Excess Molar Volumes, Relative Permittivities, and Refractive Indexes of 1,1,2,2-Tetrachloroethane + Pyridine, +Anisole, +Methyl Ethyl Ketone, and +1,4-Dioxane at 303.15 K

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Excess molar volumes, V_m^E, relative permittivities, ϵ_r, and refractive indexes, n_D, have been measured for binary mixtures of 1,1,2,2-tetrachloroethane ($CHCl_2CHCl_2$) with pyridine (C_5H_5N), anisole ($C_6H_5OCH_3$), methyl ethyl ketone ($CH_3COC_2H_5$), and 1,4-dioxane (1,4-$C_4H_8O_2$) at 303.15 K. V_m^E has been found to be negative throughout the entire mole fraction range for all these mixtures. The values of the deviations of ϵ_r and n_D from a mole fraction average, which are represented respectively by $\Delta\epsilon_r$ and Δn_D have been calculated. The results of V_m^E, Δn_D, and $\Delta\epsilon_r$ for the various mixtures have been fitted by the method of least squares to smoothing equations.

Introduction

Binary mixtures of 1,1,2,2-tetrachloroethane ($CHCl_2$-$CHCl_2$) with pyridine (C_5H_5N), anisole ($C_6H_5OCH_3$), methyl ethyl ketone ($CH_3COC_2H_5$), and 1,4-dioxane (1,4-$C_4H_8O_2$) are of considerable interest from the viewpoint of the existence of an electron donor−acceptor interaction leading to the formation of intermolecular complexes between the components. The specific interaction of C_5H_5N with $CHCl_2$-$CHCl_2$ can be thought of as being due to the presence of lone-pair electrons on the nitrogen atom of C_5H_5N, on account of which it can act as an n-donor toward $CHCl_2$-$CHCl_2$. On the other hand, the presence of lone-pair electrons on the oxygen atoms of $CH_3COC_2H_5$ and 1,4-$_8O_2$ can make these compounds act as n-donors, and as described by Mulliken (1963), $C_6H_5OCH_3$ which contains OCH_3 group and an aromatic ring system can act as an nπ-type donor toward $CHCl_2CHCl_2$, which can be involved in the formation of hydrogen bonds with and act σ-acceptor toward C_5H_5N, $C_6H_5OCH_3$, $CH_3COC_2H_5$, and 1,4-$C_4H_8O_2$. Although Nath and Tripathi (1983, 1984, 1986) have made measurements of excess molar volumes, , ultrasonic velocities, u, relative permittivities, ϵ_r, refractive indexes, n_D, and dynamic viscosities, η, for mixtures of $CHCl_2CHCl_2$ with dimethyl ketone and Chadha and Tripathi (1995) have measured excess molar enthalpies, H_m^E, for mixtures of $CHCl_2CHCl_2$ with 2-methylfuran, tetrahydrofuran, 1,4-dioxane, and cyclopentanone, extensive studies concerning interactions between components of binary mixtures of $CHCl_2CHCl_2$ with n-donor components have not been made. Hence, we have measured excess molar volume, V_m^E, relative permittivities, ϵ_r, and refractive indexes, n_D, of $CHCl_2CHCl_2$ + C_5H_5N, +$C_6H_5OCH_3$, +$CH_3COC_2H_5$, and +1,4-$C_4H_8O_2$, and the results of these measurements are reported and interpreted here.

