.327	2.071	1.798
0.5036	2.184	1.942	1.697
0.5965	2.030	1.813	1.601
0.7081	1.842	1.648	1.471
0.7931	1.697	1.531	1.377
0.8504	1.604	1.448	1.314
0.8979	1.538	1.381	1.250
0.9360	1.481	1.334	1.198

of experimental data points. The calculated values of the coefficients, A_i, along with standard deviations, σ, are reported in Table 4.

The results obtained for viscosity of binary mixtures were also used to test the semiempirical relations of viscosity: one-parameter relations (Grunberg−Nissan,[7] Tamura−Kurata,[8]

Table 4. Coefficients A_i of Redlich−Kister eq 6 along with Standard Deviations (σ) for Binary Mixtures at Different Temperatures

T/K	A_0	A_1	A_2	A_3	A_4	σ
			$V_m^E \cdot 10^6$ (m^3·mol^{-1})			
			x_1 $NH_2CH_2CH_2NH_2$ (EDA) + x_2 $C_4H_{10}O$ (2-Methyl-1-propanol)			
293.15	−3.9777	2.8454	0.2027	−1.6794	−1.4623	0.0188
298.15	−3.9687	2.7988	0.2570	−1.6493	−1.5572	0.0189
303.15	−3.9617	2.7546	0.3098	−1.6220	−1.6472	0.0192
308.15	−3.9612	2.7156	0.3711	−1.5928	−1.7583	0.0194
313.15	−3.9641	2.6746	0.4139	−1.5430	−1.8443	0.0197
			x_1 $NH_2CH_2CH_2NH_2$ (EDA) + x_2 C_3H_8O (2-Propanol)			
293.15	−3.6307	1.7531	−0.4722	−1.0474	-	0.0155
298.15	−3.6299	1.7161	−0.4710	−1.0359	-	0.0159
303.15	−3.6387	1.6751	−0.4835	−0.9911	-	0.0159
308.15	−3.6590	1.6487	−0.5002	−0.9653	-	0.0164
313.15	−3.6868	1.6244	−0.5227	−0.9123	-	0.0166
			x_1 $NH_2CH_2CH_2NH_2$ (EDA) + x_2 C_4H_8O (1-Butanol)			
293.15	−3.4905	2.4717	−0.4986	−2.0877	−0.7800	0.0162
298.15	−3.4533	2.5882	−0.5868	−1.8875	−0.7467	0.0156
303.15	−3.4302	2.5141	−0.5349	−1.8020	−0.8799	0.0170
308.15	−3.4226	2.5291	−0.5366	−1.8859	−0.8727	0.0155
313.15	−3.4234	2.5965	−0.5637	−2.0583	−0.7934	0.0179
			$\Delta\kappa_S$ (TPa^{-1})			
			x_1 $NH_2CH_2CH_2NH_2$ (EDA) + x_2 $C_4H_{10}O$ (2-Methyl-1-propanol)			
293.15	−35.0029	7.7412	−1.8290	−2.1095	−3.6364	0.0437
298.15	−36.4505	8.0653	−1.9434	−2.1097	−3.6799	0.0471
303.15	−37.9526	8.5560	−1.9460	−2.4164	−3.7844	0.0470
308.15	−39.5416	9.0988	−2.2409	−3.2459	−3.4218	0.0524
313.15	−41.2990	9.5828	−2.1305	−3.7826	−4.0317	0.0493
			x_1 $NH_2CH_2CH_2NH_2$ (EDA) + x_2 C_3H_8O (2-Propanol)			
293.15	−43.0252	11.5773	−3.5962	−1.2568	-	0.0526
