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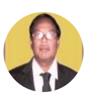

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Viscosity, Density, and Speed of Sound for the Binary Mixtures of Formamide with 2-Methoxyethanol, Acetophenone, Acetonitrile, 1,2-Dimethoxyethane, and Dimethylsulfoxide at Different Temperatures

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Densities and viscosities of binary liquid mixtures of formamide with 2-methoxyethanol, acetophenone, acetonitrile, 1,2-dimethoxyethane, and dimethylsulfoxide have been measured at (298.15, 308.15, and 318.15) K and over the entire composition range at $p = 0.1$ MPa. Ultrasonic speeds of these binary liquid mixtures have also been measured at 298.15 K and the same pressure. From the experimental data, values of excess molar volume V_m^E, viscosity deviation $\Delta\eta$, and deviation in isentropic compressibility ΔK_S have been determined. These results were fitted to the Redlich−Kister-type polynomial equation. The density and viscosity data have been analyzed in terms of some semiempirical viscosity models.

Introduction

The derived deviation or excess properties from the experimental measurements of density, viscosity, and ultrasonic speed provide valuable information that allows us to have a better understanding of the structure of liquids and intermolecular interactions in liquid mixtures.[1−3] Formamide and its derivatives are good solvents for many organic and inorganic compounds and are also used as a plasticizer. Acetophenone, a typical ketone, has been used in perfumery and as a hypnotic under the name “hypnone”. It is also used as a solvent for cellulose ethers. Acetonitrile is widely used for dissolving inorganic and organic compounds. 1,2-Dimethoxyethane is used in selected reactions as an alternative to diethyl ether. Dimethylsulfoxide is a powerful broad spectrum solvent for a wide variety of inorganic and organic reactants. Having low toxicity, it can be used in biology and medicine, especially for low-temperature preservation.[4] 2-Methoxyethanol finds a wide range of applications of technological importance, namely, as solvents and solubilizing agents in organic synthesis, reaction kinetics, and electrochemical studies.[5] Hence, we report in this paper the experimental values of densities ρ and viscosities η for the binary mixtures of formamide with 2-methoxyethanol, acetophenone, acetonitrile, 1,2-dimethoxyethane, and dimethylsulfoxide over the entire range of compositions at $T = (298.15, 308.15, \text{and } 318.15)$ K and $p = 0.1$ MPa. Also, the experimental values of ultrasonic speeds of sound u have been reported at $T = 298.15$ K and at the same pressure. From the experimental values of density ρ, viscosity η, and ultrasonic speed of sound u, the values of excess molar volume V_m^E, viscosity deviation $\Delta\eta$, and deviation in isentropic compressibility ΔK_S have been calculated. This work also provides a test of various empirical equations to correlate viscosity and acoustic data of binary mixtures in terms of pure component properties.

Experimental Section

Materials. Extrapure grade formamide, 2-methoxyethanol, acetophenone, acetonitrile, 1,2-dimethoxyethane, and dimethylsulfoxide were procured from S.d.fine-Chem Limited, Mumbai, India. 2-Methoxyethanol was dried with potassium carbonate and distilled twice in an all-glass distillation set immediately before use, and the middle fraction was collected.[5] Acetophenone was dried over anhydrous potassium carbonate for 3 days, filtered, and then distilled. The middle fraction of the distillates was retained.[6] Acetonitrile was distilled from P_2O_5 and then from CaH_2 in an all-glass distillation apparatus.[7] The middle fraction was collected. 1,2-Dimethoxyethane was purified by double-fractional distillation over $LiAlH_4$ to eliminate traces of acids, peroxides, and water, and the middle portion was collected for the preparation of mixtures.[8] Formamide and dimethylsulfoxide were purified according to the standard procedures.[9] All solvents, pure or mixed, were stored over 3 Å molecular sieves for 3 days before use. The purity of the purified liquids was ascertained by GLC and also by comparing experimental values of densities, viscosities, and ultrasonic speeds of sound with their literature values,[5−7,10−17] whenever available as listed in Table 1.

Apparatus and Procedure. The mixtures were prepared by mixing the required volume of pure liquids in airtight stoppered bottles with adequate precautions to reduce evaporation losses during mixing. The densities were measured with an Ostwald−Sprengel-type pycnometer having a bulb volume of 25 cm^3 and a capillary of internal diameter of about 0.1 cm. The pycnometer was calibrated at the experimental temperatures with doubly distilled water. The pycnometer with the test solution was equilibrated in a thermostatic water bath maintained at ± 0.01 K of the desired temperature by means of a mercury-in-glass thermoregulator. The pycnometer was then removed from the thermostatic bath, properly dried, and weighed in a digital electronic analytical balance (Mettler, AG 285, Switzerland). The mass measurements were accurate to ± 0.01 mg. The evaporation losses were insignificant during the time of actual measurements, and averages of triplicate measurements were taken into account.

The viscosity was measured by means of a suspended Ubbelohde-type viscometer, calibrated at the experimental temperatures with triply distilled water and purified methanol

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Table 1. Densities ρ, Viscosities η, and Speeds of Sound u of the Pure Liquids at Different Temperatures

		ρ/(kg·m^{-3})		η/(mPa·s)		u/(m·s^{-1})	
pure liquids	T/K	exptl	lit.	exptl	lit.	exptl	lit.
formamide	298.15	1129.2	1129.2[10]	3.302	3.302[10]	1591.3	–
	308.15	1120.5	1120.2[11]	2.542	2.59[11]		1580[12]
	318.15	1111.1	1111.9[13]	2.001	2.00[13]		
2-methoxyethanol	298.15	959.7	959.79[5]	1.543	1.543[5]	1339.4	1339.31[5]
	308.15	951.5	952.51[5]	1.257	1.257[5]		
	318.15	945.9	946.23[5]	1.051	1.050[5]		
acetophenone	298.15	1023.1	1023.1[6]	1.653	1.652[6]	1296.4	–
	308.15	1014.8	–	1.412	–		
	318.15	1007.5	–	1.117	–		
acetonitrile	298.15	776.0	776.86[7]	0.344	0.3446[7]	1260.2	–
	308.15	766.3	765.64[7]	0.313	0.3125[7]		1237.2[14]
	318.15	755.0	754.98[7]	0.289	0.2893[7]		
1,2-dimethoxyethane	298.15	861.5	861.09[15]	0.409	0.4089[15]	1146.2	1146.0[16]
	308.15	850.7	850.01[15]	0.366	0.3659[15]		
	318.15	839.6	838.85[15]	0.330	0.3302[15]		
dimethylsulfoxide	298.15	1095.4	1095.4[17]	2.042	2.0418[17]	1493.0	1493.0[17]
	308.15	1085.9	1085.9[17]	1.568	1.5682[17]		
	318.15	1076.9	1076.7[17]	1.485	1.4847[17]		

using density and viscosity values from the literature.[18−20] The flow times were accurate to ± 0.1 s.