Experimental Section

Materials. 1,1,2,2-Tetrachloroethane, methyl ethyl ketone and anisole, all three of AR quality, and 1,4-dioxane of uv spectral grade quality were obtained from Sisco Research Laboratories, Pvt. Ltd., Bombay. 1,1,2,2-Tetrachloroethane and methyl ethyl ketone were shaken with potassium carbonate solution, separated, and then dried over anhydrous potassium carbonate, followed by fractional distillations. Anisole was distilled from sodium. Pyridine of HPLC quality was obtained from S. D. Fine Chemicals Ltd. and was used without further purification. 1,4-Dioxane was stored over sodium wire and was used. The densities of these purified samples of the various liquids were measured using a single-capillary pycnometer and have been reported recently (Nath, 1996) as 0.972 86, 0.984 64, 0.794 49, 1.022 32, and 1.578 57 g·cm^{-3} for C_5H_5N, $C_6H_5OCH_3$, $CH_3COC_2H_5$, 1,4-$C_4H_8O_2$, and $CHCl_2CHCl_2$, respectively, at T = 303.15 K, as compared with the corresponding literature (Timmermans, 1950) values 0.972 81, 0.984 62, 0.794 52, 1.022 30, and 1.578 60 g·cm^{-3} for the various liquids in the same order.

Table 1. Refractive Indexes, n_D^*, and Relative Permittivities, ϵ_r^*, of the Pure Component Liquids at T = 303.15 K

	n_D^*		ϵ_r^*	
compd	obs	lit.	obs	lit.
C_5H_5N	1.5040	1.504 66[a]	12.164	12.17[e]
$C_6H_5OCH_3$	1.5118	1.511 60[b]	4.249	4.250[f]
$CH_3COC_2H_5$	1.3740	1.374 12[c]	17.664	17.65[g]
1,4-$C_4H_8O_2$	1.4180	1.418 09[d]	2.200	
$CHCl_2CHCl_2$	1.4885	1.488 45[d]	7.460	7.464[h]

[a] From Riddick and Bunger (1970). [b] Value obtained by extrapolation of data on n_D^* from Timmermans (1950). [c] From Timmermans (1950). [d] Value obtained by extrapolation of data on n_D^* from Riddick and Bunger (1970). [e] Nath (1995). [f] Value obtained by extrapolation on data on ϵ_r^* from Riddick and Bunger (1970) and Jacobs et al. (1944). [g] Nath and Saini (1990). [h] Value obtained by interpolation of data on ϵ_r^* from Nath and Tripathi (1984) and Riddick and Bunger (1970).

Methods. (i) Excess molar volumes, V_m^E, were measured with an imprecision of the order of ±0.002 cm^3·mol^{-1}, using a two-limbed Pyrex glass dilatometer, as described earlier (Nath and Chaudhary, 1992). Known amounts of the two liquid components were confined separately over mercury in the absence of air spaces in the two limbs of the dilatometer. The dilatometer (mounted on a stand) was immersed in a water thermostat, at (303.15 ± 0.01 K). The mixing of the liquid components was achieved by rocking the cell back and forth through a definite angle, and the mercury levels in the capillary of the dilatometer were noted with an accuracy of ±0.001 cm using a cathetometer.

(ii) Relative permittivities, ϵ_r, were measured at (303.15 ± 0.01) K and at 1.8 MHz with a decameter type DK_{03} (Wissenschaftlich-Technische, Werkstätten, Germany), using one cell MFL 1/S, no. 2078, for mixtures having ϵ_r <

S0021-9568(96)00054-4 CCC: $12.00

Table 2. Experimental Values of Excess Molar Volumes, V_m^E, for $CHCl_2CHCl_2$ + C_5H_5N, $C_6H_5OCH_3$, $CH_3COC_2H_5$, and 1,4-$C_4H_8O_2$ at T = 303.15 K

