Dielectric Behavior of the Ternary Systems of Toluene, Chlorobenzene, and 1-Hexanol with *n*-Hexane–Benzyl Alcohol as Their Partially Miscible Binary Subsystem

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Mixture dielectric constants ϵ_m were measured for the partially miscible ternary systems of toluene, chlorobenzene, and 1-hexanol with their partially miscible binary subsystem *n*-hexane–benzyl alcohol at 30, 40, 50, and 60 °C. Also the values of ϵ_m were calculated by an equation based on significant liquid structure (SLS) theory using pure component properties only for the systems studied. A comparison of the calculated and experimental data showed that the SLS equation is sufficiently useful for predicting the dependence of ϵ_m on the composition falling in the miscible region as well as on the binodal curves of these systems at the temperatures studied.

Introduction

Experimental dielectric constant–composition–temperature data for completely miscible binary (*1–3*) and ternary (*4*, *5*) systems of *n*-hexane, toluene, chlorobenzene, 1-hexanol, and benzyl alcohol as well as their optimal portrayal using significant liquid structure (SLS) theory (*6*) were presented earlier. Following a logical sequence in our investigation program, we now report the dielectric constants of the mixture compositions falling in the miscible regions as well as on the binodal curves of the ternary systems, *n*-hexane (1)–benzyl alcohol (2)–toluene (3), *n*-hexane (1)–benzyl alcohol (2)–chlorobenzene (3), *n*-hexane (1)–benzyl alcohol (2)–1-hexanol (3) and their partially miscible binary subsystem *n*-hexane (1)–benzyl alcohol (2) at 30, 40, 50, and 60 °C.

Experimental Section

Materials. Samples of similar liquids as used before (*1–5*, *7*) were purified by fractional distillation and drying and their purity was rechecked by measuring their densities, viscosities, and refractive indices at 25 ± 0.1 °C. The experimental values as reported in the preceding paper (*8*) agreed with the corresponding published literature values (*9–11*) within allowable limits.

Experimental Measurements. Appropriate quantities of the pure components were weighed and mixed in a thoroughly cleansed and dried bottle which was then stoppered, and the contents were shaken well and thermostated to prepare ternary liquid mixtures with compositions falling in the miscible region at the desired temperature. The component concentrations in the ternary mixtures, so prepared, were obtained from the respective weights. The estimated uncertainty in weighing was ±0.0001 g.

In order to prepare ternary liquid mixtures which fall on the binodal curve when plotted on a triangular composition diagram, appropriate quantities by weight of the pure liquids were taken in a thoroughly cleansed and dried separatory funnel and a closed rubber tube was put over its outlet to keep out the thermostat liquid. The separatory funnel was then placed in a thermostat set at 30 °C with frequent shaking. After the contents came to equilibrium, sufficient time was permitted for complete phase separation. The component concentrations of the phases, so separated, were determined chromatographically. It may be pointed out here that a ternary mixture com-

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Table I. Experimental Dielectric Constants ϵ_m for Data Points in the Miscible Region of the Ternary System *n*-Hexane (1)–Benzyl Alcohol (2)–Toluene (3) at Different Temperatures

		ϵ_m			
X_1	X_2	30 °C	40 °C	50 °C	60 °C
0.0487	0.3508	4.157	4.115	4.084	4.037
0.1151	0.4183	4.694	4.589	4.537	4.469
0.1671	0.2763	3.552	3.516	3.485	3.422
0.2179	0.5754	6.267	6.199	6.095	6.017
0.2889	0.6482	7.033	6.981	6.919	6.851
0.3455	0.5053	5.142	5.095	5.043	4.975
0.4065	0.1574	2.625	2.614	2.598	2.588
0.4843	0.2280	2.922	2.880	2.823	2.802
0.5476	0.0696	2.213	2.198	2.177	2.156

Table II. Experimental Dielectric Constants ϵ_m for Data Points in the Miscible Region of the Ternary System *n*-Hexane (1)–Benzyl Alcohol (2)–Chlorobenzene (3) at Different Temperatures

