

\bar{V}_e	decrease in volume due to electrostriction
\bar{V}_f°	limiting effective flow volume
\bar{V}_{int}	intrinsic volume of the amino acid
V_s	shrinkage volume
V_v	volume associated with void
V_{vw}	van der Waals volume
V_{sh}	volume of solvation sheath

Greek Letters

ϕ_v	apparent molar volume
ϕ_v°	limiting apparent molar volume or infinite-dilution partial molar volume
$\phi_v^\circ(\text{tr})$	change in partial molar volume when amino acid is transferred from water to aqueous NH_4Cl solution
η	viscosity of solution
η_0	viscosity of pure solvent
η_r	relative viscosity (η/η_0)
ρ	density of solution
ρ_0	density of pure solvent

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Supplementary Material Available: Tables of detailed density and viscosity data for different amino acids as a function of concentration in aqueous ammonium chloride solutions (15 pages). Ordering information is given on any current masthead page.

Viscosity and Density of Ternary Mixtures of Toluene, Bromobenzene, 1-Hexanol, and 1-Octanol

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Mixture viscosities and densities of the ternary mixtures of toluene, bromobenzene, 1-hexanol, and 1-octanol were measured at 30, 40, 50, and 60 °C. The nonidealities reflected in mixture viscosities are expressed and discussed in terms of excess viscosities, which were negative in most of the cases. The ternary η -X-T data were fitted in a Redlich-Kister-type equation along with a ternary contribution term.

Introduction

Extending our earlier work (1-8) on viscosities and dielectric constants of liquid mixtures, the present paper reports the

viscosities and the densities for the ternary mixtures of toluene, bromobenzene, 1-hexanol, and 1-octanol in the temperature range from 30 to 60 °C.

Experimental Section

Materials. Toluene (BDH), 1-hexanol (BDH), and 1-octanol (Ferak Berlin) were fractionally distilled and dried while bromobenzene (E. Merck) after repeated fractional distillation was collected at 156 ± 0.5 °C and retained for use. The mean values of repeat density, viscosity, and refractive index measurements of the liquids so purified did not deviate from the corresponding literature values beyond allowable limits (Table IV). Redistilled deionised and degassed water (electrical conductivity $< 7.0 \times 10^{-7} \Omega^{-1} \text{ cm}^{-1}$) was used in each case for checking the instruments and calibrating the pycnometers.

Experimental Measurements. Ternary mixtures were prepared by weight with an accuracy of 0.0001 g, taking care that the resulting ternary compositions represent the data points suitably distributed away from the vertices and also located in

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Table I. Experimental Densities ρ_m and Viscosities η_m for the Ternary Mixture Toluene (1)-Bromobenzene (2)-1-Octanol (3) at Different Temperatures

no.	X_1	X_2	t, °C	$\rho_m, \text{g mL}^{-1}$	η_m, cP
1	0.9033	0.0580	30	0.89481	0.5587
			40	0.88885	0.5034
			50	0.88285	0.4584
			60	0.87813	0.4255
2	0.0561	0.9058	30	1.4217	0.9434
			40	1.4143	0.8550
			50	1.4111	0.7844
			60	1.4092	0.7109
3	0.0782	0.0796	30	0.86259	4.293
			40	0.85812	3.201
			50	0.85545	2.485
			60	0.85349	1.970
4	0.7518	0.1488	30	0.94019	0.7752
			40	0.93583	0.6775
			50	0.93258	0.6087
			60	0.92914	0.5585
5	0.1446	0.7552	30	1.3117	1.055
			40	1.3046	0.9341
			50	1.3010	0.8397
			60	1.2991	0.7585
6	0.1814	0.1847	30	0.92221	2.541
			40	0.91670	2.024
			50	0.91429	1.637
			60	0.91185	1.355
7	0.3707	0.3774	30	1.0587	1.111
			40	1.0522	0.9722
			50	1.0482	0.8615
			60	1.0448	0.7766
8	0.5960	0.2423	30	0.99243	0.8902
			40	0.98594	0.7820
			50	0.98165	0.6958
			60	0.97847	0.6252
9	0.4567	0.4650	30	1.1364	0.7753
			40	1.1294	0.6802
			50	1.1235	0.6360
			60	1.1187	0.6051
10	0.2364	0.6029	30	1.2017	1.022
			40	1.1940	0.8939
			50	1.1904	0.8023
			60	1.1880	0.7275
11	0.1294	0.5222	30	1.1237	1.495
			40	1.1173	1.253
			50	1.1133	1.084
			60	1.1114	0.9470
12	0.2687	0.2737	30	0.97863	1.700
			40	0.97194	1.408
			50	0.96799	1.192
			60	0.96544	1.008
13	0.5164	0.1326	30	0.91379	1.245
			40	0.90844	1.045
			50	0.90597	0.8901
			60	0.90351	0.7809