x	V_m^E/ $cm^3 \cdot mol^{-1}$	x	V_m^E/ $cm^3 \cdot mol^{-1}$	x	V_m^E/ $cm^3 \cdot mol^{-1}$	x	V_m^E/ $cm^3 \cdot mol^{-1}$
$xCHCl_2CHCl_2 + (1 - x)C_5H_5N$							
0.0475	−0.014	0.2020	−0.045	0.5549	−0.076	0.7857	−0.051
0.0664	−0.017	0.2661	−0.057	0.6000	−0.076	0.8378	−0.042
0.0925	−0.028	0.3551	−0.068	0.6577	−0.070	0.8798	−0.030
0.1530	−0.039	0.4615	−0.074	0.7348	−0.063	0.9269	−0.017
$xCHCl_2CHCl_2 + (1 - x)C_6H_5OCH_3$							
0.0587	−0.025	0.2476	−0.065	0.6061	−0.070	0.8899	−0.027
0.0769	−0.032	0.3022	−0.076	0.6765	−0.065	0.9309	−0.018
0.0986	−0.039	0.3449	−0.080	0.7254	−0.059		
0.1533	−0.047	0.4622	−0.083	0.7802	−0.053		
0.2009	−0.056	0.5291	−0.079	0.8131	−0.047		
$xCHCl_2CHCl_2 + (1 - x)CH_3COC_2H_5$							
0.0759	−0.195	0.2007	−0.439	0.5399	−0.685	0.8397	−0.315
0.0789	−0.201	0.2479	−0.506	0.6274	−0.636	0.8988	−0.196
0.1338	−0.323	0.3097	−0.578	0.6944	−0.570	0.9609	−0.071
0.1521	−0.353	0.3750	−0.638	0.8085	−0.378		
$xCHCl_2CHCl_2 + (1 - x)(1,4\text{-}C_4H_8O_2)$							
0.0615	−0.030	0.2620	−0.136	0.5322	−0.229	0.8489	−0.162
0.1118	−0.060	0.3002	−0.154	0.6082	−0.234	0.8970	−0.120
0.1624	−0.083	0.3353	−0.171	0.6564	−0.232	0.9213	−0.096
0.1913	−0.100	0.4033	−0.192	0.7532	−0.213	0.9539	−0.055
0.2310	−0.123	0.4813	−0.220	0.8150	−0.185		

Table 3. Relative Permittivities, ϵ_r, for $CHCl_2CHCl_2$ + $_5$N, $C_6H_5OCH_3$, $CH_3COC_2H_5$, and 1,4-$C_4H_8O_2$ at T = 303.15 K

x	ϵ_r	x	ϵ_r	x	ϵ_r	x	ϵ_r
$xCHCl_2CHCl_2 + (1 - x)C_5H_5N$							
0.0437	12.310	0.2448	12.589	0.4833	12.095	0.7438	10.418
0.0798	12.412	0.2893	12.569	0.5371	11.857	0.8154	9.730
0.1186	12.488	0.3360	12.520	0.5830	11.602	0.8822	8.998
0.1607	12.556	0.3847	12.422	0.6328	11.297	0.9287	8.428
0.1982	12.579	0.4332	12.284	0.6952	10.836		
$xCHCl_2CHCl_2 + (1 - x)C_6H_5OCH_3$							
0.0542	4.412	0.3007	5.106	0.5619	5.832	0.8498	6.802
0.1062	4.569	0.3568	5.257	0.6046	5.954	0.8945	6.990
0.1516	4.701	0.3996	5.378	0.6622	6.134	0.9499	7.226
0.2045	4.848	0.4474	5.508	0.7085	6.289		
0.2570	4.988	0.5100	5.680	0.7539	6.448		
$xCHCl_2CHCl_2 + (1 - x)CH_3COC_2H_5$							
0.0447	17.728	0.2727	16.938	0.5159	14.528	0.7653	11.052
0.0890	17.718	0.3169	16.601	0.5578	13.997	0.8375	9.953
0.1354	17.617	0.3649	16.180	0.6120	13.266	0.8858	9.212
0.1765	17.473	0.4113	15.713	0.6661	12.508	0.9439	8.321
0.2255	17.230	0.4587	15.196	0.7151	11.806		
$xCHCl_2CHCl_2 + (1 - x)(1,4\text{-}C_4H_8O_2)$							
0.0128	2.258	0.1800	3.020	0.4522	4.196	0.7332	5.603
0.0321	2.352	0.2215	3.200	0.5094	4.455	0.7591	5.752
0.0726	2.539	0.2589	3.360	0.5507	4.650	0.8151	6.099
0.1083	2.701	0.2882	3.486	0.6086	4.929	0.8727	6.487
0.1347	2.820	0.3558	3.776	0.6769	5.286	0.9314	6.912
0.1501	2.888	0.3994	3.966	0.7044	5.438	0.9715	7.224

