

Viscosities and Densities of Binary Liquid Mixtures of Dimethyl Sulfoxide with Chlorobenzene, Pyridine, and Methyl Ethyl Ketone at 25, 35, 45, and 55 °C

G. S. Gokavi, J. R. Raju, T. M. Aminabhavi,* R. H. Balundgi, and M. V. Muddapur†

Department of Chemistry, Karnatak University, Dharwad 580003, India

Density and viscosity data are presented for three binary solvent mixtures of dimethyl sulfoxide with chlorobenzene, pyridine, or methyl ethyl ketone at 25, 35, 45, and 55 °C over the whole range of compositions. The experimental data of viscosities are fitted to theoretical relations proposed by McAllister, Auslander, and Heric and also to a polynomial equation. An attempt is also made to predict thermodynamic functions such as excess volume, excess enthalpy, and excess entropy.

Introduction

Due to the recent developments made in the theories of liquid mixtures and experimental techniques, the study of binary liquid mixtures has attracted several researchers in the field (1). A familiar approach is the hypothesis that there is a direct correlation between the viscosity and the thermodynamic behavior of the solution. Quite a few attempts have been made in the literature from time to time to theoretically predict viscosities of binary liquid mixtures (2-5). Among these, theoretical relations proposed by McAllister (2), Auslander (3), and Heric (4, 5) and a polynomial relation (6) have been used extensively to calculate viscosities of binary mixtures.

The present investigation is a continuation of the effort to understand the transport properties of binary liquid mixtures (6, 7). Among the nonequilibrium properties of liquid mixtures, viscosity has been studied here in order to examine several empirical relations. The closeness of the fit between each of the above relations and the experiment is examined. The binary mixtures consisting of dimethyl sulfoxide (Me₂SO) with others like chlorobenzene (CB), pyridine (PY), and methyl ethyl ketone (MEK) were selected and their viscosities and densities at 25, 35, 45, and 55 °C were measured. The solvent dimethyl sulfoxide was particularly chosen due to its high polarity and its importance as a versatile solvent in synthetic chemistry (8, 9). The other components like chlorobenzene, pyridine, and methyl ethyl ketone range from low to high polarity. An attempt has also been made to compute excess thermodynamic functions such as volume, entropy, enthalpy, and free energy of mixing; these results are discussed in relation to the nature of mixing species.

Experimental Section

All the solvents used were of reagent grade and were purified by standard procedures (10). Me₂SO, CB, and MEK were Sisco samples and pyridine was a BDH sample. The purity of each liquid was checked (11) by its density, viscosity, and refractive index (see Table I).

Solvent mixtures were prepared over the entire range of 0-100 vol %. In the first and the last step, the increase in composition was 5% and over the remaining middle range, the increases were in steps of 10%. All the binary mixtures studied consisted of Me₂SO as the first solvent and one of the other

three liquids as the second solvent. To minimize preferential evaporation of one of the solvents in the mixture, each binary mixture was prepared on the same day on which it was studied. The mole fractions (X_i) of the liquids in the binary mixtures were calculated from the volumes and densities.

The densities of pure liquids and binary mixtures at different temperatures were measured with a pycnometer. The viscosities were obtained with an Ostwald viscometer by comparing the flow times of a pure liquid or a binary mixture with that of water at different temperatures. All numerical calculations were performed on a DCM microcomputer with programs written in machine language.

Results and Discussion

The experimental data of densities and viscosities at 25, 35, 45, and 55 °C for three binary systems, namely, Me₂SO (1)-CB (2), Me₂SO (1)-PY (2), and Me₂SO (1)-MEK (2), are respectively presented in Tables II-IV. For each temperature, the viscosities of the mixtures are plotted against mole fraction (X_2) and smoothed curves are drawn (Figures 1-3). In the three systems studied here, the graphs of viscosity vs. mixture composition deviate somewhat from linearity thus suggesting a nonideal behavior.

In the theoretical analysis of our data, the binary viscosities were predicted by using the relations of McAllister (2), Auslander (3), and Heric (4, 5) and a polynomial relation (6) which are shown below.

