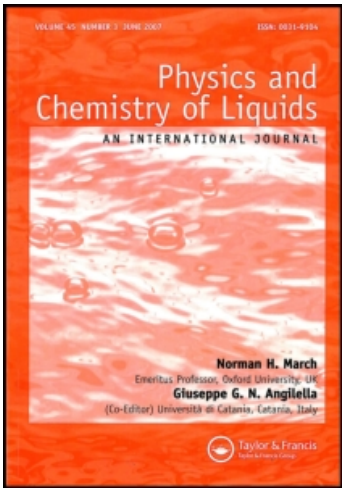

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Densities, speeds of sound and refractive indices for binary and ternary mixtures of {diethylcarbonate (1) + *p*-chloroacetophenone (2) + 1-hexanol (3)} at 303.15 K for the liquid region and at ambient pressure

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Densities, speeds of sound and refractive indices for binary and ternary mixtures of {diethylcarbonate (1) + *p*-chloroacetophenone (2) + 1-hexanol (3)} at 303.15 K for the liquid region and at ambient pressure

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Densities (ρ), speeds of sound (u) and refractive indices (n_D), of the ternary mixture (diethylcarbonate + *p*-chloroacetophenone + 1-hexanol) and the involved binary mixtures (diethylcarbonate + *p*-chloroacetophenone, diethylcarbonate + 1-hexanol, and *p*-chloroacetophenone + 1-hexanol) have been measured over the whole composition range at 303.15 K for the liquid region and at ambient pressure. The data obtained are used to calculate isentropic compressibilities k_s, isentropic compressibility deviations Δk_s and refractive index deviations Δn_D, of the binary and ternary mixtures. The data of isentropic compressibility deviations and refractive index deviations of the binary systems were fitted to the Redlich–Kister equation while the best correlation method for the ternary system was found using the Cibulka equation. The experimental data of the constitute binaries and ternaries are analysed to discuss the nature and strength of intermolecular interactions in these mixtures.

Keywords: diethylcarbonate; chloroacacetophenone; 1-hexanol; speed of sound; isentropic compressibility; refractive index

1. Introduction

This article is a continuation of our earlier work related to the study of thermodynamic and acoustic properties of binary mixtures [1–7]. In recent years, measurements of acoustic and refractive indices have been adequately employed in understanding the nature of molecular systems and physico-chemical behaviour in liquid mixtures. The non-rectilinear behaviour of above mentioned properties of liquid mixtures with changing mole or volume fractions is attributed to the difference in size of the molecules and strength of interactions. Here, we have reported densities (ρ), speeds of sound (u), refractive indices (n_D) and isentropic compressibilities (k_s), of the ternary mixture (diethylcarbonate + *p*-chloroacetophenone + 1-hexanol) and the involved binary mixtures (diethylcarbonate + *p*-chloroacetophenone), (diethylcarbonate + 1-hexanol) [7], and (*p*-chloroacetophenone + 1-hexanol) at 303.15 K and over the whole composition range

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for the liquid region and ambient pressure. The derived properties (deviations in isentropic compressibility $\Delta\kappa_s$, and deviations in the refractive index Δn_D,) in combination with other mixing properties, provide valuable information for qualitatively analysing the molecular interactions between molecules.

The deviation quantities of binary mixtures have been fitted to the Redlich–Kister [8] equation to determine the coefficients.

2. Experimental section

2.1. *Materials*

Diethylcarbonate (mass fraction $\geq 99.5\%$), *p*-chloroacetophenone (mass fraction > 98%), and 1-hexanol (mass fraction $\geq 99\%$) were purchased from Fluka. Compounds were purified by distillation using a 1 m fractionation column and stored in brown glass bottles. Purity of each compound was ascertained by the constancy of the density and also from refractive index. The resultant values were in good agreement with values found in the literature [9–13] reported in Table 1.