298.15	−45.8291	12.1297	−3.7252	−1.4721	-	0.0578
303.15	−47.1927	12.8468	−3.9570	−1.3312	-	0.0586
308.15	−49.5851	13.5686	−4.4025	−1.1710	-	0.0571
313.15	−52.4449	14.4449	−4.9603	−1.0853	-	0.0514
			x_1 $NH_2CH_2CH_2NH_2$ (EDA) + x_2 C_4H_8O (1-Butanol)			
293.15	−25.0682	5.5059	−2.7355	−3.3423	-	0.0543
298.15	−26.1027	5.3854	−2.7194	−2.2093	-	0.0650
303.15	−27.1764	5.7122	−2.9782	−2.2390	-	0.0652
308.15	−28.3123	6.2411	−2.8716	3.0011	-	0.0565
313.15	−29.5994	6.9690	−3.1465	−4.1678	-	0.0290
			Δu (m·s^{-1})			
			x_1 $NH_2CH_2CH_2NH_2$ (EDA) + x_2 $C_4H_{10}O$ (2-Methyl-1-propanol)			
293.15	79.3131	−46.9868	−23.4379	27.7972	48.4410	0.4844
298.15	81.0906	−45.5366	−23.9274	28.2407	47.8785	0.4769
303.15	82.7518	−46.3050	−25.3757	29.4124	49.4850	0.5017
308.15	84.3923	−46.8581	−25.9560	35.8531	53.1447	0.4931
313.15	86.4210	−47.3483	−25.1118	42.9559	52.8930	0.4759
			x_1 $NH_2CH_2CH_2NH_2$ (EDA) + x_2 C_3H_8O (2-Propanol)			
293.15	67.9048	−10.5830	0.5622	18.9189	-	0.5238
298.15	69.3888	−10.5378	−0.3341	18.7975	-	0.5413
303.15	71.3168	−10.3637	−0.2887	19.1060	-	0.5394
308.15	73.4490	−10.6521	2.0245	21.0924	-	0.5045
313.15	77.8678	−10.5914	3.6523	22.3098	-	0.5158
			x_1 $NH_2CH_2CH_2NH_2$ (EDA) + x_2 C_4H_8O (1-Butanol)			
293.15	49.6001	−42.9766	2.4562	33.7307	-	0.5407
298.15	51.9780	−39.0844	−0.0653	19.9721	-	0.5868
303.15	54.4182	−38.7096	1.0083	19.1492	-	0.5790
308.15	56.8469	−39.6957	−1.1393	22.6872	-	0.5457
313.15	59.4387	−42.9529	1.8920	34.5491	-	0.2185
			$\Delta\eta$ (mPa·s)			
			x_1 $NH_2CH_2CH_2NH_2$ (EDA) + x_2 $C_4H_{10}O$ (2-Methyl-1-propanol)			
298.15	0.5478	−1.5413	−0.9790	0.5446	-	0.0035
303.15	0.4921	−1.4231	−0.7227	0.8288	-	0.0049
308.15	0.6126	−1.3253	0.0038	0.2224	-	0.0095
			x_1 $NH_2CH_2CH_2NH_2$ (EDA) + x_2 C_3H_8O (2-Propanol)			
298.15	0.2943	−0.1353	−0.6150	−0.3346	0.7722	0.0195
303.15	0.2487	−0.2307	−0.1362	−0.0900	-	0.0014
308.15	0.2907	0.0756	−0.2724	−0.4067	-	0.0067
			x_1 $NH_2CH_2CH_2NH_2$ (EDA) + x_2 C_4H_8O (1-Butanol)			
298.15	1.0952	−1.9010	1.3484	0.0889	−0.8901	0.0114
303.15	0.7577	−0.5721	0.0328	−0.2290	0.4107	0.0032
308.15	0.6617	−0.3217	0.3366	−0.3336	0.3975	0.0033

