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### Excess Properties of (Methanol Toluene Or *p*-xylene) Binary Liquid Mixture

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## EXCESS PROPERTIES OF (METHANOL + TOLUENE OR *p*-XYLENE) BINARY LIQUID MIXTURE

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Density, viscosity and surface tension of two binary liquid systems: methanol + toluene, and methanol + *p*-xylene, have been determined at 20, 25, 35 and 45°C, over the whole compositional range. The excess values of molar volume, viscosity, Gibbs free energy for the activation of flow and surface tension were evaluated. The Grunberg–Nissan parameter was also calculated. The excess values were fitted to Redlich–Kister type of equation. The viscosity data were fitted to the models of McAllister, Heric and third degree polynomial.

KEY WORDS: Excess molar volume, viscosity, surface tension

### INTRODUCTION

As an extension to our previous work<sup>1–3</sup>, in the present study, we have measured densities, viscosities and surface tensions of the binary organic liquid mixtures of substituted benzene (toluene and *p*-xylene) and aliphatic alcohol (methanol) at 20, 25, 35 and 45°C. From the experimental results, excess properties have been determined and studied in terms of sign and magnitude of the physical properties in the respective mixtures. A valuable idea about the presence of secondary bonding forces in a particular system is derived from the study. Besides, the entire data on excess properties, have been fitted to the Redlich–Kister type of equation. The mixture viscosity data have been fitted to the models of McAllister, Heric and a third degree polynomial.

### EXPERIMENTAL SECTION

Toluene, *p*-xylene and methanol (S.D.'s grade) were purified by the standard procedures<sup>4–5</sup>. The purity of all the components was checked by comparing their experimental and literature values<sup>6</sup>. For each run, a fresh liquid mixture was prepared on mass basis, using a mettler balance (precision of  $1 \times 10^{-5}$  g). The purity of each component with respect to the corresponding literature values is recorded in Table 1.

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**Table 1** Physical constants of pure components at 20°C

Property	Toluene		<i>p</i> -Xylene		Methanol	
	Lit <sup>6,16</sup>	Sample used	Lit <sup>6,16</sup>	Sample used	Lit <sup>6,16</sup>	Sample used
Refractive index	1.4969	1.4950	1.4958	1.4956	1.3288	1.3289
Density (kg/m <sup>3</sup> )	866.00	866.00	861.00	861.00	792.00	792.01
Viscosity × 10 <sup>4</sup> (N – s/m <sup>2</sup> )	5.86	5.86	6.4	6.4	5.84	5.84

Densities of the pure components and their mixtures were measured with a thoroughly cleaned pycnometer calibrated at 20, 25, 35 and 45°C with mercury<sup>7</sup>. At each temperature, the densities were determined with a precision of about ±0.01 percent.

Viscosities were determined with the help of a modified Ubbelohode Viscometer<sup>8</sup>. At each temperature the viscometer was calibrated against the known viscosities of benzene and carbon tetrachloride<sup>7</sup>. The constants of the viscometer at each temperature were determined from the following equation:

$$v = \eta/\rho = at - b/t \quad (1)$$

where,  $a$  and  $b$  are the temperature dependent constants of the viscometer. At a particular temperature, an average value of the efflux time ( $t$ ) of the mixture and corresponding values of  $a$  and  $b$  were used in Eq. (1) for the calculation of the kinematic viscosity ( $v$ ). Using the density values of the respective mixtures, the absolute viscosity ( $\eta$ ) was thus calculated. The accuracy of the viscosity measurements is of the order of 0.001 cP.

Similarly, surface tension of the pure components and that of their mixtures was determined by the differential capillary rise method<sup>7</sup>. The difference ( $\Delta H$ ), in the level of liquid in two capillaries was measured with a cathetometer reading to 0.05 mm. An average reproducibility of better than 0.01 mm was obtained. The difference in the levels of liquid in two capillaries of different radii is related to the surface tension of the liquid by the relation:

$$h_1 - h_2 = \Delta H = (2\sigma/\rho g)A + B - C \quad (2)$$

The constants  $A$ ,  $B$  and  $C$  were calculated using the standard values of surface tension of benzene and ethylacetate at 20°C. For each mixture of the respective systems,  $\Delta H$  was recorded at four different temperatures over the entire compositional range. The surface tension of each mixture was thus calculated from Eq. (2).