extreme corners near the vertices of the triangular composition diagram.

Ostwald viscometers were used with necessary precautions for viscometric measurements (9). The standard deviations in the time of flow was found not to exceed 0.1%. For density measurements (9), the pyconometers used were calibrated with water with $0.997\ 07\ \text{g mL}^{-1}$ as its density at $25\ ^\circ\text{C}$.

For temperature control, a Toshniwal Model GL-15 precision thermostat, with the bath temperature monitored to $0.01\ ^\circ\text{C}$ with a standardized Beckmann thermometer, was used. During experiments, the bath temperature did not fluctuate beyond $\pm 0.1\ ^\circ\text{C}$ and also the evaporation of experimental liquids was checked to a minimum and remained insignificant.

The viscosities and densities were considered significant to four figures.

Table II. Experimental Densities ρ_m and Viscosities η_m for the Ternary Mixture Toluene (1)-1-Hexanol (2)-1-Octanol (3) at Different Temperatures

no.	X_1	X_2	t, °C	$\rho_m, \text{g mL}^{-1}$	η_m, cP
1	0.9479	0.0290	30	0.85869	0.5190
			40	0.85263	0.4586
			50	0.84732	0.4209
			60	0.84204	0.3911
2	0.0389	0.9347	30	0.81805	3.587
			40	0.81345	2.655
			50	0.81127	2.167
			60	0.80974	1.757
3	0.0479	0.0411	30	0.82397	5.337
			40	0.81951	3.927
			50	0.81680	3.026
			60	0.81522	2.367
4	0.0825	0.6372	30	0.82079	3.646
			40	0.81637	2.833
			50	0.81355	2.270
			60	0.81121	1.776
5	0.4176	0.0717	30	0.83464	1.894
			40	0.82986	1.541
			50	0.82655	1.281
			60	0.82362	1.081
6	0.6442	0.3071	30	0.84286	0.8856
			40	0.83709	0.7670
			50	0.83207	0.6770
			60	0.82813	0.5943
7	0.3941	0.3382	30	0.83147	1.841
			40	0.82519	1.519
			50	0.82213	1.312
			60	0.82001	1.141
8	0.1655	0.4972	30	0.82377	3.171
			40	0.81837	2.435
			50	0.81503	1.924
			60	0.81290	1.578
9	0.2384	0.5456	30	0.82626	2.507
			40	0.82167	1.937
			50	0.81867	1.543
			60	0.81526	1.285
10	0.3331	0.2144	30	0.83123	2.181
			40	0.82655	1.715
			50	0.82356	1.422
			60	0.82138	1.178
11	0.4811	0.1376	30	0.83669	1.555
			40	0.83134	1.315
			50	0.82885	1.116
			60	0.82619	0.9454
12	0.5182	0.3812	30	0.83752	1.211
			40	0.83179	1.008
			50	0.82763	0.8550
			60	0.82441	0.7372
13	0.5939	0.2548	30	0.84018	1.054
			40	0.83314	0.8946
			50	0.82794	0.7658
			60	0.82478	0.6768

Results and Discussions

The viscosity and density data for the ternary mixtures toluene (1)-bromobenzene (2)-1-octanol (3), toluene (1)-1-hexanol (2)-1-octanol (3), and bromobenzene (1)-1-hexanol (2)-1-octanol (3) at 30, 40, 50, and 60 °C are presented in Tables I-III. The component mole fractions were chosen in such a way that the experimental region so identified was expected to provide convenient and meaningful data leading to suitable correlations and useful conclusions.