McAllister's relation for the kinematic viscosity of a liquid mixture is

$$\ln \eta = X_1^3 \ln \eta_1 + 3X_1^2 X_2 \ln \eta_{12} + 3X_1 X_2^2 \ln \eta_{21} + X_2^3 \ln \eta_2 + L \quad (1)$$

where

$$L = -\ln(X_1 + X_2 M_2 / M_1) + 3X_1^2 X_2 \ln(2/3 + M_2 / 3M_1) + 3X_1 X_2^2 \ln(1/3 + 2M_2 / 3M_1) + X_2^3 \ln(M_2 / M_1) \quad (1a)$$

Here, η , η_1 , and η_2 are respectively the viscosities of the mixture and pure components 1 and 2. The above eq 1 contains two adjustable parameters, namely, $\ln \eta_{12}$ and $\ln \eta_{21}$, which were determined by a least-squares method. The term L was computed from a knowledge of the composition of the mixture and of the molecular weights M_1 and M_2 of the pure components.

Auslander's equation is

$$X_1(X_1 + B_{12}X_2)(\eta - \eta_1) + A_{21}X_2(X_2 + B_{21}X_1)(\eta - \eta_2) = 0 \quad (2)$$

where A_{21} , B_{12} , and B_{21} are the three binary interaction parameters which were calculated again from a least-squares fit of the experimental data.

Heric's expression is

$$\ln \eta = X_1 \ln \eta_1 + X_2 \ln \eta_2 + X_1 \ln M_1 + X_2 \ln M_2 - \ln(X_1 M_1 + X_2 M_2) + \Delta 12 \quad (3)$$

*Department of Statistics, Karnatak University, Dharwad, India, 580003.

Table I. Some Physical Properties of Pure Liquids at 25 °C

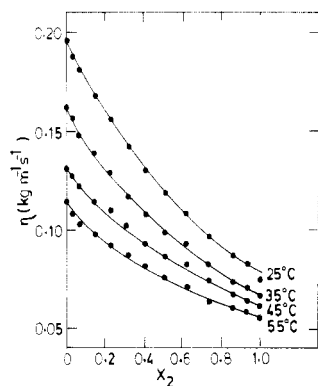
liquid	density, g mL ⁻¹		viscosity, kg m ⁻¹ s ⁻¹		refract. index		bp, °C	
	obsd	lit.	obsd	lit.	obsd	lit.	obsd	lit.
dimethyl sulfoxide	1.0986	1.1014	0.1959	0.1960	1.4745	1.4783	189.0	189.0
chlorobenzene	1.1060	1.1010	0.0757	0.0757	1.5250	1.5248	131.0	131.7
pyridine	0.9811	0.9780	0.0903	0.0899	1.5100	1.5092	115.0	115.5
methyl ethyl ketone	0.8020	0.7995	0.0407	0.0409	1.3760	1.3761	79.5	79.5

Table II. Density and Viscosity Data of the Dimethyl Sulfoxide (1)-Chlorobenzene (2) System

X ₂	density, g mL ⁻¹				viscosity, kg m ⁻¹ s ⁻¹			
	25 °C	35 °C	45 °C	55 °C	25 °C	35 °C	45 °C	55 °C
0.0	1.098	1.093	1.089	1.080	0.1959	0.1605	0.1310	0.1143
0.035	1.098	1.093	1.089	1.080	0.1865	0.1559	0.1273	0.1083
0.072	1.099	1.094	1.089	1.081	0.1803	0.1474	0.1224	0.1036
0.148	1.100	1.094	1.090	1.081	0.1682	0.1390	0.1142	0.0980
0.230	1.103	1.095	1.090	1.082	0.1564	0.1290	0.1098	0.0915
0.317	1.101	1.095	1.091	1.0829	0.1421	0.1170	0.1021	0.0865
0.411	1.103	1.096	1.091	1.0831	0.1299	0.1077	0.0927	0.0808
0.511	1.103	1.096	1.091	1.0836	0.1193	0.0986	0.0865	0.0755
0.619	1.103	1.097	1.092	1.084	0.1085	0.0929	0.0833	0.0712
0.736	1.104	1.097	1.092	1.0846	0.0966	0.0823	0.0742	0.0632
0.863	1.104	1.098	1.093	1.0846	0.0866	0.0733	0.0668	0.0600
0.936	1.104	1.098	1.093	1.0846	0.0826	0.0704	0.0643	0.0580
1.00	1.106	1.098	1.093	1.0845	0.0757	0.0666	0.0615	0.0556