The entire experimental data for both the systems is recorded in Table 2. All the measurements were made at a constant temperature with the help of a circulating

**Table 2** Mole fraction ( $x_1$ ), density ( $\rho$ ), dynamic viscosity ( $\eta$ ), surface tension ( $\sigma$ ) and Grunberg-Nissan parameter ( $d$ ) for various binary mixtures

$x_1$	<i>Toluene-Methanol 20°C</i>				<i>Toluene-Methanol 25°C</i>			
	$\rho$ (gm/mL)	$\eta$ (cP)	$\sigma$ (dyne/cm)	$d$	$\rho$ (gm/mL)	$\eta$ (cP)	$\sigma$ (dyne/cm)	$d$
0.0000	0.792010	0.584020	22.60	—	0.787481	0.547010	22.48	—
0.0405	0.798656	0.590310	23.08	0.27	0.793682	0.551980	22.85	0.22
0.0868	0.805877	0.595140	23.60	0.23	0.800750	0.557200	23.33	0.22
0.1401	0.813692	0.600940	24.17	0.23	0.808270	0.562800	23.82	0.22
0.2022	0.822123	0.606730	24.79	0.23	0.816407	0.567880	24.26	0.22
0.2755	0.831143	0.612140	25.46	0.23	0.824594	0.573010	24.83	0.22
0.3632	0.840608	0.616720	26.17	0.23	0.835897	0.577890	25.57	0.22
0.4701	0.850192	0.619400	26.91	0.23	0.842395	0.580340	26.24	0.21
0.6033	0.859129	0.618380	26.85	0.23	0.851961	0.579650	26.92	0.21
0.7739	0.865708	0.609780	28.26	0.23	0.860090	0.580850	27.54	0.29
1.0000	0.866002	0.586100	28.53	—	0.861318	0.554020	27.92	—

$x_1$	<i>Toluene-Methanol 35°C</i>				<i>Toluene-Methanol 45°C</i>			
	$\rho$ (gm/mL)	$\eta$ (cP)	$\sigma$ (dyne/cm)	$d$	$\rho$ (gm/mL)	$\eta$ (cP)	$\sigma$ (dyne/cm)	$d$
0.0000	0.778485	0.480310	21.35	—	0.769490	0.422990	20.50	—
0.0405	0.784421	0.485500	21.70	0.23	0.775208	0.429820	20.76	0.25
0.0868	0.791113	0.490840	22.08	0.23	0.781421	0.437220	21.05	0.25
0.1401	0.799829	0.496500	22.51	0.23	0.788301	0.445190	21.38	0.24
0.2022	0.806084	0.502220	22.98	0.23	0.796055	0.453760	21.75	0.24
0.2755	0.814518	0.507920	23.50	0.22	0.803879	0.462850	22.17	0.24
0.3632	0.823505	0.513190	24.08	0.22	0.812605	0.472650	22.66	0.24
0.4701	0.832822	0.517000	24.72	0.22	0.821797	0.481700	23.23	0.23
0.6033	0.841981	0.518820	25.40	0.22	0.831017	0.489180	23.89	0.22
0.7739	0.849656	0.502100	26.11	0.07	0.839305	0.480860	24.66	0.05
0.0000	0.853080	0.500120	26.75	—	0.843841	0.493900	25.57	—

$x_1$	<i>p-Xylene-Methanol 20°C</i>				<i>p-Xylene-Methanol 25°C</i>			
	$\rho$ (gm/mL)	$\eta$ (cP)	$\sigma$ (dyne/cm)	$d$	$\rho$ (gm/mL)	$\eta$ (cP)	$\sigma$ (dyne/cm)	$d$
0.0000	0.792010	0.584020	22.60	—	0.787481	0.547010	22.48	—
0.0352	0.797640	0.608700	23.15	1.12	0.792746	0.559600	22.81	0.57
0.0758	0.803852	0.635300	23.75	1.10	0.798541	0.573180	23.19	0.56
0.1233	0.810725	0.663490	24.40	1.08	0.804975	0.587700	23.61	0.55
0.1795	0.818306	0.692920	25.12	1.05	0.812106	0.603120	24.08	0.54
0.2470	0.826829	0.722690	25.90	1.02	0.819979	0.619100	24.61	0.53
0.3298	0.835649	0.750890	26.73	1.00	0.828607	0.634780	25.21	0.52
0.4336	0.845132	0.773130	27.57	0.98	0.837822	0.648300	25.86	0.52
0.5673	0.854177	0.780340	28.33	0.97	0.846897	0.655990	26.57	0.51
0.7470	0.861407	0.752640	28.79	0.98	0.854826	0.648890	27.27	0.51
1.0000	0.861001	0.640210	28.31	—	0.856700	0.604520	27.76	—

(continued)

Table 2 (continued)

$x_1$	<i>p</i> -Xylene–Methanol 35°C				<i>p</i> -Xylene–Methanol 45°C			
	$\rho$ (gm/mL)	$\eta$ (cP)	$\sigma$ (dyne/cm)	$d$	$\rho$ (gm/mL)	$\eta$ (cP)	$\sigma$ (dyne/cm)	$d$
0.0000	0.778485	0.480310	21.35	—	0.769490	0.422990	20.50	—
0.0352	0.783599	0.489280	21.68	0.42	0.774348	0.429600	20.82	0.32
0.0758	0.789269	0.499350	22.04	0.43	0.779719	0.436900	21.17	0.32
0.1233	0.795560	0.510290	22.45	0.43	0.785702	0.444930	21.57	0.32
0.1795	0.802535	0.521990	22.92	0.42	0.792363	0.453600	22.02	0.31
0.2470	0.810258	0.534190	23.44	0.42	0.799765	0.462790	22.53	0.31
0.3298	0.818753	0.546500	24.02	0.41	0.807950	0.472500	23.10	0.30
0.4336	0.827877	0.557990	24.68	0.40	0.816824	0.482300	23.74	0.30
0.5673	0.836962	0.566890	25.38	0.40	0.825813	0.489900	24.43	0.29
0.7470	0.845120	0.564890	26.11	0.40	0.834235	0.493300	25.16	0.29
1.0000	0.847750	0.540010	26.67	—	0.838010	0.483030	25.73	—