The ultrasonic speeds of sound were determined using a multifrequency ultrasonic interferometer (Mittal Enterprises, New Delhi, India) working at 2 MHz and calibrated with purified water and methanol at 298.15 K. The temperature was maintained within ± 0.01 K by circulating thermostated water around the cell with the aid of a circulating pump. The uncertainties in the mole fraction, density, viscosity, and ultrasonic speed of sound measurements were estimated to be ± 0.0002, ± 3·10^{-4} g·cm^{-3}, ± 0.003 mPa·s, and ± 0.2 m·s^{-1}. The details of the methods and techniques of the measurements have been described earlier.[2,3,21−23]

Results and Discussion

The experimental densities, viscosities, excess molar volumes, and viscosity deviations for the binary mixtures studied at (298.15, 308.15, and 318.15) K and p = 0.1 MPa as a function of mole fraction x_1 are listed in Table 2.

Excess Molar Volume. The excess molar volume, V_m^E, was calculated using the relation[24]

$$V_m^E = (x_1M_1 + x_2M_2)/\rho - x_1M_1/\rho_1 - x_2M_2/\rho_2 \qquad (1)$$

where x_1, x_2; ρ_1, ρ_2; and M_1, M_2 are the mole fractions, densities, and molecular weights of pure components 1 (formamide) and 2 of the binary liquid mixtures, respectively.

The values of excess molar volume, V_m^E, have been represented in Table 2. The estimated uncertainty for excess molar volume, V_m^E, is ± 0.005·10^6 m^3·mol^{-1}. A perusal of Table 2 shows that the values of V_m^E are negative for all the binary mixtures over the entire range of compositions and temperatures. The negative values of excess molar volume for the three systems are in the order: formamide + 1,2-dimethoxyethane > formamide + acetonitrile > formamide + 2-methoxyethanol > formamide + dimethylsulfoxide > formamide + acetophenone.

The plots of excess molar volume, V_m^E, vs mole fraction of formamide, x_1, at 298.15 K are presented in Figure 1.

The negative values of excess molar volume, V_m^E, suggest specific interactions[24] between the mixing components in the mixtures, whereas its positive values suggest dominance of dispersion forces[26,27] between them. The negative V_m^E values indicate the specific interactions such as intermolecular hydrogen bonding between the mixing components and also interstitial accommodation of the mixing components because of the difference in molar volumes. The negative V_m^E values may also be due to the difference in the dielectric constants of the liquid components of the binary mixtures.[25] The negative V_m^E values for all the systems studied may be attributed to dipole-induced dipole interactions between the component liquids of the mixtures resulting in the formation of electron donor−acceptor complexes.[28]

Thus, it is seen that the strength of interaction between the mixing components follows the order: formamide + 1,2-dimethoxyethane > formamide + acetonitrile > formamide + 2-methoxyethanol > formamide + dimethylsulfoxide > formamide + acetophenone.

A perusal of Table 2 also shows that the V_m^E values become more negative as the temperature is increased from (298.15 to 318.15) K. This indicates a gradually decreasing trend in the degree of the inter hydrogen bonds in the associated formamide molecules as the experimental temperature increases and dipole−dipole interactions between the hetero molecules are increased leading to greater contraction in the mixture volumes.[14]

Viscosity Deviation. The viscosity deviation, $\Delta\eta$, was calculated by using the relation[24]

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

where η is the viscosity of the binary mixtures. x_1,x_2 and η_1,η_2 are the mole fractions and viscosities of pure components 1 (formamide) and 2 of the binary liquid mixtures, respectively. The estimated uncertainty for viscosity deviation, $\Delta\eta$, is ± 0.004 mPa·s.

A perusal of Table 2 shows that the values of viscosity deviation, $\Delta\eta$, are positive for all the studied binary mixtures except those of 1,2-dimethoxyethane and acetonitrile over the entire composition range and at all the experimental temperatures, and they have been presented in Figure 2 as a function of the mole fraction of formamide at 298.15 K. The negative values imply the presence of dispersion forces[29] between the mixing components in the mixtures, and the positive values may be attributed to the presence of specific interactions[29] between them.

It is also evident from Table 2 that $\Delta\eta$ values of the mixtures decrease as the experimental temperatures increases, thereby suggesting an increase in the fluidity of the mixtures.[30]

Viscosity Models and Interaction Parameters. Several semiempirical models have been proposed to estimate the

Table 2. Values of Density ρ, Viscosity η, Excess Molar Volume V_m^E, Viscosity Deviation $\Delta\eta$, and Various Interaction Parameters d_{12}, T_{12}, and H_{12} for the Binary Mixtures of Formamide from T = (298.15 to 308.15) K as a Function of Mole Fraction x_1 at p = 0.1 MPa

Formamide (1) + 2-Methoxyethanol (2)