Table III. Density and Viscosity Data of the Dimethyl Sulfoxide (1)-Pyridine (2) System

X ₂	density, g mL ⁻¹				viscosity, kg m ⁻¹ s ⁻¹			
	25 °C	35 °C	45 °C	55 °C	25 °C	35 °C	45 °C	55 °C
0.0	1.0989	1.093	1.086	1.082	0.1944	0.1601	0.1310	0.1140
0.041	1.0928	1.0872	1.080	1.076	0.1859	0.1535	0.1246	0.1107
0.089	1.0871	1.081	1.075	1.069	0.1803	0.1470	0.1191	0.1053
0.180	1.075	1.069	1.063	1.0575	0.1600	0.1318	0.1098	0.0969
0.274	1.063	1.0568	1.050	1.045	0.1488	0.1200	0.1042	0.0891
0.369	1.0516	1.045	1.039	1.034	0.1380	0.1130	0.0972	0.0853
0.468	1.0391	1.0337	1.027	1.022	0.1280	0.1044	0.0922	0.0797
0.569	1.0284	1.022	1.015	1.009	0.1185	0.0984	0.0858	0.0758
0.673	1.0160	1.010	1.003	0.998	0.1097	0.0913	0.0795	0.0706
0.779	1.005	0.998	0.992	0.986	0.1029	0.0849	0.0753	0.0663
0.888	0.993	0.986	0.982	0.974	0.0969	0.0791	0.0723	0.0625
0.943	0.990	0.9798	0.978	0.967	0.0975	0.0760	0.0715	0.0606
1.0	0.9811	0.9749	0.968	0.962	0.0903	0.0750	0.0675	0.0596

Figure 1. Dependence of viscosity on mole fraction (X₂) and temperature for the Me₂SO (1)-CB (2) system.

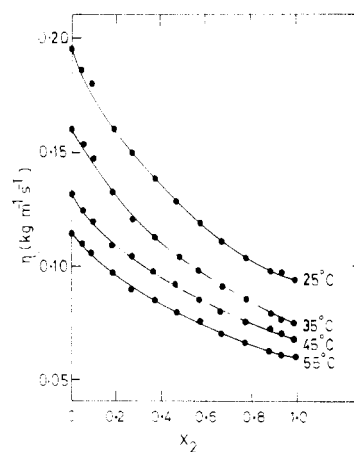
where $\Delta_{12} = \alpha_{12}X_1X_2$ is a term representing departure from a noninteracting system and $\alpha_{12} = \alpha_{21}$ is the interaction parameter. Either α_{12} or α_{21} can be expressed as a linear function of composition

$$\alpha_{12} = \beta_{12}' + \beta_{12}''(X_1 - X_2) \quad (3a)$$

From an initial guess of the values of the coefficients β_{12}' and β_{12}'' , the values of α_{12} were computed.

A polynomial equation of the type

$$\eta = \eta_1X_1 + \eta_2X_2 + X_1X_2[a + b(X_1 - X_2) + c(X_1 - X_2)^2 + \dots] \quad (4)$$

Figure 2. Same as in Figure 1 for the Me₂SO (1)-PY (2) system.

has also been frequently used to predict viscosity data for the mixtures. In eq 4, the coefficients a , b , and c were calculated from the least-squares method.

The results of the computer analysis of all the relations (i.e., eq 1-4) are summarized in Table V which shows the values of several coefficients of eq 1-4 for the systems included in this study. From the coefficients of Table V, the binary viscosities were back calculated. Judging from the closeness of the fit between the computed results and the experimental data, it can be inferred that all the theories were found to be satisfactory.

