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# Optical Studies of Binary Mixture of Chloro Substituted Benzene and *n*-Hexane or Cyclohexane or 1,4-Dioxane

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Thermophysical properties of binary liquid mixtures are highly beneficial for getting information about the intermolecular interactions and geometrical effects in the system. The chloro-substituted benzene compounds like 2-chlorotoluene, 4-chlorotoluene, 1,3-dichlorobenzene also have wide range of application in industrial and biomedical areas. In present work, the refractive indices (*n*) of haloarenes, hydrocarbons, ether, and respective possible binary mixtures were experimentally determined over the entire compositions at T= (298.15-318.15) K. The mixtures selected were 2-chlorotoluene or 4-chlorotoluene or 1,3-dichlorobenzene (1) + *n*-hexane or cyclohexane or 1,4-dioxane (2) with its possible combinations. The $\Delta n$ is positive for all binary mixtures at all investigated compositions. Different rules of mixing like Lorentz-Lorentz, Erying-John, Arago-Biot, *etc*. were also used to predict *n* values. The $\Delta n$ values were also analyzed in terms of ongoing intermolecular interactions among the components of the selected systems.

**Keywords: Refractive index, Chlorotoluene, 1,3-Dichlorobenzene, 1,4-Dioxane, Hydrocarbons, Mixing rules.**

## INTRODUCTION

The thermophysical properties of binary liquid mixtures are highly beneficial to know about the intermole-cular interactions and geometrical effects in the system [1-5]. The chloro-substituted benzene compounds like 2-chlorotoluene, 4-chlorotoluene, 1,3-dichlorobenzene also have a wide range of industrial applications [6-8]. The presence of a chloro group in the toluene molecule may affect the extent of intermolecular interactions. 2-Chlorotoluene is used as an intermediate and solvent in the chemical industries as well as a solvent for the formulation of agricultural pesticides. 4-Chlorotoluene is a suitable intermediate for organic synthesis, dyes and pesticide industries. 1,3-Dichlorobenzene is used as an intermediate in pesticide, pharmaceutical and dye industries. Hexane is used as non-polar liquid/ solvent to compare the dipolar stabilization of polar molecules. In industries, hexane is used in the formulation of glues for shoes, leather products and roofing [9]. Cyclohexane is mainly used for the industrial production of precursors used for nylon production.

The current study was undertaken to analyze the intermolecular interactions among the constituents of binary mixtures of chloro substituted benzene with *n*-hexane & dioxane and how they affect the sign and magnitude of deviation in refractive index of binary mixture. From chemical thermodynamics, it is known that ideality never exists in this real world. There always exists some deviation from ideality [10,11]. In continuation of earlier work on optical studies [12-14], here we reported the measured refractive index (*n*) data and calculated deviations in refractive index ($\Delta n$) values of binary mixtures containing 2-chlorotoluene or 4-chlorortoluene or 1,3-dichlorobenzene (1) with *n*-hexane or cyclohexane or 1,4-dioxane (2) as function of mole fractions and temperatures *T* = (298.15 - 318.15) K at atmospheric pressure. The reported measurements clearly show deviation when one component comes in contact with another component in liquid binary mixtures due to ongoing interactions between their molecules, which makes it non-ideal. In this work, an attempt is made to explore the type of interactions and their magnitude prevailing in between polar and non-polar compounds, which are responsible for these deviations from ideality. These values have their significant importance for engineers in chemical designing the industrial processes and also for chemists to study intermolecular inter-

actions. Different mixing rules are also applied for theoretical approximation. In our best of knowledge, no attempt has been made to measure the refractive index for the present mixtures.