type ultra cryostat (type MK 70, MLW, Germany) to within  $\pm 0.02$  K. Grunberg–Nissan parameter<sup>9</sup>,  $d$ , was determined using the following expression:

$$d = \left[ \frac{\ln \eta_{\text{mix}} - x_1 \ln \eta_1 - x_2 \ln \eta_2}{x_1 x_2} \right] \quad (3)$$

From the experimental data, excess properties viz;  $V^E$ ,  $\eta^E$ ,  $G^E$  and  $\sigma^E$  were calculated using the following expressions<sup>10</sup>:

$$V^E = V_{\text{mix}} - [x_1 V_1 + x_2 V_2] \quad (4)$$

$$\log(1/\eta^0) = x_1 \log(1/\eta_1) + x_2 \log(1/\eta_2) \quad (5)$$

$$\eta^E = \eta_{\text{mix}} - \eta^0$$

$$G^E/RT = [\ln \eta_{\text{mix}} V_{\text{mix}} - \{x_1 \ln \eta_1 V_1 + x_2 \ln \eta_2 V_2\}] \quad (6)$$

$$\sigma^E = \sigma_{\text{mix}} - [x_1 \sigma_1 + x_2 \sigma_2] \quad (7)$$

The values of  $V^E$ ,  $\eta^E$ ,  $G^E$  and  $\sigma^E$  were fitted with a Redlich–Kister type of equation<sup>11</sup>, using least square method.

$$F^E = x_1 x_2 \sum_1^n A_{j-1} (2x_1 - 1)^{j-1} \quad (8)$$

where  $F^E$  represents the excess property,  $A_j$  is the polynomial coefficient and  $n$  the polynomial degree. The results thus obtained are recorded in Table 3.

The following theoretical models for mixture viscosities have been tested with the

**Table 3** Coefficients of Eq. (8) and standard deviations (SD) determined by the method of least squares

<i>System: Toluene + Methanol</i>						
<i>Temp. (°C)</i>	<i>Property</i>	<i>A</i> <sub>0</sub>	<i>A</i> <sub>1</sub>	<i>A</i> <sub>2</sub>	<i>A</i> <sub>3</sub>	<i>SD</i>
20	<i>V</i> <sup>E</sup>	-2.367	-3.5279	0.0904	-0.1087	0.0005
	<i>G</i> <sup>E</sup>	1577.78	-402.76	176.811	-129.405	1.269
	<i>η</i> <sup>E</sup>	0.1383	0.0027	-0.001	-0.0101	0.0003
	<i>σ</i> <sup>E</sup>	4.5816	-4.959	5.434	13.0875	0.173
25	<i>V</i> <sup>E</sup>	-1.5074	-2.4523	-1.3374	-3.0656	0.0574
	<i>G</i> <sup>E</sup>	1577.986	-377.114	499.90	228.731	3.253
	<i>η</i> <sup>E</sup>	0.1172	0.004	0.0843	0.0971	0.0006
	<i>σ</i> <sup>E</sup>	4.8406	0.7312	-0.7192	-1.4549	0.0382
35	<i>V</i> <sup>E</sup>	-0.9234	-3.0462	-0.05	0.4283	0.0310
	<i>G</i> <sup>E</sup>	1705.81	-522.403	-532.69	-848.633	4.737
	<i>η</i> <sup>E</sup>	0.1155	-0.0149	-0.1413	-0.1547	0.0009
	<i>σ</i> <sup>E</sup>	3.3154	0.0035	-0.0013	-0.0096	0.00027
45	<i>V</i> <sup>E</sup>	-0.2735	-2.7919	-0.0571	-0.3062	0.006
	<i>G</i> <sup>E</sup>	1800.31	-625.725	-660.952	-964.508	4.043
	<i>η</i> <sup>E</sup>	0.106	-0.0245	-0.1576	-0.1582	0.00079
	<i>σ</i> <sup>E</sup>	0.3821	-0.0016	0.0053	0.0082	0.0001
<i>System: p-Xylene + Methanol</i>						
<i>Temp. (°C)</i>	<i>Property</i>	<i>A</i> <sub>0</sub>	<i>A</i> <sub>1</sub>	<i>A</i> <sub>2</sub>	<i>A</i> <sub>3</sub>	<i>SD</i>
20	<i>V</i> <sup>E</sup>	-2.1674	-14.018	0.1242	-1.5146	0.0084
	<i>G</i> <sup>E</sup>	6609.61	-2345.47	2042.77	-2507.72	9.242
	<i>η</i> <sup>E</sup>	0.6703	-0.0022	0.0012	0.00431	0.0004
	<i>σ</i> <sup>E</sup>	10.158	-0.0347	0.0538	0.0971	0.0009
25	<i>V</i> <sup>E</sup>	4.6226	-12.6302	0.2487	-1.0866	0.0072
	<i>G</i> <sup>E</sup>	5613.752	-2350.06	1881.67	-2501.39	9.206
	<i>η</i> <sup>E</sup>	0.311	0.000	0.0019	0.0025	0.0003
	<i>σ</i> <sup>E</sup>	4.4477	-0.0305	0.04530	0.09090	0.001
35	<i>V</i> <sup>E</sup>	5.5479	-12.3373	0.2556	-1.0824	0.007
	<i>G</i> <sup>E</sup>	5535.62	-2402.25	1864.17	-2640.11	8.501
	<i>η</i> <sup>E</sup>	0.2131	0.0053	-0.0092	-0.0119	0.0002
	<i>σ</i> <sup>E</sup>	4.1316	-0.0355	0.0584	-0.1161	0.0011
45	<i>V</i> <sup>E</sup>	6.9399	-11.5744	0.2386	-1.0508	0.0073
	<i>G</i> <sup>E</sup>	5446.96	-2502.77	1948.49	-2647.36	9.912
	<i>η</i> <sup>E</sup>	0.1345	-0.0012	0.0008	-0.0042	0.0001
	<i>σ</i> <sup>E</sup>	3.9294	-0.0204	0.0446	0.0703	0.0009

