Densities, Viscosities, and Excess Molar Volumes of Ternary Liquid Mixtures of Bromobenzene + 1,4-Dioxane + (Benzene or + Toluene or + Carbon Tetrachloride) and Some Associated Binary Liquid Mixtures

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Densities and viscosities of ternary liquid mixtures of bromobenzene + 1,4-dioxane + (benzene or + toluene or + carbontetrachloride) and binary mixtures of bromobenzene + 1,4-dioxane + benzene, bromobenzene + benzene, 1,4-dioxane + toluene, bromobenzene + toluene, 1,4-dioxane + carbon tetrachloride, and bromobenzene + carbon tetrachloride have been measured at 303.15 K. From density and viscosity data, the values of viscosity deviations ($\Delta\eta$) and the excess molar volume (V^E) for the ternary systems have been determined and fitted to Cibulka's equation.

Introduction

This paper is a continuation of our work related to the study of excess thermodynamic and transport properties of binary mixtures.[1−3] A survey of literature reveals that data on densities, viscosities, and excess thermodynamic properties in regard to ternary mixtures involving bromobenezene and nonpolar solvents are scarce. The present paper therefore deals with the measurement of densities, viscosities, viscosity deviations, and excess molar volumes V^E at 303.15 K for the following ternary mixtures: bromobenzene + 1,4-dioxane + benzene, bromobenzene + 1,4-dioxane + toluene, and bromobenzene + 1,4-dioxane + carbon tetrachloride.

Experimental Section

Materials. The chemicals used were of analytical grade. Bromobenzene (> 99.2 %), 1,4-dioxane (> 99.4 %), benzene (> 99.3 %), toluene (> 99.5 %), and carbon tetrachloride (> 99.3 %) were obtained from Merck. The organic liquids were further purified according to the procedure described in the literature,[4,5] and the purity was better than 99 %.

Measurements. Liquid mixtures of various compositions were prepared by mass in a 25 cm^3 flask using a Mettler analytical balance. The average uncertainty in the mole fraction of the mixtures was estimated to be less than ± 0.0001. Density and viscosity measurements were carried out using a thermostatically controlled, well-stirred water bath, where a constant temperature was measured with a digital thermometer with an uncertainty of ± 0.01 K.

Densities of pure liquids and their binary and ternary liquid mixtures were measured at 303.15 K with an Anton Parr digital vibrating-tube densimeter (model 60/602, Anton Parr, Austria). The densimeter was calibrated with degassed water and dehumidified air at atmospheric pressure. The uncertainty of the density measurements was estimated to be less than 1×10^{-4} g·cm^{-3}.

Table 1. Comparison of Experimental Density and Viscosity of Pure Liquids with Literature Values at 303.15 K

	ρ/g·cm^{-3}		η/mPa·s	
pure liquid	exptl	lit	exptl	lit
bromobenzene	1.4814	1.4889[a]	0.9915	0.9850[a]
1,4-dioxane	1.0195	1.0233[b]	1.0500	1.0505[c]
benzene	0.8650	0.8659[d]	0.5640	0.5643[e]
toluene	0.8582	0.8583[f]	0.5229	0.5227[g]
carbon tetrachloride	1.5775	1.5852[h,i]	0.8300	0.9128[h,i]

[a] Ref 13. [b] Ref 18. [c] Ref 14. [d] Ref 15. [e] Ref 16. [f] Ref 17. [g] Ref 18. [h] Ref 19. [i] Reference at 298.15 K.

The viscosities η of pure organic liquids and their binary and ternary mixtures were determined using an Ostwald viscometer, which was suspended in a thermostat maintained at (303.15 ± 0.01) K. The details of the procedure have been reported in an earlier publication.[1] The uncertainty of calculated absolute viscosities was ± 1×10^{-4} mPa·s. The excess molar volumes were calculated from density data, and the uncertainties were estimated to be within ± 5×10^{-3} cm^3·mol^{-1}.

Results and Discussion

The experimental values of densities and viscosities of the pure components have been compared with the literature values and presented in Table 1. It is seen that the experimental values compare fairly well with the literature values.

The densities ρ and viscosities η of binary and ternary mixtures were measured at (303.15 ± 0.01) K as a function of the composition of the corresponding ternary mixtures. The results of the study have been presented in Tables 2 and 3.