2.2. *Apparatus and procedure*

The density of the compounds and their binary mixtures were measured with Anton Paar DMA 4500 oscillating u-tube densitometer with a certified precision, of better than $\pm 1 \times 10^{-5}\,\mathrm{g\,cm^{-3}}$ operated in the static mode and in the cell was regulated to ± 0.01 K with solid-state thermostat. The apparatus was calibrated once a day with dry air and double-distilled, freshly degassed water. Airtight stoppered bottles were used for the preparation of the mixtures. The mass of the dry bottle was first determined. The less volatile component of the mixture was introduced in the bottle, and the total mass was recorded. Subsequently, the other component was introduced, and the mass of bottle along with the two components was determined. Each mixture was immediately used after it was well-mixed by shaking. All the weightings were performed on an electronic balance (AB 204-N Mettler) accurate to 0.1 mg. Speeds of sound at 303.15 K at a frequency of 1 MHz were determined using a quartz crystal interferometer (Mittal Enterprises, New Delhi, India). The uncertainty in the speed of sound measurement was $\pm 1\,\mathrm{m\,s^{-1}}$. The temperature in the cell was regulated to ± 0.01 K. Refractive indices were measured using a digital Abbe-type refractometer. The uncertainty of refractive index measurement was estimated to be less than ± 0.0001 units.

Table 1. Source, purity grades, densities (ρ) and refractive indices (n_D) of the pure components at 293.15 K.

			ρ ($\mathrm{g\,cm^{-3}}$)		n_D	
Component	Source	Purity (mass %)	Expt.	Lit.	Expt.	Lit.
Diethylcarbonate	Fluka	99.5	0.97465	0.97468[a]	1.3845	1.3845[d]
p-Chloroacetophenone	Fluka	98	1.19209	1.192[b]	1.5552	1.5550[b]
1-Hexanol	Fluka	99	0.81884	0.81880[c]	1.4178	1.4181[e]

Notes: [a]Reference [9]; [b]Reference [10]; [c]Reference [11]; [d]Reference [12]; [e]Reference [13].

3. Results and discussion

3.1. *Speeds of sound and isentropic compressibility deviations*

Isentropic compressibility was calculated using Laplace–Newton equation

$$\kappa_S = \frac{1}{\rho u^2}. \tag{1}$$

Deviation in isentropic compressibilities $\Delta\kappa_s$, n-component were calculated using the following relation:

$$\Delta\kappa_s = \kappa_s - \sum_i \left(\kappa^*_{s,i}\right)\phi_i, \tag{2}$$

where $\kappa^*_{s,i}$, k_s, and ϕ_i are the isentropic compressibility of pure components, mixtures and volume fraction average, respectively, and

$$\phi_i = \frac{x_i V_i}{\sum_{i=1}^{N} x_i V_i}, \tag{3}$$

where V_i is the molar volume of the pure component i.

The densities, speeds of sound, isentropic compressibilties and isentropic compressibility deviations for (diethylcarbonate + *p*-chloroacetophenone), (diethylcarbonate + 1-hexanol) and (*p*-chloroacetophenone + 1-hexanol) are recorded in Table 2 and variation of isentropic compressibility deviations with mole fraction were graphically represented in Figure 1.

Each set of results were fitted using Redlich−Kister expression [8] for the binary mixtures as follows:

$$Y = x_1(1 - x_1)\sum_{k=0}^{N} A_k(2x_1 - 1)^k, \tag{4}$$

where $Y \equiv (\Delta k$ or $\Delta n_D)$ and x_1 is the mole fraction of the first component and A_k are the fitting parameters obtained by the un-weighted least-squares method. In each case, the optimum number of coefficients was ascertained from an examination of the variation of standard deviation $\sigma(Y)$ with

$$\sigma(Y) = \left[\frac{\sum\left(Y^E_{m\,\mathrm{exp.},i} - Y^E_{m\,\mathrm{cal.},i}\right)^2}{n - p}\right]^{1/2}, \tag{5}$$

where n and p are the number of experimental points and number of parameters retained in the respective equation. The adjustable parameters A_k, and standard deviations $\sigma(\Delta k_s)$, are given in Table 3.

