



## THERMODYNAMIC PARAMETERS FOR BINARY MIXTURES OF DMF AND DMSO AT VARIOUS COMPOSITIONS.

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**ABSTRACT** Density and viscosity of DMF, DMSO and their binary mixtures have been measured as a function of their weight percent composition at different temperature. Excess volume  $V_E$ , deviation in viscosity  $\Delta\eta$ , excess Gibb's free energy  $\Delta G^*E$ ,  $W_{vis}$ ,  $d_{12}$ ,  $H_{12}$  and  $T_{12}$  parameters have been calculated as a function of composition (DMF-DMSO system) over the temperature range from 298.15 K to 313.15 K. The main thrust of the study is to co-relate the excess properties and the relevant interaction parameters with the nature of molecular interactions between the mixing components.

**KEYWORDS :** Tobacco smoking, Nicotine dependence, Intention to quit smoking

### INTRODUCTION

Mixed solvents, rather than single pure liquids, are extensively used in industrial and chemical processes. The physical property data on mixed solvents are important for theoretical and applied areas of research and extensively used to design new bulk processes and process equipments. Experimental and theoretical studies of solvent-solvent interaction in mixed solvent systems are very important to understand molecular interaction either in aqueous or non-aqueous medium [1-5].

Dimethyl sulfoxide (DMSO; mol. wt. = 78.13 g/mol, B.P. = 189°C (372°F), sp.gr.= 1.0955 g/cm<sup>3</sup> at 25°C) is a widely used solvent capable of dissolving both hydrophobic and hydrophilic solutes [6]. DMSO is a poly functional molecule with a highly polar S=O group and two hydrophobic -CH<sub>3</sub> groups. Its polar site can interact with water forming strong hydrogen bonds, and its non-polar site can affect hydrophobic hydration and hydrophobic association of DMSO molecules. Brown [7] on the basis of solid state X-ray spectroscopy data proposed that DMSO molecule can bind to anions by forming hydrogen bond between its methyl hydrogen and free electron pair in anion. The self-association is found to influence the S=O stretching vibration. The strong basic character of DMSO makes it an excellent probe to study H-bond interactions. DMSO is a widely used cryoprotectant for biological structures such as membranes and proteins [8]. The behavior of DMSO molecules in its associated forms and in the monomeric state is of very much interest.

Dimethyl formamide (DMF; mol. wt. = 73.095 g/mol, B. P. = 153°C (307°F), sp. gr. = 0.9445 g/cm<sup>3</sup> at 25°C) is a colorless, polar, high-boiling, non-hydrogen bonded liquid. It is freely miscible with water, alcohols, ethers, ketones, esters, carbon disulfide, and chlorinated and aromatic hydrocarbons. It is either immiscible or only partly miscible with aliphatic hydrocarbons. DMF have very little tendency to hydrolyze. DMF is an aprotic protophilic, potentially basic medium, and important to obtain information about protein systems. DMF is used as industrial solvent and in a variety of industrial processes, for the manufacture of synthetic fibers, leathers, films, and surface coatings [9]. DMF is a stable compound with a strong electron – pair donating and accepting ability and widely used in settings such as solvent reactivity relationships [10–13]. DMF is of particular interest because any significant structural effects are absent due to the lack of

hydrogen bonds and applied as an aprotic as well as protophilic solvent. It has large dipole moment ( $\mu = 3.86$  D at 25°C), a high dielectric constant (37.51 at 25°C) [14, 15] and good donor–acceptor properties.

Thus, a study of thermodynamic properties on the binary mixture of DMF and DMSO has attracted considerable interest in our present study.

### Experimental:

**Materials:** The chemicals DMSO and DMF employed were of analytical grade and were purchased from E. Merck, Germany (99.5%), were used as such without further purification.

**Density measurements:** The bi-capillary pycnometer was calibrated by measuring the densities of triple distilled water. The densities of distilled organic liquids like acetone, alcohol, benzene, carbon tetra chloride, aniline, and nitrobenzene were evaluated with respect to density of water. The density was measured with an uncertainty of  $\pm 1.48 \times 10^{-4}$  g.cm<sup>-3</sup>.

**Viscosity measurements:** The viscosities of pure liquids and their binary mixtures were measured with an uncertainty of  $\pm 2.4 \times 10^{-4}$  mPa.s by using by using Ubbelohde viscometer. The viscometer was calibrated with triply distilled water. The viscometer containing the test liquid was allowed to stand for about 30 minutes in a thermostatic water bath so that the thermal fluctuations in the viscometer were minimized. The temperature of thermostat was maintained to desired temperature, by using demerstat with an accuracy of  $\pm 0.1$  K. The times of flow were recorded with a digital stopwatch with an accuracy of  $\pm 0.01$  second.

