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Refractive Indices and Excess Properties of Binary Mixtures of p-Cymene with Fluorobenzene, Chlorobenzene and Bromobenzene at T = 303.15, 308.15 and 313.15 K

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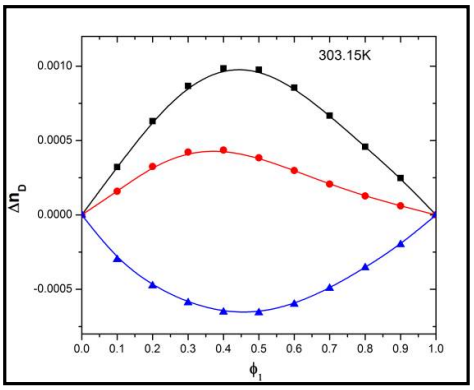
ABSTRACT

Refractive indices (n_D) of binary mixtures of p-cymene with fluorobenzene, chlorobenzene and bromobenzene were measured using Abbe refractometer at 303.15, 308.15 and 313.15 K along the whole composition range and at normal pressure. From the experimental data, deviation in refractive indices (Δn_D), molar refraction (ΔR_m) and deviation in molar refraction (ΔR_m) were calculated. Excess properties were satisfactorily fitted using the Redlich-Kister polynomial equation to derive the standard deviations (σ). These values were reported as a function of volume fraction (ϕ_1) or mole fraction (X_1) of p-cymene. Theoretical study of nine mixing rules of refractive index has been carried out to investigate their validity for these mixtures over the whole mole fraction of p-cymene at all studied temperatures. Results indicated that there is a strong dipole-dipole interaction present in the p-cymene + fluorobenzene binary mixture when compared to chlorobenzene and bromobenzene binary mixtures.

Graphical Abstract



Deviation in refractive index (Δn_D)as a function of volume fraction (ϕ_1) for p-Cymene (1) + Fluorobenzene (2) (■), p-Cymene (1) + Chlorobenzene (2) (●), p-Cymene (1) + Bromobenzene (2) (▲) at *T*= 303.15K..

Keywords: Refractive Index, Molar Refraction, Theoretical mixing rules, Intermolecular interaction.

INTRODUCTION

p-Cymene (1-Methyl-4-(Propan-2-yl) benzene) is a naturally obtained from oils of cypress and essential oils in various plant species. It is also found in citrus juices, pea oils, guava, papaya, pine apple, blue barry, cumine seed, pepper, peppermint oil etc. Due to its odour and taste, it is widely used in daily food products [1]. P-Cymene is also used in many natural therapies due to its medicinal characteristics [2].

The study of thermo physical and transport properties of p-Cymene with organic solvents helps us to understand the nature and behaviour of p-Cymene in binary mixture. Refractive index measurements are very useful in industry and also for common substance like oils, wax and syrup. The prediction of refractive index is essential for the determination of composition of binary mixtures [3, 4]. The study of these parameters helps us in various separation techniques, mass transfer phenomena as well as in various analytical techniques [5, 6].

Literature survey indicates that there is no information is available regarding behaviour of p-cymene with organic solvents. So, it was decided to study refractometric behaviour of the binary mixtures of p-cymene with halobenzenes.

MATERIALS AND METHODS

Materials: p-Cymene (with > 95% purity, Tokyo chemical industry Co. Ltd., Tokyo, Japan), Fluorobenzene (with 99% purity, S. D. Fine Chem. Ltd., India), Chlorobenzene (with 99% purity, S. D. Fine Chem. Ltd., India), Bromobenzene (with 99% purity, S. D. Fine Chem. Ltd., India) were used in this study after purification using standard methods [7]. All the components are purified by the standard methods available in literature [8, 9]. The purification of the liquids was checked by comparing Refractive indices (n_D) with their corresponding literature values reported in table 1.