7.0, and another cell MFL 2/S, no. 2084, for mixtures having ϵ_r > 7.0, as described earlier (Nath and Singh, 1987). The imprecision in ϵ_r is ~0.002 units for mixtures having ϵ_r < 7.0, and is ~0.005 units for mixtures having ϵ_r > 7.0.

(iii) The refractive indexes (sodium D line), n_D, were measured at (303.15 ± 0.01) K, with an accuracy of ±0.0002, using a thermostated Abbe refractometer.

Results and Discussion

The values of n_D^* and ϵ_r^* of the pure liquids C_5H_5N, $C_6H_5OCH_3$, $CH_3COC_2H_5$, 1,4-$C_4H_8O_2$, and $CHCl_2CHCl_2$ are given in Table 1. Values of V_m^E for $xCHCl_2CHCl_2 + (1 - x)C_5H_5N$, $xCHCl_2CHCl_2 + (1 - x)C_6H_5OCH_3$, $xCHCl_2CHCl_2 + (1 - x)CH_3COC_2H_5$, and $xCHCl_2CHCl_2 + (1 - x)(1,4\text{-}C_4H_8O_2)$ are given in Table 2, whereas those of the relative permittivities ϵ_r for these mixtures are given in Table 3, and values of n_D are given in Table 4. The values of $V_m^E/cm^3 \cdot mol^{-1}$ for the various mixtures of $CHCl_2CHCl_2$ have been plotted against the mole fraction of $CHCl_2CHCl_2$, x, in Figure 1, whereas the values of refractive indexes n_D of these mixtures have been plotted against x in Figure 2. The values of x have the uncertainty of ±0.0001. The values of the changes of refractive index on mixing, Δn_D,

Table 4. Refractive Indexes, n_D, for $CHCl_2CHCl_2$ + C_5H_5N, $C_6H_5OCH_3$, $CH_3COC_2H_5$, and 1,4-$C_4H_8O_2$ at T = 303.15 K

x	n_D	x	n_D	x	n_D	x	n_D
$xCHCl_2CHCl_2 + (1 - x)C_5H_5N$							
0.0491	1.5024	0.2899	1.4988	0.5292	1.4952	0.8111	1.4914
0.1294	1.5010	0.3318	1.4982	0.5843	1.4945	0.8518	1.4905
0.1619	1.5005	0.3774	1.4975	0.6459	1.4935	0.8633	1.4904
0.2035	1.4998	0.4384	1.4966	0.7038	1.4928	0.9328	1.4895
0.2579	1.4992	0.4656	1.4962	0.7560	1.4922		
$xCHCl_2CHCl_2 + (1 - x)C_6H_5OCH_3$							
0.0802	1.5102	0.3261	1.5045	0.5294	1.4996	0.8556	1.4920
0.1077	1.5095	0.3439	1.5038	0.5982	1.4982	0.9468	1.4900
0.1702	1.5080	0.3846	1.5030	0.6915	1.4960		
0.2097	1.5072	0.4456	1.5015	0.7766	1.4940		
0.2579	1.5060	0.4948	1.5005	0.8204	1.4930		
$xCHCl_2CHCl_2 + (1 - x)CH_3COC_2H_5$							
0.1037	1.3895	0.3634	1.4240	0.5633	1.4468	0.7559	1.4665
0.2221	1.4052	0.3918	1.4272	0.6059	1.4510	0.8102	1.4715
0.2683	1.4120	0.4543	1.4345	0.6618	1.4570	0.8752	1.4770
0.3070	1.4168	0.4990	1.4398	0.7089	1.4620	0.9275	1.4815
$xCHCl_2CHCl_2 + (1 - x)(1,4\text{-}C_4H_8O_2)$							
0.0289	1.4210	0.1710	1.4335	0.4737	1.4576	0.8210	1.4796
0.0424	1.4220	0.2531	1.4408	0.5931	1.4655	0.8291	1.4800
0.0528	1.4232	0.3128	1.4454	0.6040	1.4660	0.8991	1.4845
0.0917	1.4265	0.3205	1.4460	0.6768	1.4710	0.9048	1.4846
0.1198	1.4290	0.4000	1.4520	0.6980	1.4722		