		ϵ_m			
X_1	X_2	30 °C	40 °C	50 °C	60 °C
0.0474	0.3414	6.413	6.293	6.163	5.976
0.1127	0.4096	6.575	6.445	6.293	6.121
0.1630	0.2694	5.376	5.314	5.236	5.121
0.2158	0.5700	7.002	6.914	6.762	6.606
0.2880	0.6464	7.726	7.633	7.534	7.336
0.3432	0.5019	5.824	5.757	5.632	5.496
0.3986	0.1543	3.933	3.902	3.855	3.771
0.4780	0.2251	3.761	3.745	3.688	3.609
0.5352	0.0684	3.250	3.193	3.151	3.115

Table III. Experimental Dielectric Constants ϵ_m for Data Points in the Miscible Region of the Ternary System *n*-Hexane (1)–Benzyl Alcohol (2)–1-Hexanol (3) at Different Temperatures

		ϵ_m			
X_1	X_2	30 °C	40 °C	50 °C	60 °C
0.0534	0.3845	11.22	10.81	10.19	9.446
0.1235	0.4489	10.09	9.811	9.321	8.612
0.1819	0.3007	9.144	8.925	8.419	7.929
0.2246	0.5933	8.607	8.352	8.065	7.622
0.2915	0.6543	8.070	7.935	7.700	7.309
0.3532	0.5166	6.742	6.606	6.445	6.268
0.4342	0.1680	5.131	4.996	4.868	4.860
0.5055	0.2381	4.615	4.501	4.444	4.329
0.5799	0.0739	3.678	3.620	3.500	3.412

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Table IV. Experimental Dielectric Constants ϵ_m for Data Points in the Miscible Region of the Binary System *n*-Hexane (1)-Benzyl Alcohol (2) at Different Temperatures

X_1	ϵ_m 30 °C	40 °C	50 °C	60 °C
	Binary Rich in Benzyl Alcohol			
0.0000	11.92	11.02	10.29	9.811
0.0312	11.45	10.60	9.911	9.451
0.0645	10.95	10.15	9.550	9.003
0.0976	10.50	9.753	9.154	8.654
0.1313	9.952	9.300	8.753	8.201
0.1653	9.503	8.852	8.253	7.805
0.2056	8.851	8.252	7.682	7.300
0.2550[a]	8.201	7.653	7.140	6.783
	Binary Rich in *n*-Hexane			
0.8792[a]	2.222	2.182	2.166	2.161
0.9550	2.020	1.993	1.975	1.968
0.9699	1.982	1.950	1.940	1.930
0.9799	1.953	1.925	1.915	1.908
0.9899	1.925	1.900	1.894	1.881
1.0000	1.895	1.875	1.865	1.858

[a] Binary saturated at 30 °C.

Table V. Experimental Dielectric Constants ϵ_m for Data Points Which Fall on Binodal Curve at 30 °C and in the Miscible Region at 40, 50, and 60 °C of the Ternary System *n*-Hexane (1)-Benzyl Alcohol (2)-Toluene (3)

X_1	X_2	ϵ_m 30 °C	40 °C	50 °C	60 °C
0.3306	0.6043	6.966	6.773	6.362	6.267
0.4672	0.4288	4.720	4.401	4.240	4.024
0.6099	0.2755	3.396	3.234	3.141	3.104
0.6954	0.2093	3.078	2.967	2.796	2.781
0.8267	0.1429	2.472	2.392	2.323	2.308
0.2883	0.6944	8.163	7.538	7.124	6.812
0.8149	0.1215	2.429	2.382	2.312	2.307
0.2722	0.6929	8.020	7.482	7.020	6.715
0.7576	0.1544	2.726	2.698	2.667	2.635
0.3124	0.6201	7.012	6.862	6.556	6.314

position corresponding to a point falling on the binodal curve represents a dissolved ternary mixture saturated with its partially miscible component. The details of phase separation, chromatographic method, and gas chromatograph as well as the column used remained the same as employed in the preceding paper (*8*).