The viscosities of both the pure liquids and their mixtures were measured from 298.15 K to 313.15 K at atmospheric pressure with an Ubbelohde viscometer.

### Results and Discussion:-

1) The densities and viscosities of mixtures of various compositions are listed in Table 1. It is observed that the density as well as viscosity increases with increase in mole fraction of DMSO and decreases with increases temperature. This might be due to strengthening of solvent-solvent interactions.

**Table: 1.** Experimental density  $d^0$  and Experimental viscosity  $\eta^0$  (N.s.m<sup>-2</sup>) for the DMSO- DMF mixtures at different temperatures.

DMSO in Wt %	Mole Fraction of DMSO	Experimental density $d_0$				Experimental viscosity $\eta_0$			
		Temperature (K)				Temperature (K)			
		298.15	303.15	308.15	313.15	298.15	303.15	308.15	313.15
0	0.0000	0.94402	0.94151	0.93527	0.93123	0.80384	0.76272	0.71602	0.68260
10	0.1142	0.96045	0.95787	0.95142	0.94738	0.87578	0.83459	0.77818	0.76154
20	0.2248	0.97682	0.97372	0.96731	0.96341	0.94409	0.88912	0.82784	0.80053
30	0.3321	0.99410	0.99045	0.98397	0.97961	1.02923	0.98875	0.90606	0.86483
40	0.4361	1.00974	1.00623	0.99994	0.99559	1.12740	1.08263	0.98816	0.93672
50	0.5371	1.02658	1.02197	1.01570	1.01186	1.21692	1.11941	1.03828	0.99650
60	0.6351	1.04024	1.03547	1.02966	1.02600	1.37410	1.27497	1.14399	1.09309

70	0.7302	1.05372	1.04966	1.04353	1.04044	1.55119	1.40813	1.26492	1.20680
80	0.8227	1.06752	1.06346	1.05739	1.05421	1.68133	1.52514	1.37928	1.30945
90	0.9126	1.08105	1.07743	1.07157	1.06851	1.84024	1.65899	1.49430	1.41197
100	1.0000	1.09513	1.09124	1.08548	1.08267	1.99070	1.79717	1.61501	1.51078

1) The excess molar volume  $V^E$  of binary liquid mixtures is evaluated using the following equation.

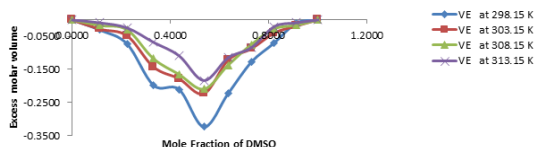
$$V^E = \left( \frac{X_1 M_1 + X_2 M_2}{d_{12}} \right) - \left( \frac{X_1 M_1}{d_1} + \frac{X_2 M_2}{d_2} \right)$$

Where  $x_1$  and  $x_2$  are the mole fraction of components 1 (DMSO) and 2 (DMF) respectively.  $M_1$  and  $M_2$  are the molar mass of components 1 and 2 respectively.  $d_{12}$  is the density of binary liquid mixtures,  $d_1$  and  $d_2$  are the density of component 1 and component 2 respectively.

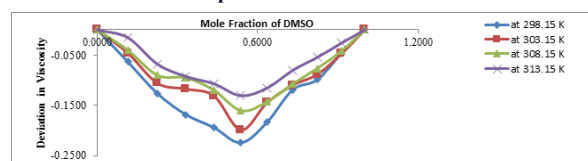
Table 2 shows the values of  $V^E$  for DMSO and DMF mixtures over the whole composition range studied at different temperatures. The excess molar volume show negative deviation over the whole mole fraction range for binary mixtures of DMSO and DMF at different temperatures. The excess molar volumes give the important information for physical, chemical, and geometrical contributions [16]. The variation of  $V^E$  with mole fraction of DMSO at four temperatures is shown in Figure 1. The sign and magnitude of  $V^E$  play vital role in assessing for the molecular rearrangement as a result of molecular interactions between component molecules in the liquid mixtures [17]. The negative  $V^E$  values indicate the presence of strong intermolecular interaction between unlike molecules. The negative values of  $V^E$  over the entire mole fraction of DMSO in mixture may be attributed mainly due to the association through intermolecular hydrogen bonds between the oxygen atom of sulfoxide group and the hydrogen atom of amide group. Another effect is the differences in