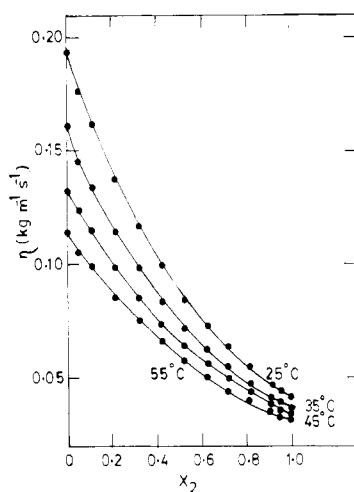
Table IV. Density and Viscosity Data of the Dimethyl Sulfoxide (1)-Methyl Ethyl Ketone (2) System

X_2	density, g mL ⁻¹				viscosity, kg m ⁻¹ s ⁻¹			
	25 °C	35 °C	45 °C	55 °C	25 °C	35 °C	45 °C	55 °C
0.0	1.099	1.0929	1.087	1.0803	0.1926	0.1604	0.1325	0.1138
0.054	1.085	1.0793	1.073	1.067	0.1762	0.1448	0.1230	0.1050
0.107	1.071	1.0657	1.059	1.053	0.1615	0.1335	0.1142	0.0978
0.213	1.0429	1.0377	1.029	1.025	0.1372	0.1138	0.0982	0.0854
0.317	1.0143	1.0090	1.000	0.9963	0.1162	0.0979	0.0852	0.0749
0.419	0.985	0.980	0.973	0.9660	0.0988	0.0830	0.0734	0.0655
0.520	0.955	0.949	0.941	0.9360	0.0835	0.0712	0.0639	0.0574
0.619	0.925	0.919	0.912	0.9050	0.0717	0.0621	0.0561	0.0507
0.716	0.896	0.903	0.881	0.8760	0.0627	0.0547	0.0496	0.0446
0.812	0.865	0.858	0.850	0.8433	0.0540	0.0473	0.0436	0.0398
0.907	0.834	0.828	0.8186	0.8126	0.0462	0.0407	0.0383	0.0353
0.954	0.819	0.801	0.803	0.7930	0.0438	0.0379	0.0361	0.0330
1.0	0.802	0.796	0.7938	0.7799	0.0407	0.0363	0.0343	0.0315

Table V. Computer Analysis of Parameters of Eq 1-4

temp, °C	McAllister formula ^a		Auslander formula ^b			Heric formula, ^c	polynomial formula ^d		
	η_{12}	η_{21}	A_{21}	B_{21}	B_{12}	α_{12}	a	b	c
A. Dimethyl Sulfoxide (1)-Chlorobenzene (2): $M_1 = 78, M_2 = 112.5$									
25	1.4076	1.0287	-0.7033	-2.4096	-0.4193	-0.0143	-0.6256	-0.1218	-0.0238
35	1.1248	0.8897	-1.6742	-1.0757	-0.9490	-0.0570	-0.5316	-0.2133	-0.0452
45	0.9958	0.7809	-0.9925	-1.7820	-0.5615	-0.0679	-0.4098	-0.1245	-0.0421
55	0.8117	0.6838	-1.5605	-1.4052	-0.6803	-0.1647	-0.3393	-0.1869	-0.3437
B. Dimethyl Sulfoxide (1)-Pyridine (2): $M_1 = 78, M_2 = 79.1$									
25	1.3432	1.1116	-0.2229	-9.0233	-0.1263	-0.2441	-0.7332	-0.2903	0.0974
35	1.1028	0.9108	-0.7885	-2.7563	-0.3501	-0.2847	-0.6008	-0.2463	-0.1863
45	0.9344	0.8193	-0.2966	-7.0706	-0.0843	-0.2177	-0.3894	-0.2205	-0.1688
55	0.8352	0.7012	-1.1254	-1.7906	-0.6453	-0.2231	-0.3390	-0.1329	-0.1251
C. Dimethyl Sulfoxide (1)-Methyl Ethyl Ketone (2): $M_1 = 78, M_2 = 72$									
25	1.1139	0.6647	-0.7482	-2.9874	-0.3447	-0.0986	-1.2215	-0.3849	-0.1688
35	0.9223	0.5774	-0.5030	-4.4938	-0.2262	-0.1312	-0.9646	-0.3480	-0.2219
45	0.8285	0.5148	0.3869	5.0404	0.1153	-0.0963	-0.7100	-0.1558	0.0179
55	0.7230	0.4746	-0.5415	-3.6340	-0.2768	-0.0633	-0.5408	-0.1505	-0.0985

^aEquation 1. ^bEquation 2. ^cEquation 3a. ^dEquation 4.