x_1	ρ / kg·m^{-3}	η / mPa·s	$V_m^E \cdot 10^6$ / m^3·mol^{-1}	$\Delta\eta$ / mPa·s	d_{12}	T_{12}	H_{12}	x_1	ρ / kg·m^{-3}	η / mPa·s	$V_m^E \cdot 10^6$ / m^3·mol^{-1}	$\Delta\eta$ / mPa·s	d_{12}	T_{12}	H_{12}
T = 298.15 K															
0.0000	959.7	1.543	0.000	0.000	–	–	–	0.7171	1060.2	3.331	−0.265	0.527	1.104	4.053	3.721
0.1581	977.0	1.912	−0.200	0.091	0.707	3.319	2.764	0.7977	1076.7	3.395	−0.191	0.449	1.126	4.058	3.813
0.2969	994.2	2.291	−0.325	0.226	0.812	3.525	2.964	0.8711	1093.8	3.399	−0.129	0.324	1.133	4.030	3.868
0.4199	1010.9	2.659	−0.368	0.377	0.922	3.738	3.196	0.9383	1111.1	3.348	−0.057	0.155	1.051	3.881	3.759
0.5297	1027.6	2.963	−0.374	0.488	1.001	3.892	3.402	1.0000	1129.2	3.302	0.000	0.000	–	–	–
0.6282	1044.1	3.173	−0.339	0.525	1.040	3.965	3.546								
T = 308.15 K															
0.0000	951.5	1.257	0.000	0.000	–	–	–	0.7171	1052.3	2.485	−0.300	0.307	0.871	2.925	2.656
0.1581	969.1	1.515	−0.224	0.055	0.567	2.507	2.106	0.7977	1068.7	2.541	−0.220	0.259	0.880	2.921	2.702
0.2969	986.2	1.759	−0.351	0.121	0.609	2.587	2.189	0.8711	1085.7	2.562	−0.153	0.186	0.879	2.900	2.728
0.4199	1002.8	1.997	−0.390	0.200	0.685	2.696	2.309	0.9383	1102.9	2.550	−0.076	0.088	0.807	2.808	2.656
0.5297	1019.4	2.206	−0.395	0.268	0.759	2.797	2.437	1.0000	1120.5	2.542	0.000	0.000	–	–	–
0.6282	1035.9	2.367	−0.361	0.303	0.816	2.867	2.548								
T = 318.15 K															
0.0000	945.9	1.051	0.000	0.000	–	–	–	0.7171	1045.3	1.929	−0.333	0.197	0.718	2.223	2.012
0.1581	963.5	1.225	−0.248	0.024	0.387	1.899	1.616	0.7977	1061.5	1.978	−0.261	0.169	0.735	2.228	2.049
0.2969	980.3	1.394	−0.373	0.061	0.437	1.958	1.672	0.8711	1078.1	2.016	−0.189	0.137	0.804	2.271	2.136
0.4199	996.5	1.561	−0.407	0.111	0.514	2.037	1.754	0.9383	1094.6	2.003	−0.104	0.061	0.707	2.179	2.053
0.5297	1012.8	1.709	−0.410	0.155	0.583	2.106	1.837	1.0000	1111.1	2.001	0.000	0.000	–	–	–
0.6282	1029.1	1.831	−0.388	0.183	0.644	2.162	1.918								

Formamide (1) + Acetophenone (3)

x_1	ρ / kg·m^{-3}	η / mPa·s	$V_m^E \cdot 10^6$ / m^3·mol^{-1}	$\Delta\eta$ / mPa·s	d_{12}	T_{12}	H_{12}	x_1	ρ / kg·m^{-3}	η / mPa·s	$V_m^E \cdot 10^6$ / m^3·mol^{-1}	$\Delta\eta$ / mPa·s	d_{12}	T_{12}	H_{12}
T = 298.15 K															
0.0000	1023.1	1.653	0.000	0.000	–	–	–	0.8001	1084.4	3.819	−0.009	0.847	1.775	5.457	5.125
0.2286	1032.9	2.234	−0.012	0.204	0.811	4.157	3.056	0.8616	1095.3	3.735	−0.006	0.662	1.838	5.367	5.253
0.4001	1042.9	2.795	−0.021	0.482	1.034	4.582	3.481	0.9143	1106.4	3.590	−0.004	0.430	1.827	5.180	5.222
0.5334	1053.1	3.267	−0.023	0.735	1.255	4.955	3.954	0.9600	1117.7	3.439	−0.003	0.204	1.788	4.983	5.134
0.6401	1063.3	3.596	−0.019	0.888	1.452	5.223	4.405	1.0000	1129.2	3.302	0.000	0.000	–	–	–
0.7273	1073.8	3.778	−0.013	0.926	1.631	5.396	4.812								
T = 308.15 K															
0.0000	1014.8	1.412	0.000	0.000	–	–	–	0.8001	1076.3	2.779	−0.031	0.463	1.292	3.764	3.424
0.2286	1024.9	1.762	−0.042	0.092	0.495	2.972	2.237	0.8616	1087.0	2.748	−0.022	0.363	1.338	3.718	3.499
0.4001	1035.1	2.077	−0.061	0.213	0.628	3.153	2.420	0.9143	1097.9	2.685	−0.013	0.240	1.343	3.634	3.508
0.5334	1045.3	2.355	−0.064	0.341	0.797	3.354	2.662	0.9600	1109.1	2.608	−0.006	0.112	1.288	3.508	3.435
0.6401	1055.5	2.587	−0.056	0.452	0.995	3.564	2.958	1.0000	1120.5	2.542	0.000	0.000	–	–	–
0.7273	1065.8	2.724	−0.041	0.490	1.157	3.694	3.212								
T = 318.15 K															
0.0000	1007.5	1.117	0.000	0.000	–	–	–	0.8001	1068.2	2.141	−0.055	0.317	1.152	2.844	2.550
0.2286	1017.5	1.377	−0.052	0.058	0.431	2.281	1.724	0.8616	1078.7	2.111	−0.044	0.232	1.124	2.761	2.532
0.4001	1027.5	1.607	−0.078	0.136	0.542	2.399	1.843	0.9143	1089.4	2.068	−0.030	0.143	1.059	2.662	2.472
0.5334	1037.6	1.825	−0.085	0.236	0.722	2.569	2.033	0.9600	1100.1	2.027	−0.016	0.061	0.939	2.541	2.354
0.6401	1047.7	1.988	−0.079	0.305	0.882	2.697	2.221	1.0000	1111.1	2.001	0.000	0.000	–	–	–
0.7273	1057.9	2.100	−0.067	0.340	1.045	2.806	2.416								

Formamide (1) + Acetonitrile (4)