The isentropic compressibility deviations Δk_s, for (diethylcarbonate + 1-hexanol) are positive over the whole composition range. The positive Δk_s, values are due to the depolymerisation effects of diethylcarbonate on 1-hexanol and the dispersion interactions between unlike molecules.

The isentropic compressibility deviations Δk_s, for the systems (*p*-chloroacetophenone + 1-hexanol) and (diethylcarbonate + *p*-chloroacetophenone) are negative over the entire range of mole fraction. The negative Δk_s, for these binary mixtures

Table 2. Densities, ρ in $g\,cm^{-3}$, speeds of sound, u in ms^{-1}, refractive indices, n_D, isentropic compressibilities, κ_s in TPa^{-1}, isentropic compressibility deviations, $\Delta\kappa_s$ in TPa^{-1}, and deviation in refractive index deviations, Δn_D, for binary mixtures at 303.15 K.

x	ρ	u	n_D	κ_s	$\Delta\kappa_s$	Δn_D
x diethylcarbonate + $(1-x)$*p*-chloroacetophenone						
0.0511	1.17269	1390	1.5426	441	−4	0.0004
0.1131	1.16055	1376	1.5331	455	−8	0.0009
0.1611	1.15106	1365	1.5257	466	−11	0.0013
0.2221	1.13879	1351	1.5161	481	−14	0.0017
0.2808	1.12690	1337	1.5068	496	−17	0.0021
0.3435	1.11401	1324	1.4967	512	−19	0.0025
0.4256	1.09666	1305	1.4832	535	−21	0.0028
0.5032	1.07988	1288	1.4702	558	−22	0.0029
0.6089	1.05635	1265	1.4519	592	−21	0.0028
0.6807	1.04006	1250	1.4393	615	−20	0.0026
0.7618	1.02131	1234	1.4247	643	−18	0.0022
0.8127	1.00930	1224	1.4152	661	−16	0.0018
0.8641	0.99688	1214	1.4057	681	−13	0.0014
0.9197	0.98331	1203	1.3953	703	−9	0.0010
0.9678	0.97155	1193	1.3861	723	−4	0.0005
x diethylcarbonate + $(1-x)$1-hexanol						
0.0502	0.81855	1267	1.4131	761	7	0.0008
0.1123	0.82716	1256	1.4115	766	13	0.0013
0.1499	0.83248	1249	1.4106	770	18	0.0016
0.2103	0.84104	1240	1.4088	773	22	0.0018
0.2508	0.84679	1234	1.4074	776	26	0.0018
0.3121	0.85569	1225	1.4051	779	30	0.0016
0.3576	0.86223	1217	1.4033	783	34	0.0013
0.4113	0.87021	1210	1.4011	785	37	0.0009
0.4632	0.87788	1204	1.3989	786	39	0.0005
0.5111	0.88513	1200	1.3969	785	39	0.0002
0.5520	0.89135	1197	1.3952	783	38	−0.0001
0.6008	0.89887	1194	1.3931	780	35	−0.0006
0.6498	0.90642	1193	1.3911	775	31	−0.0009
0.7108	0.91593	1192	1.3887	768	25	−0.0012
0.7554	0.92308	1191	1.3870	764	22	−0.0013
0.8112	0.93203	1190	1.3851	758	17	−0.0013
0.8501	0.93829	1189	1.3838	754	14	−0.0012
0.9002	0.94657	1188	1.3823	749	9	−0.0010
0.9555	0.95577	1187	1.3809	743	4	−0.0005
x *p*-chloroacetophenone + $(1-x)$1-hexanol						
0.0521	0.83186	1282	1.4216	731	−6	0.0002
0.1009	0.85072	1288	1.4287	709	−11	0.0005
0.1668	0.87605	1296	1.4383	680	−18	0.0008
0.2325	0.90116	1305	1.4477	652	−24	0.0011
0.3002	0.92687	1314	1.4574	625	−29	0.0014
0.4028	0.96545	1326	1.4719	589	−32	0.0017
0.5001	1.00167	1338	1.4854	558	−31	0.0019
0.5708	1.02779	1346	1.4951	537	−29	0.0020
0.6410	1.05356	1354	1.5046	518	−26	0.0020
0.6998	1.07502	1361	1.5124	502	−23	0.0019
0.7655	1.09882	1370	1.5209	485	−19	0.0016
0.8376	1.12480	1380	1.5301	467	−14	0.0012
0.9007	1.14740	1389	1.5381	452	−9	0.0008
0.9513	1.16545	1395	1.5444	441	−4	0.0004