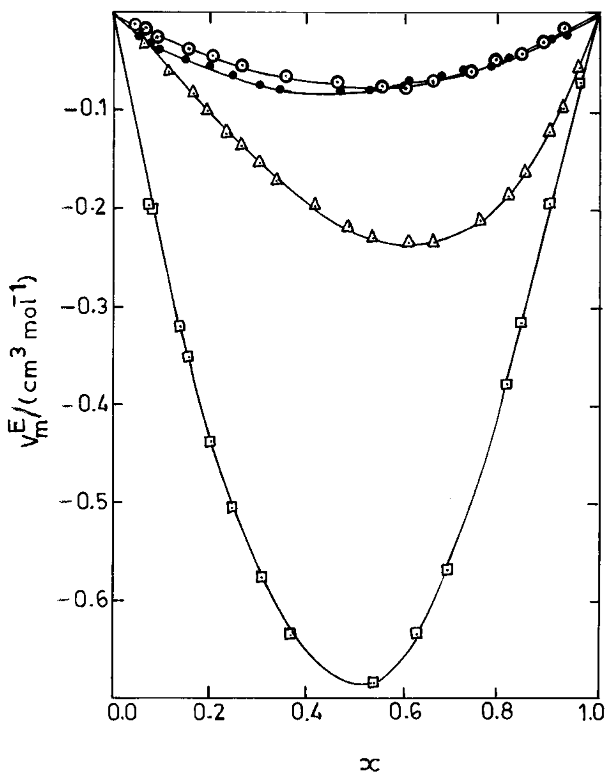

Figure 1. V_m^E plotted versus x for the following systems at 303.15 K: (⊙) $xCHCl_2CHCl_2 + (1 - x)C_5H_5N$; (●) $xCHCl_2CHCl_2 + (1 - x)C_6H_5OCH_3$; (⊡) $xCHCl_2CHCl_2 + (1 - x)CH_3COC_2H_5$; (△) $xCHCl_2CHCl_2 + (1 - x)(1,4\text{-}C_4H_8O_2)$. The smoothed curves are based on the parameters A_1, A_2, A_3, and A_4 given in Table 5.

+ $(1 - x)CH_3COC_2H_5$, and $xCHCl_2CHCl_2 + (1 - x)(1,4\text{-}C_4H_8O_2)$ are given in Table 2, whereas those of the relative permittivities ϵ_r for these mixtures are given in Table 3, and values of n_D are given in Table 4. The values of $V_m^E/cm^3 \cdot mol^{-1}$ for the various mixtures of $CHCl_2CHCl_2$ have been plotted against the mole fraction of $CHCl_2CHCl_2$, x, in Figure 1, whereas the values of refractive indexes n_D of these mixtures have been plotted against x in Figure 2. The values of x have the uncertainty of ±0.0001. The values of the changes of refractive index on mixing, Δn_D,

Table 5. Values of the Parameters A_i of eq 3 and the Standard Deviations, δ, for the Various Mixtures