x_1	ρ / kg·m^{-3}	η / mPa·s	$V_m^E \cdot 10^6$ / m^3·mol^{-1}	$\Delta\eta$ / mPa·s	d_{12}	T_{12}	H_{12}	x_1	ρ / kg·m^{-3}	η / mPa·s	$V_m^E \cdot 10^6$ / m^3·mol^{-1}	$\Delta\eta$ / mPa·s	d_{12}	T_{12}	H_{12}
T = 298.15 K															
0.0000	776.0	0.344	0.000	0.000	–	–	–	0.5775	965.2	1.168	−0.467	−0.884	−0.343	0.261	0.011
0.0919	803.8	0.402	−0.175	−0.214	−0.626	0.608	0.541	0.6802	1002.9	1.507	−0.412	−0.849	−0.282	0.179	−0.128
0.1856	832.8	0.485	−0.302	−0.408	−0.507	0.567	0.473	0.7847	1042.6	1.945	−0.311	−0.720	−0.251	0.070	−0.308
0.2809	863.4	0.593	−0.401	−0.582	−0.452	0.506	0.382	0.8913	1085.1	2.540	−0.193	−0.440	−0.169	0.008	−0.448
0.3779	895.8	0.733	−0.469	−0.729	−0.419	0.431	0.272	1.0000	1129.2	3.302	0.000	0.000	–	–	–
0.4768	929.2	0.921	−0.471	−0.833	−0.374	0.353	0.153								
T = 308.15 K															
0.0000	766.3	0.313	0.000	0.000	–	–	–	0.5775	956.6	0.978	−0.527	−0.622	−0.286	0.343	0.152
0.0919	794.9	0.366	−0.237	−0.151	−0.400	0.583	0.523	0.6802	994.4	1.239	−0.463	−0.589	−0.219	0.305	0.073
0.1856	824.1	0.432	−0.381	−0.294	−0.429	0.532	0.454	0.7847	1034.7	1.569	−0.377	−0.492	−0.182	0.251	−0.029
0.2809	855.1	0.522	−0.495	−0.416	−0.373	0.496	0.396	0.8913	1076.9	1.999	−0.228	−0.300	−0.126	0.212	−0.121
0.3779	887.4	0.635	−0.560	−0.520	−0.354	0.446	0.321	1.0000	1120.5	2.542	0.000	0.000	–	–	–
0.4768	921.2	0.786	−0.566	−0.589	−0.307	0.402	0.247								
T = 318.15 K															
0.0000	755.0	0.289	0.000	0.000	–	–	–	0.5775	950.7	0.847	−0.780	−0.431	−0.176	0.411	0.262
0.0919	785.6	0.339	−0.378	−0.108	−0.242	0.552	0.498	0.6802	989.4	1.046	−0.731	−0.408	−0.141	0.385	0.207
0.1856	815.8	0.396	−0.579	−0.211	−0.298	0.513	0.447	0.7847	1029.3	1.295	−0.590	−0.338	−0.114	0.356	0.145
0.2809	847.4	0.472	−0.719	−0.298	−0.265	0.490	0.407	0.8913	1071.6	1.615	−0.417	−0.200	−0.043	0.359	0.113
0.3779	879.9	0.565	−0.774	−0.371	−0.260	0.458	0.356	1.0000	1111.1	2.001	0.000	0.000	–	–	–
0.4768	914.0	0.687	−0.783	−0.418	−0.227	0.431	0.307								

Table 2. (Continued)

Formamide (1) + 1,2-Dimethoxyethane (5)

x_1	ρ / $kg \cdot m^{-3}$	η / mPa·s	$V_m^E \cdot 10^6$ / $m^3 \cdot mol^{-1}$	$\Delta\eta$ / mPa·s	d_{12}	T_{12}	H_{12}	x_1	ρ / $kg \cdot m^{-3}$	η / mPa·s	$V_m^E \cdot 10^6$ / $m^3 \cdot mol^{-1}$	$\Delta\eta$ / mPa·s	d_{12}	T_{12}	H_{12}
							T = 298.15 K								
0.0000	861.5	0.409	0.000	0.000	–	–	–	0.7501	1022.2	2.296	−0.974	−0.283	0.846	2.144	1.100
0.1819	893.1	0.603	−1.101	−0.332	0.058	1.196	0.739	0.8236	1048.3	2.627	−0.752	−0.164	0.964	2.351	1.291
0.3334	920.1	0.848	−1.411	−0.525	0.151	1.281	0.674	0.8889	1073.7	2.899	−0.462	−0.081	1.034	2.501	1.445
0.4616	946.0	1.164	−1.464	−0.580	0.331	1.449	0.688	0.9474	1100.3	3.120	−0.201	−0.029	1.072	2.605	1.564
0.5715	971.8	1.545	−1.395	−0.517	0.554	1.695	0.799	1.0000	1129.2	3.302	0.000	0.000	–	–	–
0.6668	997.3	1.925	−1.229	−0.413	0.704	1.915	0.926								
							T = 308.15 K								
0.0000	850.7	0.366	0.000	0.000	–	–	–	0.7501	1013.6	1.806	−1.074	−0.192	0.761	1.731	0.942
0.1819	883.5	0.522	−1.252	−0.240	0.014	1.014	0.647	0.8236	1039.4	2.045	−0.809	−0.113	0.856	1.867	1.065
0.3334	910.4	0.713	−1.536	−0.378	0.096	1.081	0.603	0.8889	1065.7	2.243	−0.536	−0.057	0.914	1.966	1.165
0.4616	936.3	0.952	−1.563	−0.418	0.249	1.202	0.613	0.9474	1092.7	2.408	−0.264	−0.019	0.964	2.048	1.263
0.5715	962.3	1.243	−1.487	−0.366	0.472	1.393	0.706	1.0000	1120.5	2.542	0.000	0.000	–	–	–
0.6668	988.5	1.536	−1.339	−0.281	0.639	1.574	0.821								
							T = 318.15 K								
0.0000	839.6	0.330	0.000	0.000	–	–	–	0.7501	1007.5	1.427	−1.348	−0.156	0.600	1.368	0.749
0.1819	874.6	0.459	−1.527	−0.175	0.005	0.879	0.576	0.8236	1033.3	1.614	−1.038	−0.092	0.709	1.478	0.849
0.3334	902.6	0.609	−1.874	−0.278	0.053	0.923	0.540	0.8889	1059.2	1.775	−0.707	−0.040	0.815	1.582	0.963
0.4616	929.4	0.785	−1.927	−0.316	0.140	0.992	0.529	0.9474	1085.5	1.903	−0.376	−0.010	0.895	1.663	1.065
0.5715	955.9	0.994	−1.834	−0.291	0.296	1.107	0.572	1.0000	1111.1	2.001	0.000	0.000	–	–	–
0.6668	982.2	1.218	−1.647	−0.226	0.469	1.245	0.657								