Figure 3. Same as in Figure 1 for the Me₂SO (1)-MEK (2) system.

As a further analysis of the experimental data, an attempt has also been made to predict excess thermodynamic functions, such as enthalpy of mixing (ΔH_{mix}^*) and entropy of mixing (ΔS_{mix}^*) by using the following equation which was developed earlier by Eyring and co-workers (12-14):

$$\ln \nu M = \left[\ln hN - \frac{\Delta S_{\text{mix}}^*}{R} \right] + \frac{\Delta H_{\text{mix}}^*}{RT} \quad (5)$$

Here, ν ($\equiv \eta/\rho$) refers to kinematic viscosity of the mixture, M ($=M_1 + M_2$) is the algebraic sum of the component molecular weights, and the rest of the symbols have their usual meanings.

Table VI. Thermodynamic Excess Mixing Functions for the Me₂SO (1)-CB (2) System

X_2	ΔH_{mix}^* , kJ mol ⁻¹	$-\Delta S_{\text{mix}}^*$, J mol ⁻¹ deg ⁻¹	ΔG_{mix}^* , kJ mol ⁻¹			
			25 °C	35 °C	45 °C	55 °C
0.035	14.293	46.412	28.036	28.500	28.964	29.428
0.072	14.272	45.800	27.920	28.378	28.836	29.294
0.148	13.984	46.196	27.750	28.212	28.674	29.136
0.230	13.894	46.481	27.545	28.010	28.475	28.940
0.318	12.724	48.941	27.309	27.798	28.287	28.777
0.410	12.137	50.149	27.082	27.583	28.085	28.586
0.511	11.783	50.589	26.858	27.364	27.870	28.376
0.619	10.708	53.502	26.652	27.187	27.722	28.257
0.736	10.723	52.472	26.360	26.865	27.409	27.934
0.863	9.373	56.021	26.068	26.628	27.188	27.748
0.932	8.919	57.117	25.940	26.511	27.082	27.653

Table VII. Thermodynamic Excess Mixing Functions for the Me₂SO (1)-PY (2) System

X_2	ΔH_{mix}^* , kJ mol ⁻¹	$-\Delta S_{\text{mix}}^*$, J mol ⁻¹ deg ⁻¹	ΔG_{mix}^* , kJ mol ⁻¹			
			25 °C	35 °C	45 °C	55 °C
0.041	13.930	45.587	27.515	27.971	28.427	28.883
0.089	14.419	43.717	27.446	27.884	28.321	28.758
0.180	13.277	46.647	27.178	27.645	28.111	28.576
0.274	13.064	46.812	27.014	27.482	27.950	28.418
0.370	12.507	48.151	26.856	27.338	27.820	28.301
0.470	12.131	48.879	26.597	27.185	27.674	28.162
0.569	11.503	50.448	26.536	27.041	27.545	28.050
0.672	11.401	50.244	26.374	26.877	27.379	27.882
0.779	11.216	50.406	26.237	26.741	27.245	27.749
0.888	10.977	50.791	26.112	26.620	27.128	27.636
0.944	11.529	48.909	26.104	26.598	27.082	27.571

The plots of $\ln \nu M$ vs. $1/T$ were found to be linear in the temperature range studied and the values of ΔH_{mix}^* and ΔS_{mix}^*

Table VIII. Thermodynamic Excess Mixing Functions for Me₂SO (1)-MEK (2) System

X ₂	ΔH _{mix} [*] , kJ mol ⁻¹	-ΔS _{mix} [*] , J mol ⁻¹ deg ⁻¹	ΔG _{mix} [*] , kJ mol			
			25 °C	35 °C	45 °C	55 °C
0.054	13.503	46.260	27.289	27.752	28.214	28.677
0.108	13.044	47.185	27.105	27.577	28.049	28.521
0.213	12.277	48.601	26.760	27.245	27.732	28.218
0.317	11.444	50.282	26.428	26.981	27.434	27.936
0.418	10.515	52.218	26.076	26.598	27.120	27.642
0.520	9.479	54.566	25.740	26.285	26.831	27.377
0.619	8.703	56.192	25.448	26.010	26.572	27.134
0.717	8.369	56.434	25.186	25.750	26.315	26.879
0.813	7.422	58.678	24.908	25.495	26.082	26.569
0.907	6.991	61.315	24.609	25.216	25.829	26.443
0.954	6.506	60.401	24.506	25.110	25.714	26.318