Hind et al.,[9] and Katti−Chaudhry[10]) and the three-parameter Heric−Brewer relation.[11]

(1) Grunberg and Nissan suggested a logarithmic relation between the viscosity of a liquid mixture and that of its pure components:

$$\eta = \exp\left[\sum_{i=1}^{2}(x_i \ln \eta_i) + G_{12}\prod_{i=1}^{2} x_i\right] \quad (8)$$

where G_{12} is a constant that is regarded as a measure of the strength of the molecular interactions between the mixing components and is a parameter proportional to the interchange energy.

(2) Tamura and Kurata developed the following equation for the viscosity of binary liquid mixtures:

$$\eta = \sum_{i=1}^{2} x_i\varphi_i\eta_i + 2T_{12}\prod_{i=1}^{2}(x_i\varphi_i)^{1/2} \quad (9)$$

where T_{12} is an adjustable parameter.

(3) Hind et al. suggested the following equation for the viscosity of binary liquid mixtures:

$$\eta = \sum_{i=1}^{2} x_i^2\eta_i + 2H_{12}\prod_{i=1}^{2} x_i \quad (10)$$

where H_{12} is the interaction parameter.

(4) Katti and Chaudhry derived the following equation:

$$\ln \eta V_m = \sum_{i=1}^{2} x_i \ln(\eta_i V_i) + x_1x_2(W_{vis}/RT) \quad (11)$$

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Table 5. Values of the Interaction Parameters along with the Percent Standard Deviations for Binary Mixtures at Different Temperatures

semiempirical relations	parameter	σ %
x_1 $NH_2CH_2CH_2NH_2$ (EDA) + x_2 $C_4H_{10}O$ (2-Methyl-1-propanol)		
	298.15 K	
Grunberg−Nissan	G_{12} = 0.4728	4.1749
Tamura−Kurata	T_{12} = 2.3379	5.3385
Hind et al.	H_{12} = 0.5480	9.3566
Katti−Chaudhry	W_{vis}/RT = 1.8649	39.4495
Heric−Brewer (three-parameter)	a = 0.500; b = −0.400; c = −0.600	0.0051
	303.15 K	
Grunberg−Nissan	G_{12} = 0.4774	3.6948
Tamura−Kurata	T_{12} = 1.9924	4.9494
Hind et al.	H_{12} = 2.0919	4.8518
Katti−Chaudhry	W_{vis}/RT = 1.8091	37.3121
Heric−Brewer (three-parameter)	a = −0.520; b = −0.430; c = −0.530	0.0166
	308.15 K	
Grunberg−Nissan	G_{12} = 0.5599	4.5732
Tamura−Kurata	T_{12} = 1.7798	5.9589
Hind et al.	H_{12} = 1.8523	5.6957
Katti−Chaudhry	W_{vis}/RT = 1.8724	33.9015
Heric−Brewer (three-parameter)	a = 0.570; b = −0.540; c = −0.210	0.0076
x_1 $NH_2CH_2CH_2NH_2$ (EDA) + x_2 C_3H_8O (2-Propanol)		
	298.15 K	
Grunberg−Nissan	G_{12} = −1.2511	15.0873
Tamura−Kurata	T_{12} = 2.7513	13.8631
Hind et al.	H_{12} = 1.8498	1.2468
Katti−Chaudhry	W_{vis}/RT = 1.4223	16.8160
Heric−Brewer (three-parameter)	a = 0.200; b = −0.018; c = 0.210	0.0238
	303.15 K	
Grunberg−Nissan	G_{12} = −2.1408	17.6277
Tamura−Kurata	T_{12} = 2.3674	11.2832
Hind et al.	H_{12} = 1.5096	1.7185
Katti−Chaudhry	W_{vis}/RT = 1.2700	15.5256
Heric−Brewer (three-parameter)	a = 0.050; b = −0.150; c = −0.064	0.0109
	308.15 K	
Grunberg−Nissan	G_{12} = −2.0695	17.0497
Tamura−Kurata	T_{12} = 2.1925	10.1572
Hind et al.	H_{12} = 1.4311	1.2577
Katti−Chaudhry	W_{vis}/RT = 1.4644	16.0233
Heric−Brewer (three-parameter)	a = −0.008; b = 0.190; c = −0.240	0.1074
x_1 $NH_2CH_2CH_2NH_2$ (EDA) + x_2 C_4H_8O (1-Butanol)		
	298.15 K	
Grunberg−Nissan	G_{12} = 0.8527	6.8293
Tamura−Kurata	T_{12} = 2.4508	7.0919
Hind et al.	H_{12} = 2.5115	6.5069
Katti−Chaudhry	W_{vis}/RT = 1.9810	30.3343
Heric−Brewer (three-parameter)	a = 0.690; b = 0.560; c = 0.540	0.0099
	303.15 K	
Grunberg−Nissan	G_{12} = 0.7172	4.6058
Tamura−Kurata	T_{12} = 2.0807	3.1805
Hind et al.	H_{12} = 2.4529	12.7113
Katti−Chaudhry	W_{vis}/RT = 0.5938	30.8810
Heric−Brewer (three-parameter)	a = −0.640; b = −0.170; c = −0.079	0.0062
	308.15 K	
Grunberg−Nissan	G_{12} = 0.7286	6.3367
Tamura−Kurata	T_{12} = 1.8211	2.6198
Hind et al.	H_{12} = 1.9037	2.1479
Katti−Chaudhry	W_{vis}/RT = 0.5768	29.4395
Heric−Brewer (three-parameter)	a = 0.180; b = 0.090; c = 0.320	0.0066