Formamide (1) + Dimethylsulfoxide (6)

x_1	ρ / $kg \cdot m^{-3}$	η / mPa·s	$V_m^E \cdot 10^6$ / $m^3 \cdot mol^{-1}$	$\Delta\eta$ / mPa·s	d_{12}	T_{12}	H_{12}	x_1	ρ / $kg \cdot m^{-3}$	η / mPa·s	$V_m^E \cdot 10^6$ / $m^3 \cdot mol^{-1}$	$\Delta\eta$ / mPa·s	d_{12}	T_{12}	H_{12}
							T = 298.15 K								
0.0000	1095.4	2.042	0.000	0.000	–	–	–	0.7224	1115.8	3.652	−0.016	0.700	1.168	4.562	4.417
0.1616	1099.3	2.457	−0.032	0.212	0.794	3.943	3.454	0.8019	1119.1	3.626	−0.010	0.574	1.189	4.533	4.478
0.3025	1102.8	2.868	−0.047	0.445	0.921	4.193	3.726	0.8740	1122.5	3.540	−0.007	0.397	1.183	4.453	4.475
0.4264	1106.3	3.214	−0.047	0.635	1.017	4.381	3.969	0.9398	1125.8	3.426	−0.004	0.200	1.165	4.358	4.442
0.5363	1109.5	3.468	−0.039	0.751	1.094	4.513	4.182	1.0000	1129.2	3.302	0.000	0.000	–	–	–
0.6343	1112.6	3.608	−0.026	0.767	1.140	4.564	4.325								
							T = 308.15 K								
0.0000	1085.9	1.568	0.000	0.000	–	–	–	0.7224	1107.6	2.653	−0.050	0.382	0.883	3.154	3.007
0.1616	1090.7	1.859	−0.082	0.134	0.681	2.901	2.549	0.8019	1110.7	2.639	−0.032	0.291	0.841	3.078	2.971
0.3025	1094.9	2.135	−0.127	0.272	0.769	3.034	2.699	0.8740	1113.9	2.613	−0.021	0.194	0.804	3.010	2.936
0.4264	1098.5	2.365	−0.129	0.382	0.838	3.134	2.836	0.9398	1117.2	2.571	−0.010	0.088	0.717	2.895	2.833
0.5363	1101.6	2.520	−0.106	0.430	0.866	3.169	2.919	1.0000	1120.5	2.542	0.000	0.000	–	–	–
0.6343	1104.6	2.621	−0.077	0.435	0.893	3.189	2.993								
							T = 318.15 K								
0.0000	1076.9	1.485	0.000	0.000	–	–	–	0.7224	1099.5	2.118	−0.103	0.260	0.696	2.484	2.391
0.1616	1082.7	1.640	−0.152	0.072	0.379	2.223	2.009	0.8019	1102.3	2.083	−0.074	0.184	0.624	2.394	2.322
0.3025	1087.3	1.823	−0.212	0.182	0.545	2.393	2.174	0.8740	1105.3	2.054	−0.049	0.118	0.579	2.334	2.279
0.4264	1090.9	1.968	−0.214	0.263	0.631	2.477	2.280	0.9398	1108.2	2.021	−0.024	0.051	0.493	2.246	2.194
0.5363	1093.9	2.059	−0.182	0.297	0.670	2.503	2.340	1.0000	1111.1	2.001	0.000	0.000	–	–	–
0.6343	1096.7	2.112	−0.140	0.300	0.704	2.516	2.389								

dynamic viscosity, η, of the binary liquid mixtures in terms of pure component data.[31,32] Some of them we examined are as follows:

(i) Grunberg and ***Nissan***[33] have suggested the following logarithmic relation between the viscosity of binary liquid mixtures and of pure components

$$\ln \eta = x_1 \ln \eta_1 + x_2 \ln \eta_2 + x_1 x_2 d_{12} \tag{3}$$

where d_{12} is a constant, proportional to interchange energy, and the other symbols have their usual significance. It may be regarded as an approximate measure of the strength of molecular interactions between the mixing components. The calculated d_{12} values of the binary mixtures at different temperatures are listed in Table 2. Table 2 shows that the values of d_{12} are positive for all the binary liquid mixtures except that of formamide with acetonitrile over the entire range of compositions and at all the experimental temperatures. The positive values of d_{12} may be attributed to the presence of specific interactions[29,34] between the mixing components in the mixtures, whereas the negative values of d_{12} indicate the presence of dispersion forces[29] between them.

(ii) Tamura and ***Kurata***[35] put forward the following equation for the viscosity of binary liquid mixtures

$$\eta = x_1 \phi_1 \eta_1 + x_2 \phi_2 \eta_2 + 2(x_1 x_2 \phi_1 \phi_2)^{0.5} \cdot T_{12} \tag{4}$$

where T_{12} is the interaction parameter which depends on temperature and composition of the mixture and ϕ_1 [$\phi_1 = x_1V_1/(x_1V_1 + x_2V_2)$] and ϕ_2 [$\phi_2 = 1 - \phi_1$] are the volume fractions of pure components 1 and 2, respectively.

(iii) Hind et al.[36] suggested the following equation for the viscosity of the binary liquid mixture

$$\eta = x_1^2 \eta_1 + x_2^2 \eta_2 + 2x_1 x_2 H_{12} \tag{5}$$

where H_{12} is the Hind interaction parameter and is attributed to the unlike pair interactions.[37]

In the present study, the values of interaction parameter T_{12} and H_{12} have been calculated from eqs 4 and 5, respectively,