Table 1. Purification method and comparison of the Refractive indices (n_D) of p-Cymene, Fluorobenzene, Chlorobenzene and Bromobenzene with Literature Data at $T = 303.15$ K

Chemical Name	Supplier	CAS No.	Purification method	(n_D)	
				Exp.	Lit.
p-Cymene	TCI Co.Ltd.	99-87-6	Distillation	1.4860	--
Fluorobenzene	S.D.Fine Chemicals Ltd.	462-06-6	Fractional distillation	1.4612	--
Chlorobenzene	S.D.Fine Chemicals Ltd.	108-90-7	Fractional distillation	1.5201	1.5210[25] 1.5166[26] 1.5150[25]
Bromobenzene	S.D.Fine Chemicals Ltd.	108-86-1	Fractional distillation	1.5550	1.5542[26] 1.5514[26] 1.5500[25]

Apparatus and procedure: Binary mixtures were fresh prepared by gravimetrically with an electronic balance Reptech RA-2012 (Supplied by Reptech India) with an accuracy of ±0.0001 g. The uncertainty in the mole fraction of the mixtures was estimated to less than ± 0.0001.

Measurement of refractive indexes of pure components and binary mixtures for sodium D-line were measured by thermostated Abbe's Refractometer SER. No. 995033. The Refractometer was calibrated using 1-bromonaphthalene, methanol and double distilled water. A water pump was attached to Refractometer for circulating water through Refractometer to maintain the temperature. A thermostatic water bath (Model No. 14L-SS supplied by Equiptron, India) having accuracy of ± 0.01°C with range of 20°C to 90°C has been used for this study.

Calculations

Deviation in refractive index(Δn_D) and deviation in molar refraction(ΔR_m): Table 2 represents the data of refractive indexes (n_D) of pure components and their binary mixtures at 303.15, 308.15 and 313.15 K. Deviation in refractive index (Δn_D) and deviation in molar refraction (ΔR_m) were calculated using following equations and their values are also reported in table 2.

$$\Delta n_D = n_{D_{12}} - \sum_{i=1}^{2} \phi_i \, n_{D_i} \quad (1)$$

$$\Delta R_m = R_{m_{12}} - \sum_{i=1}^{2} R_{m_i} n_{D_i} \quad (2)$$

where, ϕ_i, n_{D_i} and R_{m_i} are volume fraction, refractive index and molar refraction of i[th] component. $n_{D_{12}}$ and $R_{m_{12}}$ are refractive index and molar refraction of the mixture. ϕ_i and R_{m_i} can be calculated using following equations:

$$\phi_i = \frac{x_i v_i}{\sum_{i=1}^{2} x_i v_i} \quad (3)$$

$$R_{m_i} = \frac{n_{D_i}^2 - 1}{n_{D_i}^2 + 2} v_i \quad (4)$$

$$v_i = \frac{M_i}{\rho_i} \quad (5)$$

where, x_i, v_i and M_i are mole fraction, molar volume and molecular mass of i[th] component.

Theoretical Refractive Index Relations:

Arago-biot (A-B) [15]

$$n_D = n_{D_1}\phi_1 + n_{D_2}\phi_2 \quad (6)$$

Dale-Glastone (D-G) [16]

$$n_D - 1 = (n_{D_1} - 1)\phi_1 + (n_{D_2} - 1)\phi_2 \quad (7)$$

Lorentz-lorenz (L-L) [17]

$$\frac{n_D^2 - 1}{n_D^2 + 2} = \left(\frac{n_{D_1}^2 - 1}{n_{D_1}^2 + 2}\right)\phi_1 + \left(\frac{n_{D_2}^2 - 1}{n_{D_2}^2 + 2}\right)\phi_2 \quad (8)$$

Eykman (Eyk) [18]

$$\frac{n_D^2 - 1}{n_D^2 + 0.4} = \left(\frac{n_{D_1}^2 - 1}{n_{D_1}^2 + 0.4}\right)\phi_1 + \left(\frac{n_{D_2}^2 - 1}{n_{D_2}^2 + 0.4}\right)\phi_2 \quad (9)$$

Weiner (w) [19]

$$\frac{n_D^2 - n_{D_1}^2}{n_D^2 + 2n_{D_1}^2} = \left(\frac{n_{D_2}^2 - n_{D_1}^2}{n_{D_2}^2 + 2n_{D_1}^2}\right)\phi_2 \quad (10)$$

Heller (Hr) [20]

$$\frac{n_D - n_{D_1}}{n_{D_1}} = \left(\frac{(n_{D_2} - n_{D_1})^2 - 1}{(n_{D_2} - n_{D_1})^2 + 2}\right)\phi_2 \quad (11)$$

