CHAPTER IX

Excess Properties of Binary Mixtures of N,N – Dimethylacetamide + 2-Methoxyethanol, and N,N – Dimethylacetamide + Water at 298.15, 308.15 and 318.15 K

Thermodynamic excess properties of binary liquid mixtures have been very helpful to obtain information on the intermolecular interactions and geometrical effects in these systems.[1,2] Solvents like 2-methoxyethanol (ME) and N,N-dimethylacetamide (DMA) find a wide range of applications of technological importance, namely, as solvents and solubilizing agents in organic synthesis, reaction kinetics and electrochemical studies. Hence the recent spurt in research in these areas.[3-6]

The amide group in DMA is a good model of a peptide bond. A systematic study of the structural and energetic consequences of the interaction between DMA and water will enable us to understand how water exercises thermodynamic and kinetic control over the chemical activities of polypeptides in aqueous media.

We have measured densities, viscosities and ultrasonic velocities for the liquid mixtures of DMA + ME and DMA + water over the entire range of their compositions at 298.15, 308.15 and 318.15 K. We have also evaluated the κ_s and τ values for DMA + ME and DMA + water mixtures at these three temperatures, as mentioned in the previous Chapter. The excess molar volumes (V^E), viscosity deviations ($\Delta\eta$) and isentropic compressibility changes ($\Delta\kappa_s$) of these systems have been calculated at different temperatures.

9.1 EXPERIMENTAL SECTION

The purification of the solvents used and the experimental procedure for the measurement of the above properties have been described earlier under the Experimental Section. The measurements of densities, viscosities and ultrasonic velocities have also been described earlier. In all cases, the experiments were performed at least in five replicates for each composition and the results were averaged.

9.2 RESULTS

9.2.1 Solvent Parameters of the Mixtures

The solvent properties (density, viscosity, and ultrasonic velocity) of the above mixtures as a function of mole fractions of DMA, at 298.15, 308.15 and 318.15 K, have been presented in Tables 1-2.

To obtain information about the nature of DMA + ME and DMA + water solvent systems, at various temperatures and compositions, the values of density, viscosity and ultrasonic velocity are plotted against mole fractions of DMA. These plots are shown in Figures 1-3.

Two derived parameters, namely κ_s and τ , have been evaluated and these have been reported in Tables 3-4. The variation of κ_s and τ values with the composition of the mixtures (mole fraction of DMA) have been shown in Figures 4-5.

$$\tau = 4.\eta \kappa_s/3 \qquad (1)$$

9.2.2 Excess Properties of the Mixtures

The excess functions have been evaluated using the following equations:

$$V^E = V - (V_1x_1 + V_2x_2) \qquad (2)$$

$$\Delta\eta = \eta - (\eta_1x_1 + \eta_2x_2) \qquad (3)$$

$$\Delta\kappa_s = \kappa_s - (\kappa_{s\,1}x_1 + \kappa_{s\,2}x_2) \qquad (4)$$

where x_1 and x_2 are the mole fractions of DMA and ME / water, respectively. V, η and κ_s are the respective solution properties, V_1 and V_2 , η_1 and η_2, and κ_{s1} and κ_{s2} are the molar volumes, the coefficients of viscosities and, the isentropic compressibilities of DMA and ME / water in the mixture respectively.

The molar volume V is defined by the relation, $V = (M_1x_1 + M_2x_2)/\rho$, where, M_1 and M_2 are the molecular masses of pure substances and ρ is the density of the mixture.

The excess functions at 298.15, 308.15 and 318.15 K have been presented in Tables 5-6.

Graphical representations of V^E, $\Delta\eta$ and $\Delta\kappa_s$ as functions of mole fractions of DMA are given in Figures 6-8.

As earlier, the excess properties Y^E were fitted to the Redlich- Kister equation:

$$Y^E = x_1 (1 - x_1) \Sigma A_j (1 - 2x_1)^j, \qquad (5)$$

where Y^E is V^E / (cm^3mol^{-1}) or $\Delta\eta$ / (mPa.s) or $\Delta\kappa_s$ / (bar^{-1}), and A_0, A_1, A_2, ... etc., are adjustable parameters. These parameters were evaluated by fitting Y^E / x_1 (1- x_1) to equation (5) by the method of least squares. The values of these parameters along with the standard deviation σ are recorded in Tables 7-8, for DMA + ME and DMA + water systems respectively.

9.3 DISCUSSION

9.3.1 Solvent Properties of DMA + ME and DMA + Water Systems

The plots of densities, viscosities, and ultrasonic velocities of DMA + ME, and DMA + water mixtures of various compositions and at different temperatures offer some information about the nature of these systems. For both the systems, the variations of density, viscosity, and ultrasonic velocity as functions of the mole fractions of DMA are non-linear (Fig 1-3) at all three temperatures. In the case of DMA + ME, there is an uniform and gradual decrease in density and viscosity (the curves deviate only slightly from linearity and there are no minima or maxima) as the mole fraction of DMA increases;

however, the ultrasonic velocity increases rather sharply with the mole fraction of DMA. In terms of temperatures, all the three parameters (ρ, η and 'u') decrease with increase in temperature for the DMA + ME system.

In the case of DMA + water system, the non-linear behaviour of all these parameters is very well pronounced. In fact, the variation of these parameters with an increase in the mole fraction of DMA is not uniform and gradual as in the case of the former system but very distinct and characteristic of each parameter. The density at 298.15 K initially increases slightly with the mole fraction of DMA but falls rapidly around mole fraction 0.30 until it reaches mole fraction 0.60 and falls off less rapidly thereafter. At higher temperatures density decreases almost from the start, but the shape and behaviour are the same as those at 298.15 K but below that curve. The viscosity and ultrasonic velocity of this system increase very steeply with the increase in the percentage of DMA, reach the maxima (around mole fraction 0.20 - 0.25) and fall off sharply after that. The viscosity curve is almost symmetrical whereas the ultrasonic one is not. All these three parameters decrease with an increase of temperature.

The plots of the two derived parameters, namely, isentropic compressibility (κ_s) and shear relaxation time(τ) versus the mole composition of DMA (Figures 4-5) for the two systems show exactly the similar trend as their respective density, viscosity and ultrasonic velocity plots. In DMA + ME, κ_s and τ plots give slightly non-linear curves which fairly resemble each other and wherein the κ_s and τ values decrease gradually with mole fraction of DMA as well as with temperature. However, in DMA + water, the κ_s plots give very well pronounced curves with clear minima (around mole fraction 0.20 in DMA) while the τ plots give very well pronounced curves with sharp maxima (around mole fraction 0.30 in DMA) at each of the temperature studied.

9.3.2 Excess Properties of DMA + ME and DMA + Water Systems

9.3.2.1 Excess Molar Volume

It is well known that the sign and magnitude of V^E give a good estimate of the strength of the unlike interactions in binary mixtures.[7] Large positive V^E values are taken as indicative of weak intermolecular interactions whereas large negative values of V^E are usually found when these interactions are strong and intermolecular association "complexes" are believed to be present.

The systems DMA + ME and DMA + water show negative V^E values over the entire range of mole fraction and over the entire range of temperatures studied (Figures 6-8). Both the systems show clear minima at a mole fraction of about 0.40 of DMA.

DMA + ME System

DMA is a dipolar aprotic solvent and due to the moderate dielectric constant (ϵ = 37.8 at 298.15 K) it is likely to be moderately structured. On the other hand, intermolecular hydrogen bonding exists in ME molecules in the liquid state. Moreover, the values of the Kirkwood correlation factors, g_k, for pure ME in the temperature range studied are not much greater than unity. This indicates that 2-methoxyethanol is a relatively unstructured liquid and that there are strong but not specific dipole-dipole forces.

Thus the interaction between a structured DMA and a relatively unstructured ME results in a negative V^E. This may be due to the disruption of (i) intermolecular hydrogen bonds in the ME molecules, and (ii) intermolecular dipolar interactions in DMA. Secondly, the existence of hydrogen bonds between the unlike molecules is likely to offer a negative contribution to V^E.

Considering the molar volumes of DMA and ME (93.02 and 79.27 cm^3 mol^{-1} respectively), the interstitial accommodation of one component into the other is ruled out.