experimental data. McAllister<sup>12</sup> equation derived on the basis of Eyring's absolute reaction rate theory is:

$$\begin{aligned}
 \ln \eta_{\text{mix}} = & x_1^3 \ln \eta_1 + x_2^3 \ln \eta_2 + 3x_1^2x_2 \ln \eta_{12} + 3x_1x_2^2 \ln \eta_{21} \\
 & - \ln[x_1 + x_2(M_2/M_1)] + 3x_1^2x_2 \ln [2/3 + (M_2/3M_1)] \\
 & + 3x_1x_2^2 \ln [1/3 + 2M_2/3M_1] + x_2^3 \ln [M_2/M_1] \quad (9)
 \end{aligned}$$

Where,  $\eta_{12}$  and  $\eta_{21}$  are the interaction parameters. The Heric's<sup>13</sup> equation has the following form:

$$\ln \eta_{\text{mix}} = x_1 \ln \eta_1 + x_2 \ln \eta_2 + x_1 \ln M_1 + x_2 \ln M_2 - \ln(x_1 M_1 + x_2 M_2) + \delta_{12} \quad (10)$$

where  $\delta_{12}$  is a deviation function expressed as:

$$\delta_{12} = x_1 x_2 [\beta_{12} + \beta_{21}(x_1 - x_2)] \quad (11)$$

The viscosity data has also been fitted to a third degree polynomial:

$$\eta_{\text{mix}} = A + Bx_1 + Cx_1^2 \quad (12)$$

The parameters corresponding to Eqs. (9), (11) and (12) have been determined using a least square method and are reported in Table 4.

**Table 4** Interaction parameters for McAllister model Eq. (9), Heric's model Eq. (11) the coefficients of the polynomial Eq. (12) and the standard deviations (SD) determined by least square method

Model	System: Toluene + Methanol			
	20°C	25°C	35°C	45°C
McAllister				
$\eta_{12}$	0.635906	0.605431	0.516125	0.485692
$\eta_{21}$	0.708550	0.657267	0.598688	0.553740
SD	0.0017	0.0032	0.0025	0.0028
Heric				
$\beta_{12}$	0.7730	0.7692	0.7384	0.7396
$\beta_{21}$	-0.2039	-0.1695	-0.2827	-0.3141
SD	0.0018	0.0034	0.0027	0.0030
Polynomial				
A	0.584272	0.546549	0.481341	0.424200
B	0.139243	0.131678	0.122806	0.163308
C	-0.13737	-0.12199	-0.10807	-0.09834
SD	0.0002	0.002	0.0039	0.0044
		System: p-Xylene + Methanol		
	20°C	25°C	35°C	45°C
McAllister				
$\eta_{12}$	0.700539	0.571805	0.491154	0.421633
$\eta_{21}$	1.603287	1.269162	1.076033	0.918758
SD	0.0251	0.0205	0.0175	0.0152
Heric				
$\beta_{12}$	2.9372	2.4667	2.3553	2.2473
$\beta_{21}$	-1.5310	-1.4898	-1.4785	-1.4782
SD	0.0269	0.0219	0.0187	0.0162
A	0.584046	0.546999	0.480458	0.422950
B	0.726753	0.368672	0.271518	0.194755
C	-0.67065	-0.31111	-0.21195	-0.13468
SD	0.00008	0.00005	0.0008	0.0001

RESULTS AND DISCUSSION

The experimental values of physical and excess properties of both the systems are represented graphically in Figures (1-8). From Figures (1-6), it is observed that the density, viscosity and surface tension vary nonlinearly with the increase in concentration of toluene and *p*-xylene in the respective mixtures. The degree of nonlinearity