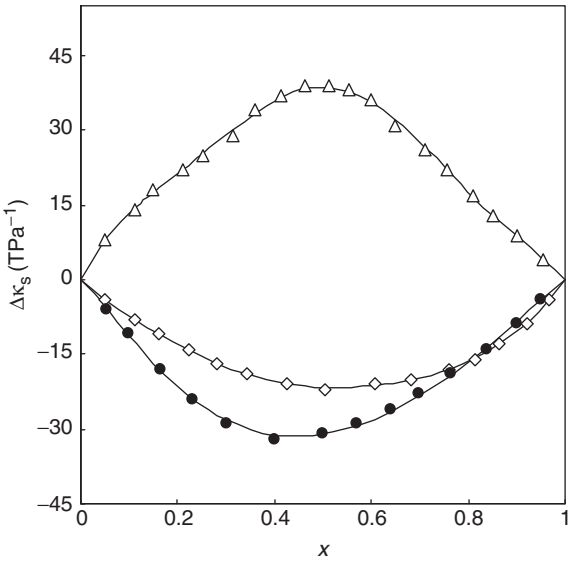

Figure 1. Deviation in the isentropic compressibility of the binary mixtures *vs.* mole fraction x at $T = 303.15$ K. Notes: Experimental results: (◇) x diethylcarbonate + $(1-x)$*p*-chloroacetophenone, (Δ) x diethylcarbonate + $(1-x)$ 1-hexanol, (●) x *p*-chloroacetophenone + $(1-x)$1-hexanol. Solid curves were calculated from the Redlich–Kister equation.

Table 3. Values of adjustable coefficients A_k, in Equation (4) and standard deviation σ, for isentropic compressibility deviations, $\Delta\kappa_s$, and refractive index deviations, Δn_D, for the binary mixtures at 303.15 K.

	A_0	A_1	A_2	A_3	A_4	σ
x diethylcarbonate + $(1-x)$ *p*-chloroacetophenone						
$\Delta\kappa_s$ (TPa^{-1})	−86.75	−11.47	−11.30	−16.44	−11.81	0.24
Δn_D	0.0118	0.0004	−0.0053	0.0039	0.0063	0.00006
x diethylcarbonate + $(1-x)$1-hexanol						
$\Delta\kappa_s$ (TPa^{-1})	154.79	−1.95	−142.64	−46.33	141.50	0.86
Δn_D	0.0009	−0.0165	0.0015	0.0011		0.00005
x *p*-chloroacetophenone + $(1-x)$1-hexanol						
$\Delta\kappa_s$ (TPa^{-1})	−124.43	33.00	7.20	−22.41	22.53	0.47
Δn_D	0.0077	0.0027	0.0006	−0.0005	−0.0027	0.00004

are due to the interstitial accommodation of molecules in a network of bonded 1-hexanol molecules leading to more dense packing of unlike molecules.

Also, the corresponding $\Delta k_{s,123}$, values of the ternary mixture (diethylcarbonate + *p*-chloroacetophenone + 1-hexanol) are shown in Table 4. The isentropic compressibility deviations $\Delta k_{s,123}$, and refractive index deviations Δn_{123}, were correlated with ternary composition using the equation proposed by Cibulka equation [14].

$$Y_{123} = Y_{bin} + x_1x_2(1 - x_1 - x_2)(B_0 + B_1x_1 + B_2x_2), \tag{6}$$

where $Y \equiv (\Delta k_{s,123}$ or $\Delta n_{123})$ and $Y_{bin} = Y_{12} + Y_{13} + Y_{23}$ are the so-called binary contribution.