The mixture dielectric constants were measured with the help of a Toshniwal dipolemeter Type RL 09, working on the heterodyne beat principle. The experimental procedure followed was as described elsewhere (*1*).

For temperature control, a thermostat, whose bath temperature was monitored to ±0.01 °C with a standard Beckmann thermometer, was used for equilibrating the liquids at the desired temperature during phase separation experiments and to circulate thermostated water around the cell used for the dielectric constant measurements. The temperature fluctuations inside the cell did not exceed ±0.1 °C in any experiment. The accuracy of the measured dielectric constants was estimated to be within 2% at 30, 40, and 50 °C and within 3% at 60 °C whereas the measurements were precise to ±0.002. The molar volumes as calculated from the experimental densities (*12*) were considered significant to four figures whereas refractive indices were obtained at desired temperatures from smoothened literature data significant to five figures (*9-11*).

Results and Discussion

Experimental dielectric constant–composition–temperature data for mixture compositions in the miscible region at 30, 40, 50, and 60 °C are listed in Tables I–IV. Similar data for mixture compositions which fall on the binodal curves at 30 °C and in the miscible region at 40, 50, and 60 °C are listed in Tables V–VII. With a view to assess the applicability of the SLS theory, which proved quite useful (*1-5*) in predicting the mixture dielectric constant data for completely miscible binary as well as ternary liquid systems, the following SLS equation, used earlier, was extended to the partially miscible liquid systems as well

$$\frac{3\epsilon_m(\epsilon_m - n_m^2)}{(2\epsilon_m + n_m^2)} = \frac{4\pi N}{V_m}\left[\frac{\epsilon_m(n_m^2 + 2)}{(2\epsilon_m + n_m^2)}\right]^2\left[\frac{V_{sm}}{V_m}\left(\sum_i^3 \frac{X_i^2\mu_i^2 G_i}{kT} + \sum_{i\neq j}^3 \frac{2X_iX_j\mu_i\mu_j G_{ij}}{kT}\right) + \frac{V_m - V_{sm}}{V_m}\left(\sum_i^3 \frac{X_i\mu_i^2}{3kT}\right)\right] \quad (1)$$

where ϵ is the dielectric constant, k is the Boltzmann constant, μ is the dipole moment of the free molecule, and V_s is the molar volume of the substance just before melting. The value of V_s was obtained by the method described earlier (1).

The mixture parameters n_m, V_m, V_{sm}, and G_{ij} were obtained with the help of the corresponding pure component values employing the following mixture rules (*13*, *14*)

$$Q_m = \sum_i^3 X_iQ_i \quad (2)$$

$$n_m = \sum_i^3 X_i^2 n_i + \sum_{i\neq j}^3 2X_iX_jn_{ij} \quad (3)$$

$$P_{ij} = (P_iP_j)^{1/2} \quad (4)$$

where Q stands for V and V_s and P stands for n and G. The subscript m stands for mixture and G is the correlation parameter. For each pure liquid, the value of G at each temperature was calculated by using an equation obtained by putting $X_2 = X_3 = 0$ and dropping the subscripts in eq 1. The values of V, μ, and G for the ternary components were taken from an earlier work (*1*, *3*) and are listed in Table VIII while the values of V_s and n were taken from the preceding paper (*8*).

Table VI. Experimental Dielectric Constants ϵ_m for Data Points Which Fall on Binodal Curve at 30 °C and in the Miscible Region at 40, 50, and 60 °C of the Ternary System *n*-Hexane (1)-Benzyl Alcohol (2)-Chlorobenzene (3)

X_1	X_2	ϵ_m 30 °C	40 °C	50 °C	60 °C
0.3883	0.5499	6.127	6.028	5.908	5.835
0.5568	0.3535	4.058	3.948	3.891	3.855
0.5578	0.3634	4.188	4.136	4.079	4.016
0.7731	0.1831	2.557	2.495	2.469	2.416
0.3494	0.6171	7.502	7.204	7.073	6.712
0.8080	0.1590	2.317	2.276	2.234	2.203
0.2841	0.6933	8.056	7.443	7.145	6.984