Table 2 Refractive Index (n_D) and Deviation in Refractive Index (Δn_D) Vs Volume Fraction (ϕ_1) for p-Cymene (1) + Fluoro-, Chloro- and Bromobenzene (2) at 303.15, 308.15 and 313.15 K

ϕ_1	n_D			Δn_D			ΔR_m(cm^3·mol^{-1})		
	303.15 K	308.15 K	313.15 K	303.15 K	308.15 K	313.15 K	303.15 K	308.15 K	313.15 K
p-Cymene (1) + Fluorobenzene (2)									
0.0000	1.4612	1.4590	1.4575	0.00000	0.00000	0.00000	0.0000	0.0000	0.0000
0.1000	1.4640	1.4617	1.4601	0.00032	0.00028	0.00027	-0.7247	-0.7242	-0.7230
0.2000	1.4668	1.4644	1.4628	0.00064	0.00061	0.00060	-1.3424	-1.3390	-1.3362
0.3000	1.4695	1.4671	1.4653	0.00086	0.00083	0.00082	-1.8436	-1.8386	-1.8347
0.4000	1.4721	1.4696	1.4678	0.00098	0.00096	0.00094	-2.2113	-2.2053	-2.2007
0.5000	1.4746	1.4720	1.4701	0.00097	0.00095	0.00094	-2.4258	-2.4182	-2.4125
0.6000	1.4770	1.4743	1.4724	0.00087	0.00085	0.00083	-2.4577	-2.4500	-2.4454
0.7000	1.4792	1.4765	1.4745	0.00067	0.00063	0.00062	-2.2740	-2.2686	-2.2638
0.8000	1.4815	1.4787	1.4767	0.00045	0.00043	0.00043	-1.8346	-1.8297	-1.8249
0.9000	1.4838	1.4809	1.4788	0.00025	0.00023	0.00019	-1.0943	-1.0921	-1.0924
1.0000	1.4860	1.4831	1.4809	0.00000	0.00000	0.00000	0.0000	0.0000	0.0000
p-Cymene (1) + Chlorobenzene (2)									
0.0000	1.5201	1.5170	1.5144	0.00000	0.00000	0.00000	0.0000	0.0000	0.0000
0.1000	1.5169	1.5138	1.5112	0.00016	0.00014	0.00012	-0.4654	-0.4658	-0.4673
0.2000	1.5136	1.5105	1.5080	0.00032	0.00031	0.00029	-0.8586	-0.8575	-0.8595
0.3000	1.5103	1.5073	1.5048	0.00043	0.00042	0.00041	-1.1738	-1.1725	-1.1740
0.4000	1.5069	1.5039	1.5014	0.00044	0.00043	0.00042	-1.4034	-1.4016	-1.4034
0.5000	1.5034	1.5004	1.4980	0.00037	0.00036	0.00035	-1.5332	-1.5317	-1.5338
0.6000	1.5000	1.4970	1.4946	0.00031	0.00030	0.00029	-1.5396	-1.5379	-1.5399
0.7000	1.4964	1.4935	1.4911	0.00020	0.00019	0.00018	-1.4101	-1.4087	-1.4105
0.8000	1.4930	1.4900	1.4877	0.00013	0.00012	0.00011	-1.1245	-1.1236	-1.1253
0.9000	1.4895	1.4866	1.4843	0.00006	0.00006	0.00005	-0.6641	-0.6631	-0.6645
1.0000	1.4860	1.4831	1.4809	0.00000	0.00000	0.00000	0.0000	0.0000	0.0000
p-Cymene (1) + Bromobenzene (2)									
0.0000	1.5550	1.5521	1.5490	0.00000	0.00000	0.00000	0.0000	0.0000	0.0000
0.1000	1.5478	1.5449	1.5419	-0.00030	-0.00032	-0.00033	-0.3661	-0.3666	-0.3688
0.2000	1.5407	1.5378	1.5349	-0.00047	-0.00049	-0.00050	-0.6720	-0.6718	-0.6752
0.3000	1.5337	1.5308	1.5280	-0.00060	-0.00061	-0.00062	-0.9170	-0.9157	-0.9201
0.4000	1.5268	1.5238	1.5211	-0.00065	-0.00066	-0.00067	-1.0912	-1.0894	-1.0945
0.5000	1.5199	1.5169	1.5143	-0.00065	-0.00066	-0.00067	-1.1858	-1.1838	-1.1891
0.6000	1.5130	1.5101	1.5075	-0.00060	-0.00061	-0.00062	-1.1840	-1.1819	-1.1872
0.7000	1.5062	1.5033	1.5008	-0.00050	-0.00051	-0.00052	-1.0739	-1.0721	-1.0770
0.8000	1.4995	1.4965	1.4942	-0.00035	-0.00036	-0.00037	-0.8493	-0.8481	-0.8522
0.9000	1.4927	1.4898	1.4875	-0.00020	-0.00022	-0.00023	-0.4994	-0.4999	-0.5027
1.0000	1.4860	1.4831	1.4809	0.00000	0.00000	0.00000	0.0000	0.0000	0.0000