DMA + Water System

Consistent with some recent studies in amide + water mixtures,[8,9] the excess molar volumes at higher temperature is negative over the whole range of compositions (Figure 6). The magnitude of V^E slightly decreases with temperature. It must be pointed out here that the mixing of DMA with water was a highly exothermic process.

The negative V^E values at 298.15, 308.15 and 318.15 K for this mixture and the clear minima in the curves indicate strong association through multiple hydrogen bonding between the polar group of DMA and water, although the pure liquids are presumed to be highly structured. Compared to the V^E values of DMA + ME, the V^E values here are five times more negative which may be a sign of strong solvation by hydrogen bonding accompanied by a minor destruction of water structure, besides the interstitial accommodation of DMA molecules inside the water cavities. In aqueous mixtures, the magnitude of V^E values reflects the strength of the hydrogen bonding between the polar groups of the amides and water. Due to the presence of the third methyl group in DMA, the molecule becomes more polar than DMF, thereby increasing its hydrogen bonding ability and thus giving more negative V^E in the DMA + water mixture than in the DMF + water mixture.[10]

From Figure 6 we see that, as the temperature rises there is a slight decrease in V^E in the water-rich portion. A rise in temperature leads to the breaking of the less dense hydrogen bonded structures and/or breakdown of self-associated DMA aggregates. This may result in a denser packing of molecules than before. It may also be accompanied by the release of free water and an increase in the fraction of unbounded molecules. This is consistent with the view that the excess molar volumes of aqueous mixtures reflect the structural changes of water introduced by the non-aqueous molecules.[11]

All this suggests that the hydrogen bonding between water and DMA is stronger and more compact than that between water molecules themselves, and hence the interaction between these unlike molecules gives a negative contribution to the V^E values.

9.3.2.2 Viscosity Deviations

Both DMA + ME and DMA + water systems show positive deviation of $\Delta\eta$ from ideality over the entire mole fraction range and over the whole range of studied temperatures. In DMA + ME, the maxima correspond to a mole fraction of about 0.36 in DMA whereas in DMA + water the maxima occur at a mole fraction of about 0.30 in DMA as shown in Figure 7.

The positive deviation indicates the predominance of specific hydrogen bonding interactions between the unlike molecules over the dissociation effects in both the systems. However, compared to DMA + ME, $\Delta\eta_{max}$ in DMA + water is about 20 to 40 times larger, indicating very strong specific hydrogen bond interactions between DMA and water molecules. Even, η versus the mole fraction of DMA shows a large maximum (Figure 2) suggesting high association or complex formation between water and the peptido dipole in DMA. There are no such maxima in the other system.

As the temperature increases the magnitude of the viscosity deviation decreases only slightly in the case of the DMA + ME system, whereas in DMA + water the decrease is rather rapid. The sharp decrease may be due to the more rapid breaking up of the extensive hydrogen bonds in the latter than in the former.

9.3.2.3 Isentropic Compressibility Changes

The results of deviations in isentropic compressibility versus mole fraction at three different temperatures are given in Figures 8, for DMA + ME, and DMA + water systems. For both the systems, the values of $\Delta\kappa_s$ are negative over the entire composition range and at all the three temperatures investigated. Negative $\Delta\kappa_s$ means that the mixture is less compressible than the corresponding ideal mixture suggesting a predominant hydrogen bond interaction between DMA and water, thereby causing an increase in the ultrasonic velocity and a decrease in the compressibility of these solutions. This process continues until the mole fraction reaches around 0.20 in DMA, at all the three temperatures studied, because at this composition water cavities are filled up by interstitial solvation, or due to the formation of hydrophobic DMA aggregates, or as a consequence of both. It is thus apparent that the interstitial accommodation of DMA into water multimer structure becomes more effective and the positive contribution from the breaking of hydrogen bonds becomes less predominant, giving negative $\Delta\kappa_s$ values over the entire mole fraction range for these mixtures.

It is very interesting to note the variation of ultrasonic velocity with the mole fraction of DMA in the DMA + water mixture. The ultrasonic velocities at the three temperatures converge at a particular mole fraction (around 0.065 in DMA) and then diverge in a reverse manner, after passing through maxima (Figure 3). A very similar trend is observed when κ_s is plotted against the mole fraction of DMA in the same mixture, the transition taking place at the mole fraction of about 0.05 in DMA, after which the curves diverge in the reverse manner and pass through minima and increase (Figure 4).

This phenomenon can be explained,[12] as follows. On increasing the amount of DMA, the number of free water molecules around the DMA molecules decrease gradually until a situation is reached where all the free water molecules are involved in the solvation. Such a condition may be correlated to the primary solvation (or hydration) of DMA since the water molecules are not further compressed and both the ultrasonic velocity and isentropic compressibility are independent of temperature (Figures 3-4). At this condition, which is the convergence point in the above figures, κ_s becomes $\kappa_{s,h}$.[13] The magnitude of this value for each system should indicate the level of interaction between the two components. The smaller the $\kappa_{s,h}$ value for a system in comparison to free water, the stronger will be the interaction. However, here, along with the process of solvation, interstitial accommodation of DMA inside the water cavities is taking place simultaneously, along with a possible rupture of water structure. So the $\kappa_{s,h}$ obtained in this manner is only a qualitative indicator of the interaction, and it may as well be called $\kappa_{s,c}$, isentropic compressibility at convergence point. Further, in an aqueous mixture of an organic solvent, we can suggest that if $\kappa_{s,c} < \kappa_s^0$, where κ_s^0 is the isentropic compressibility of pure water, there is strong interaction between the two components. However, further investigations are required to quantify the relationship between the two.

Considering the experimental data of the two systems, it is apparent that, in the DMA + water system strong hetero-association is present through multiple hydrogen bonding between the polar groups of the amide and water. Large negative V^E and $\Delta\kappa_s$ values, and large positive $\Delta\eta$ values are indicators for this phenomenon. On the other hand, there is a weak interaction and solvent structure effect in the DMA + ME mixture, very akin to that in the DMA and acetonitrile system.[14]

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TABLE 1. Density (ρ), Viscosity (η) and Ultrasonic Velocity (u) of N,N-Dimethylacetamide + 2-Methoxyethanol mixtures at different mole fractions (x) of DMA, at 298.15, 308.15 and 318.15 K.

	ρ (g cm^{-3})			η (mPa s)			u (m s^{-1})		
x (DMA)	298.15	308.15	318.15	298.15	308.15	318.15	298.15	308.15	318.15 (K)
0.0000	0.959790	0.952515	0.946235	1.5436	1.2577	1.0500	1339.31	1303.85	1269.05
0.0301	0.959279	0.952208	0.946145	1.5359	1.2545	1.0487	1344.60	1308.91	1273.95
0.0600	0.958755	0.951754	0.945835	1.5285	1.2508	1.0469	1349.92	1314.07	1278.68
0.0871	0.958265	0.951321	0.945357	1.5207	1.2470	1.0452	1354.65	1318.87	1283.23
0.1250	0.957562	0.950609	0.944507	1.5078	1.2394	1.0413	1361.02	1325.37	1289.40
0.1767	0.956566	0.949517	0.943178	1.4881	1.2280	1.0345	1369.15	1333.95	1297.74
0.2250	0.955560	0.948387	0.941863	1.4662	1.2145	1.0261	1376.15	1341.51	1305.29
0.2902	0.954130	0.946778	0.940049	1.4326	1.1929	1.0116	1385.07	1350.76	1314.63
0.3640	0.952412	0.944894	0.937971	1.3903	1.1646	0.9916	1394.65	1360.30	1324.27
0.4602	0.950124	0.942394	0.935219	1.3294	1.1222	0.9604	1406.55	1371.81	1335.76
0.5200	0.948662	0.940816	0.933477	1.2891	1.0929	0.9384	1413.59	1378.50	1342.40
0.5611	0.947644	0.939726	0.932269	1.2600	1.0709	0.9218	1418.27	1382.83	1346.66
0.6200	0.946167	0.938152	0.930506	1.2168	1.0387	0.8972	1424.60	1388.66	1352.30
0.6654	0.944995	0.936940	0.929134	1.1815	1.0118	0.8761	1429.13	1393.07	1356.32
0.7250	0.943474	0.935341	0.927288	1.1354	0.9770	0.8491	1434.83	1398.76	1361.49
0.7732	0.942252	0.934068	0.925764	1.0990	0.9495	0.8271	1439.49	1403.31	1365.63
0.8250	0.940939	0.932644	0.924130	1.0595	0.9194	0.8033	1444.16	1408.11	1369.92
0.8847	0.939455	0.931006	0.922243	1.0143	0.8850	0.7759	1449.22	1413.49	1374.74
0.9501	0.937873	0.929186	0.920119	0.9651	0.8474	0.7460	1454.89	1419.33	1380.09
1.0000	0.936690	0.927760	0.918495	0.9271	0.8190	0.7232	1459.18	1423.75	1384.11

TABLE 2. Density (ρ), Viscosity (η) and Ultrasonic Velocity (u) of N,N-Dimethylacetamide + Water mixtures at different mole fractions (x) of DMA, at 298.15, 308.15 and 318.15 K.