## EXPERIMENTAL

2-Chlorotoluene (Loba, > 98% purity), 4-chlorotoluene (Loba, > 99% purity) and 1,3-dichlorobenzene (Loba, > 98% purity) were purified by their standard procedures [15,16], while *n*-hexane, cyclohexane, 1,4-dioxane (Merck, > 99% purity) were distilled twice. All the chemicals were kept over molecular sieves (4 Å) for more than 72 h after their purification and their chemical specifications are mentioned in Table-1. The 'ρ' values of all compounds were measured using vibrating tube densimeter (DSA 5000 M Anton Paar) at (T = 298.15-318.15) K and $p$ = 0.1 MPa. The $n$ values of all compounds and binary mixtures were measured using refractometer (Anton Paar Abbemat 200) having temperature-controlled within ±0.01 K. The densimeter as well as refractometer was calibrated by measuring the ρ and $n$ values of doubly distilled deionized degassed water as a function of temperature. Binary mixtures were prepared using a sensitive weighing balance (AR224CN, OHAUS) of accuracy up to 0.1 mg and uncertainty in mole fraction was 0.0002. Practically considering the factors like the purity of chemicals and calibration of instruments, the standard uncertainty ($u$) in measured ρ and $n$ values were found to be 0.05 kg.m$^{-3}$ and 0.0002, respectively. The measured ρ and $n$ values of pure components are in accordance with their literature [7-9,17-31] values as shown in Table-2.

## RESULTS AND DISCUSSION

The deviation in refractive index ($\Delta n$) have been evaluated using eqn. 1:

$$\Delta n = n - \sum_{i=1}^{2} x_i n_i \quad (1)$$

where, $x_i$ and $n_i$ shows the mole fraction and refractive index of the i$^{th}$ component, respectively and $n$ is the refractive index of the binary mixture formed after mixing. Experimental $n$ and $\Delta n$ values of the defined systems are depicted in Table-3. The polynomial Redlich-Kister (eqn. 2) for fitting $\Delta n$ values is used [32]:

$$\Delta n = x_1(1-x_1)\left[\sum_{n=1}^{4} A^{(n)}(2x_1-1)^{(n-1)}\right] \quad (2)$$

where $A^{(n)}$ represents adjustable parameters and their standard deviations $\sigma(\Delta n)$ was calculated using eqn. 2:

$$\sigma(\Delta n) = \left\{\left[\sum\left(\Delta n_{exptl.} - \Delta n_{calcd.(eqn.\ 2)}\right)^2\right] \Big/ (m-n)\right\}^{1/2} \quad (3)$$

TABLE-1
CHEMICAL SOURCE, SPECIFICATION, ANALYSIS METHOD OF CHEMICALS AND PURITY

| Compound | Source | Stated purity (mass fraction) | CAS number | Analysis method[a] | Confirmed purity[a] (mass fraction) |
|---|---|---|---|---|---|
| 2-Chlorotoluene | Loba | ≥ 0.98 | 95-49-8 | GC | 0.998 |
| 4-Chlorotoluene | Loba | ≥ 0.99 | 106-43-4 | GC | 0.995 |
| 1,3-Dichlorobenzene | Loba | ≥ 0.98 | 541-73-1 | GC | 0.995 |
| *n*-Hexane | Merck | ≥ 0.99 | 110-54-3 | GC | 0.997 |
| Cyclohexane | Merck | ≥ 0.99 | 110-82-7 | GC | 0.996 |
| 1,4-Dioxane | Merck | ≥ 0.99 | 123-91-1 | GC | 0.997 |

[a]Determined by gas chromatography analysis after purification.

TABLE-2
MEASURED DENSITIES (ρ, g cm$^{-3}$) AND REFRACTIVE INDICES ($n$) OF THE PURE COMPONENTS AT VARIOUS TEMPERATURES (T = 298.15-318.15 K)