Standard uncertainties U are U(T)= ±0.01 K, U(n_D) = ±0.0001, U(ϕ) = ±0.0001. All physical quantities are measured at atmospheric pressure

Newton (Nw) [21]

$$n_{D_1}^2 - 1 = \left(n_{D_1}^2 - 1\right)\phi_1 + \left(n_{D_2}^2 - 1\right)\phi_2 \qquad (12)$$

Oster (Os) [22]

$$\left[\frac{(n_D^2-1)\,(2n_D^2+1)}{n_D^2}\right]V = \left[\frac{(n_{D_1}^2-1)\,(2n_{D_1}^2+1)}{n_{D_1}^2}\right]x_1v_1 + \left[\frac{(n_{D_2}^2-1)\,(2n_{D_2}^2+1)}{n_{D_2}^2}\right]x_2v_2 \qquad (13)$$

Eyring and John (EJ) [23]

$$n_D = n_{D_1}\phi_1^2 + 2(n_{D_1}n_{D_2})^{1/2}\phi_1\phi_2 + n_{D_2}\phi_2^2 \quad (14)$$

The experimentally determined values of Δn_D, and ΔR_m for binary mixtures were fitted to a following Redlich-Kisters' equation [24],

$$\Delta_{12} = x_1 x_2 \sum_{i=0}^{i-n} A_i\,(2x_1 - 1)^i \quad (15)$$

Where, Δ_{12} is given excess or deviation function X_1 and $X_2$are the mole fraction of component 1 and 2 respectively. The coefficients A_i were determined by a multiple regression analysis based on least square method. The number of coefficient for a given set of experimental point of any function were optimized by keeping the ratio between experimental points and number of coefficient to a maximum value which in case was not less than 3.

The standard deviation (σ) was calculated using following expression,

$$\sigma = [\sum(\Delta_{obs} - \Delta_{cal})^2/(N-n)]^{1/2} \quad (16)$$

Where, N is the number of measurement and n is the number of parameters. Fitting coefficients, A_i, and the standard deviations, σ, are represented in table 3.

Table 3. Fitting coefficients,(A_0 ,A_1,A_2,A_3,A_4 with Standard deviation σ for Least square representation of Δn_D and ΔR_m of p-cymene (1) + fluoro-, chloro- and bromobenzene (2) binary mixtures