**Table: 2. Excess volume  $V^E$  ( $\text{cm}^3 \text{mol}^{-1}$ ) and Deviation in viscosity  $\Delta\eta$  ( $\text{kg/m}^3$ ) for the System DMSO-DMF mixtures at different temperatures.**

DMSO in Wt %	Mole Fraction of DMSO	Excess volume $V^E$ Temperature (K)				Deviation in viscosity $\Delta\eta$ Temperature (K)			
		298.15	303.15	308.15	313.15	298.15	303.15	308.15	313.15
10	0.1142	-0.0307	-0.0280	-0.0170	-0.0089	-0.0636	-0.0463	-0.0405	-0.0156
20	0.2248	-0.0728	-0.0480	-0.0298	-0.0242	-0.1266	-0.1062	-0.0903	-0.0683
30	0.3321	-0.1980	-0.1425	-0.1175	-0.0678	-0.1688	-0.1175	-0.0953	-0.0928
40	0.4361	-0.2106	-0.1782	-0.1649	-0.1087	-0.1941	-0.1312	-0.1199	-0.1071
50	0.5371	-0.3240	-0.2234	-0.2096	-0.1835	-0.2243	-0.1989	-0.1605	-0.1309
60	0.6351	-0.2232	-0.1219	-0.1370	-0.1148	-0.1835	-0.1447	-0.1429	-0.1154
70	0.7302	-0.1275	-0.0872	-0.0743	-0.0839	-0.1193	-0.1100	-0.1076	-0.0806
80	0.8227	-0.0698	-0.0398	-0.0269	-0.0204	-0.0990	-0.0886	-0.0763	-0.0545
90	0.9126	-0.0098	-0.0183	-0.0162	-0.0089	-0.0467	-0.0478	-0.0421	-0.0264



**Figure: 1- Plot of  $V^E$  Excess molar volume versus mole fraction of DMSO at different temperatures**



**Figure: 2- Plot of deviation in viscosities verses mole fraction of DMSO at different temperatures.**

On mixing, a DMSO molecule would induce a small dipole moment in the aromatic hydrocarbon molecule, which might lead to weak dipole-induced dipole interaction between the component molecules, in addition to donor-acceptor interaction between  $\pi$ -electrons of benzene ring of the hydrocarbons and highly electronegative oxygen of S=O group of DMSO, resulting in negative  $\Delta\eta$  values. Negative deviation in  $\Delta\eta$  values may also be observed due to the difference in the molecular size of the component molecules, [20, 21] as in the present mixtures.

4. The excess Gibb's free energy of activation of flow  $\Delta G^{\ddagger}$  for the binary liquid mixtures were calculated from the equation.

molecular sizes between the two components in binary mixtures. The difference in free volumes between two components in binary mixtures would facilitate the penetration of one component into the other. The decrease in negative values of  $V^E$  with increase in temperature indicates the decrease in strength of molecular interaction with temperature.

3) The deviation in viscosity was calculated from the following equation.

$$\eta \Delta\eta = \eta_1 \eta_2 - (X_1 M_1 + X_2 M_2)$$

Where  $\eta_1$  and  $\eta_2$  are viscosities of component 1 and component 2 respectively. Table 2 also summarizes the deviation in viscosity ( $\Delta\eta$ ) in DMSO + DMF mixture and decreases with increase in temperature. The negative values of DMSO with DMF will induce the partial dissociation and loosening of dipolar interaction between DMSO molecules.

Thus, the apparent intermolecular interaction in DMSO + DMF mixture may be due to weak dipole induced forces resulting from the polarization of DMSO molecules by the dipoles of the surrounding DMF molecules. This effect can be superimposed upon dipole-dipole interaction and lead to a slight increase in attraction giving less negative  $\Delta\eta$  values at higher temperatures. The negative  $\Delta\eta$  values are generally observed [18, 19] for the systems where dispersion or weak dipole-dipole forces are primarily responsible for the interaction between the component molecules.

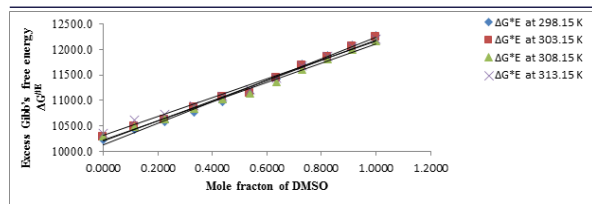
$$\Delta G^{\ddagger} = RT [\ln(\eta V) - X_1 \ln(\eta_1 V_1) - X_2 \ln(\eta_2 V_2)] \quad (3)$$

Where  $V_1$ ,  $V_2$  and  $V$  are the molar volume of DMSO, DMF and their mixture respectively.  $\eta_1$ ,  $\eta_2$  and  $\eta$  are the viscosities of DMSO, DMF and their mixture respectively. R and T have their usual meanings.