The coefficients B_p, of Equation (6) and standard deviations $\sigma(\Delta k_{s,123})$ of Equation (5) obtained by the least-squares method are gathered in Table 5.

The ternary isentropic compressibility deviations $\Delta k_{s,123}$, are negative and for few compositions are positive over the composition range. In Figure 2 the isoline curves for $\Delta k_{s,123}$, for the ternary mixture (diethylcarbonate + *p*-chloroacetophenone + 1-hexanol) are shown.

Table 4. Densities, ρ in $g\,cm^{-3}$, speeds of sound, u in ms^{-1}, refractive indices, n_D, isentropic compressibilities, κ_s in TPa^{-1}, isentropic compressibility deviations, $\Delta\kappa_{s,123}$ in TPa^{-1}, and refractive index deviations, $\Delta n_{D,123}$, for the ternary system diethylcarbonate (1) + *p*-chloroacetophenone (2) + 1-hexanol (3) at 303.15 K.

x_1	x_2	ρ	u	n_D	κ_s	$\Delta\kappa_{s,123}$	$\Delta n_{D,123}$
0.0507	0.0514	0.83889	1281	1.4196	727	−9	0.00001
0.0505	0.2485	0.91451	1297	1.4481	650	−21	0.0009
0.0501	0.4409	0.98733	1317	1.4750	584	−23	0.0012
0.0552	0.6475	1.06442	1353	1.5032	513	−27	0.0014
0.0498	0.8444	1.13521	1380	1.5297	463	−15	0.0013
0.0502	0.9000	1.15527	1390	1.5372	448	−12	0.0014
0.0513	0.6972	1.08221	1351	1.5101	506	−19	0.0015
0.0509	0.5024	1.01030	1328	1.4836	561	−26	0.0014
0.0503	0.3000	0.93430	1306	1.4555	628	−26	0.0011
0.0501	0.1002	0.85775	1287	1.4266	704	−16	0.0001
0.0998	0.0507	0.84550	1269	1.4176	735	−1	−0.0003
0.1005	0.2494	0.92249	1289	1.4464	653	−17	0.0006
0.0993	0.4489	0.99771	1322	1.4744	574	−30	0.0010
0.0991	0.6463	1.07103	1346	1.5018	515	−25	0.0015
0.0998	0.8496	1.14517	1374	1.5287	463	−12	0.0011
0.2018	0.0526	0.86077	1254	1.4143	739	6	−0.0004
0.2003	0.2524	0.93828	1299	1.4441	632	−35	0.0012
0.2010	0.4610	1.01785	1307	1.4732	575	−23	0.0013
0.1998	0.6504	1.08833	1335	1.5003	516	−21	0.0025
0.2038	0.7490	1.12225	1351	1.5115	488	−17	0.0005
0.1991	0.5524	1.05196	1316	1.4861	549	−19	0.0016
0.2002	0.3506	0.97580	1285	1.4580	621	−13	0.0014
0.2014	0.1500	0.89890	1262	1.4290	698	−2	0.0005
0.3004	0.0506	0.87433	1233	1.4110	752	20	−0.0001
0.2874	0.2777	0.96117	1270	1.4442	645	−11	0.0005
0.2984	0.4481	1.02818	1290	1.4686	585	−16	0.0016
0.3002	0.6479	1.10367	1322	1.4963	519	−16	0.0019
0.3978	0.0504	0.88874	1225	1.4069	750	19	−0.0009
0.3978	0.2505	0.96755	1258	1.4372	653	−11	0.0010
0.4019	0.4484	1.04462	1287	1.4650	578	−20	0.0012
0.3994	0.5497	1.08287	1300	1.4806	547	−18	0.0027
0.4006	0.3505	1.00683	1267	1.4513	619	−11	0.0011
0.3998	0.1509	0.92877	1237	1.4219	704	7	−0.0002
0.4993	0.0522	0.90459	1208	1.4036	757	29	−0.0011
0.5003	0.2487	0.98288	1241	1.4333	661	−1	0.0007
0.4991	0.4490	1.06059	1277	1.4628	578	−18	0.0020
0.5999	0.0505	0.91942	1202	1.4000	752	25	−0.0010
0.5919	0.2458	0.99632	1229	1.4298	665	3	0.0006
0.6006	0.3496	1.03863	1252	1.4455	614	−13	0.0019
0.7007	0.0501	0.93513	1188	1.3968	758	32	−0.0007
0.7015	0.2482	1.01514	1226	1.4271	656	−3	0.0012
0.8494	0.0504	0.95935	1170	1.3921	762	39	−0.0003
0.8497	0.1001	0.97977	1183	1.3999	730	23	0.0003
0.8999	0.0506	0.96773	1172	1.3908	752	30	0.0001