	ρ (g cm^{-3})			η (mPa s)			u (m s^{-1})		
x (DMA)	298.15	308.15	318.15	298.15	308.15	318.15	298.15	308.15	318.15 (K)
0.0000	0.997093	0.994108	0.990268	0.8894	0.7211	0.6001	1497.16	1519.35	1535.49
0.0250	0.997543	0.993767	0.988502	1.2654	0.9975	0.7841	1566.07	1589.74	1584.83
0.0650	0.998544	0.993766	0.987144	1.8717	1.4283	1.1069	1650.72	1652.48	1641.76
0.1011	0.999425	0.992753	0.985621	2.4138	1.8107	1.3873	1702.24	1685.26	1664.61
0.1200	0.999811	0.992283	0.984955	2.7546	2.0225	1.5336	1717.94	1696.17	1671.75
0.1495	0.999386	0.991701	0.984051	3.1609	2.3078	1.7182	1732.84	1704.88	1675.75
0.2004	0.998206	0.989909	0.981554	3.7281	2.6612	1.9444	1740.25	1706.21	1671.72
0.2512	0.995426	0.987225	0.978260	3.9404	2.7900	2.0500	1735.61	1695.32	1657.44
0.3002	0.991300	0.983007	0.974050	3.9415	2.7847	2.0565	1722.59	1678.70	1637.94
0.3501	0.986651	0.978300	0.969257	3.6927	2.6670	1.9865	1700.48	1655.93	1613.90
0.4009	0.981926	0.973377	0.964332	3.3419	2.4743	1.8537	1672.08	1631.39	1590.24
0.5018	0.972715	0.963790	0.954933	2.6064	1.9295	1.5539	1619.80	1582.81	1540.91
0.6031	0.963803	0.954865	0.946066	1.9958	1.5686	1.2742	1577.93	1539.75	1497.25
0.6505	0.959770	0.950945	0.942076	1.7779	1.4247	1.1784	1560.65	1522.46	1480.39
0.7005	0.955897	0.947069	0.938220	1.5921	1.3059	1.0850	1543.71	1505.54	1464.59
0.7550	0.951848	0.943109	0.934167	1.4190	1.1831	0.9991	1525.94	1488.73	1447.76
0.8048	0.948387	0.939635	0.930728	1.2846	1.0859	0.9327	1510.83	1472.92	1433.31
0.8501	0.945457	0.936612	0.927793	1.1851	1.0032	0.8681	1497.74	1460.26	1421.06
0.9049	0.941928	0.933131	0.924215	1.0830	0.9224	0.8038	1483.33	1445.84	1407.41
1.0000	0.936527	0.927710	0.918779	0.9332	0.8165	0.7213	1461.90	1423.71	1385.17

TABLE 3. Isentropic Compressibility (κ_s) and Shear Relaxation Time (τ) of the mixtures of N,N-Dimethylacetamide + 2-Methoxyethanol, at different mole fractions, at 298.15, 308.15 and 318.15 K.

	$\kappa_s \times 10^5$ (bar^{-1})			$\tau \times 10^{13}$ (s)		
x (DMA)	298.15K	308.15 K	318.15 K	298.15K	308.15 K	318.15 K
0.0000	5.8085	6.1755	6.5621	11.95	10.36	9.19
0.0301	5.7660	6.1298	6.5123	11.81	10.25	9.11
0.0600	5.7237	6.0847	6.4664	11.67	10.15	9.03
0.0871	5.6867	6.0433	6.4238	11.53	10.05	8.95
0.1250	5.6378	5.9885	6.3682	11.33	9.90	8.84
0.1767	5.5768	5.9186	6.2955	11.06	9.69	8.68
0.2250	5.5260	5.8590	6.2316	10.80	9.49	8.53
0.2902	5.4632	5.7889	6.1552	10.44	9.21	8.30
0.3640	5.3982	5.7194	6.0793	10.01	8.88	8.04
0.4602	5.3200	5.6387	5.9928	9.43	8.44	7.67
0.5200	5.2752	5.5935	5.9447	9.07	8.15	7.44
0.5611	5.2461	5.5649	5.9149	8.81	7.95	7.27
0.6200	5.2077	5.5276	5.8767	8.45	7.66	7.03
0.6654	5.1811	5.4997	5.8506	8.16	7.42	6.83
0.7250	5.1484	5.4644	5.8178	7.79	7.12	6.59
0,7732	5.1217	5.4364	5.7921	7.51	6.88	6.39
0.8250	5.0957	5.4077	5.7660	7.20	6.63	6.18
0.8847	5.0682	5.3760	5.7374	6.85	6.34	5.94
0.9501	5.0373	5.3423	5.7061	6.48	6.04	5.68
1.0000	5.0140	5.3174	5.6831	6.20	5.81	5.48

TABLE 4. Isentropic Compressibility (κ_s) and Shear Relaxation Time (τ) of the mixtures of N,N-Dimethylacetamide + Water, at different mole fractions, at three temperatures.

	$\kappa_s \times 10^5$ (bar^{-1})			$\tau \times 10^{13}$ (s)		
x (DMA)	298.15K	308.15 K	318.15 K	298.15K	308.15 K	318.15 K
0.0000	4.4743	4.3576	4.2831	5.31	4.19	3.43
0.0250	4.0874	3.9816	4.0277	6.90	5.30	4.21
0.0650	3.6753	3.6851	3.7584	9.17	7.02	5.55
0.1011	3.4531	3.5467	3.6615	11.11	8.56	6.77
0.1200	3.3890	3.5029	3.6328	12.45	9.45	7.43
0.1495	3.3324	3.4692	3.6188	14.04	10.68	8.29
0.2004	3.3079	3.4701	3.6455	16.44	12.31	9.45
0.2512	3.3349	3.5244	3.7211	17.52	13.11	10.17
0.3002	3.3996	3.6099	3.8267	17.87	13.40	10.49
0.3501	3.5051	3.7277	3.9610	17.26	13.26	10.49
0.4009	3.6426	3.8601	4.1006	16.23	12.73	10.13
0.5018	3.9182	4.1415	4.4103	13.62	10.65	9.14
0.6031	4.1671	4.4173	4.7151	11.09	9.24	8.01
0.6505	4.2778	4.5368	4.8436	10.14	8.62	7.61
0.7005	4.3899	4.6583	4.9689	9.32	8.11	7.19
0.7550	4.5119	4.7842	5.1072	8.54	7.55	6.80
0.8048	4.6194	4.9055	5.2300	7.91	7.10	6.50
0.8501	4.7150	5.0070	5.3373	7.45	6.70	6.18
0.9049	4.8251	5.1264	5.4625	6.97	6.31	5.85
1.0000	4.9963	5.3180	5.6726	6.22	5.79	5.46

TABLE 5. Excess Molar Volumes (V^E), Viscosity Deviations ($\Delta\eta$) and Isentropic Compressibility Changes ($\Delta\kappa_s$) for the N,N-Dimethylacetamide + 2-Methoxyethanol mixtures at different mole fractions, at three temperatures.