were estimated in each case from the slope and intercept of the linear plot. The values of ΔH_{mix}^{*} and ΔS_{mix}^{*} were used to calculate the excess free energies (ΔG_{mix}^{*}) by using

$$\Delta G_{\text{mix}}^* = \Delta H_{\text{mix}}^* - T\Delta S_{\text{mix}}^* \quad (6)$$

The numerical values of the three excess functions are shown in Tables VI-VIII. In each of these tables, the negative entropy is due to the attraction between two components of the binary mixtures indicating the nonideal behavior of each mixture. It is seen that the magnitude of entropy increases with an increase in composition of the second component in the mixture. The calculated values of ΔG_{mix}^{*} tend to increase by about 1.3-1.8 kJ/mol with temperature, but decrease by about 1.3-2.8 kJ/mol with increasing concentration of the second component in the mixture. A similar trend is also noticeable in case of ΔH_{mix}^{*} values (see Tables VI-VIII). In general, it may be noted that the values of all the three thermodynamic excess functions have the same order of magnitude which agree closely with the recent literature findings (15, 16).

Recently, Aminabhavi and Munk (17) have developed theoretical relations to treat the changes in volume during mixing of a two-component system using density data on binary mixtures; the validity of these relations were also tested by Aminabhavi et al. (18-20) on a number of binary mixtures. The relations have been derived based on the volume fraction

$$\Delta V_{\text{mix}}^* = \sum_i (N_i V_i) V_{12} \phi_i \phi_2 \quad (7)$$

where N_i and V_i are the number of moles and molar volume of the ith component in the mixture; φ_i represents the volume fraction [defined as φ_i ≡ V_iX_i/(V₁X₁ + V₂X₂)]; V₁₂ is excess volume parameter. This parameter can be calculated from the experimental densities of the mixtures and of pure components by using

$$V_{12} = (\phi_1 \rho_1 + \phi_2 \rho_2 - \rho) / \phi_1 \phi_2 \rho \quad (8)$$

where ρ's represent the densities. The calculated values of V₁₂

Table IX. Computer Analysis of Parameters of Eq 9

system	temp, °C	parameters		
		10 ² a ₀	10 ² a ₁	10 ² a ₂
Me ₂ SO (1)-CB (2)	25	-0.5569	1.0606	2.4941
	35	-0.0959	-0.1928	-0.1684
	45	-0.0011	-0.3029	0.0020
	55	-0.6308	-0.3755	0.1087
Me ₂ SO (1)-PY (2)	25	0.5289	-1.9612	-2.4127
	35	-0.0902	0.5791	1.1173
	45	0.6518	-1.9464	-4.1764
Me ₂ SO (1)-MEK (2)	25	-11.0531	-5.0579	-3.5095
	35	-12.9621	-4.3317	0.4193
	45	-11.5962	2.3482	9.5095
	55	-12.8804	-3.7034	-0.7862

(as given by eq 8) are further fitted to a quadratic equation of the type

$$V_{12} = a_0 + a_1(\phi_2 - \phi_1) + a_2(\phi_2 - \phi_1)^2 \quad (9)$$

The least-squares analysis of eq 9 yields the parameters a₀, a₁, and a₂ which are given in Table IX. The scatter of points throughout the best calculated line does not justify any attempt at interpreting these parameters. It appears, therefore, that no structure correlation can be made between the mixing species and the excess volume parameter V₁₂.

Note Added in Proof. The values reported for viscosity in the tables and figures are in kg m⁻¹ s⁻¹. However, when the data were fitted to theoretical relations viscosities in centipoise (cP) were used.

Registry No. CB, 108-90-7; MEK, 78-93-3; Py, 110-86-1; Me₂SO, 67-68-5.

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