It can be observed from Table 4 that the  $\Delta G^{\ddagger}$  values are positive for all mixtures over entire composition range at all temperatures. The values  $\Delta G^{\ddagger}$  increases with increase in mole fraction of DMSO at different temperature.

**Table: 4. Excess Gibb's free energy  $\Delta G^{\ddagger}$  ( $\text{kJ.mole}^{-1}$ ) for DMSO-DMF mixtures at various temperatures.**

DMSO in Wt %	Mole Fraction of DMSO	Excess Gibb's free energy $\Delta G^{\ddagger} \times 10^3$ Temperature			
		298.15 K	303.15 K	308.15 K	313.15 K
0	0.0000	10.2305	10.2767	10.3015	10.3559
10	0.1142	10.4198	10.4802	10.4912	10.6166
20	0.2248	10.5827	10.6173	10.6266	10.7224
30	0.3321	10.7712	10.8602	10.8326	10.8991
40	0.4361	10.9755	11.0664	11.0313	11.0829
50	0.5371	11.1404	11.1284	11.1351	11.2191
60	0.6351	11.4245	11.4393	11.3649	11.4406
70	0.7302	11.7083	11.6709	11.6039	11.6779
80	0.8227	11.8905	11.8541	11.8070	11.8716
90	0.9126	12.0973	12.0477	11.9927	12.0477
100	1.0000	12.2735	12.2310	12.1727	12.2038



**Figure 3-Plot of Excess Gibbs Free Energy  $\Delta G^*E$  versus mole fraction of DMSO at different temperatures.**

The  $\Delta G^{*E}$  values [22] were positive for the mixtures indicate a sign of strong association by H-bonding or as a sign of fitting of one molecule into the cavities of the other molecule.

5) The several models as well as equations have been put forward to study the molecular interaction between components in binary mixtures. Katti-Chaudhary [23] was suggested the following equation.

$$\ln(\eta_m V_m) = X_1 \ln(\eta_1 V_1) + X_2 \ln(\eta_2 V_2) + (X_1 X_2 W_{vis}/RT) \quad (4)$$

$W_{vis}$  is the interchange energy and may be regarded as an approximate measure of interaction parameters.  $X_1$  and  $X_2$  are the mole fractions of component 1 and component 2 respectively.  $\eta_1$  and  $\eta_2$  are the viscosity of the component 1 and component 2 respectively.  $V_1$  and  $V_2$  are volume of pure component 1 component 2 respectively.

$W_{vis}$  is the interchange energy and may be regarded as an approximate

**Table 5.  $W_{vis}$  and Gruenberg and Nissan's parameters for DMSO-DMF water system at various temperatures.**

DMSO in Wt %	Mole Fraction of DMSO	$W_{vis}$				Gruenberg and Nissan's parameter ( $d_{12}$ )			
		Temperature (K)				Temperature (K)			
		298.15	303.15	308.15	313.15	298.15	303.15	308.15	313.15
10	0.1142	1791.30	2137.52	2300.51	3023.59	-17.1833	-18.3516	-18.5653	-17.6606
20	0.2248	3086.53	3238.93	3412.96	3621.64	-10.8113	-11.5653	-11.7580	-11.1555
30	0.3321	3341.44	3768.42	3795.17	3805.11	-9.0726	-9.6472	-9.8572	-9.2535
40	0.4361	3394.15	3832.31	3866.63	3810.80	-8.6585	-9.1945	-9.4119	-8.8233
50	0.5371	3163.52	3219.06	3453.15	3546.89	-9.0688	-9.7074	-9.8726	-9.4048
60	0.6351	3303.15	3509.21	3436.36	3515.52	-10.0503	-10.8411	-11.0375	-10.3734
70	0.7302	3317.13	3321.07	3294.17	3407.52	-12.1750	-13.2106	-13.4494	-12.6662
80	0.8227	2329.94	2364.91	2480.24	2644.38	-17.1587	-18.5163	-18.8725	-17.8271
90	0.9126	-1002.96	-1096.03	-986.65	-605.22	-32.4978	-35.1084	-35.8186	-33.7977

Hind et al. [27] have suggested the following equation for the viscosity of binary liquid mixtures.

$$\eta_1 = X_1 \eta_1 + X_2 \eta_2 + 2X_1 X_2 H_{12} \quad (7)$$

It can be seen from Table 6 that the interaction parameters  $H_{12}$  and  $T_{12}$