Y	A_1	A_2	A_3	A_4	δ
		xCHCl$_2$CHCl$_2$ + (1 − x)C$_5$H$_5$N			
V_m^E/cm^3·mol^{-1}	−0.3088	−0.0484	0.0351	0.0999	0.0016
Δn_D	−0.001 503	0.000 047	−0.007 823	0.011 166	0.000 17
$\Delta\epsilon_r$	8.8607	0.6924	0.1696	−0.0693	0.0047
		xCHCl$_2$CHCl$_2$ + (1 − x)C$_6$H$_5$OCH$_3$			
V_m^E/cm^3·mol^{-1}	−0.3154	0.0310	−0.0587	0.0891	0.0031
Δn_D	0.000 397	0.000 476	0.004 481	−0.000 585	0.000 12
$\Delta\epsilon_r$	−0.8070	−0.7943	−0.0460	0.0669	0.0028
		xCHCl$_2$CHCl$_2$ + (1 − x)CH$_3$COC$_2$H$_5$			
V_m^E/cm^3·mol^{-1}	−2.7611	−0.0101	0.4705	0.6190	0.0032
Δn_D	0.034 741	−0.000 647	−0.005 584	−0.014 552	0.000 30
$\Delta\epsilon_r$	8.6164	−3.7388	0.1685	−0.0205	0.0046
		xCHCl$_2$CHCl$_2$ + (1 − x)(1,4-C$_4$H$_8$O$_2$)			
V_m^E/cm^3·mol^{-1}	−0.9040	−0.4651	−0.0336	0.0419	0.0035
Δn_D	0.022 737	−0.001 136	0.010 382	0.005 723	0.000 27
$\Delta\epsilon_r$	−1.6547	−1.3992	−0.2055	0.1136	0.0031

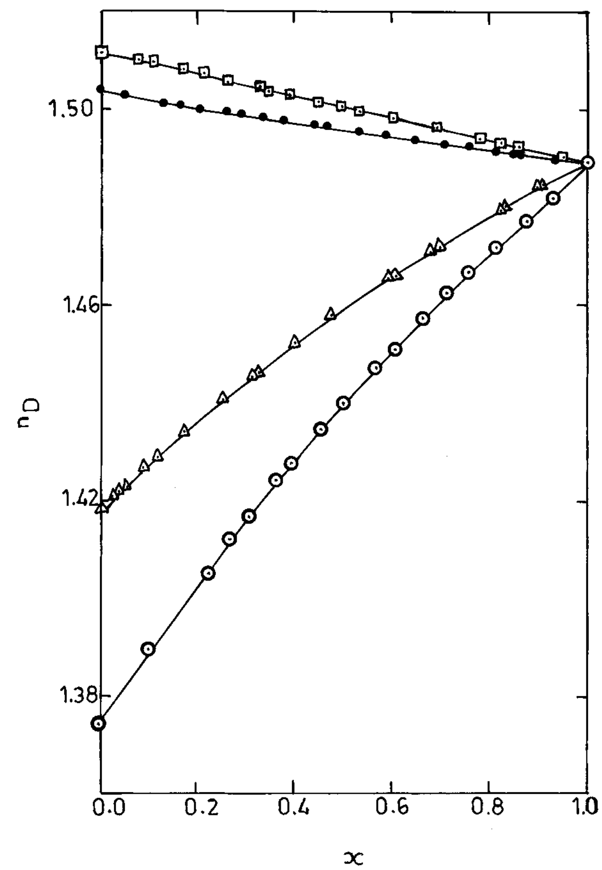

Figure 2. n_D plotted versus x for the following systems at 303.15 ●) xCHCl$_2$CHCl$_2$ + (1 − x)C$_5$H$_5$N; (⊡) xCHCl$_2$CHCl$_2$ + (1 − H$_5$OCH$_3$; (⊙) xCHCl$_2$CHCl$_2$ + (1 − x)CH$_3$COC$_2$H$_5$; (△) xCHCl$_2$-CHCl$_2$ + (1 − x)(1,4-C$_4$H$_8$O$_2$). The smoothed curves are based on the values of n_D obtained from eq 1, using parameters A_1, A_2, A_3, and A_4 given in Table 5.