The viscosity deviations $\Delta\eta$ and excess molar volumes V^E for binary and ternary mixtures were determined using the following equations:[6,7]

$$\Delta\eta = \eta - \sum_{i=1}^{n} x_i\eta_i \quad (1)$$

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Table 2. Densities (ρ), Viscosities (η), Viscosity Deviations ($\Delta\eta$), and Excess Molar Volume (V^E) of Binary Liquid Mixtures at 303.15 K

x_1	ρ / g·cm^{-3}	η / mPa·s	$\Delta\eta$ / mPa·s	V^E / cm^3·mol^{-1}	x_1	ρ / g·cm^{-3}	η / mPa·s	$\Delta\eta$ / mPa·s	V^E / cm^3·mol^{-1}	x_1	ρ / g·cm^{-3}	η / mPa·s	$\Delta\eta$ / mPa·s	V^E / cm^3·mol^{-1}
Bromobenzene (1) + 1,4-Dioxane (2)														
0.000	1.020	1.050	0.000	0.000	0.352	1.150	1.131	0.102	4.372	0.766	1.348	1.079	0.074	3.060
0.083	1.051	1.075	0.030	1.200	0.449	1.193	1.125	0.102	4.557	0.880	1.411	1.034	0.035	1.800
0.170	1.081	1.078	0.038	2.607	0.550	1.243	1.115	0.098	4.189	1.000	1.481	0.992	0.000	0.000
0.259	1.117	1.122	0.087	3.345	0.655	1.292	1.108	0.096	3.900					
1,4-Dioxane (2) + Benzene (3)														
0.000	0.865	0.564	0.000	0.000	0.411	0.895	0.733	−0.031	3.207	0.807	0.968	0.937	−0.019	1.906
0.104	0.870	0.599	−0.016	1.050	0.511	0.908	0.779	−0.033	3.302	0.904	0.991	0.991	−0.012	1.110
0.207	0.877	0.644	−0.021	1.914	0.610	0.925	0.828	−0.033	3.137	1.000	1.020	1.050	0.000	0.000
0.309	0.882	0.686	−0.028	2.950	0.709	0.945	0.882	−0.027	2.591					
Bromobenzene (1) + Benzene (3)														
0.000	0.865	0.564	0.000	0.000	0.362	1.101	0.742	0.023	0.960	0.774	1.349	0.909	0.014	0.700
0.086	0.923	0.610	0.009	0.330	0.460	1.162	0.785	0.024	0.960	0.884	1.415	0.951	0.009	0.330
0.176	0.982	0.653	0.014	0.640	0.561	1.223	0.827	0.023	0.930	1.000	1.481	0.992	0.000	0.000
0.268	1.040	0.700	0.022	0.900	0.665	1.286	0.867	0.019	0.829					
1,4-Dioxane (2) + Toluene (3)														
0.000	0.858	0.523	0.000	0.000	0.453	0.888	0.729	−0.033	3.781	0.833	0.962	0.939	−0.023	2.354
0.122	0.861	0.573	−0.014	1.616	0.554	0.902	0.779	−0.036	3.947	0.918	0.990	0.995	−0.012	1.228
0.237	0.868	0.622	−0.026	2.706	0.650	0.917	0.831	−0.034	3.834	1.000	1.020	1.050	0.000	0.000
0.347	0.877	0.675	−0.031	3.363	0.744	0.937	0.882	−0.033	3.316					
Bromobenzene (1) + Toluene (3)														
0.000	0.858	0.523	0.000	0.000	0.403	1.114	0.740	0.028	−0.660	0.802	1.362	0.918	0.019	−0.391
0.101	0.922	0.581	0.011	−0.211	0.503	1.176	0.788	0.029	−0.600	0.901	1.422	0.957	0.012	−0.241
0.202	0.987	0.635	0.017	−0.471	0.603	1.239	0.831	0.026	−0.601	1.000	1.481	0.992	0.000	0.000
0.303	1.051	0.689	0.024	−0.565	0.703	1.301	0.877	0.025	−0.510					
1,4-Dioxane (2) + Carbon Tetrachloride (3)														
0.000	1.578	0.830	0.000	0.000	0.430	1.368	1.137	0.213	−0.920	0.819	1.137	1.132	0.122	−0.463
0.112	1.527	0.922	0.067	−0.320	0.530	1.312	1.163	0.216	−0.932	0.910	1.079	1.092	0.062	−0.281
0.220	1.475	1.016	0.138	−0.570	0.629	1.255	1.164	0.196	−0.881	1.000	1.020	1.050	0.000	0.000
0.326	1.421	1.097	0.195	−0.740	0.725	1.196	1.161	0.172	−0.686					
Bromobenzene (1) + Carbon Tetrachloride (3)														
0.000	1.578	0.830	0.000	0.000	0.380	1.544	0.595	−0.296	−0.321	0.787	1.503	0.778	−0.179	−0.180
0.092	1.570	0.727	−0.118	−0.117	0.479	1.535	0.619	−0.288	−0.335	0.892	1.492	0.881	−0.093	−0.095
0.187	1.562	0.653	−0.207	−0.206	0.580	1.524	0.656	−0.268	−0.307	1.000	1.481	0.992	0.000	0.000
0.283	1.553	0.624	−0.252	−0.278	0.682	1.514	0.700	−0.240	−0.255					