	V^E			$\Delta\eta$ (mPa s)			10^5 $\Delta\kappa_s$ (bar^{-1})		
x (DMA)	298.15K	308.15 K	318.15 K	298.15K	308.15 K	318.15 K	298.15K	308.15 K	318.15 K
0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.0301	-0.0250	-0.0476	-0.0760	0.0109	0.0100	0.0085	-0.0186	-0.0199	-0.0233
0.0600	-0.0480	-0.0820	-0.1325	0.0219	0.0194	0.0165	-0.0371	-0.0393	-0.0430
0.0871	-0.0672	-0.1110	-0.1666	0.0308	0.0275	0.0237	-0.0526	-0.0575	-0.0617
0.1250	-0.0920	-0.1420	-0.1978	0.0413	0.0365	0.0321	-0.0714	-0.0797	-0.0840
0.1767	-0.1215	-0.1725	-0.2240	0.0534	0.0478	0.0422	-0.0913	-0.1053	-0.1113
0.2250	-0.1412	-0.1900	-0.2402	0.0613	0.0555	0.0496	-0.1037	-0.1234	-0.1327
0.2902	-0.1595	-0.2038	-0.2555	0.0679	0.0625	0.0564	-0.1147	-0.1376	-0.1518
0.3640	-0.1682	-0.2100	-0.2662	0.0711	0.0666	0.0606	-0.1211	-0.1438	-0.1628
0.4602	-0.1698	-0.2079	-0.2692	0.0695	0.0664	0.0608	-0.1229	-0.1419	-0.1648
0.5200	-0.1639	-0.2008	-0.2640	0.0661	0.0633	0.0583	-0.1202	-0.1358	-0.1603
0.5611	-0.1572	-0.1937	-0.2575	0.0623	0.0594	0.0552	-0.1166	-0.1291	-0.1540
0.6200	-0.1438	-0.1801	-0.2425	0.0554	0.0530	0.0498	-0.1082	-0.1159	-0.1404
0.6654	-0.1285	-0.1678	-0.2274	0.0481	0.0460	0.0436	-0.0987	-0.1048	-0.1266
0.7250	-0.1078	-0.1484	-0.2002	0.0388	0.0374	0.0360	-0.0841	-0.0890	-0.1070
0.7732	-0.0898	-0.1325	-0.1725	0.0321	0.0310	0.0298	-0.0725	-0.0756	-0.0904
0.8250	-0.0685	-0.1078	-0.1405	0.0245	0.0236	0.0229	-0.0573	-0.0599	-0.0709
0.8847	-0.0443	-0.0768	-0.0998	0.0161	0.0154	0.0150	-0.0374	-0.0403	-0.0470
0.9501	-0.0190	-0.0368	-0.0451	0.0072	0.0065	0.0065	-0.0163	-0.0179	-0.0209
1.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000

TABLE 6. Excess Molar Volumes (V^E), Viscosity Deviations ($\Delta\eta$) and Isentropic Compressibility Changes ($\Delta\kappa_s$) for the N,N-Dimethylacetamide + Water mixtures, at different mole fractions, at three temperatures.

	V^E			$\Delta\eta$ (mPa s)			$10^5\,\Delta\kappa_s$ (bar^{-1})		
x (DMA)	298.15K	308.15 K	318.15 K	298.15K	308.15 K	318.15 K	298.15K	308.15 K	318.15 K
0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.0250	-0.1502	-0.1500	-0.1355	0.3750	0.2740	0.1810	-0.4000	-0.3400	-0.2901
0.0650	-0.4001	-0.3999	-0.3730	0.9795	0.7010	0.4990	-0.8330	-0.7350	-0.6150
0.1011	-0.6298	-0.5998	-0.5730	1.5800	1.0800	0.7750	-1.0740	-0.9050	-0.7620
0.1200	-0.7498	-0.7040	-0.6781	1.8600	1.2900	0.9190	-1.1480	-0.9700	-0.8170
0.1495	-0.9100	-0.8685	-0.8425	2.2800	1.5725	1.1000	-1.2200	-1.032	-0.8720
0.2004	-1.1680	-1.1210	-1.0861	2.7800	1.9210	1.3200	-1.2710	-1.076	-0.9160
0.2512	-1.3600	-1.3275	-1.2810	3.0200	2.0450	1.4195	-1.2705	-1.068	-0.9110
0.3002	-1.4691	-1.4264	-1.4032	3.0400	2.0350	1.4200	-1.2314	-1.028	-0.8735
0.3501	-1.5302	-1.5098	-1.4725	2.7880	1.9125	1.3440	-1.1520	-0.9625	-0.8085
0.4009	-1.5570	-1.5350	-1.5025	2.4600	1.7150	1.2050	-1.0410	-0.8820	-0.7395
0.5018	-1.5110	-1.4800	-1.4660	1.6980	1.1605	0.8930	-0.8180	-0.6900	-0.5700
0.6031	-1.3400	-1.3150	-1.3120	1.0800	0.7900	0.6010	-0.6220	-0.5075	-0.4060
0.6505	-1.2198	-1.2050	-1.2000	0.8600	0.6415	0.4995	-0.5360	-0.4325	-0.3434
0.7005	-1.0872	-1.0750	-1.0740	0.6720	0.5180	0.4000	-0.4500	-0.3590	-0.2875
0.7550	-0.9200	-0.9175	-0.9115	0.4965	0.3900	0.3075	-0.3565	-0.2840	-0.2250
0.8048	-0.7551	-0.7540	-0.7525	0.3600	0.2880	0.2350	-0.2750	-0.2190	-0.1714
0.8501	-0.5990	-0.5920	-0.5995	0.2585	0.2010	0.1650	-0.2030	-0.1660	-0.1270
0.9049	-0.3820	-0.3810	-0.3810	0.1540	0.1150	0.0940	-0.1215	-0.1000	-0.0780
1.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000

TABLE 7. Coefficients of Least –Square Fit by equation (5) for Excess Molar Volumes, Viscosity Deviations and Isentropic Compressibility Changes of N,N-Dimethylacetamide + 2-Methoxyethanol mixtures at 298.15, 308.15 and 318.15 K.

Property	Temp. (K)	A_0	A_1	A_2	A_3	A_4	$\sigma(Y^E)$
V^E	298.15	-0.6672	0.2748	0.0155	-0.0237	0.0458	0.001
	308.15	-0.8147	0.2460	-0.2749	0.2092	-0.1750	0.001
	318.15	-1.0667	0.0977	-0.1514	0.8689	-0.7636	0.003
$\Delta\eta$	298.15	0.2683	-0.1674	0.0054	0.0457	-0.0103	0.001
	308.15	0.2563	-0.1375	-0.0329	0.0277	0.0170	0.001
	318.15	0.2366	-0.1135	-0.0321	0.0321	0.0068	0.000
$\Delta\kappa_s$	298.15	-0.4824	0.1631	-0.1264	0.0172	0.1370	0.002
	308.15	-0.5499	0.2881	-0.1114	-0.1326	0.1586	0.001
	318.15	-0.6518	0.2322	0.0729	-0.0523	-0.0275	0.001

TABLE 8. Coefficients of Least –Square Fit by equation (5) for Excess Molar Volumes, Viscosity Deviations and Isentropic Compressibility Changes of N,N-Dimethylacetamide + Water mixtures at 298.15, 308.15 and 318.15 K.