Least Square	T(K)	A_0	A_1	A_2	A_3	A_4	σ
p-Cymene (1) + Fluorobenzene (2)							
Δn_D	303.15	0.00390	-0.00141	-0.00169	0.00140	0.00085	0.000009
	308.15	0.00381	-0.00149	-0.00182	0.00175	0.00048	0.000013
	313.15	0.00372	-0.00133	-0.00122	0.00121	-0.00091	0.000012
ΔR_m ($cm^3 \cdot mol^{-1}$)	303.15	-9.70277	-2.56133	-0.62504	-0.00797	-0.00648	0.000239
	308.15	-9.67426	-2.55726	-0.62759	0.00363	-0.03438	0.000577
	313.15	-9.65552	-2.54028	-0.58320	-0.03599	-0.13596	0.001213
p-Cymene (1) + Chlorobenzene (2)							
Δn_D	303.15	0.00154	-0.00148	-0.00014	0.00125	-0.00056	0.000009
	308.15	0.00149	-0.00156	-0.00014	0.00158	-0.00073	0.000009
	313.15	0.00146	-0.00157	-0.00021	0.00169	-0.00094	0.000009
ΔR_m ($cm^3 \cdot mol^{-1}$)	303.15	-6.12796	-1.40906	-0.13430	0.04838	-0.15004	0.000832
	308.15	-6.12134	-1.41527	-0.13108	0.07196	-0.16303	0.000767
	313.15	-6.12909	-1.41616	-0.13431	0.07496	-0.17875	0.000884
p-Cymene (1) + Bromobenzene (2)							
Δn_D	303.15	-0.00263	0.00054	0.00050	0.00024	-0.00113	0.000007
	308.15	-0.00267	0.00058	0.00053	0.00019	-0.00162	0.000007
	313.15	-0.00271	0.00058	0.00053	0.00019	-0.00179	0.000007
ΔR_m ($cm^3 \cdot mol^{-1}$)	303.15	-4.74254	-0.93812	0.05753	0.02235	-0.25041	0.000898
	308.15	-4.73457	-0.93179	0.06107	0.01349	-0.28892	0.001004
	313.15	-4.75608	-0.93436	0.06042	0.01094	-0.30310	0.001028

RESULTS AND DISCUSSION

Δn_D values are positive for p-cymene + fluorobenzene and + chlorobenzene binary mixtures and negative for p-cymene + bromobenzene binary mixtures. The temperature dependence of this property is extremely systematic and Δn_D values show decrease with increase of temperature for all binary

mixtures. The trends in Δn_D values are positive to less positive and then negative for fluorobenzene, chlorobenzene and bromobenzene binary mixtures respectively. ΔR_m values are negative for all binary mixtures at all temperatures. With increase in temperatures the negative values of ΔR_m become less negative. The values of ΔR_m are become less negative when binary mixtures changes from fluorobenzene to bromobenzene.

The deviation in refractive index (Δn_D) is representing the electronic perturbation due to orbital mixing of molecule. Positive (Δn_D) values indicate that there is a strong interaction present between the unlike molecules [10]. The negative values of ΔR_m indicate for presence of strong interaction [11, 12]. The refractive index, (n_D) of mixtures increases due to the increase in polarizability of mixtures comes from the dipole-dipole interactions between the unlike molecules [13]. Data shows that there is a strong dipole-dipole interaction present between the p-cymene + fluorobenzene binary mixture when compared to chlorobenzene and bromobenzene binary mixtures. The graphical veriation ofΔn_D and ΔR_mwere presented in figure 1 and 2, respectively.

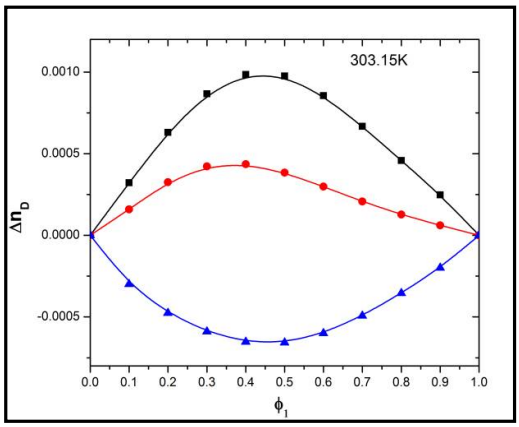


Figure 1. Deviation in refractive index (Δn_D) as a function of volume fraction (ϕ_1) for p-Cymene (1) + Fluorobenzene (2) (■), p-Cymene (1) + Chlorobenzene (2) (●), p-Cymene (1) + Bromobenzene (2) (▲) at *T*= 303.15K.

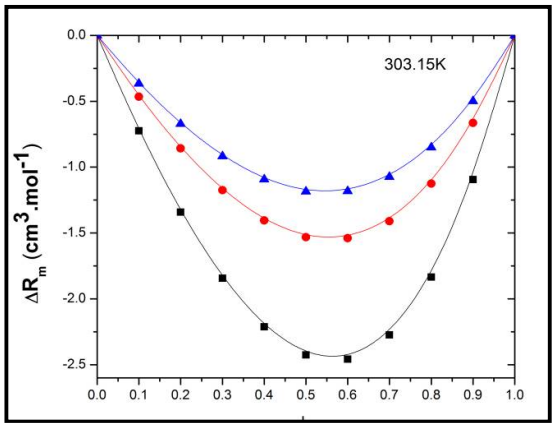


Figure 2. Deviation in molar refraction (ΔR_m) as a function of volume fraction (ϕ_1) for p-Cymene (1) + Fluorobenzene (2) (■), p-Cymene (1) + Chlorobenzene (2) (●), p-Cymene (1) + Bromobenzene (2) (▲) at *T*= 303.15K.