Table 3. Densities (ρ),Viscosities (η), Viscosity Deviations ($\Delta\eta$), and Excess Molar Volume (V^E) of Ternary Liquid Mixtures at 303.15 K

x_1	x_2	x_3	ρ / g·cm^{-3}	η / mPa·s	$\Delta\eta$ / mPa·s	V^E / cm^3·mol^{-1}	x_1	x_2	x_3	ρ / g·cm^{-3}	η / mPa·s	$\Delta\eta$ / mPa·s	V^E / cm^3·mol^{-1}
Bromobenzene (1) + 1,4-Dioxane (2) + Benzene (3)													
0.043	0.053	0.905	0.953	0.583	−0.024	−4.723	0.261	0.535	0.204	1.106	0.772	−0.164	1.754
0.041	0.911	0.048	0.951	0.606	−0.418	7.707	0.357	0.329	0.314	1.140	0.848	−0.028	1.498
0.083	0.819	0.098	0.992	0.678	−0.319	5.173	0.355	0.436	0.208	1.149	0.830	−0.098	1.961
0.086	0.106	0.808	0.980	0.614	−0.038	−3.545	0.458	0.113	0.430	1.213	0.882	0.068	−1.947
0.086	0.210	0.705	0.968	0.630	−0.073	−1.016	0.556	0.227	0.217	1.259	0.924	0.012	0.544
0.085	0.417	0.499	0.988	0.659	−0.144	0.048	0.553	0.339	0.108	1.260	0.946	−0.019	1.650
0.174	0.213	0.613	1.042	0.708	−0.034	−2.007	0.659	0.231	0.111	1.305	0.977	0.019	1.729
0.170	0.730	0.100	1.056	0.731	−0.260	3.458	0.769	0.118	0.113	1.349	1.010	0.060	1.840
0.171	0.629	0.200	1.053	0.734	−0.209	2.246	0.826	0.060	0.114	1.386	0.994	0.048	0.750
0.264	0.323	0.413	1.097	0.795	−0.039	−0.066	0.883	0.060	0.057	1.430	1.006	0.035	−0.208
Bromobezene (1) + 1,4-Dioxane (2) + Toluene (3)													
0.050	0.062	0.889	0.872	0.545	−0.034	3.028	0.270	0.553	0.178	1.084	0.662	−0.279	3.708
0.041	0.917	0.041	1.017	0.575	−0.451	1.549	0.376	0.346	0.278	1.145	0.693	−0.189	0.946
0.085	0.832	0.084	1.028	0.591	−0.410	1.900	0.367	0.451	0.181	1.143	0.689	−0.244	2.464
0.098	0.122	0.780	0.917	0.567	−0.066	2.187	0.491	0.121	0.388	1.208	0.760	−0.057	−1.911
0.096	0.236	0.667	0.929	0.593	−0.099	2.649	0.576	0.235	0.189	1.245	0.806	−0.111	1.598
0.092	0.453	0.456	0.913	0.594	−0.211	7.749	0.563	0.345	0.092	1.243	0.795	−0.173	3.006
0.193	0.236	0.571	1.020	0.629	−0.109	−0.477	0.671	0.235	0.095	1.298	0.869	−0.092	2.228
0.173	0.742	0.085	1.015	0.609	−0.386	7.302	0.783	0.120	0.097	1.335	0.939	−0.014	2.938
0.177	0.649	0.174	1.015	0.613	−0.335	5.973	0.842	0.061	0.098	1.366	0.977	0.028	2.344
0.282	0.346	0.372	1.080	0.670	−0.168	1.241	0.890	0.061	0.049	1.390	1.022	0.050	2.815
Bromobenzene (1) + 1,4-Dioxane (2) + Carbon Tetrachloride (3)													
0.046	0.056	0.898	1.130	0.874	0.024	35.702	0.265	0.543	0.192	1.268	0.888	−0.104	0.112
0.041	0.914	0.045	1.109	0.863	−0.175	−3.060	0.365	0.336	0.298	1.334	0.904	−0.059	2.776
0.084	0.825	0.092	1.143	0.891	−0.134	−1.675	0.361	0.443	0.196	1.318	0.895	−0.091	−0.165
0.091	0.113	0.796	1.190	0.868	−0.002	26.253	0.473	0.116	0.411	1.378	0.905	−0.027	6.947
0.090	0.222	0.688	1.169	0.891	−0.002	23.560	0.565	0.231	0.205	1.374	0.922	−0.050	2.532
0.088	0.433	0.479	1.164	0.888	−0.051	14.504	0.557	0.342	0.101	1.385	0.920	−0.075	−2.305
0.183	0.223	0.594	1.240	0.897	−0.012	16.107	0.664	0.232	0.104	1.396	0.967	−0.021	0.202
0.172	0.735	0.094	1.195	0.892	−0.127	−2.054	0.775	0.119	0.106	1.407	1.025	0.044	2.751
0.174	0.638	0.188	1.202	0.892	−0.106	1.632	0.833	0.060	0.107	1.436	1.056	0.078	2.317
0.272	0.334	0.394	1.286	0.888	−0.059	7.118	0.886	0.061	0.053	1.428	1.143	0.157	2.567