where W_{12} is the interaction term and V_i is the molar volume of pure component i.

(5) The Heric and Brewer (three-parameter) model is as follows:

$$\ln \nu = x_1 \ln \nu_1 + x_2 \ln \nu_2 + x_1 \ln M_1 + x_2 \ln M_2 - \ln(x_1 M_1 + x_2 M_2) + x_1 x_2 [a + b(x_1 - x_2) + c(x_1 - x_2)^2] \quad (12)$$

where a, b, and c are adjustable parameters.

The correlating ability of each of the eqs 8 to 12 was tested, and their adjustable parameters and percent standard deviations, σ %, calculated using eq 13 are given in Table 5.

$$\sigma\% = \left[100/(n-p) \sum_{i=1}^{2} ((\eta_{exptl} - \eta_{cal})/\eta_{exptl})^2 \right]^{1/2} \quad (13)$$

where n represents the number of data points and p is the number of adjustable parameters.

Figures 1 to 3 show that the values of excess molar volume of the binary liquid mixtures of ethylenediamine with 2-methyl-1-propanol, 2-propanol, and 1-butanol are negative over the whole composition ranges at all of the studied temperatures. The negative trend in the values of V_m^E is observed in all of the three binary mixtures with minima at $x_1 \approx 0.4$. The negative V_m^E values show the presence of strong intermolecular forces of attraction. The present results can be interpreted qualitatively by taking into account the fact that several expansion and contraction processes proceed simultaneously when amine−alkanol mixtures are formed. The following effects[1] can be considered: (i) expansion due to depolymerization of alcohol and amine by one another, (ii) contraction due to free volume difference of unlike molecules, and (iii) contraction due to hydrogen bond formation between amine and alcohol through $NH_2 \cdots OH$ and $OH \cdots NH_2$. This interaction can be considered the reaction between alkanol as a Lewis acid and amine as a Lewis base. There is no significant change observed in the values of V_m^E with temperature as noted by others.[12−14] Further, it is also observed from

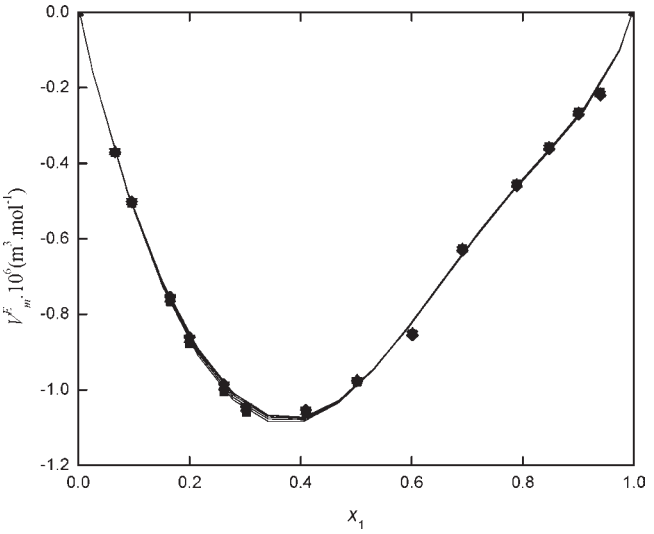

Figure 1. Excess molar volumes against mole fractions x_1 $C_2H_8N_2 + x_2$ $C_4H_{10}O$ (2-methyl-1-propanol) at ■, T = 293.15 K; ●, T = 298.15 K; ▲, T = 303.15 K; ▼, T = 308.15 K; ♦, T = 313.15 K. The solid curves have been derived from eq 6.