Property	Temp. (K)	A_0	A_1	A_2	A_3	A_4	$\sigma(Y^E)$
V^E	298.15	-6.0140	2.5359	-0.8871	-1.5370	2.1658	0.009
	308.15	-5.9784	2.3413	-0.1007	-1.4837	1.1110	0.011
	318.15	-5.8716	2.1625	-0.3684	-1.5317	1.6935	0.007
$\Delta\eta$	298.15	7.0443	-15.3472	12.1404	8.5762	-12.6279	0.035
	308.15	4.9385	-9.9039	8.0347	4.8343	-8.0417	0.028
	318.15	3.5502	-6.6516	6.0496	3.0762	-6.4072	0.011
$\Delta\kappa_s$	298.15	-3.4297	3.7710	-2.3595	4.2384	-4.3034	0.022
	308.15	-2.8559	3.2534	-1.8254	3.5924	-4.0838	0.019
	318.15	-2.3793	2.8886	-1.4153	2.8869	-3.6520	0.018

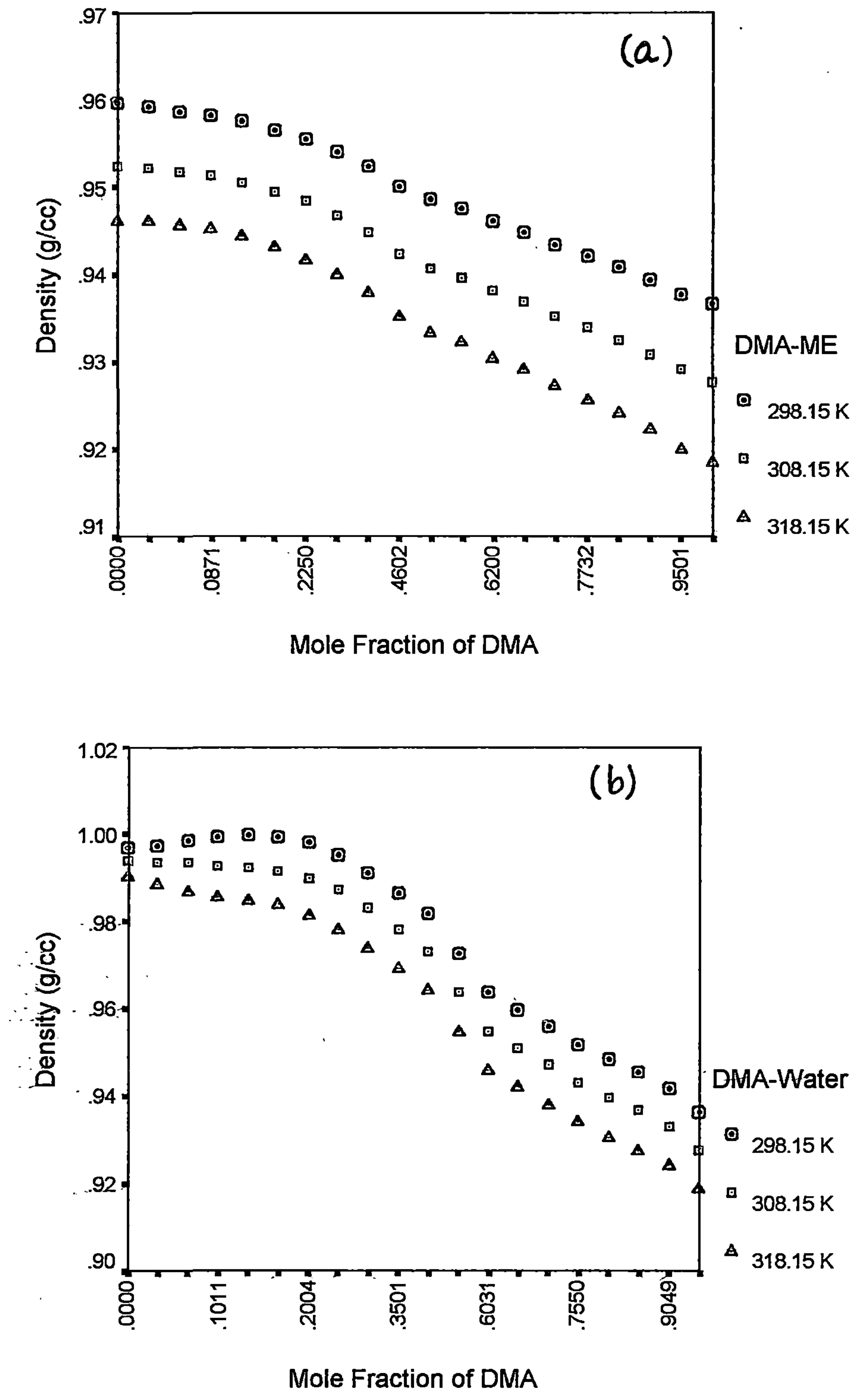


Figure 1. (a) Density (ρ) versus mole fraction of DMA in DMA + ME mixture at three temperatures.

(b) Density (ρ) versus mole fraction of DMA in DMA + Water mixture at three temperatures.

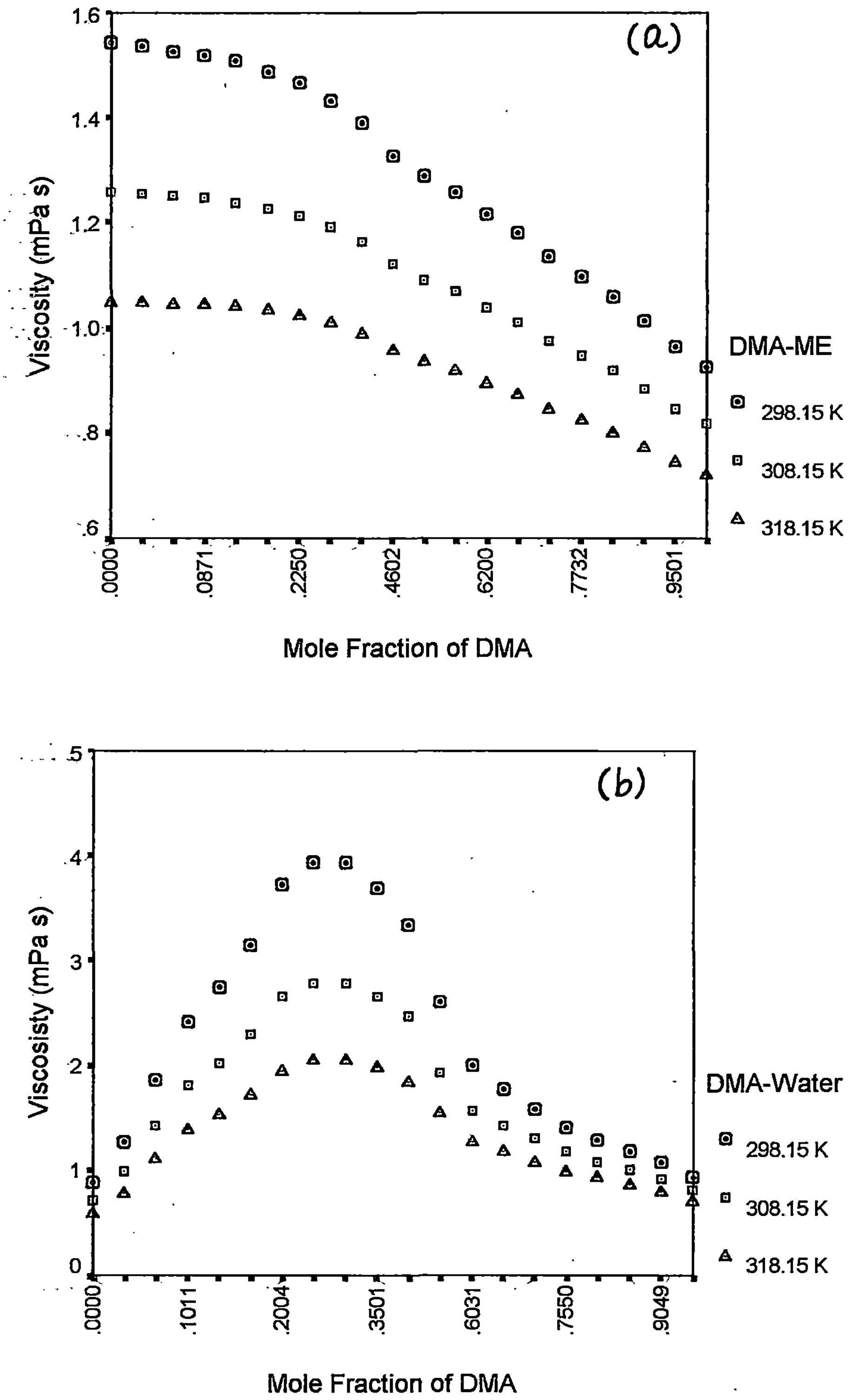


Figure 2. (a) Viscosity (η) versus mole fraction of DMA in DMA + ME mixture at three temperatures.
(b) Viscosity (η) versus mole fraction of DMA in DMA + Water mixture at three temperatures.