Table 4. Coefficients of Redlich−Kister's Equation and the Corresponding Standard Deviation (σ) for the Binary Liquid Mixtures at 303.15 K[a]

	A_0	A_1	A_2	A_3	A_4	A_5	σ
			Bromobenzene + 1,4-Dioxane				
$\Delta\eta$/mPa·s	0.393	0.005	0.803	−1.267	−3.411	6.447	0.0002
V^E/cm^3·mol^{-1}	−17.58	9.78	−11.45	−244.61	−9433.74	5950.88	0.0041
			1,4-Dioxane + Benzene				
$\Delta\eta$/mPa·s	−0.131	−0.039	−0.047	0.429	0.407	−1.362	0.0001
V^E/cm^3·mol^{-1}	13.13	0.87	6.74	−33.16	−49.32	140.80	0.0016
			Bromobenzene + Benzene				
$\Delta\eta$/mPa.s	0.095	0.006	−0.006	−0.495	0.019	1.829	0.0004
V^E/cm^3·mol^{-1}	3.816	−0.339	1.460	−8.698	4.271	39.257	0.0020
			1,4-Dioxane + Toluene				
$\Delta\eta$/mPa·s	−0.137	−0.031	−0.100	−0.030	0.073	0.280	0.0009
V^E/cm^3·mol^{-1}	15.553	3.454	3.742	1.355	−4.765	−29.122	0.0035
			Bromobenzene + Toluene				
$\Delta\eta$/mPa·s	0.114	−0.039	0.015	0.430	−0.098	−1.125	0.0006
V^E/cm^3·mol^{-1}	−2.507	0.819	−0.844	−6.330	0.855	23.011	0.0025
			1,4-Dioxane + Carbon Tetrachloride				
$\Delta\eta$/mPa·s	0.861	−0.107	0.199	0.550	−1.367	−0.212	0.0030
V^E/cm^3·mol^{-1}	−3.802	−0.216	3.008	−2.875	−5.484	18.663	0.0015
			Bromobenzene + Carbon Tetrachloride				
$\Delta\eta$/mPa·s	−1.146	0.511	−0.428	−4.011	1.404	13.488	0.0034
V^E/cm^3·mol^{-1}	−1.323	0.307	0.434	−0.296	−0.308	0.382	0.0014

[a] $\Delta\eta$, viscosity deviations; V^E, excess volumes.