Using mixing rules, the refractive indices of the mixture can be predicated from the experimental densities of mixture and pure components, and the refractive indices of pure component [14]. So, nine mixing rules are applied and the average percentage deviation, (σ %) for these relations are given in table 4 which gives as following characteristics

(i) Arago-biot, (A-B) and Dale-Gladstone, (G-D) show the similar values for σ% for all three binary mixtures.

(ii) Weiner, (WR) relation shows positive σ % for p-cymene + fluorobenzene and negative σ % for p-cymene + chlorobenzene and + bromobenzene binary mixtures and also have lowest σ % values among all the relations.

(iii) Heller relation shows positive σ % values for p-cymene + fluorobenzene and + chlorobenzene binary mixtures and negative for p-cymene + bromobenzene binary mixture and also have the highest σ % values among all relation for all binary mixtures.

Table 4. Average deviation (σ) in the Refractive index from Nine different mixing relations of p-cymene (1) + fluoro-, chloro- and bromobenzene (2) binary mixtures

Parameters	*T* (K)		
	303.15	308.15	313.15
p-Cymene (1) + Fluorobenzene (2)			
A–B	0.000546	0.000524	0.000513
G–D	0.000546	0.000524	0.000513
L–L	0.000580	0.000556	0.000543
WR	0.000281	0.000273	0.000276
Heller	0.000580	0.000556	0.000543
Newton	0.000515	0.000494	0.000485
Eyring–John	0.000562	0.000539	0.000527
Eykman	0.000556	0.000533	0.000521
Oster	0.000527	0.000506	0.000496
p-Cymene (1) + Chlorobenzene (2)			
A–B	0.000220	0.000212	0.000202
G–D	0.000220	0.000212	0.000202
L–L	0.000285	0.000276	0.000264
WR	-0.000282	-0.000286	-0.000285
Heller	0.000287	0.000278	0.000267
Newton	0.000162	0.000154	0.000146
Eyring–John	0.000249	0.000241	0.000230
Eykman	0.000237	0.000229	0.000219
Oster	0.000183	0.000175	0.000166
p-Cymene (1) + Bromobenzene (2)			
A–B	-0.000393	-0.000404	-0.000412
G–D	-0.000393	-0.000404	-0.000412
L–L	-0.000124	-0.000135	-0.000151
WR	-0.002449	-0.002463	-0.002421
Heller	-0.000110	-0.000120	-0.000136
Newton	-0.000628	-0.000639	-0.000641
Eyring–John	-0.000275	-0.000286	-0.000297
Eykman	-0.000324	-0.000334	-0.000344
Oster	-0.000547	-0.000558	-0.000562

A-B = Arago-Biot, D-G = Dale-Gladstone, L-L = Lorentz-Lorentz, WR = Weiner

APPLICATION

The polarity is decrease from fluorobenzene to bromobenzene and it is clear from result that the interactions become weaker when the binary mixture changes from fluorobenzene to bromobenzene.

CONCLUSIONS

Small positive value of deviation in $\Delta n_D$and negative values of molar refraction ΔR_m show that the interactions present in p-cymene + fluorobenzene binaries are stronger when compared to other binaries. Nine mixing relations for refractive index studies also supports that the interaction becomes weaker when moving from fluorobenzene to bromobenzene in these binaries because of the polarity of the halobenzenes. Because of the polarity of the molecules, the interactions are either decrease or increase depends on the structure of the component. In present study fluorobenzene is a highly polar than the other two halobenzenes, so it gives higher interaction with p-cymene molecules. The polarity is decrease from fluorobenzene to bromobenzene and it is clear from result that the interaction becomes weaker when the binary mixture changes from fluorobenzene to bromobenzene.

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