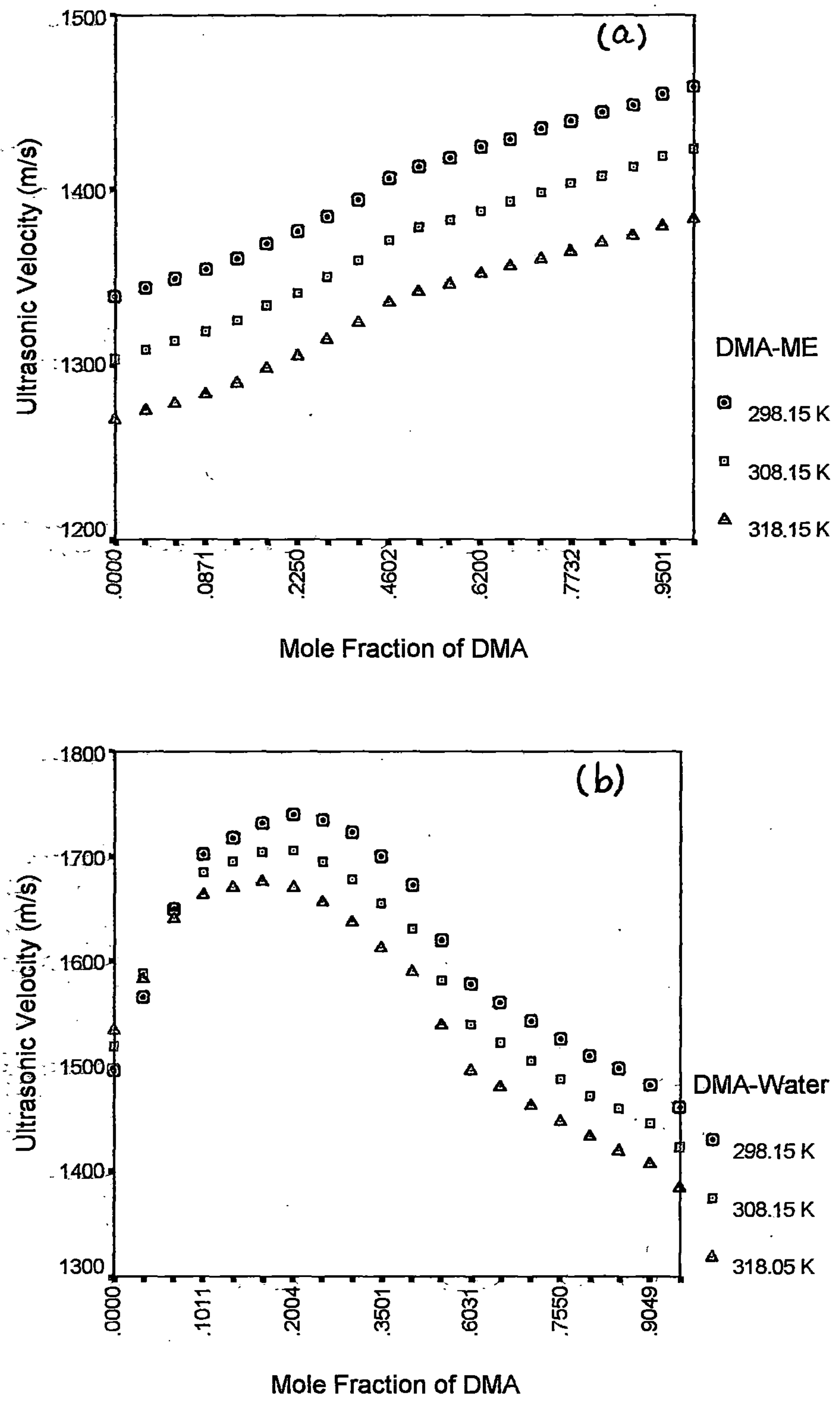


Figure 3. (a) Ultrasonic Velocity (u) versus mole fraction of DMA in DMA + ME mixture at three temperatures.

(b) Ultrasonic Velocity (u) versus mole fraction of DMA in DMA + Water mixture at three temperatures.

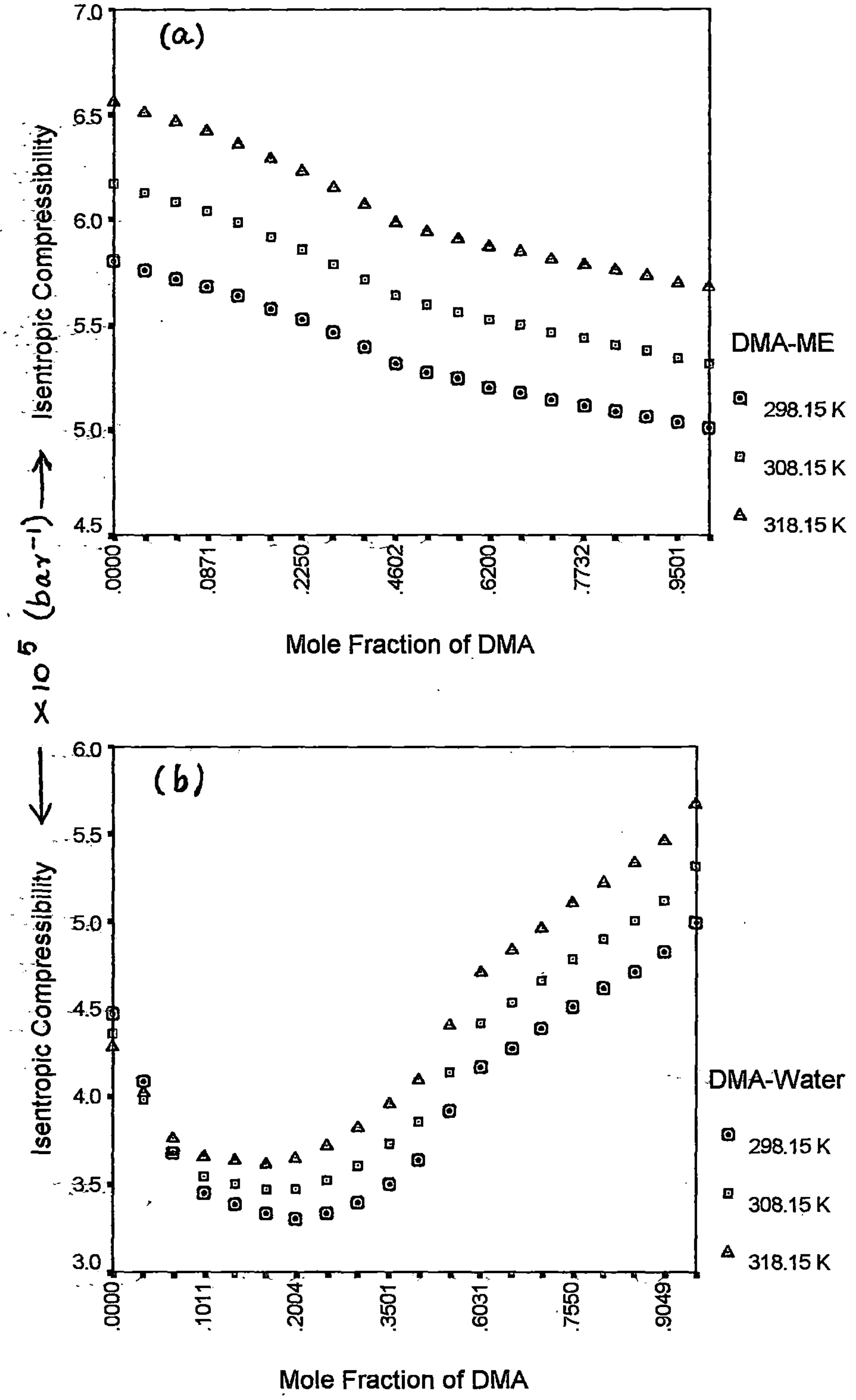


Figure 4. (a) Isentropic Compressibility (κ_s) versus mole fraction of DMA in DMA + ME mixture at three temperatures.

(b) Isentropic Compressibility (κ_s) versus mole fraction of DMA in DMA + Water mixture at three temperatures.