Table 5. Coefficient of Cibulka Equation and the Corresponding Standard Deviation (σ) for Binary Liquid Mixtures at 303.15 K[a]

	B_1	B_2	B_3	σ
	Bromobenzene + 1,4-Dioxane + Benzene			
$\Delta\eta$/mPa.s	4.386	2.571	−26.724	0.074
V^E/cm^3·mol^{-1}	−322.012	487.006	144.116	0.161
	Bromobenzene + 1,4-Dioxane + Toluene			
$\Delta\eta$/mPa·s	5.857	−5.961	−35.474	0.097
V^E/cm^3·mol^{-1}	−176.46	166.21	844.00	0.283
	Bromobenzene + 1,4-Dioxane + Carbon Tetrachloride			
$\Delta\eta$/mPa·s	8.103	−1.704	−30.143	0.071
V^E/cm^3·mol^{-1}	−272.36	139.60	199.38	0.175

[a] $\Delta\eta$, viscosity deviations; V^E, excess volumes.

where x_i, η_i, and η refer respectively to the mole fraction and viscosities of ith pure components and of the binary and ternary mixtures:

$$V^E = V - \sum_{i=1}^{n} x_i V_i \tag{2}$$

where V_i represents the molar volume and x_i represents the mole fraction of the ith component. The quantity V refers to the molar volume of the binary and ternary mixtures, which can be calculated from the mixture density ρ and the component molecular weights M_i as given below:

$$V = \sum_{i=1}^{n} x_i M_i / \rho \tag{3}$$

where the letters have their usual significance.

The experimentally determined values of $\Delta\eta$ and V^E from eqs 1 and 2 have been presented in Table 3. The experimentally determined viscosity deviations ($\Delta\eta$) and excess molar volumes (V^E) for the following associated binary mixtures were fitted to a Redlich−Kister's equation:[8]

$$Y_{ij}^E = x_i x_j \sum_{p=0}^{p} A_p (x_i - x_j)^p \tag{4}$$

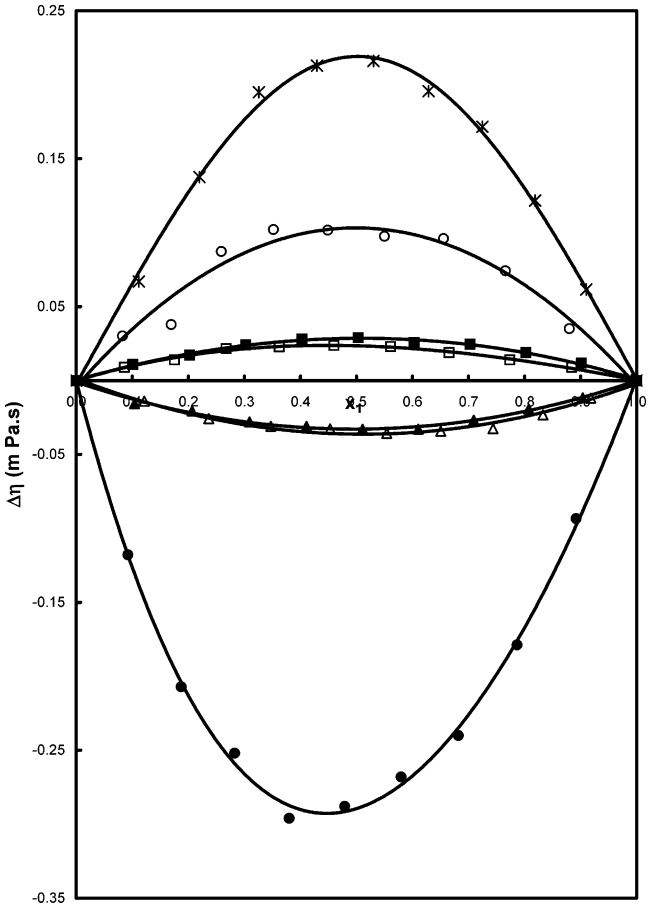

Figure 1. Variation of viscosity deviations $\Delta\eta$ with mole fraction x_1 of the first component in binary liquid mixtures of ○, bromobenzene + 1,4-dioxane; ▲, 1,4-dioxane + benzene; □, bromobenezene + benzene; ■, bromobenzene + toluene; △, 1,4-dioxane + toluene; ●, bromobenzene + carbon tetrachloride; and ∗, 1,4-dioxane + carbon tetrachloride at 303.15 K.

where Y_{ij}^E is $\Delta\eta$ or V^E; x_i denotes the mole fraction of component i of the i,j mixture with $x_j = 1 - x_i$; and A_p is the adjustable parameters. The values of adjustable parameters (A_p)