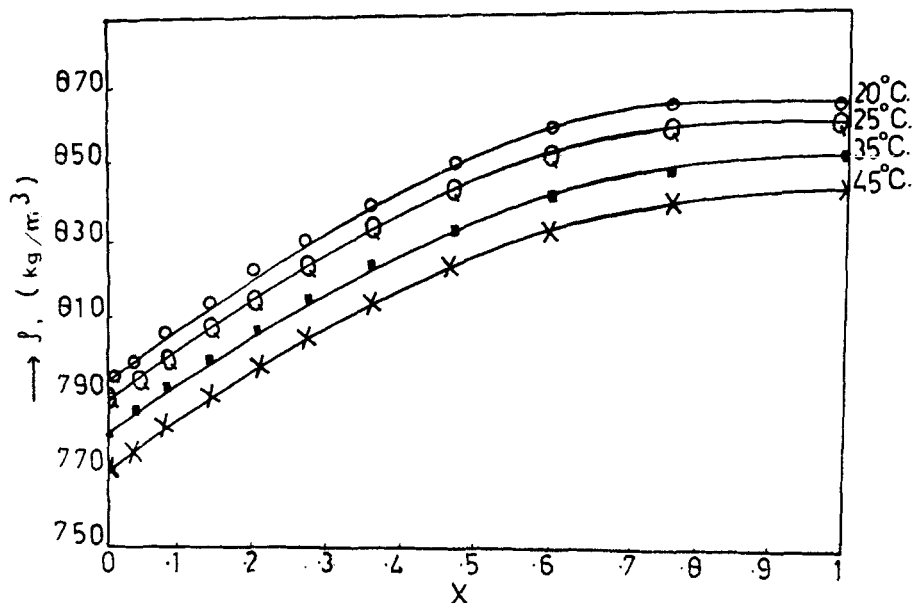


Figure 1 Composition dependence of density for the system ( $x\text{C}_6\text{H}_5\text{CH}_3 + 1 - x\text{CH}_3\text{OH}$ ).

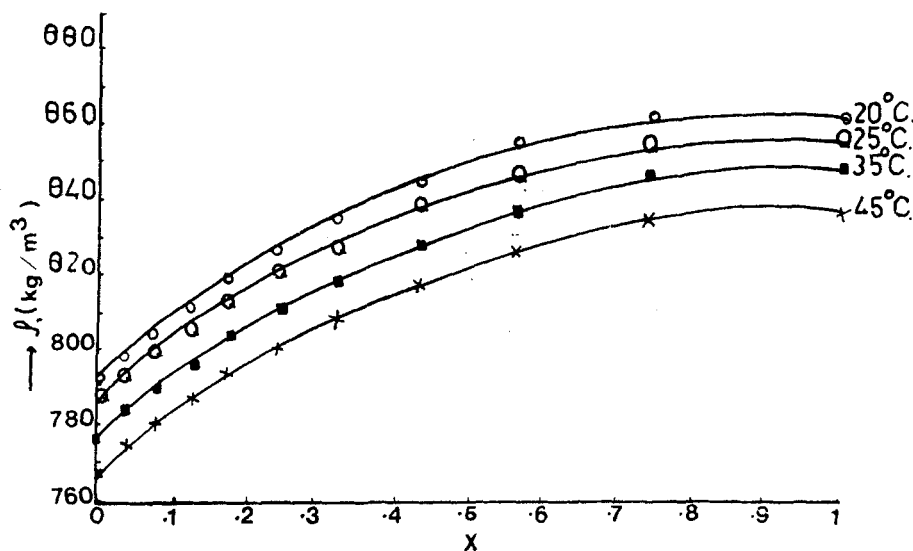


Figure 2 Composition dependence of density for the system ( $x\text{P} - \text{C}_6\text{H}_4(\text{CH}_3)_2 + 1 - x\text{CH}_3\text{OH}$ ).