The experimental measurements of viscosities and densities were made at 30, 40, 50, and 60 °C. The increment in temperature level was kept regular at 10 °C to ensure measurable effects of temperature change on experimental observations.

Table III. Experimental Densities ρ_m and Viscosities η_m for the Ternary Mixture Bromobenzene (1)-1-Hexanol (2)-1-Octanol (3) at Different Temperatures

no.	X_1	X_2	$t, ^\circ\text{C}$	$\rho_m, \text{g mL}^{-1}$	η_m, cP
1	0.9488	0.0286	30	1.4422	0.9735
			40	1.4342	0.8621
			50	1.4290	0.7812
			60	1.4219	0.7111
2	0.0396	0.9340	30	0.83923	3.618
			40	0.83435	2.760
			50	0.83234	2.166
			60	0.83165	1.734
3	0.0487	0.0411	30	0.85210	5.490
			40	0.84077	4.000
			50	0.83889	3.071
			60	0.83860	2.412
4	0.0839	0.6363	30	0.86371	3.793
			40	0.85907	2.909
			50	0.85536	2.258
			60	0.85274	1.815
5	0.4220	0.0711	30	1.0444	2.498
			40	1.0385	2.001
			50	1.0333	1.646
			60	1.0287	1.376
6	0.6483	0.3036	30	1.2200	1.371
			40	1.2136	1.178
			50	1.2099	1.019
			60	1.2072	0.9144
7	0.3984	0.3358	30	1.0423	2.156
			40	1.0331	1.735
			50	1.0249	1.463
			60	1.0166	1.217
8	0.1680	0.4957	30	0.90832	3.473
			40	0.90175	2.672
			50	0.89853	2.119
			60	0.89794	1.733
9	0.2417	0.5433	30	0.95304	2.897
			40	0.94710	2.295
			50	0.94451	1.814
			60	0.94285	1.486
10	0.3371	0.2131	30	1.0010	2.824
			40	0.99470	2.248
			50	0.99081	1.808
			60	0.98894	1.472
11	0.4856	0.1364	30	1.0889	2.071
			40	1.0838	1.719
			50	1.0800	1.432
			60	1.0776	1.219
12	0.5227	0.3776	30	1.1319	1.700
			40	1.1252	1.401
			50	1.1204	1.200
			60	1.1165	1.030
13	0.5982	0.2521	30	1.1697	1.553
			40	1.1663	1.315
			50	1.1639	1.126
			60	1.1619	0.9719

The highest temperature level was restricted to 60 °C in order to avoid errors due to evaporation losses during the experimental work.

The molecules of one or more of the components forming the ternaries are either polar or associating and accordingly show nonideal behaviors in mixtures. The nonidealities as reflected in mixture viscosities are expressed in terms of excess viscosity η^E given by the following equation

$$\eta^E = \eta_m - \sum X_i \eta_i \quad (1)$$

where η is the viscosity, X is the component mole fraction while superscript E stands for excess and subscript i and m stand for pure component and the mixture, respectively. With η^E-X-T data from Tables I-IV and eq 1, the corresponding η^E-X-T data were calculated.

Figures 1-3 show the plots of η^E vs η_m . Negative values of η^E in most of the cases are the consequence of lower viscosity