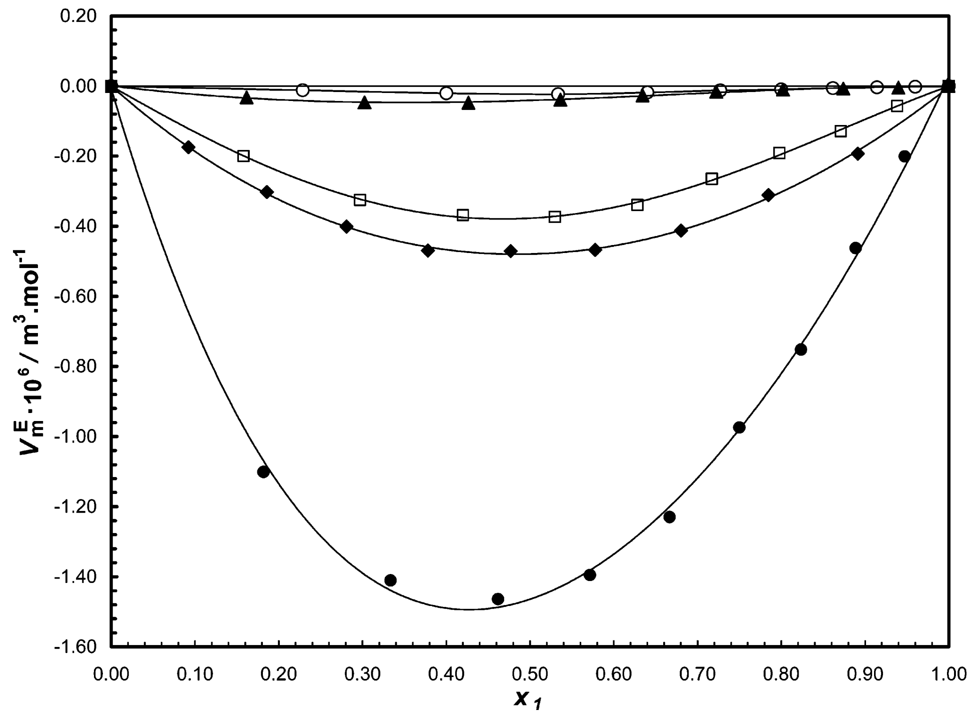

Figure 1. Excess molar volumes, V_m^E, for binary mixtures of formamide (1) at T = 298.15 K with: □, 2-methoxyethanol (2); ○, acetophenone (3); ◆, acetonitrile (4); ●, 1,2-dimethoxyethane (5); and ▲, dimethylsulfoxide (6).

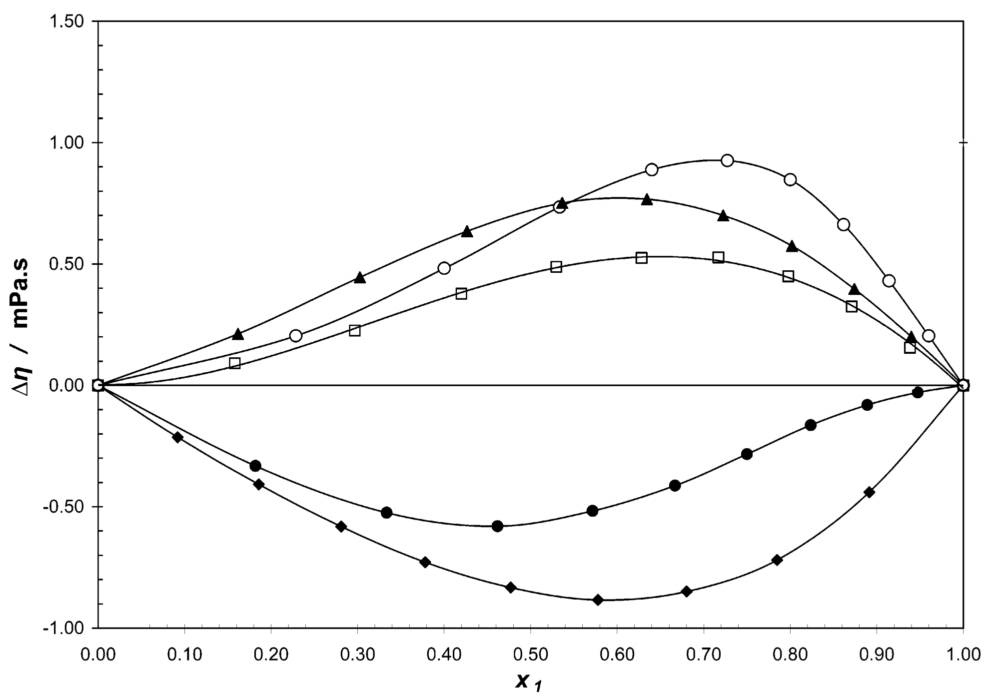

Figure 2. Viscosity deviations, $\Delta\eta$, for binary mixtures of formamide (1) at T = 298.15 K with: □, 2-methoxyethanol (2); ○, acetophenone (3); ◆, acetonitrile (4); ●, 1,2-dimethoxyethane (5); and ▲, dimethylsulfoxide (6).

and are listed in Table 2. A perusal of Table 2 shows that the T_{12} and H_{12} values do not differ appreciably with a change in the composition of the binary mixtures at all the experimental temperatures. This is in agreement with the view put forward by Fort and Moore[29] in regard to the nature of parameter T_{12} and H_{12}.

Deviation in Isentropic Compressibility. The isentropic compressibility, K_S, values were calculated using the relation[5]

$$K_S = 1/(u^2\rho) \tag{6}$$

where u and ρ are the experimental values of ultrasonic speed of sound and density, respectively, for the binary mixtures.

The deviations in isentropic compressibility, ΔK_S, for the binary mixtures have been derived using the relation[5]

$$\Delta K_S = K_S - (x_1 K_{S,1} + x_2 K_{S,2}) \tag{7}$$

where x_1, x_2 and $K_{S,1}$, $K_{S,2}$ are the mole fractions and isentropic compressibilities of pure components 1 (formamide) and 2 of the binary liquid mixtures, respectively.

The experimental values of density, ρ, ultrasonic speed of sound, u, the isentropic compressibility, K_S, and the deviation in isentropic compressibility, ΔK_S, at 298.15 K as a function of the composition of the binary mixtures have been presented in Table 3.

Table 3. Values of Density ρ, Ultrasonic Speed of Sound *u*, Isentropic Compressibility K_S, and Deviation in Isentropic Compressibility ΔK_S for the Binary Mixtures of Formamide at *T* = 298.15 K and *p* = 0.1 MPa as a Function of Mole Fraction x_1