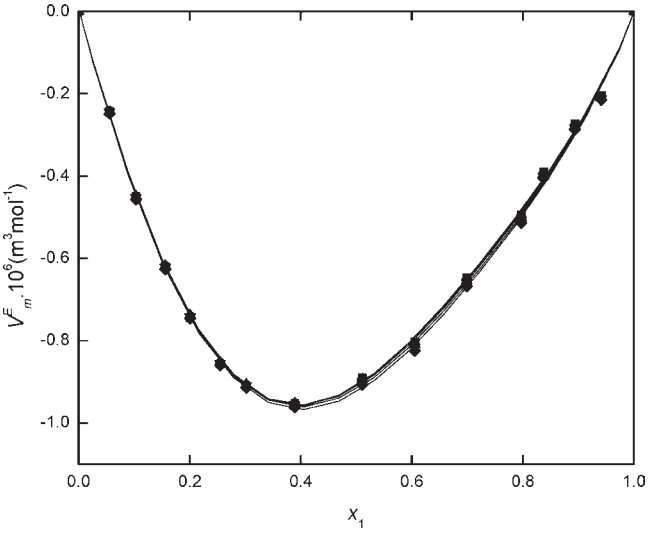

Figure 2. Excess molar volumes against mole fractions x_1 $C_2H_8N_2 + x_2$ C_3H_8O (2-propanol) at ■, T = 293.15 K; ●, T = 298.15 K; ▲, T = 303.15 K; ▼, T = 308.15 K; ♦, T = 313.15 K. The solid curves have been derived from eq 6.

Figures 1 to 3 that as we move from 2-methyl-1-propanol to 1-butanol to 2-propanol the negative values of V^E_m decrease. This shows that there are strong forces of attraction in 2-methyl-1-propanol among the three alcohols. The large negative value of V^E_m for the mixture with 2-methyl-1-propanol indicates that a most efficient packing of molecules occurs in this mixtures. The values of viscosities, η, of pure liquids and their binaries, decreases with composition as well as with increasing temperature, whereas the values of κ_S decreases with composition and increases with temperature.

The values of $\Delta\kappa_S$ and Δu are plotted in Figures 4 and 5, respectively (given in the Supporting Information). The deviations in isentropic compressibility, $\Delta\kappa_S$, show a negative trend for all of the studied systems over the entire composition range.

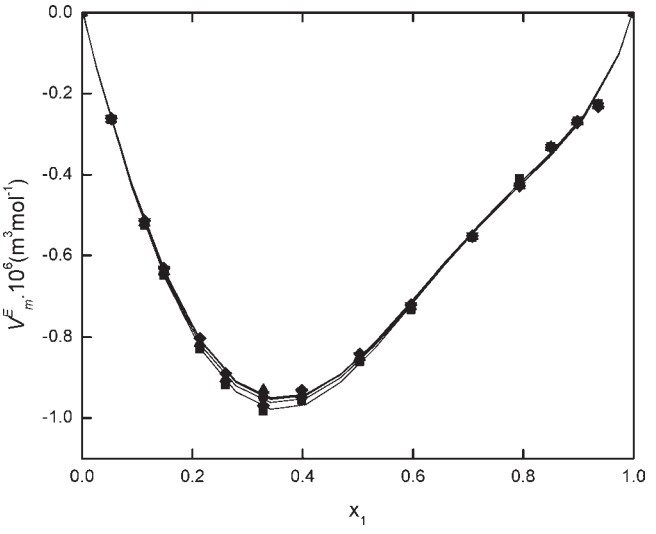

Figure 3. Excess molar volumes against mole fractions x_1 $C_2H_8N_2 + x_2$ $C_4H_{10}O$ (1-butanol) at ■, T = 293.15 K; ●, T = 298.15 K; ▲, T = 303.15 K; ▼, T = 308.15 K; ♦, T = 313.15 K. The solid curves have been derived from eq 6.