Table VII. Experimental Dielectric Constants ϵ_m for Data Points Which Fall on Binodal Curve at 30 °C and in the Miscible Region at 40, 50, and 60 °C of the Ternary System *n*-Hexane (1)-Benzyl Alcohol (2)-Hexanol (3)

X_1	X_2	ϵ_m 30 °C	40 °C	50 °C	60 °C
0.4718	0.4837	5.387	5.226	5.084	5.017
0.5203	0.4189	4.871	4.761	4.668	4.599
0.3379	0.6289	7.393	7.263	6.945	6.815
0.6549	0.3011	3.610	3.563	3.516	3.479
0.7579	0.2197	3.214	3.078	3.026	3.005
0.3599	0.6106	6.497	6.325	6.111	6.022
0.8353	0.1581	2.487	2.451	2.425	2.399
0.3001	0.6751	8.061	7.512	7.129	6.978

Table VIII. Values of Molar Volume, Dielectric Constant, Adjustable Parameter G in SLS Equation, and Dipole Moment for Selected Liquids

parameter	t, °C	n-hexane	toluene	chlorobenzene	1-hexanol	benzyl alcohol
V, cm^3 mol^{-1}	25	131.59 (131.61)[a]	106.86 (106.85)[a]	102.23 (102.23)[b]	125.22 (125.23)[a]	103.85 (103.85)[a]
	30	130.94	106.3	102.3	125.6	103.8
	40	132.2	107.8	102.9	126.1	104.1
	50	133.9	108.4	103.4	126.7	104.5
	60	135.5	108.7	103.9	127.2	104.8
ϵ	25	1.906 (1.879)[a]	2.416 (2.379)[a]	5.68 (5.621)[a]	13.4 (13.30)[a]	
	30	1.890	2.297	5.34	12.50	11.92
	40	1.879	2.281	5.27	11.46	11.02
	50	1.869	2.271	5.20	10.71	10.30
	60	1.859	2.260	5.10	10.09	9.81
G[c]	30	0.9118	0.1951	0.2770	1.356	0.7255
	40	1.399	0.2118	0.2863	1.283	0.6908
	50	1.829	0.2444	0.2952	1.242	0.6635
	60	2.207	0.2752	0.2997	1.211	0.6526
μ, D		0.085[a]	0.31[a]	1.54[a]	1.55[a]	1.66[a]

[a] Reference 9. [b] Reference 10. [c] References 1 and 3.

Table IX. Root Mean Square (rms) Deviations for Different Binary and Ternary Systems Using SLS Equation[a] at Different Temperatures for Data Points in Miscible Region

system	rms[b] dev using eq 1				mean[c]
	30 °C	40 °C	50 °C	60 °C	
1. n-hexane (1)–benzyl alcohol (2)–toluene (3)	0.1461	0.1643	0.1782	0.1851	0.1684
2. n-hexane (1)–benzyl alcohol (2)–chlorobenzene (3)	0.1204	0.1348	0.1458	0.1477	0.1372
3. n-hexane (1)–benzyl alcohol (2)–1-hexanol (3)	0.1121	0.1388	0.1524	0.1501	0.1384
4. n-hexane (1)–benzyl alcohol (2) (binary rich in benzyl alcohol)	0.0857	0.0974	0.0971	0.0969	0.0940
5. n-hexane (1)–benzyl alcohol (2) (binary rich in n-hexane)	0.0423	0.0381	0.0305	0.0236	0.0337

[a] Equation 1. [b] Rms deviation = $[\sum d_i^2/n]^{1/2}$ where n is the number of observations and $d = [(\epsilon_{exptl} - \epsilon_{calcd})/\epsilon_{exptl}]$. [c] Overall mean = 0.1143.