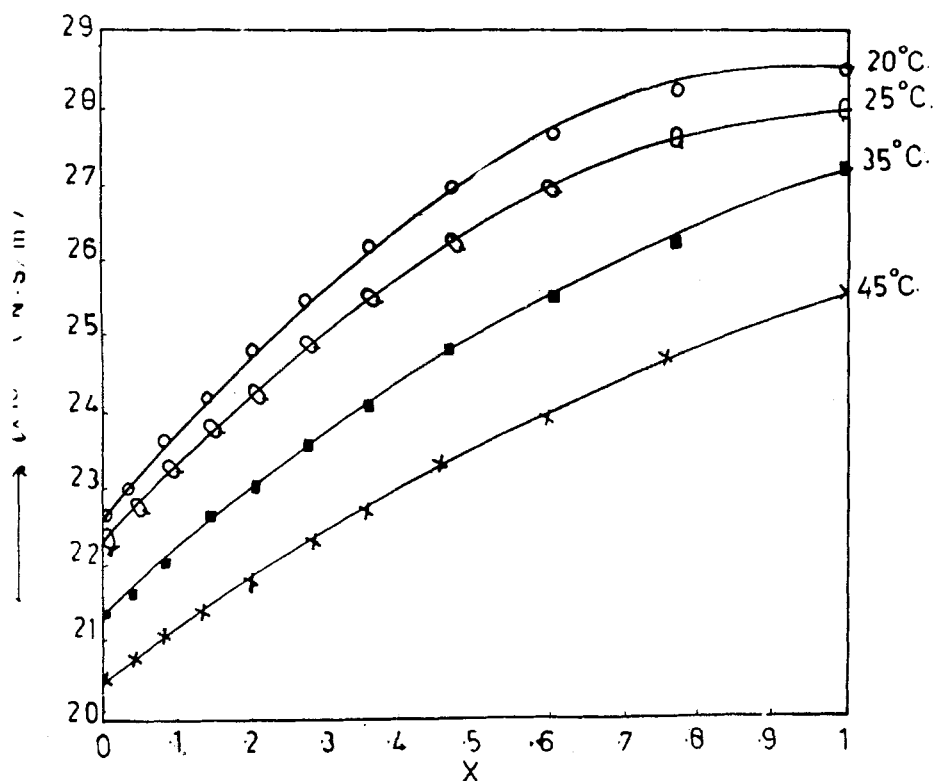


Figure 3 Composition dependence of viscosity for the system ( $x\text{C}_6\text{H}_5\text{CH}_3 + 1 - x\text{CH}_3\text{OH}$ ).

decreases with increase in temperature. Density and surface tension of the mixtures decrease with increase in concentration of toluene and *p*-xylene. However, the viscosity first increases and then gradually decreases with the increase in concentration of toluene and *p*-xylene in the respective mixtures. The elevation in the viscosity curve of *p*-xylene + methanol system (Figure 4) is more prominent as compared to that of toluene + methanol system (Figure 3), suggesting the existence of strong interactions between unlike molecules<sup>14</sup>. These interactions are prominent in the case of *p*-xylene + methanol system as compared to that of toluene + methanol system. This could be explained considering the higher polarisability of the methyl substituted benzene molecules<sup>15</sup>. The stability of the complex formation decreases with the increase in temperature. From both the plots (Figures 3 and 4), it is observed that a temporary complex formation may exist between 0.4 to 0.7 mole fraction of toluene and *p*-xylene in respective mixtures. Besides, as the temperature increases, the surface tension (Figures 5 and 6), tends to approach to linearity, indicating that the molecular interactions decrease gradually.

The increase in the molecular interactions with increase in concentration of toluene or *p*-xylene is further evident from  $V^E$  results. The variation of  $V^E$  from

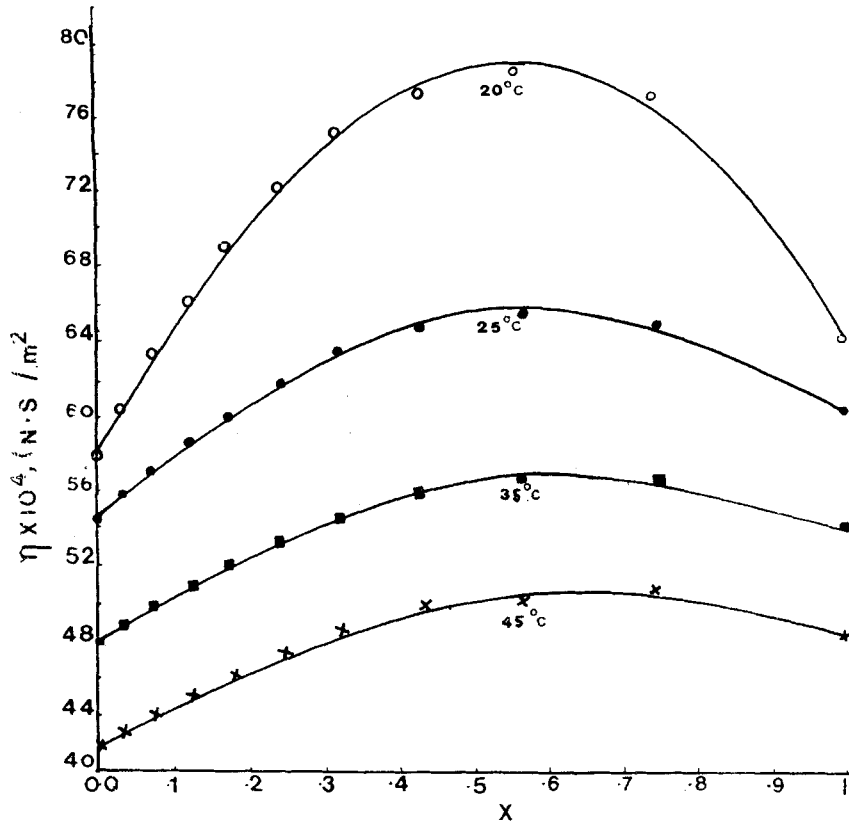


Figure 4 Composition and temperature dependence of viscosity of  $(x\text{C}_6\text{H}_4(\text{CH}_3)_2 + 1 - x\text{CH}_3\text{OH})$ .