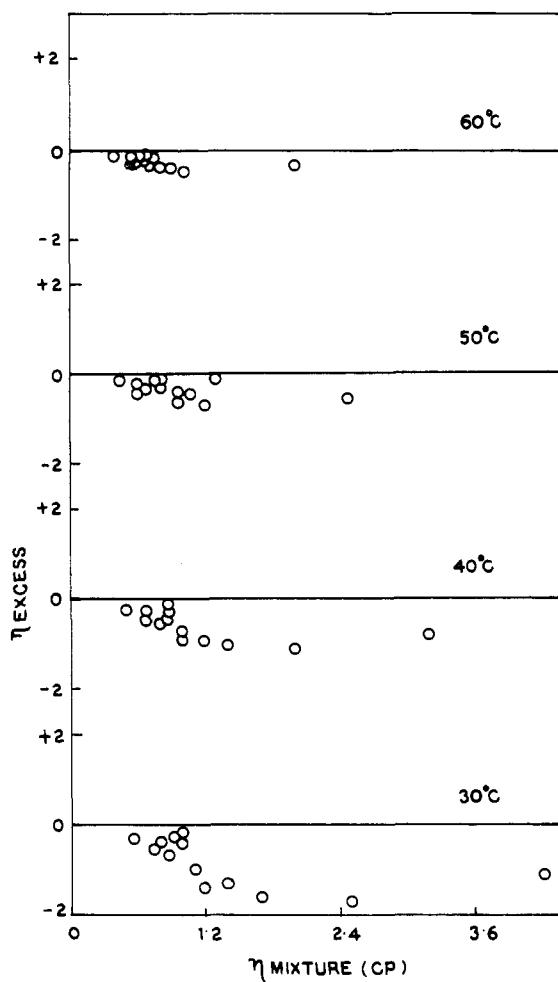


Figure 1. Plot of excess viscosity against corresponding mixture viscosity for the ternary toluene (1)-bromobenzene (2)-1-octanol (3) at 30, 40, 50, and 60 °C.

Table IV. Experimental Densities and Viscosities for Pure Components of the Ternaries

pure component	$t, ^\circ\text{C}$	$\rho, \text{g mL}^{-1}$	η, cP	ref
toluene	25	0.8623	0.5520	
		(0.86231)	(0.5516)	12
	30	0.86696	0.5372	
	40	0.85473	0.4851	
	50	0.84969	0.4272	
	60	0.84750	0.3905	
	25	1.4965	1.045	
		(1.48820)	(1.0430)	12
	30	1.4889	0.9850	
	40	1.4748	0.8744	
1-hexanol	50	1.4610	0.7819	
	60	1.4470	0.7129	
	25	0.8160	4.590	
		(0.81590)	(4.5920)	12
	30	0.81353	3.765	
	40	0.81050	2.934	
	50	0.80650	2.169	
	60	0.80340	1.655	
1-octanol	25	0.8262	6.298 ^a	
		(0.82209)	(6.125 ^a)	12
	30	0.82392	6.298	
	40	0.81926	4.577	
	50	0.81472	3.428	
	60	0.81039	2.678	

^a At 30 °C.

contributions of similar nonspecific interactions and H-bonding effects of molecular species in real mixtures rather than those in the corresponding ideal mixtures.

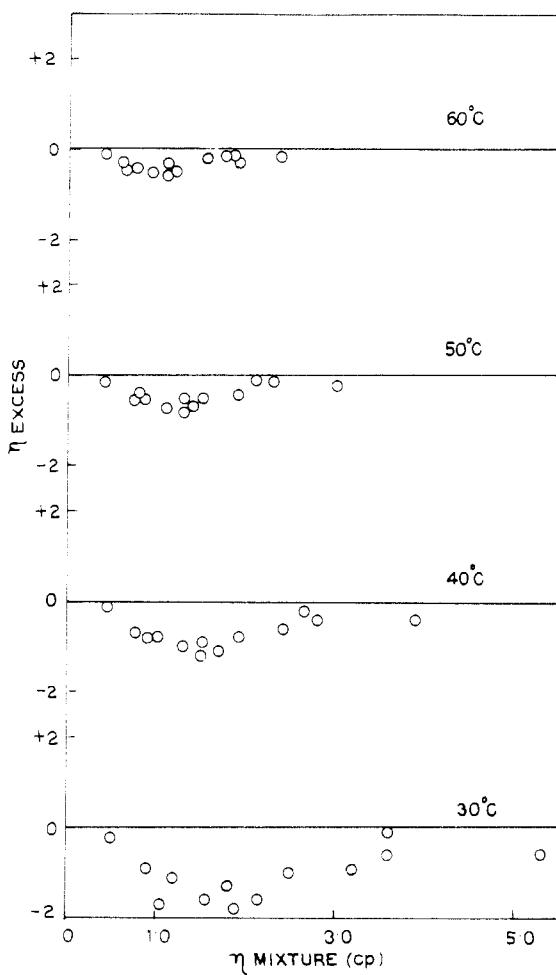


Figure 2. Plot of excess viscosity against corresponding mixture viscosity for the ternary toluene (1)-1-hexanol (2)-1-octanol (3) at 30, 40, 50, and 60 °C.