Table X. Root Mean Square (rms) Deviations for Different Ternary Systems with SLS Equation[a] for Data Points Falling on the Binodal Curves at 30 °C and in the Miscible Region at 40, 50, and 60°

ternary system	rms[b] dev using eq 1				mean[c]
	30 °C	40 °C	50 °C	60 °C	
1. n-hexane (1)–benzyl alcohol (2)–toluene (3)	0.1956	0.1968	0.1882	0.1735	0.1885
2. n-hexane (1)–benzyl alcohol (2)–chlorobenzene (3)	0.1784	0.1983	0.2027	0.2179	0.1993
3. n-hexane (1)–benzyl alcohol (2)–1-hexanol (3)	0.1826	0.2008	0.2256	0.2249	0.2084

[a] Equation 1. [b] Rms deviation = $[\sum d_i^2/n]^{1/2}$ where n is the number of $d = [(\epsilon_{exptl} - \epsilon_{calcd})/\epsilon_{exptl}]$. [c] Overall mean = 0.1987.

The experimental ϵ_m–X–T data were compared with those predicted by eq 1. The root mean square (rms) deviations for the data listed in Tables I–IV are given in Table IX. The average rms deviations for mixture compostions in the miscible region of the ternary systems n-hexane (*1*)–benzyl alcohol (2)–toluene (3), *n*-hexane(1)–benzyl alcohol (2)–chlorobenzene (3), *n*-hexane (1)–benzyl alcohol (2)–1–hexanol (3), and benzyl alcohol-rich as well as *n*-hexane-rich binaries of *n*-hexane (1)–benzyl alcohol (2), in the temperature range studied are 0.1684, 0.1372, 0.1384, 0.094, and 0.0337, respectively, with an overall average of 0.1143 for all the mixtures taken together. The rms deviations as listed in Table X for the mixture compositions falling on the binodal curves at 30 °C and in the miscible region at 40, 50, and 60 °C for the ternaries in the above order are 0.1885, 0.1993, and 0.2084, respectively, with an overall average 0.1987 for all such mixtures taken together.

As expected eq 1 predicts the binary data better than it does the ternary ones. Also eq 1 is equally useful in predicting the data for the ternary systems in the miscible region and those falling on the binodal curves. An additional advantage with eq 1 is that it needs only pure component data as input and does not involve any adjustable parameter to be evaluated by the use of experimental mixture data.

Glossary

k	Boltzmann constant
n	refractive index
G	SLS correlation parameter (eq 1)
N	Avogadro's number
T	absolute temperature
V	molar volume at the temperature of investigation, cm^3 mol^{-1}
V_s	molar volume of a substance just before melting, cm^3 mol^{-1}
X_i	mole fraction of component i
rms	root mean square deviation

Greek Letters

ϵ	dielectric constant
μ	dipole moment of the free molecule, D

Subscripts

D	for sodium light
i, j	component in a mixture
m	mixture
1, 2, 3	component numbers in a mixture

Registry No. Toluene, 108-88-3; chlorobenzene, 108-90-7; 1-hexanol, 111-27-3; *n*-hexane, 110-54-3; benzyl alcohol, 100-51-6.

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Excess Volumes for Binary Liquid Mixtures of 1,2-Dichloroethane with Benzene, Toluene, *p*-Xylene, Quinoline, and Cyclohexane

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Dilatometric measurements of excess volumes (V^E) have been made for binary liquid mixtures of 1,2-dichloroethane (CH_2ClCH_2Cl) with benzene, toluene, *p*-xylene, and quinoline at 298.15 and 308.15 K, and for mixtures of CH_2ClCH_2Cl with cyclohexane at 308.15 K. The values of V^E have been found to be positive for CH_2ClCH_2Cl–benzene, CH_2ClCH_2Cl–toluene, CH_2ClCH_2Cl–*p*-xylene, and CH_2ClCH_2Cl–cyclohexane and negative for CH_2ClCH_2Cl–quinoline.