3.2. *Refractive index deviations*

The deviation in the refractive index Δn_D, was calculated from the volume fraction average as suggested by Brocos [15] and is given by

$$\Delta n_D = n_D - \sum_{i=1}^{N} \phi_i n_{D_i}, \tag{7}$$

where n_D, n_{D_i} and ϕ_i, are the refractive indices of the mixture, the refractive index of pure component i, and the volume fraction of pure component i, respectively. V and V_i are the molar volume of the mixture and the molar volume of pure component i, respectively. The refractive indices for (diethylcarbonate + *p*-chloroacetophenone), (diethylcarbonate + 1-hexanol) [7] and (*p*-chloroacetophenone + 1-hexanol) are given in Table 2 and refractive index deviations with volume fraction were graphically represented in Figure 3.

Each set of results were fitted using Redlich–Kister expression [8] for the binary mixtures and adjustable parameters A_k, and standard deviations $\sigma(\Delta n_D)$, are also given in Table 3.

Table 5. Ternary coefficients of Cibulka equation and standard deviations, σ, for isentropic compressibility deviations, $\Delta\kappa_s$, and refractive index deviations, Δn_D, at 303.15 K.

	B_0	B_1	B_2	σ
$\Delta\kappa_s$ (TPa^{-1})	−2781.6	8771.6	1259.1	23.1
Δn_D	−0.1812	−0.1243	−0.2625	0.003

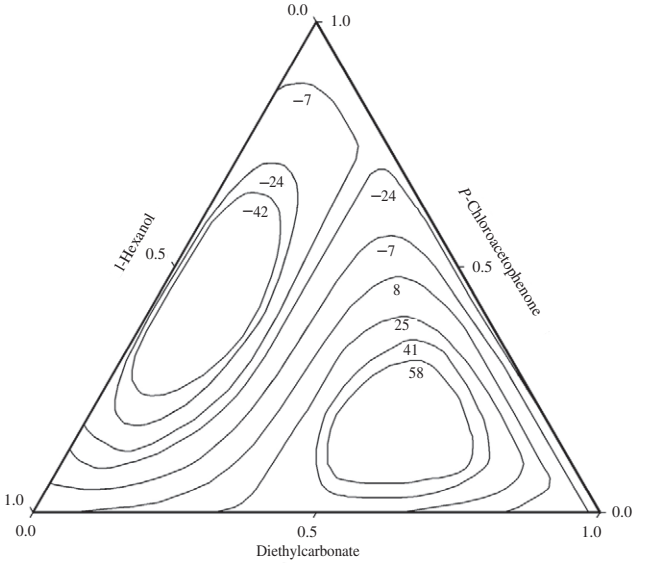

Figure 2. Isolines for deviation in the isontropic compressibility from Cibulka's Equation (6) for the ternary system diethylcarbonate (1) + *p*-chloroacetophenone (2) + 1-hexanol (3) at 303.15 K.