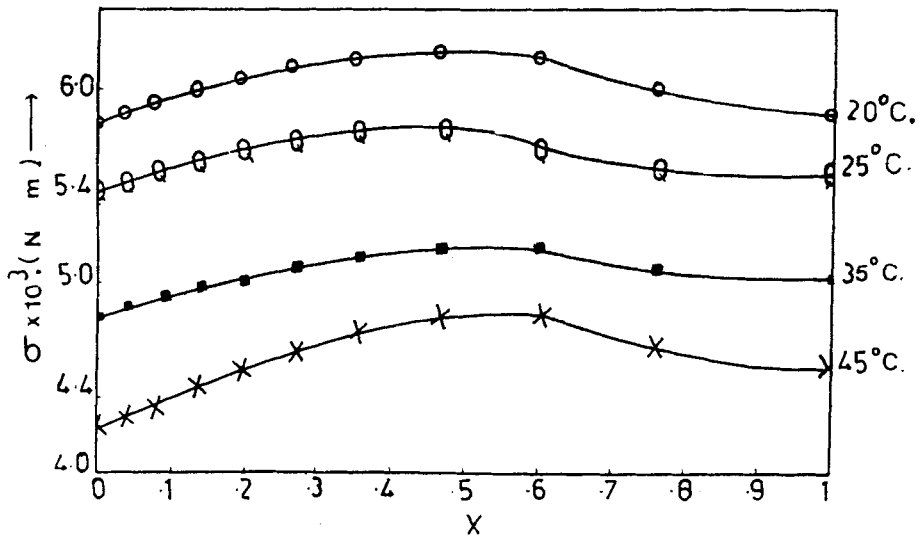


Figure 5 Composition dependence of surface tension for the system  $(x\text{C}_6\text{H}_5\text{CH}_3 + 1 - x\text{CH}_3\text{OH})$ .

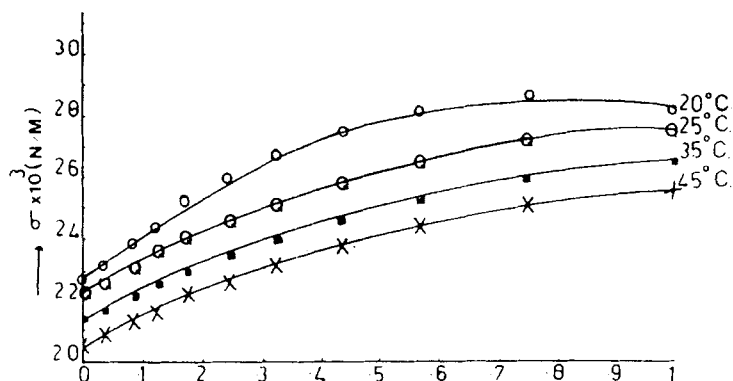


Figure 6 Composition dependence of surface tension for the system  $(xC_6H_4(CH_3) + 1 - xCH_3OH)$ .