| Compound | Temp. (K) | ρ | | $n$ | |
|---|---|---|---|---|---|
| | | This work | Literature | This work | Literature |
| 2-Chlorotoluene | 298.15 | 1.077458 | 1.07730 [7,8]<br>1.07741 [17] | 1.5227 | 1.52190 [17] |
| | 308.15 | 1.067682 | 1.06760 [7,8]<br>1.06824 [17] | 1.5175 | 1.51690 [17] |
| | 318.15 | 1.057862 | – | 1.5124 | |
| 4-Chlorotoluene | 298.15 | 1.064325 | 1.06437 [18]<br>1.06511 [17] | 1.5184 | 1.51880 [17] |
| | 308.15 | 1.054484 | 1.05390 [17] | 1.5133 | 1.51380 [17] |
| | 318.15 | 1.044607 | – | 1.5082 | |
| 1,3-Dichlorobenzene | 298.15 | 1.282835 | 1.28279 [19]<br>1.28300 [9]<br>1.27777 [20]<br>1.28276 [21] | 1.5436 | 1.54380 [21] |
| | 308.15 | 1.271529 | 1.27209 [20]<br>1.27149 [19] | 1.5383 | – |
| | 318.15 | 1.260204 | 1.26131 [20]<br>1.26017 [19] | 1.5334 | – |

| | | | | | |
|---|---|---|---|---|---|
| *n*-Hexane | 298.15 | 0.655206 | 0.65528 [9]<br>0.65493 [22]<br>0.65520 [23] | 1.3726 | 1.37207 [23]<br>1.37211 [24]<br>1.37330 [25]<br>1.37290 [26]<br>1.37220 [27] |
| | 308.15 | 0.646031 | 0.64574 [22]<br>0.64600 [23] | 1.3671 | 1.36626 [23]<br>1.36652 [24]<br>1.36790 [25]<br>1.36720 [26] |
| | 318.15 | 0.636677 | 0.63637 [22] | 1.3616 | 1.36052 [24]<br>1.36280 [25] |
| 1,4-Dioxane | 298.15 | 1.027757 | 1027.900 [8]<br>1027.850 [28]<br>1.027994 [29] | 1.4195 | 1.41967 [28] |
| | 308.15 | 1.016464 | 1016.600 [8]<br>1016.520 [28]<br>1.016663 [29] | 1.4149 | 1.41480 [28] |
| | 318.15 | 1.00511 | 1005.120 [28]<br>1.005279 [29] | 1.4102 | 1.40993 [28] |
| Cyclohexane | 298.15 | 0.773869 | 773.94 [7]<br>773.84 [30]<br>773.97 [25] | 1.4236 | 1.42370 [25]<br>1.42350 [27] |
| | 308.15 | 0.764374 | 764.45 [7]<br>764.35 [30]<br>764.49 [25] | 1.4181 | 1.41800 [25]<br>1.41750 [31] |
| | 318.15 | 0.754763 | 754.74 [30]<br>754.89 [25] | 1.4125 | 1.41230 [25] |

Standard uncertainties u: u(T) = 0.01 K; u(p) = 0.04 MPa, and the standard uncertainty $U_c$: $U_c$ (ρ) = 0.05 kg m$^{-3}$; $U_c(n)$ = 0.0002.

where m and n represent the number of measured data points and the number of adjustable parameters in eqn. 2, respectively. The choice of n to have 1-4 values was based on the consideration that the maximum deviation $\sigma_{max}(\Delta n)$ calculated using eqn. 3 from corresponding $\Delta n$ values satisfied the relation $\sigma_{max}(\Delta n) \leq 2\sigma(\Delta n)$. The adjustable parameters $A^{(n)}$ and standard deviations $\sigma(\Delta n)$ are given in Table-4.