Introduction

The binary systems of 1,2-dichloroethane (CH_2ClCH_2Cl) with aromatic hydrocarbons, quinoline, and cyclohexane are of considerable interest from the viewpoint of the presence of specific interaction between the components in the liquid state. The specific interaction of CH_2ClCH_2Cl with aromatic hydrocarbons and quinoline can be visualized to be due to the presence of two Cl atoms and four H atoms in CH_2ClCH_2Cl on account of which it can act as a σ-acceptor toward, and be involved in hydrogen-bond formation with, the aromatic hydrocarbons and quinoline. The aromatic hydrocarbons in their interaction with CH_2ClCH_2Cl will act as π-donors, whereas quinoline will act as an n-donor. The system CH_2ClCH_2Cl–cyclohexane, in which only dispersion, dipolar, and induction forces are believed to be present between the components, is of interest as it can be used as a reference system. Extensive studies of the interactions between the components of such systems have not been made. In the present program, we have made measurements of excess volumes for binary liquid mixtures of CH_2ClCH_2Cl with benzene, toluene, *p*-xylene, quinoline, and cyclohexane at various temperatures, and the results obtained are interpreted in this paper.

Experimental Section

Benzene (AR, BDH), toluene (AR, BDH), and *p*-xylene (Merck-Schuchardt) of synthesis quality were purified in a manner similar to that described by Nath and Tripathi (*1*). Cyclohexane (BDH) of reagent grade was subjected to treatments with nitrating mixture, with distilled water, and with dilute sodium hydroxide solution. Further, it was washed thoroughly with distilled water, and then dried over anhydrous calcium

Table I. Experimental Values of Excess Volumes for the Various Systems of CH_2ClCH_2Cl at 298.15 and 308.15 K

temp, K	x_1[a]	V^E, cm^3 mol^{-1}	temp, K	x_1[a]	V^E, cm^3 mol^{-1}
CH_2ClCH_2Cl–Benzene				0.6653	0.259
298.15	0.2699	0.193		0.7226	0.247
	0.2828	0.195		0.8547	0.161
	0.3399	0.213	308.15	0.1450	0.108
	0.4474	0.232		0.3411	0.217
	0.5467	0.228		0.4323	0.250
	0.6166	0.215		0.6021	0.273
	0.7359	0.175		0.6832	0.264
	0.7621	0.162		0.7584	0.237
308.15	0.1785	0.118		0.7768	0.225
	0.2040	0.131		0.8318	0.188
	0.2987	0.181			
	0.3643	0.205	CH_2ClCH_2Cl–Quinoline		
	0.4122	0.216	298.15	0.2266	−0.115
	0.4810	0.228		0.3594	−0.155
	0.6213	0.212		0.3624	−0.162
	0.7150	0.177		0.4125	−0.167
	0.7247	0.167		0.6100	−0.145
				0.7602	−0.096
CH_2ClCH_2Cl–Toluene			308.15	0.2630	−0.146
298.15	0.1482	0.061		0.2871	−0.154
	0.2351	0.092		0.4288	−0.182
	0.2385	0.095		0.5680	−0.169
	0.3548	0.127		0.6435	−0.154
	0.4812	0.157		0.7415	−0.120
	0.6687	0.155		0.7483	−0.114
	0.8418	0.102		0.8157	−0.089
	0.8939	0.073			
308.15	0.3370	0.141	CH_2ClCH_2Cl–Cyclohexane		
	0.3468	0.142	308.15	0.2045	0.750
	0.3990	0.156		0.2683	0.884
	0.5494	0.176		0.3361	0.978
	0.6726	0.166		0.4974	1.040
	0.7386	0.151		0.5275	1.031
	0.8751	0.092		0.7056	0.815
				0.7792	0.654
CH_2ClCH_2Cl–*p*-Xylene				0.8303	0.532
298.15	0.1577	0.100			
	0.2386	0.154			
	0.4482	0.254			
	0.5823	0.273			

[a] x_1 refers to the mole fraction of CH_2ClCH_2Cl.

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