for the various mixtures of CHCl$_2$CHCl$_2$ were calculated from the refractive indexes n_D of the mixtures, using the relation

$$\Delta n_D = n_D - \sum_i x_i n^*_{D,i} \qquad (1)$$

where $n^*_{D,i}$ refers to the refractive index of the pure component i and x_i is the mole fraction of the component i in the mixture. Iglesias et al. (1984) have also represented the refractive index data for mixtures, by the changes of refractive index on mixing, Δn_D. Also the values of the changes of relative permittivity on mixing $\Delta\epsilon_r$ for the various mixtures of CHCl$_2$CHCl$_2$ were calculated from the relative permittivities ϵ_r of the mixtures, using the relation

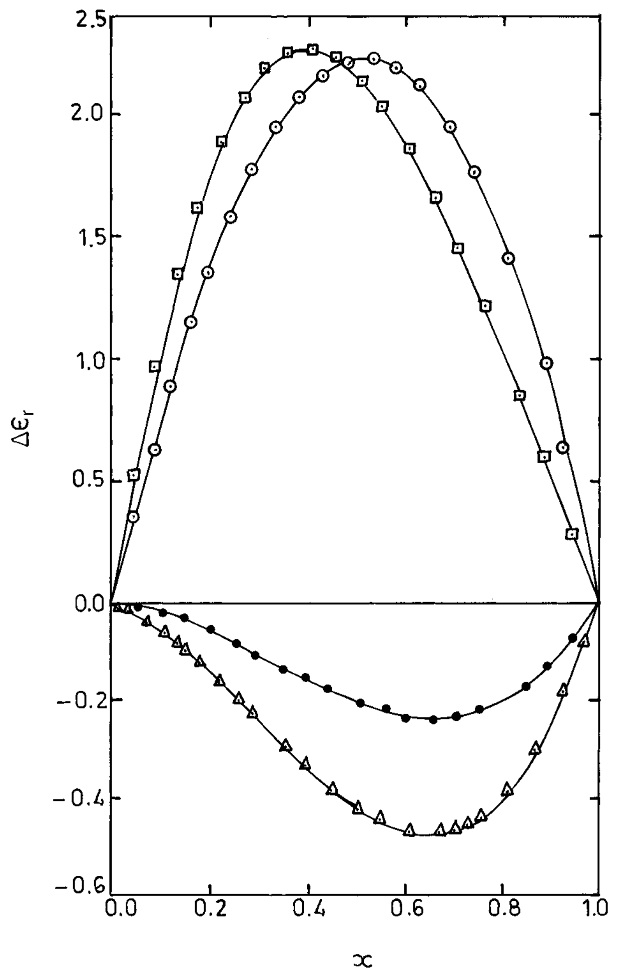

Figure 3. $\Delta\epsilon_r$ plotted versus x for the various systems at 303.15 K: (⊙) xCHCl$_2$CHCl$_2$ + (1 − x)C$_5$H$_5$N; (●) xCHCl$_2$CHCl$_2$ + (1 − x)C$_6$H$_5$OCH$_3$; (⊡) xCHCl$_2$CHCl$_2$ + (1 − x)CH$_3$COC$_2$H$_5$; (△) xCHCl$_2$-CHCl$_2$ + (1 − x)(1,4-C$_4$H$_8$O$_2$). The smoothed curves are based on the parameters A_1, A_2, A_3, and A_4 given in Table 5.