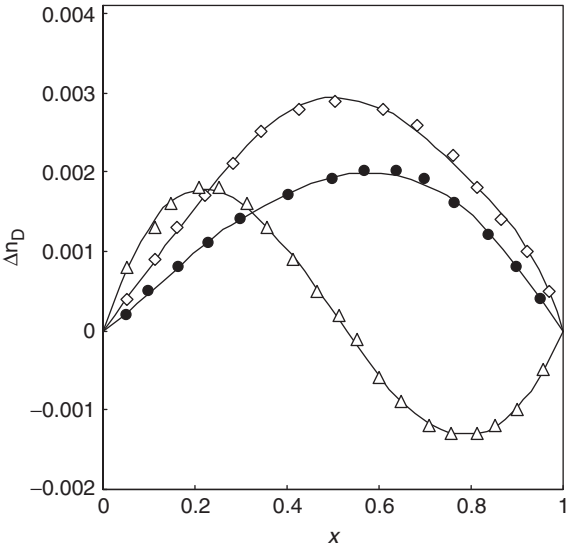

Figure 3. Deviation in refractive indices of the binary mixtures *vs*. mole fraction x at $T = 303.15$ K.
Notes: Experimental results: (◇) x diethylcarbonate + $(1-x)$*p*-chloroacetophenone, (Δ) x diethylcarbonate + $(1-x)$ 1-hexanol, (●) x *p*-chloroacetophenone + $(1-x)$ 1-hexanol. Solid curves were calculated from the Redlich–Kister equation.

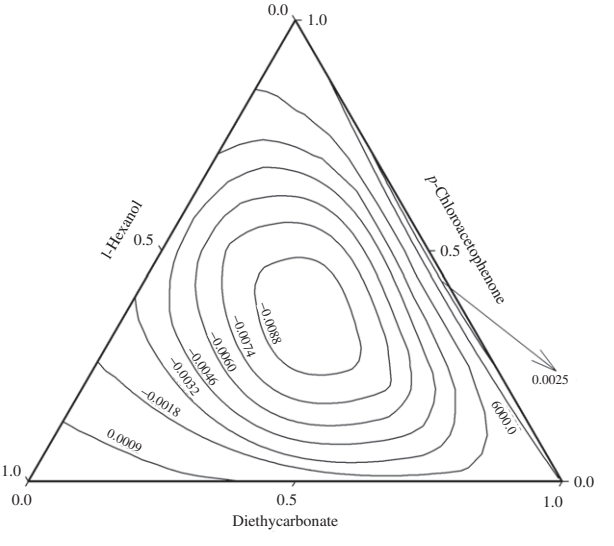

Figure 4. Isolines for deviation in refractive indices from Cibulka's Equation (6) for the ternary system diethylcarbonate (1) + *p*-chloroacetophenone (2) + 1-hexanol (3) at 303.15 K.

For the whole composition range, the Δn_D, values are positive for systems formed by (*p*-chloroacetophenone + 1-hexanol) and (diethylcarbonate + *p*-chloroacetophenone). The deviations in refractive index of (diethylcarbonate + 1-hexanol) [7] shows a S-shaped dependence on composition with positive values in the 1-hexanol-rich region and negative values at the opposite extreme. Positive values of deviations in refractive index for

{*p*-chloroacetophenone + 1-hexanol) and (diethylcarbonate + *p*-chloroacetophenone) and with an initial positive region for (diethylcarbonate + 1-hexanol) [7] shows that dispersion forces in the mixture are higher than in the pure liquids. The negative values of Δn_D, at high diethylcarbonate mole fraction for the system of (diethylcarbonate + 1-hexanol) could be explained by considering that when the packing effect decreases, the number of dipoles per unit volume diminishes and therefore, n_D, also becomes smaller, originating negative Δn_D.

Also, the corresponding $\Delta n_{D,123}$, values of the ternary mixture (diethylcarbonate + *p*-chloroacetophenone + 1-hexanol) are given in Table 4 and the isoline curves for $\Delta n_{D,123}$, are shown in Figure 4. The ternary refractive index deviations $\Delta n_{D,123}$, are positive and for few compositions are negative over the composition range.

The coefficients B_p, of Equation (6) and standard deviations $\sigma(\Delta n_{D,123})$, of Equation (5) obtained by the least-squares method are gathered in Table 5.

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