x_1	ρ / $kg \cdot m^{-3}$	u / $m \cdot s^{-1}$	$K_S \cdot 10^{10}$ / Pa^{-1}	$\Delta K_S \cdot 10^{10}$ / Pa^{-1}	x_1	ρ / $kg \cdot m^{-3}$	u / $m \cdot s^{-1}$	$K_S \cdot 10^{10}$ / Pa^{-1}	$\Delta K_S \cdot 10^{10}$ / Pa^{-1}
				Formamide (1) + 2-Methoxyethanol (2)					
0.0000	959.7	1339.4	5.808	0.000	0.7171	1060.2	1478.3	4.316	0.165
0.1581	977.0	1353.6	5.586	0.143	0.7977	1076.7	1506.0	4.095	0.130
0.2969	994.2	1373.7	5.329	0.208	0.8711	1093.8	1533.9	3.885	0.090
0.4199	1010.9	1397.5	5.065	0.227	0.9383	1111.1	1564.3	3.678	0.038
0.5297	1027.6	1423.8	4.800	0.216	1.0000	1129.2	1591.3	3.497	0.000
0.6282	1044.1	1451.1	4.548	0.192					
				Formamide (1) + Acetophenone (3)					
0.0000	1023.1	1296.4	5.816	0.000	0.8001	1084.4	1456.5	4.347	0.386
0.2286	1032.9	1304.8	5.686	0.400	0.8616	1095.3	1488.4	4.122	0.303
0.4001	1042.9	1323.3	5.475	0.587	0.9143	1106.4	1527.3	3.875	0.179
0.5334	1053.1	1345.8	5.243	0.663	0.9600	1117.7	1558.9	3.682	0.091
0.6401	1063.3	1379.3	4.943	0.611	1.0000	1129.2	1591.3	3.497	0.000
0.7273	1073.8	1413.6	4.661	0.531					
				Formamide (1) + Acetonitrile (4)					
0.0000	776.0	1260.2	8.114	0.000	0.5775	965.2	1412.8	5.190	−0.257
0.0919	803.8	1277.7	7.620	−0.069	0.6802	1002.9	1449.0	4.749	−0.225
0.1856	832.8	1299.6	7.109	−0.148	0.7847	1042.6	1488.8	4.327	−0.164
0.2809	863.4	1322.8	6.619	−0.198	0.8913	1085.1	1529.8	3.938	−0.061
0.3779	895.8	1349.6	6.129	−0.240	1.0000	1129.2	1591.3	3.497	0.000
0.4768	929.2	1380.8	5.644	−0.269					
				Formamide (1) + 1,2-Dimethoxyethane (5)					
0.0000	861.5	1146.2	8.835	0.000	0.7501	1022.2	1441.9	4.705	−0.126
0.1819	893.1	1200.3	7.771	−0.093	0.8236	1048.3	1480.5	4.352	−0.087
0.3334	920.1	1254.3	6.908	−0.147	0.8889	1073.7	1518.5	4.039	−0.051
0.4616	946.0	1306.2	6.195	−0.176	0.9474	1100.3	1556.1	3.753	−0.025
0.5715	971.8	1355.0	5.604	−0.180	1.0000	1129.2	1591.3	3.497	0.000
0.6668	997.3	1399.9	5.116	−0.160					
				Formamide (1) + Dimethylsulfoxide (6)					
0.0000	1095.4	1493.0	4.095	0.000	0.7224	1115.8	1512.7	3.917	0.253
0.1616	1099.3	1480.1	4.153	0.154	0.8019	1119.1	1526.4	3.835	0.219
0.3025	1102.8	1480.6	4.136	0.222	0.8740	1122.5	1543.7	3.738	0.165
0.4264	1106.3	1483.9	4.105	0.265	0.9398	1125.8	1561.2	3.644	0.111
0.5363	1109.5	1492.3	4.047	0.273	1.0000	1129.2	1591.3	3.497	0.000
0.6343	1112.6	1502.9	3.979	0.263					

Table 3 shows that ΔK_S values are positive for the binary mixtures of 2-methoxyethanol, dimethylsulfoxide, and acetophenone and negative for those of 1,2-dimethoxyethane and acetonitrile over the entire range of compositions at 298.15 K, and the deviations in isentropic compressibility, ΔK_S, have been plotted against mole fraction of formamide, x_1, for the binary mixtures at 298.15 K in Figure 3.

The observed values of ΔK_S can be qualitatively explained by considering the factors, namely (i) the mutual disruption of associated present in the pure liquids, (ii) the formation of weak bonds by dipole-induced dipole interaction between unlike molecules, and (iii) geometrical fitting of component molecules into each other structure. The first factor contributes to positive ΔK_S values, whereas the remaining two factors lead to negative ΔK_S values.[38] The resultant values of ΔK_S for the present mixtures are due to the net effect of the combination of (i) to (iii).[39]

In the present investigation, the negative deviation in isentropic compressibility is an indication of strong interactions, whereas positive deviation is a sign for weak interactions between component molecules.[1,29,40−42]

From Table 3, it is also seen that the values of deviation in isentropic compressibility, ΔK_S, are more negative for the formamide + acetonitrile mixture than the formamide + 1,2-dimethoxyethane mixture. Higher values of the Grunberg−Nissan interaction parameter, d_{12}, in Table 2 for the formamide + 1,2-dimethoxyethane mixture than for the formamide + acetonitrile mixture are an indication of its higher molecular interactions than the formamide + acetonitrile mixture.[6]

Therefore, from Table 3, Figure 3, and the values of d_{12} in Table 2, it is seen that the strength of interaction between the binary mixtures follows the order: formamide + 1,2-dimethoxyethane > formamide + acetonitrile > formamide + 2-methoxyethanol > formamide + dimethylsulfoxide > formamide + acetophenone. Thus, the graded behavior of this function, ΔK_S, over the entire range of compositions at 298.15 K supports the results obtained earlier.

Redlich−Kister Polynomial Equation. The excess or deviation properties were fitted to the Redlich−Kister polynomial equation[43] of the type

$$X^E = x_1 x_2 [\sum_{i=0}^{k} A_i (x_1 - x_2)^i] \tag{8}$$

where X^E represents an excess or deviation property. The coefficients, A_i, of eq 8, were evaluated using a least-squares method and have been presented in Table 4 along with their standard deviations. The standard deviation σ was evaluated by using the following relation

$$\sigma(X^E) = [(X^E{}_{(obsd)} - X^E{}_{(calcd)})^2/(n - p)]^{0.5} \tag{9}$$

where n is the total number of experimental points and p is the number of A_i coefficients considered. The σ values lie between 0.013 $m^3 \cdot mol^{-1}$ and 0.001 $m^3 \cdot mol^{-1}$ for V_m^E, between 0.022 mPa·s and 0.001 mPa·s for $\Delta\eta$, and between 0.011 Pa^{-1} and 0.001 Pa^{-1} for ΔK_S, respectively. The largest σ value