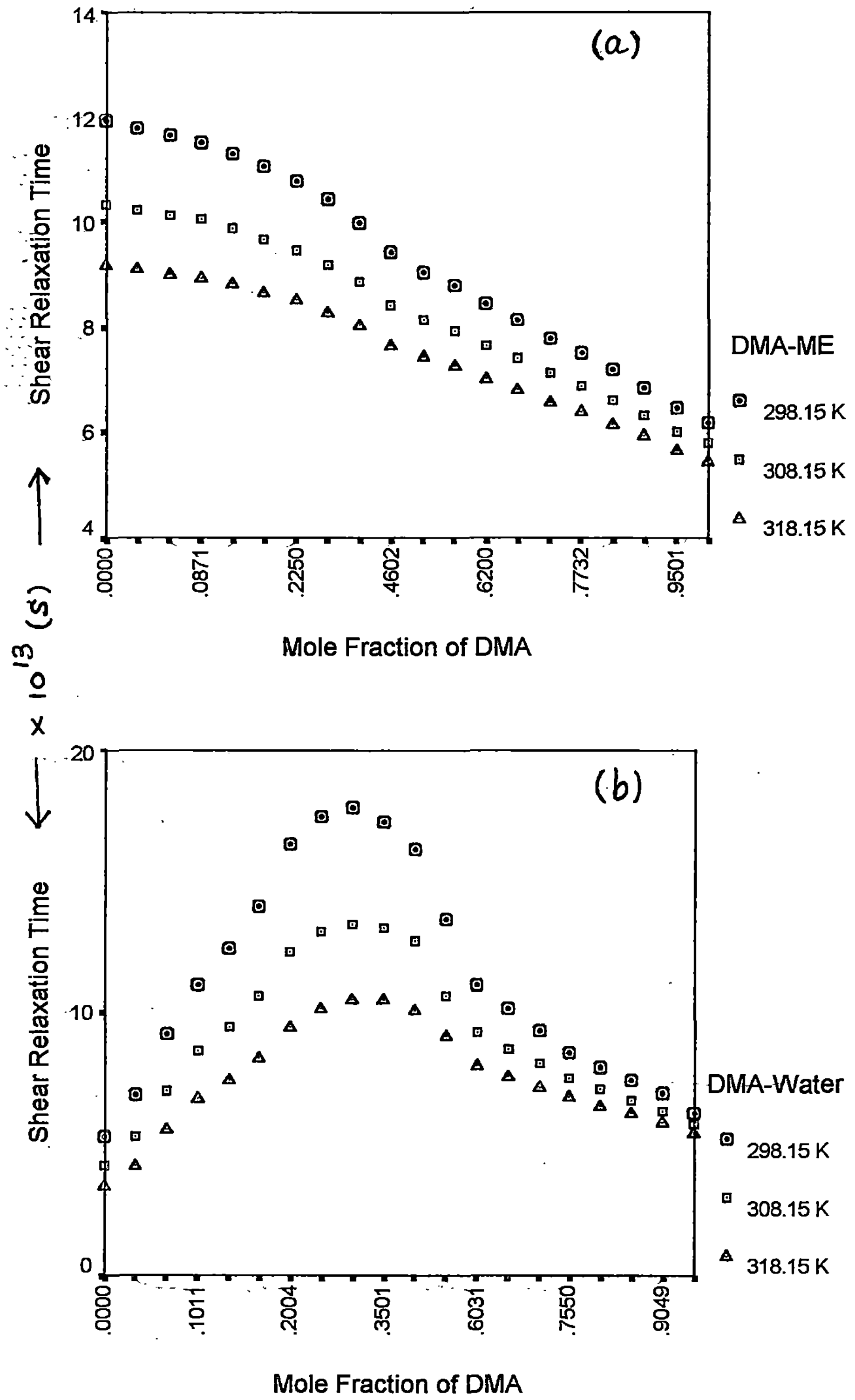
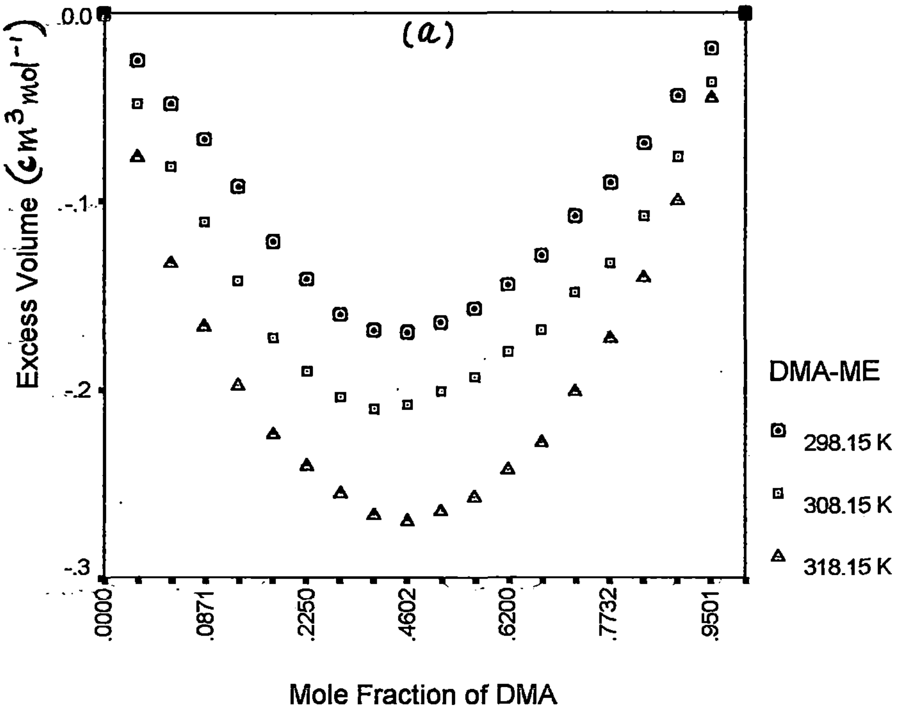


Figure 5. (a) Shear Relaxation Time (τ) versus mole fraction of DMA in DMA + ME mixture at three temperatures.

(b) Shear Relaxation Time (τ) versus mole fraction of DMA in DMA + Water mixture at three temperatures.



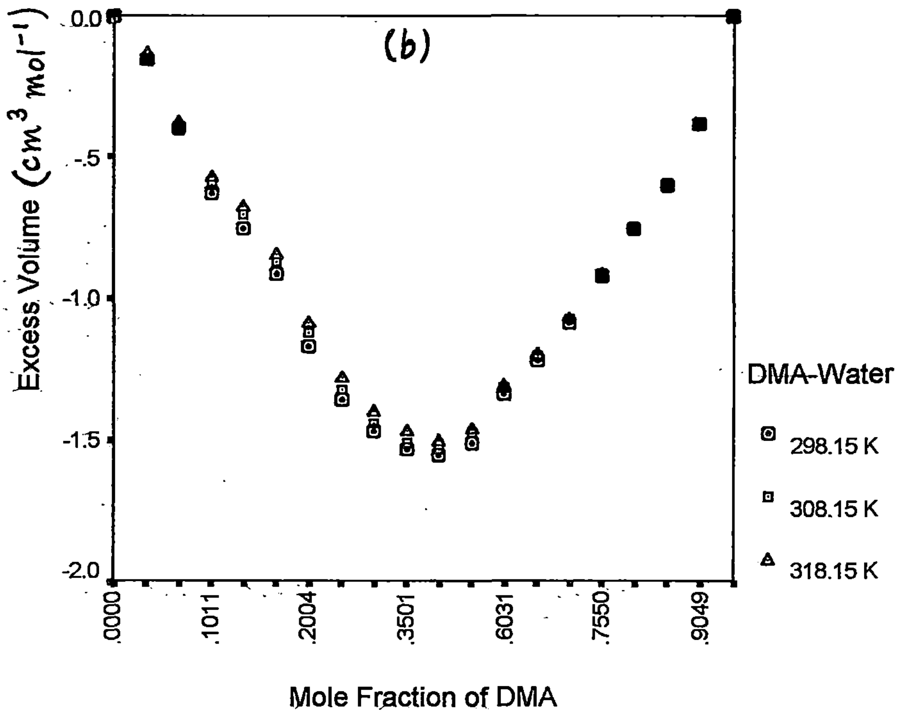


Figure 6. (a) Excess Molar Volume (V^E) versus mole fraction of DMA in DMA + ME mixtures at three temperatures.
(b) Excess Molar Volume (V^E) versus mole fraction of DMA in DMA + Water mixtures at three temperatures.