$$\Delta\epsilon_r = \epsilon_r - \sum_i x_i \epsilon^*_{r,i} \qquad (2)$$

where $\epsilon^*_{r,i}$ refers to the relative permittivity of the pure component i. $\Delta\epsilon_r$ has been plotted against x in Figure 3. The values of V_m^E, Δn_D, and $\Delta\epsilon_r$ for xCHCl$_2$CHCl$_2$ + (1 − x)C$_5$H$_5$N, xCHCl$_2$CHCl$_2$ + (1 − x)C$_6$H$_5$OCH$_3$, xCHCl$_2$CHCl$_2$ + (1 − x)CH$_3$COC$_2$H$_5$, and xCHCl$_2$CHCl$_2$ + (1 − x)(1,4-C$_4$H$_8$O$_2$) were fitted by the method of least squares to the

Redlich−Kister (1949) type equation:

$$Y = x(1 - x)\sum_{i=1}^{n} A_i(2x - 1)^{i-1} \qquad (3)$$

where Y is V_m^E/cm^3·mol^{-1} or Δn_D or $\Delta\epsilon_r$. The values of the parameters A_i of eq 3 and the standard deviations δ are given in Table 5.

The data show that V_m^E is negative throughout the entire range of x for $x$$CHCl_2CHCl_2$ + (1 − x)C_5H_5N, $x$$CHCl_2$-$CHCl_2$ + (1 − x)$C_6H_5OCH_3$, $x$$CHCl_2CHCl_2$ + (1 − x)CH_3-COC_2H_5, and $x$$CHCl_2CHCl_2$ + (1 − x)(1,4-$C_4H_8O_2$). At x = 0.5, V_m^E for the various mixtures of $CHCl_2CHCl_2$ has the sequence

$$C_5H_5N > C_6H_5OCH_3 > 1,4\text{-}C_4H_8O_2 > CH_3COC_2H_5$$

The negative values of V_m^E for the present mixtures show that $CHCl_2CHCl_2$ forms intermolecular complexes with C_5H_5N, $C_6H_5OCH_3$, $CH_3COC_2H_5$, and 1,4-$C_4H_8O_2$ in the liquid state. The relative permittivity data are found to exhibit positive deviations (Rivail and Thiebaut, 1974) from a mole fraction mixture law for chloroform + pyridine where a strong intermolecular complex is formed on account of the hydrogen-bond interaction between the components. The data show (see Figure 3) that $\Delta\epsilon_r$ is highly positive for $x$$CHCl_2CHCl_2$ + (1 − x)C_5H_5N, and $CHCl_2CHCl_2$ + (1 − x)$CH_3COC_2H_5$, and slightly negative $x$$CHCl_2CHCl_2$ + (1 − x)$C_6H_5OCH_3$, and $x$$CHCl_2CHCl_2$ (1 − x)(1,4-$C_4H_8O_2$). The positive values of $\Delta\epsilon_r$ for a system may be interpreted as being due to the specific interaction between the components. The present values $\Delta\epsilon_r$, thus, indicate that $CHCl_2CHCl_2$ forms strong intermolecular complexes with C_5H_5N and $CH_3COC_2H_5$, which is in accordance with the V_m^E data. The values of are highly negative (Chadha and Tripathi, 1995) for $CHCl_2CHCl_2$ + (1 − x)(1,4-$C_4H_8O_2$), which gives evidence in favor of the formation of strong intermolecular complexes between $CHCl_2CHCl_2$ and 1,4-$C_4H_8O_2$ in the liquid state. The negative value of $\Delta\epsilon_r$ for $x$$CHCl_2CHCl_2$ + (1 − x)C_6H_5-OCH_3 and $x$$CHCl_2CHCl_2$ + (1 − x)(1,4-$C_4H_8O_2$) may be attributed to the predominance of contributions to $\Delta\epsilon_r$ from nonspecific interactions.

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Received for review February 12, 1996. Accepted April 28, 1996.[⊗] The authors gratefully acknowledge the financial support received from the Council of Scientific and Industrial Research, New Delhi, India.

JE960054H

[⊗] Abstract published in *Advance ACS Abstracts*, June 1, 1996.