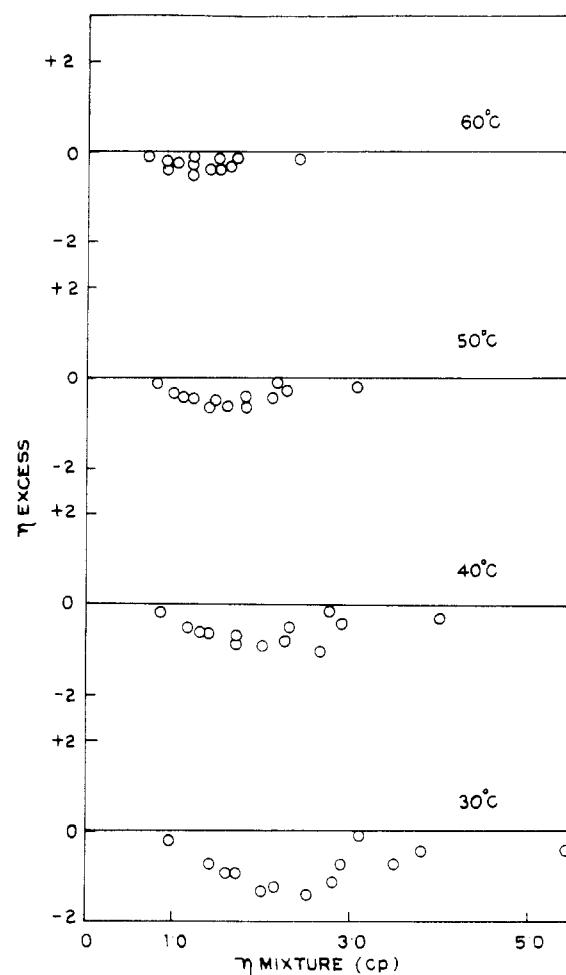


Figure 3. Plot of excess viscosity against corresponding mixture viscosity for the ternary bromobenzene (1)-1-hexanol (2)-1-octanol (3) at 30, 40, 50, and 60 °C.

Table V. Values of Binary Polynomial Constants A_{ij} , B_{ij} , C_{ij} and Additional Ternary Constant A^*_{ijk} Used in Equation 2 at Different Temperatures^a

systems	parameter	30 °C	40 °C	50 °C	60 °C
toluene (1)-bromobenzene (2) ^b	A_{12}	-0.2687	-0.2215	-0.1252	-0.1535
	B_{12}	-0.4618	-0.4316	-0.3819	-0.1741
	C_{12}	0.6807	0.6537	0.5806	-0.5685
bromobenzene (1)-1-octanol (2)	A_{12}	-7.0851	-4.7740	-3.3477	-2.6436
	B_{12}	1.2492	0.3444	-0.3452	-0.6920
	C_{12}	-3.7390	-3.5998	-2.8254	-2.4515
toluene (1)-1-octanol (2)	A_{12}	-7.3819	-4.8729	-3.3106	-2.5434
	B_{12}	3.3259	2.1651	1.3623	0.9412
	C_{12}	0.9632	-0.9081	-0.5458	-0.6506
toluene (1)-1-hexanol (2)	A_{12}	-3.8947	-2.9628	-2.0096	-1.5114
	B_{12}	1.3869	1.2676	0.7865	0.2333
	C_{12}	-3.8947	-0.6250	-0.5634	-0.0897
1-hexanol (1)-1-octanol (2)	A_{12}	-0.0023	-0.0855	-0.0801	0.0999
	B_{12}	2.2728	1.2523	1.5697	1.5038
	C_{12}	1.1830	1.1972	1.3014	0.8312
bromobenzene (1)-1-hexanol (2)	A_{12}	-3.0760	-2.2790	-1.3397	-0.7798
	B_{12}	0.8089	0.8572	0.1168	-0.1247
	C_{12}	-0.0898	-0.6156	0.1474	0.1338
toluene (1)-bromobenzene (2)-1-octanol (3)	A^*_{123}	11.652	9.620	7.836	8.425
toluene (1)-1-hexanol (2)-1-octanol (3)	A^*_{123}	-3.522	-3.564	-0.451	1.273
bromobenzene (1)-1-hexanol (2)-1-octanol (3)	A^*_{123}	1.964	2.894	3.394	3.329

^a Reference 11. ^bIf binary toluene (1)-bromobenzene (2) is changed to bromobenzene (1)-toluene (2), then the values of A_{ij} and C_{ij} will remain the same. The value of B_{ij} will remain the same but its sign will change.