The negative $\Delta\kappa_S$ values for the studied binary mixtures shows the predominance of strong forces in these mixtures, and the mixture is less compressible than the corresponding ideal mixture. In these binary mixtures the contraction in free volume makes the mixture less compressible than ideal mixtures. The trend can be seen graphically from Figure 4 of the SI. The plot of deviation in speeds of sound, Δu, shows a positive trend for all of the studied systems over the entire composition range. The effect of temperature on Δu is not so high, which shows a slight increase in its value with temperature.

The variation of the viscosity deviations, $\Delta\eta$, with the mole fraction, x_1, for the binary mixtures is presented in Figure 6 (reported in the Supporting Information). The $\Delta\eta$ values are positive for all systems but change sign from positive to negative in case of 2-methy-1-propanol and 2-propanol only at higher mole fractions. Positive values of $\Delta\eta$ are indicative of strong interactions, whereas negative values indicate weaker interactions.[15] The $\Delta\eta - x_1$ curves show a maximum positive value at $x_1 \approx 0.30$ for 2-methyl-1-propanol and 1-butanol while at $x_1 \approx$ 0.4 for 2-propanol. The positive values of viscosity deviations further support the presence of strong forces between the mixing components in these binary mixtures. Also a decrease in the magnitude of $\Delta\eta$ is observed with the increase in the temperature for each binary liquid mixture under study.

To perform a numerical comparison of the correlating capabilities of these relations, we calculated the percentage standard deviation, σ %, using the relation 13. Table 5 shows the calculated parameters and the σ % between experimental values obtained for viscosities and the predicted results using the semiempirical relations. As can be clearly seen by the results reported in Table 5 for one-parameter relations, the Grunberg−Nissan relation represents the behavior of the binary mixtures satisfactorily as compared to other equations, and the Katti−Chaudhry relation gives the maximum σ %. Further, the interaction parameter, G_{12}, gives an idea about quantitative estimation of interactions in the mixtures. It is well-known that, if $G_{12} > 0$ and higher in magnitude, there will be strong specific interactions in the mixture, and if $G_{12} < 0$, it indicates the presence of weak

interactions.[16] It is clear from Table 5 that σ % values for the three-parameter relation Heric−Brewer are in the range (0.0062 to 0.1074) %. The relation predicts the viscosity data almost equally well for all of the three mixtures investigated. The observed σ % for studied binary systems indicates that three-parameter relations have best capability of reproducing the viscosity data with lowest σ %. Thus, it can be concluded that, as the number of adjustable parameters in the correlating equation increases, the predicting ability also increases.

CONCLUSIONS

In this paper, an attempt has been made to measure densities and speeds of sound at (293.15, 298.15, 303.15, 308.15, and 313.15) K and viscosities at (298.15, 303.15, and 308.15) K over the entire range of mixture composition of ethylenediamine with 2-methyl-1-propanol, 2-propanol, and 1-butanol. Out of these measured data, the excess molar volumes, deviations in isentropic compressibility, deviations in speeds of sound, and deviations in viscosity have been calculated and correlated by a Redlich−Kister type polynomial equation to derive the coefficients and standard deviations. Negative deviations are observed in the case of V_m^E, for all three binary mixtures at all studied temperatures. From the results it is concluded that the strength of bonding is expected to decrease as we move from 2-methyl-1-propanol to 1-butanol to 2-propanol, and the negative values of V_m^E decrease. The present results of V_m^E support this fact. The negative values of $\Delta\kappa_S$ and positive values of $\Delta\eta$ also support the results. The results obtained for the viscosity of binary mixtures used to test the semiempirical Grunberg−Nissan, Tamura−Kurata, Hind et al., Katti−Chaudhry, and Heric−Brewer relations of viscosity shows that the three-parameter relation, that is, Heric−Brewer, gives the best results.

ASSOCIATED CONTENT

Supporting Information. Tables: deviations in isentropic compressibilities and speeds of sound for binary mixtures at (293.15 to 313.15) K and deviations in viscosity for binary mixtures at (298.15, 303.15, and 308.15) K. Figures show the deviations in isentropic compressibility vs mole fraction, deviations in speed of sound vs mole fraction, and deviations in viscosity vs mole fraction. This material is available free of charge via the Internet at http://pubs.acs.org.

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