**Table 6.  $H_{12}$  and  $T_{12}$  parameters for DMSO-DMF system at various temperatures.**

DMSO in Wt %	Mole Fraction of DMSO	$H_{12}$ parameters				$T_{12}$ parameters			
		Temperature (K)				Temperature (K)			
		298.15	303.15	308.15	313.15	298.15	303.15	308.15	313.15
10	0.1142	1.08275	1.05101	0.96544	1.01946	1.1120	1.0791	0.9899	1.0486
20	0.2248	1.03402	0.97539	0.90652	0.90076	1.0662	1.0040	0.9318	0.9260
30	0.3321	1.01683	1.01511	0.92097	0.88751	1.0543	1.0497	0.9508	0.9155
40	0.4361	1.00271	1.01314	0.92172	0.87901	1.0459	1.0519	0.9553	0.9101
50	0.5371	0.94613	0.88001	0.84266	0.83348	0.9953	0.9231	0.8802	0.8680
60	0.6351	1.00147	0.96779	0.85717	0.84762	1.0563	1.0152	0.8991	0.8857
70	0.7302	1.09440	1.00077	0.89251	0.89219	1.1519	1.0516	0.9377	0.9324
80	0.8227	1.05809	0.97608	0.90383	0.90985	1.1214	1.0320	0.9523	0.9521
90	0.9126	1.10446	0.98044	0.90148	0.93117	1.1696	1.0404	0.9535	0.9748

Where  $H_{12}$  is Hind interaction parameter and attributed to unlike pair interactions.  $X_1$  and  $X_2$  are the mole fraction of component 1 and component 2 respectively.  $\eta_1$  and  $\eta_2$  are the viscosity of component 1 and component 2 respectively.

#### Conclusion:-

Density, Excess molar volume  $V^E$ , Deviation in viscosity  $\Delta\eta$ , Excess Gibbs free energy  $\Delta G^{*E}$ ,  $W_{vis}$ ,  $d_{12}$ ,  $H_{12}$  &  $T_{12}$  parameter for the system DMSO-DMF have been measured over the entire range of composition at Temperature 308.15 K. The behavior of binary mixture can be explained in term of 1) physical force-dispersion 2) chemical force-dipole-dipole interaction. For binary mixture DMSO and DMF imply that the specific chemical dipole-dipole interactions between

measure of interaction parameters and are listed in Table 5. All the observed values of  $W_{vis}$  values are positive.

The evaluated Grunberg-Nissan interaction parameters are also listed in Table 5 and are found negative, which suggest weak interactions between unlike molecules [24]. Grunberg and Nissan [25] have suggested the following equation (5) which shows relation between the viscosity of the binary liquid mixture and pure components. According to Grunberg and Nissan, the adjustable binary parameter measure of the strength of interactions between the mixing species.

$$\ln \eta = X_1 \ln \eta_1 + X_2 \ln \eta_2 + X_1 X_2 d_{12} \quad (5)$$

Where  $d_{12}$  is a constant, proportional to interchange energy, called as Grunberg and Nissan parameter,  $X_1$  and  $X_2$  are the mole fraction of component 1 and component 2 respectively. The  $\eta_1$ ,  $\eta_2$  and  $\eta$  are the dynamic viscosities of component 1 component 2 and binary mixtures respectively.

6) Tamura and Kurata [26] have developed the following equation for the viscosity of binary liquid mixtures.

$$\eta = [X_1 \Phi_1 \eta_1 + X_2 \Phi_2 \eta_2 + 2X_1 X_2 \Phi_1 \Phi_2]^{0.5} T_{12} \quad (6)$$

Where  $T_{12}$  is the interaction parameter which is depends upon temperature and composition of the mixture.  $X_1$  and  $X_2$  are the mole fractions of component 1 and component 2 respectively.  $\eta_1$  and  $\eta_2$  are the viscosity of the component 1 and component 2 respectively.  $\Phi_1$  and  $\Phi_2$  are volume fractions of component 1 component 2 respectively.

are positive for DMSO-DMF mixtures at four different temperatures. The  $H_{12}$  and  $T_{12}$  parameters are positive. The positive value of  $H_{12}$  and  $T_{12}$  support the existence of weak interactions and dispersion forces. The  $H_{12}$  and  $T_{12}$  parameters are small and positive, which suggests that the weak interaction between DMSO and DMF.

unlike molecules dominate over the physical dispersive interaction between like molecules.

The values of  $\Delta G^{*E}$  increases with increase in the mole fraction of DMSO in the mixtures. This suggests that the formation of activated species necessary for viscous flow appears easy in an aromatic hydrocarbon rich region and becomes difficult as the concentration of DMSO in the mixture increases.

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