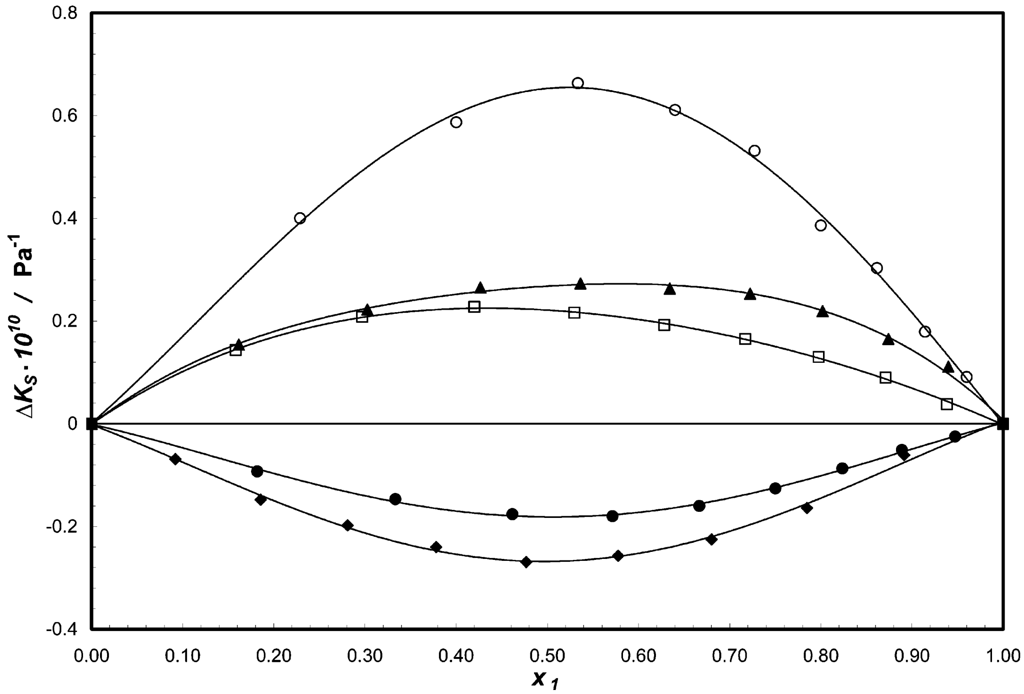

Figure 3. Deviation in isentropic compressibility, ΔK_S, for binary mixtures of formamide (1) at T = 298.15 K with: □, 2-methoxyethanol (2); ○, acetophenone (3); ◆, acetonitrile (4); ●, 1,2-dimethoxyethane (5); and ▲, dimethylsulfoxide (6).

Table 4. Coefficients A_i of the Redlich−Kister Equation and the Standard Deviation σ for the Binary Mixtures of Formamide from T = (298.15 to 318.15) K

binary mixture	excess property	T/K	A_0	A_1	A_2	A_3	σ
formamide (1) + 2-methoxyethanol (2)	$V_m^E \cdot 10^6/(m^3 \cdot mol^{-1})$	298.15	−1.508	0.252	0.391	−	0.005
		308.15	−1.595	0.230	0.142	−	0.004
		318.15	−1.659	0.154	−0.246	−	0.004
	$\Delta\eta/(mPa \cdot s)$	298.15	1.840	1.705	−0.282	0.847	0.004
		308.15	1.015	0.983	−	−	0.011
		318.15	0.569	0.819	0.358	−0.264	0.005
	$\Delta K_S \cdot 10^{10}/(Pa^{-1})$	298.15	0.886	−0.221	0.099	−	0.003
formamide (1) + acetophenone (3)	$V_m^E \cdot 10^6/(m^3 \cdot mol^{-1})$	298.15	−0.092	−0.002	0.147	0.032	0.001
		308.15	−0.261	0.017	0.204	0.058	0.001
		318.15	−0.329	−0.045	−	−	0.002
	$\Delta\eta/(mPa \cdot s)$	298.15	2.714	3.537	0.859	4.677	0.002
		308.15	1.221	2.088	2.178	−0.174	0.002
		318.15	0.941	1.428	−	−	0.022
	$\Delta K_S \cdot 10^{10}/(Pa^{-1})$	298.15	2.612	0.646	−1.141	−1.290	0.011
formamide (1) + acetonitrile (4)	$V_m^E \cdot 10^6/(m^3 \cdot mol^{-1})$	298.15	−1.917	0.112	−0.113	−	0.007
		308.15	−2.242	0.624	−0.215	−1.923	0.001
		318.15	−3.148	0.131	−1.741	−	0.013
	$\Delta\eta/(mPa \cdot s)$	298.15	−3.404	−1.339	−0.280	0.117	0.002
		308.15	−2.406	−0.818	−0.106	−	0.002
		318.15	−1.697	−0.496	−	−	0.002
	$\Delta K_S \cdot 10^{10}/(Pa^{-1})$	298.15	−1.056	−0.102	0.111	0.393	0.005
formamide (1) + 1,2-dimethoxyethane (5)	$V_m^E \cdot 10^6/(m^3 \cdot mol^{-1})$	298.15	−5.815	1.014	−1.024	2.051	0.012
		308.15	−6.196	0.929	−2.312	4.658	0.008
		318.15	−7.639	1.094	−2.352	4.833	0.006
	$\Delta\eta/(mPa \cdot s)$	298.15	−2.263	0.806	1.369	−	0.004
		308.15	−1.634	0.774	1.805	−2.595	0.001
		318.15	−1.255	0.337	1.149	−0.036	0.001
	$\Delta K_S \cdot 10^{10}/(Pa^{-1})$	298.15	−0.721	−0.152	0.262	0.828	0.001
formamide (1) + dimethylsulfoxide(6)	$V_m^E \cdot 10^6/(m^3 \cdot mol^{-1})$	298.15	−0.161	0.148	−	−	0.002
		308.15	−0.461	0.477	0.140	−0.373	0.001
		318.15	−0.775	0.649	−0.013	−0.336	0.001
	$\Delta\eta/(mPa \cdot s)$	298.15	2.881	1.807	−0.638	−0.637	0.002
		308.15	1.690	0.853	−0.636	−0.550	0.003
		318.15	1.178	0.604	−0.738	−0.364	0.004
	$\Delta K_S \cdot 10^{10}/(Pa^{-1})$	298.15	1.091	0.155	0.162	0.186	0.005

corresponds to the formamide + acetonitrile system for V_m^E and the formamide + acetophenone system for $\Delta\eta$ and ΔK_S, respectively. It is seen that the fit is quite satisfactory as revealed by the values of standard deviation.

Conclusions

The present study reveals that the order of specific interaction for the binary mixtures of formamide follows the order: formamide + 1,2-dimethoxyethane > formamide + acetonitrile

> formamide + 2-methoxyethanol > formamide + dimethylsulfoxide > formamide + acetophenone. This may be attributed to intermolecular hydrogen bonding and interstitial accommodation between the mixing components. The graded pattern can also be partly due to the difference in the dielectric constants of the mixing components.

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