The ternary η - X - T data were fitted in eq 2, which includes the contributions of each constituent binary as calculated by a three-parameter Redlich-Kister-type equation (10, 11) along with a ternary contribution term besides the contribution on an ideal mixture basis. The A_{ij} , B_{ij} , and C_{ij} were determined by

$$\eta_m = \sum_i^3 X_i \eta_i + \sum_{i \neq j}^3 X_i X_j [A_{ij} + B_{ij}(X_j - X_i) + C_{ij}(X_j - X_i)^2] + X_i X_j X_k A^*_{ijk} \quad (2)$$

least-squares method for the constituent binary using corre-

Table VI. Root Mean Square (rms) Deviations for Different Ternary Systems with Use of Equation 2 at Different Temperatures

	rms ^a				
	30 °C	40 °C	50 °C	60 °C	mean
toluene (1)-bromobenzene (2)-1-octanol (3)	0.0845	0.0787	0.0796	0.0911	0.0835
toluene (1)-1-hexanol (2)-1-octanol (3)	0.0606	0.0423	0.0351	0.0449	0.0457
bromobenzene (1)-1-hexanol (2)-1-octanol (3)	0.0465	0.0434	0.0499	0.0547	0.0485

^a rms deviation = $[\sum d_i^2/n]^{1/2}$ where n is the number of observations and $d = [(\eta_{\text{exptl}} - \eta_{\text{calcd}})/\eta_{\text{exptl}}]$.

sponding binary η_m-X-T data (11). A A^*_{ijk} of eq 2 is the additional ternary constant to be evaluated by experimental ternary η_m-X-T data by the least-squares method. Such values of A_{ij} , B_{ij} , C_{ij} , and A^*_{ijk} are listed in Table V. The parameters included in Table IV were used as input in eq 2 for calculating $\eta-X-T$ data showing root mean square deviations as listed in Table VI.

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Glossary

T	temperature, K
t	temperature, °C
X	mole fraction

Greek Letters

η	absolute viscosity, cP
ρ	density, g mL ⁻¹

Subscripts

i	component in a mixture
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m mixture
1,2,3 component number in a mixture

Superscript

E excess quantity

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NEW COMPOUNDS

Synthesis of

1-Aryl-2-hydrazino-4-phenyl-1,6-dihydro-1,3,5-triazine-6-thiones and Related Thiosemicarbazides

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Different 1-aryl-2-(benzylthio)-4-phenyl-1,6-dihydro-1,3,5-triazine-6-thiones have been synthesized by known methods. These triazines on treatment with hydrazine hydrate under suitable conditions afforded corresponding hydrazino derivatives. 1-Aryl-2-hydrazino-4-phenyl-1,6-dihydro-1,3,5-triazine-6-thione on interaction with aryl/alkyl isothiocyanates gave related thiosemicarbazides.

The presence of antituberculous (1, 2), antibacterial, antiviral (3), and antifungal (4) activity in some substituted thiosemicarbazides prompted us to search for new members of this series containing the triazinyl moiety.

The present paper deals with the synthesis of 1-aryl-2-hydrazino-4-phenyl-1,6-dihydro-1,3,5-triazine-6-thiones (II) and *N*-aryl/alkyl-*N*''-(1-aryl-4-phenyl-1,6-dihydro-6-thioxo-1,3,5-triazin-2-yl)thiosemicarbazides (III). The precursor 1-aryl-2-(benzylthio)-4-phenyl-1,6-dihydro-1,3,5-triazine-6-thione (I) was obtained by condensation of benzoyl isothiocyanate (5) and