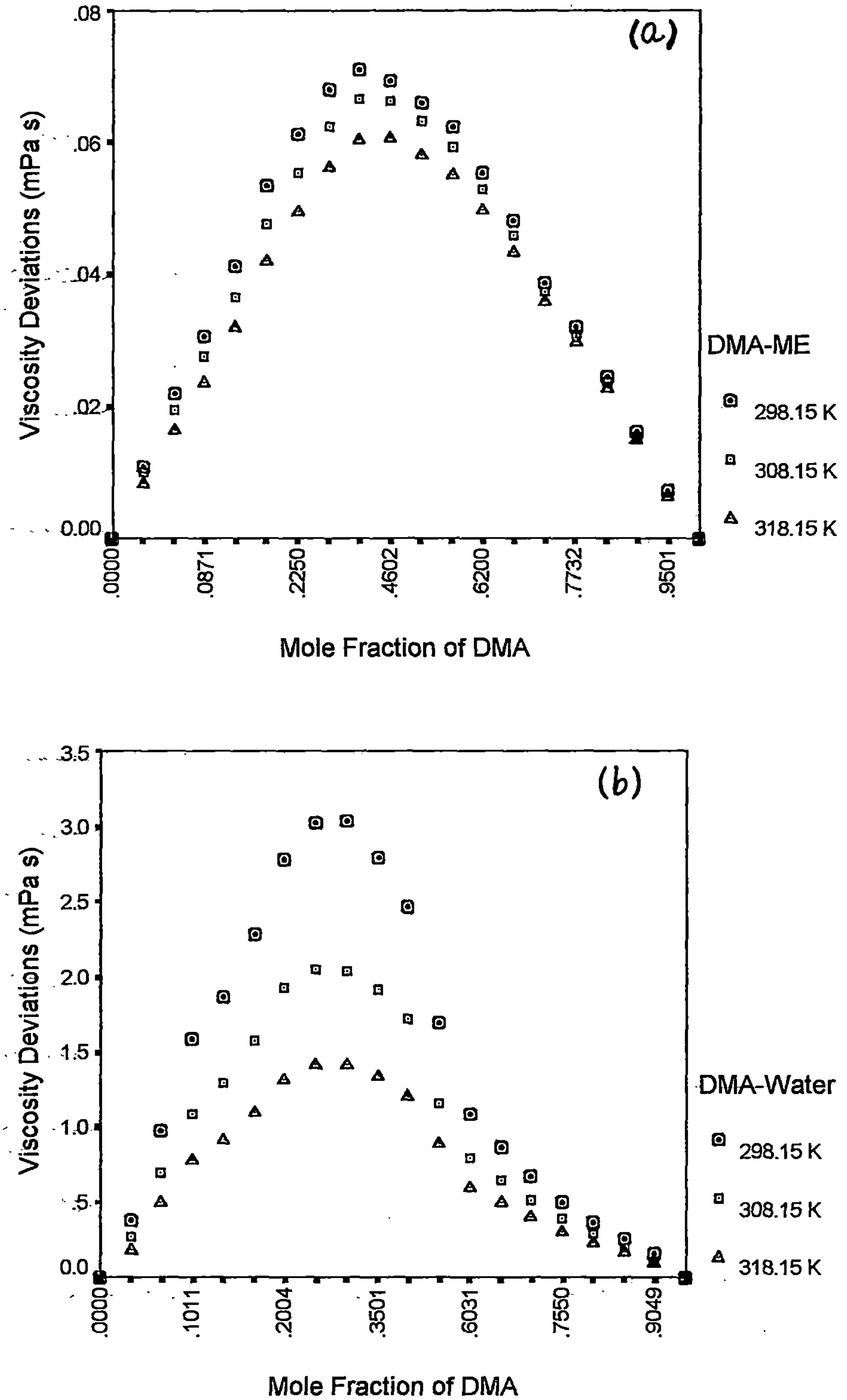


Figure 7. **(a) Viscosity Deviations (Δη) versus mole fraction of DMA in DMA + ME mixture at three temperatures.**

(b) Viscosity Deviations (Δη) versus mole fraction of DMA in DMA + Water mixture at three temperatures.

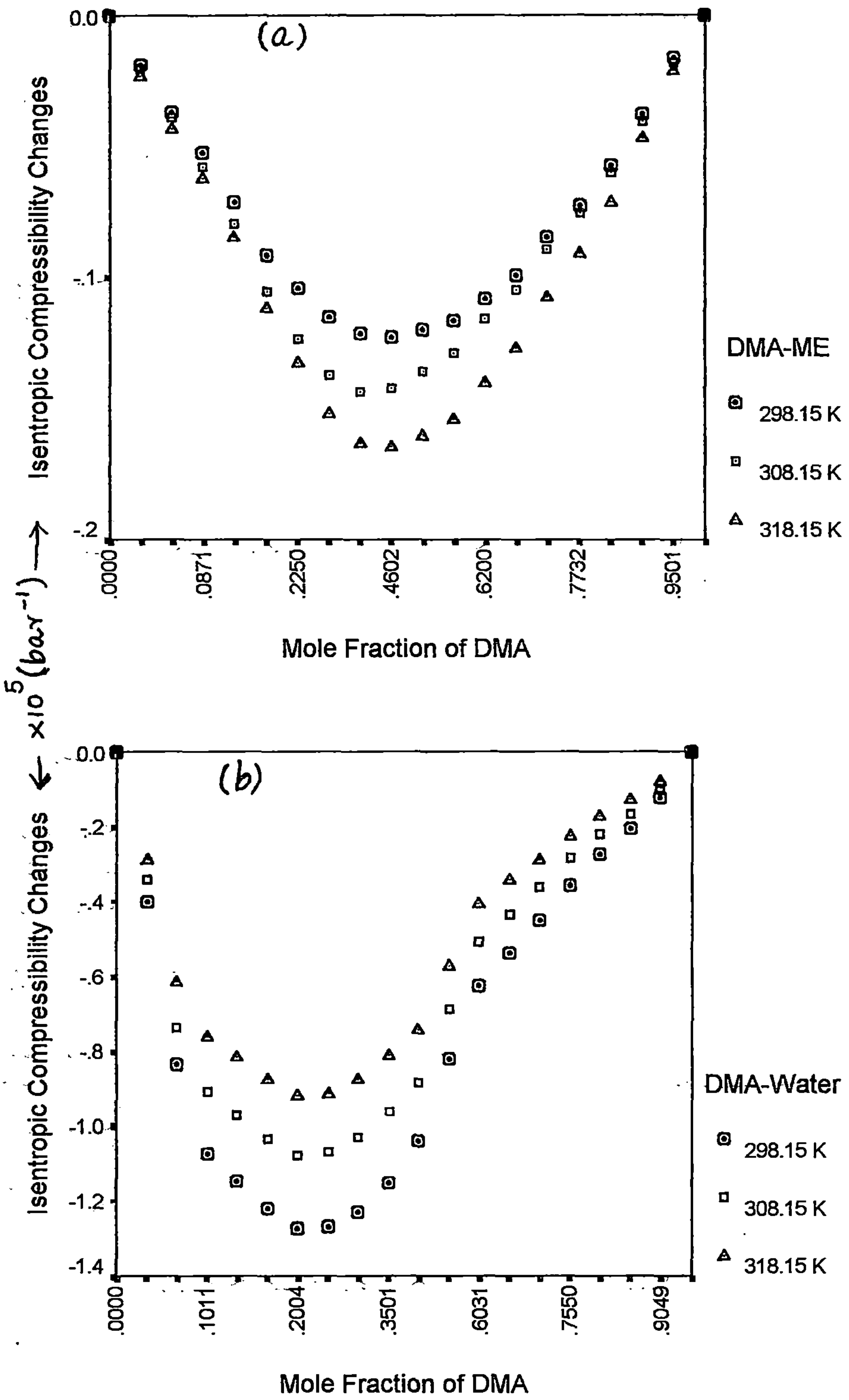


Figure 8. (a) Isentropic Compressibility Changes ($\Delta\kappa_s$) versus mole fraction of DMA in DMA + ME mixture at three temperatures.

(b) Isentropic Compressibility Changes ($\Delta\kappa_s$) versus mole fraction of DMA in DMA + Water mixture at three temperatures.