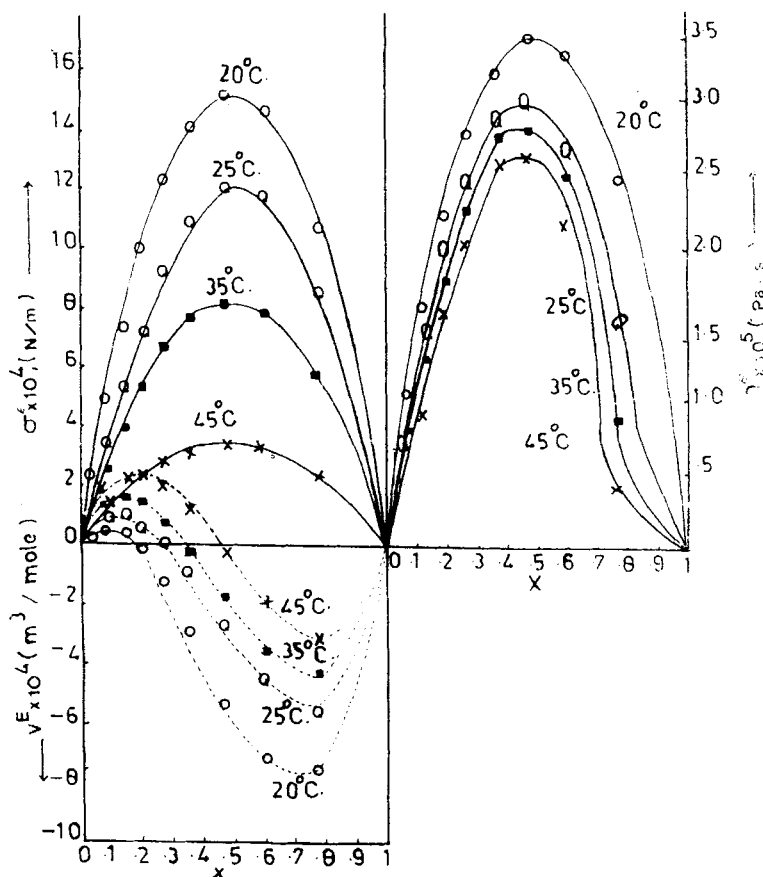
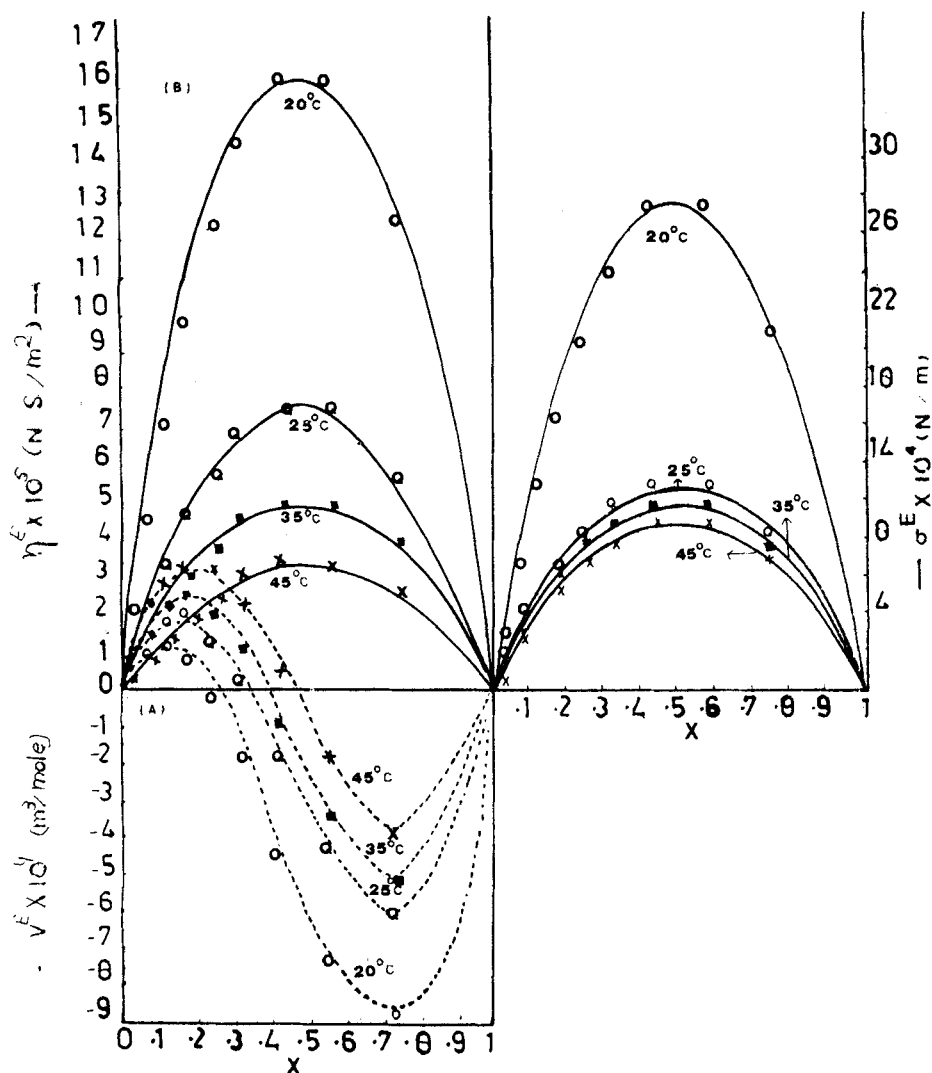


Figure 7 Composition dependence of excess volume  $v^E$ , excess viscosity  $\eta^E$  and excess surface tension  $\sigma^E$  for the system  $(xC_6H_5CH_3 + 1 - xCH_3OH)$ .



**Figure 8** Composition dependence of (A) excess volume (B) excess viscosity and (C) excess surface tension for the system ( $x\text{C}_6\text{H}_4(\text{CH}_3)_2 + 1 - x\text{CH}_3\text{OH}$ ).

positive to negative values with increase in concentration of toluene (Figure 7) and *p*-xylene (Figure 8) supports the above conclusion. The decrease in the negative values of  $V^E$  with increase in temperature, fortifies the decrease in the interactions with increasing concentration of toluene and *p*-xylene. The above depictions are also borne by  $\eta^E$  and  $\sigma^E$  values (Figures 7b, 7c, 8b, 8c) of both the systems.

Comparing the results of both the mixtures, it seems that the magnitude of excess properties indicate the deviation from ideality is observed to be more in case of *p*-xylene + methanol system as compared to that of toluene + methanol system.

Besides, with increase in the number of methyl groups on benzene ring, the polarizability also increases<sup>15</sup>. Thus an enhanced dipole-induced dipole interactions are observed in *p*-xylene-methanol system, which involves stronger interactions as compared to those present in toluene + methanol system.

Finally, the models of McAllister and Heric produce satisfactory results for the systems studied. Third degree polynomial predicts the viscosity data very well, for the *p*-xylene-methanol system.

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