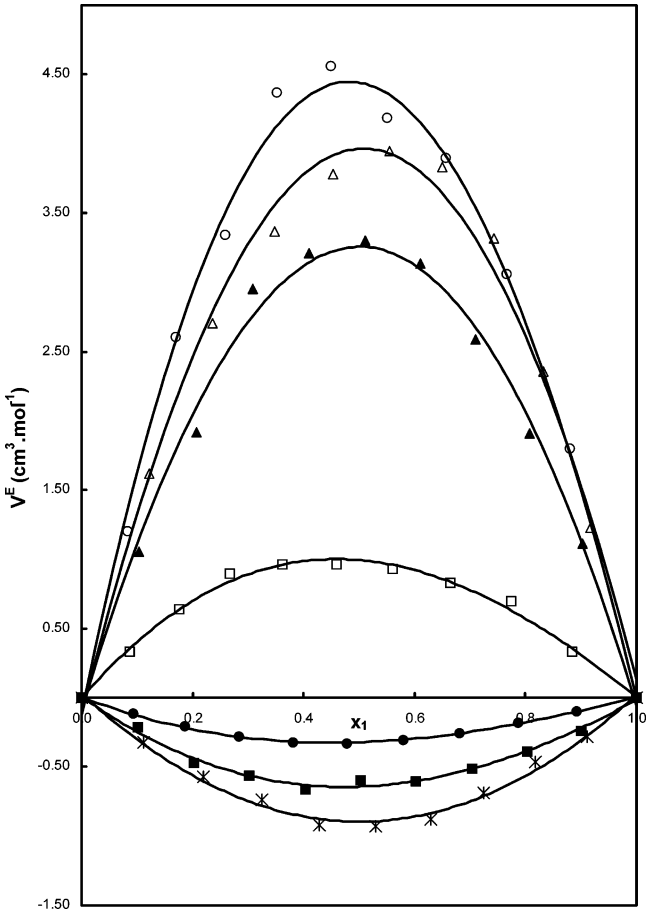

Figure 2. Variation of excess molar volumes V^E with mole fraction x_1 of the first component in binary liquid mixtures of ○, bromobenzene + 1,4-dioxane; ▲, 1,4-dioxane + benzene; □, bromobenezene + benzene; ■, bromobenzene + toluene; △, 1,4-dioxane + toluene; ●, bromobenzene + carbon tetrachloride; and ∗, 1,4-dioxane + carbon tetrachloride at 303.15 K.

of eq 4 along with the standard deviations are presented in Table 4.

The viscosity deviations ($\Delta\eta$) and the excess molar volumes (V^E) for the ternary mixtures were fitted to Cibulka's equation:[8]

$$Y^E = Y^E_{bin} + x_1x_2x_3[B_1 + B_2x_1 + B_3x_2] \quad (5)$$

where

$$Y^E_{bin} = Y^E_{12} + Y^E_{13} + Y^E_{23} \quad (6)$$

and Y^E_{ij} (for ij binary mixtures) are given by eq 4.

Using the values of A_p (eq 4), the values of Y^E_{12}, Y^E_{13}, and Y^E_{23} were obtained from eq 4 on substituting the values of corresponding mole fractions of the components. Having thus obtained the values of Y^E_{bin}, the experimental values of $\Delta\eta$ and V^E for ternary mixtures were fitted to eq 5 to obtain the values of coefficients B_1, B_2, and B_3 by the method of least squares.

After having determined the values of coefficients B_1, B_2, and B_3, the calculated values of $\Delta\eta$ and V^E were obtained from eq 5. The standard deviation in regard to experimental and theoretical values of $\Delta\eta$ and V^E for the ternary mixtures was obtained using the following expression:[10]

$$\sigma(Y^E) = \left(\frac{\sum_{i=1}^{n}(Y^E_{exptl} - Y^E_{calc})^2}{(n-p)}\right)^{1/2} \quad (7)$$

where Y^E is $\Delta\eta$ or V^E, n is the number of experimental data, and p is the number of parameters in eq 5. The values of coefficients B_1, B_2, and B_3 and $\sigma(\Delta\eta)$ and $\sigma(V^E)$ are presented in Table 5. From the values of σ, it is seen that the experimental values on $\Delta\eta$ or V^E compare fairly well with those calculated from Cibulka's equation.

The plots of $\Delta\eta$ and V^E versus x_1 in respect of the binary mixtures have been presented in Figures 1 and 2. It is seen that in each case the plots are of parabolic shape and are characterized by the presence of well-defined maxima/minima, which indicate the presence of complex formation[2,11] between the mixing components of binary mixtures. The interactions in a ternary $i + j + k$ mixtures are closely dependent[12] on the interactions in the constituent $i + j$, $j + k$, and $i + k$ mixtures. It should therefore be possible to correlate the nature of interactions occurring in ternary mixtures with those of binary mixtures.

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