Different mixing rules [33] used for deviation in the refractive index are as follows:

Arago-Biot:

$$n = n_1\phi_1 + n_2\phi_2 \quad (4)$$

Gladstone-Dale:

$$n\text{-}1 = (n_1\text{-}1)\phi_1 + (n_2\text{-}1)\phi_2 \quad (5)$$

Lorentz-Lorentz:

$$\frac{n^2-1}{n^2+2} = \left(\frac{n_1^2-1}{n^2+2}\right)\phi_1 + \left(\frac{n_2^2-1}{n^2+2}\right)\phi \quad (6)$$

Heller:

$$\frac{n-1}{n} = \frac{3}{2}\left(\frac{(n_2/n_1)^2-1}{(n_2/n_1)+2}\right)\phi_2 \quad (7)$$

TABLE-3
REFRACTIVE INDEX ($n_D$) AND DEVIATION IN REFRACTIVE INDEX ($\Delta n$) FOR BINARY SYSTEMS AT DIFFERENT TEMPERATURES (T = 298.15-318.15 K)

| $x_1$ | 298.15 K | | 308.15 K | | 318.15 K | | $x_1$ | 298.15 K | | 308.15 K | | 318.15 K | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | $n$ | $\Delta n$ | $n$ | $\Delta n$ | $n$ | $\Delta n$ | | $n$ | $\Delta n$ | $n$ | $\Delta n$ | $n$ | $\Delta n$ |
| 2-Chlorotoluene (1) + *n*-hexane (2) | | | | | | | 2-Chlorotoluene (1) + cyclohexane (2) | | | | | | |
| 0.0629 | 1.3854 | 0.0034 | 1.3802 | 0.0036 | 1.3748 | 0.0037 | 0.0559 | 1.4308 | 0.0016 | 1.4252 | 0.0015 | 1.4203 | 0.0021 |
| 0.1435 | 1.3980 | 0.0039 | 1.3933 | 0.0046 | 1.3885 | 0.0053 | 0.1311 | 1.4396 | 0.0030 | 1.4345 | 0.0033 | 1.4296 | 0.0039 |
| 0.2364 | 1.4111 | 0.0030 | 1.4070 | 0.0043 | 1.4030 | 0.0058 | 0.2251 | 1.4494 | 0.0034 | 1.4447 | 0.0042 | 1.4397 | 0.0049 |
| 0.3403 | 1.4270 | 0.0033 | 1.4233 | 0.0050 | 1.4201 | 0.0072 | 0.3191 | 1.4593 | 0.0040 | 1.4546 | 0.0047 | 1.4497 | 0.0051 |
| 0.4251 | 1.4416 | 0.0052 | 1.4380 | 0.0070 | 1.4350 | 0.0093 | 0.4059 | 1.4679 | 0.0040 | 1.4630 | 0.0045 | 1.4579 | 0.0049 |
| 0.4608 | 1.4480 | 0.0062 | 1.4445 | 0.0081 | 1.4414 | 0.0103 | 0.4469 | 1.4719 | 0.0040 | 1.4668 | 0.0042 | 1.4622 | 0.0048 |
| 0.5248 | 1.4595 | 0.0081 | 1.4560 | 0.0100 | 1.4530 | 0.0123 | 0.4920 | 1.4760 | 0.0036 | 1.4713 | 0.0042 | 1.4664 | 0.0045 |
| 0.5741 | 1.4682 | 0.0094 | 1.4647 | 0.0113 | 1.4614 | 0.0132 | 0.5430 | 1.4811 | 0.0036 | 1.4759 | 0.0038 | 1.4710 | 0.0042 |
| 0.6236 | 1.4767 | 0.0105 | 1.4728 | 0.0119 | 1.4695 | 0.0139 | 0.5977 | 1.4862 | 0.0033 | 1.4812 | 0.0036 | 1.4760 | 0.0039 |
| 0.6816 | 1.4856 | 0.0107 | 1.4817 | 0.0121 | 1.4780 | 0.0136 | 0.6503 | 1.4910 | 0.0029 | 1.4859 | 0.0031 | 1.4809 | 0.0035 |
| 0.7397 | 1.4934 | 0.0098 | 1.4894 | 0.0110 | 1.4854 | 0.0123 | 0.7010 | 1.4957 | 0.0026 | 1.4905 | 0.0027 | 1.4859 | 0.0031 |
| 0.8335 | 1.5040 | 0.0063 | 1.4996 | 0.0071 | 1.4952 | 0.0079 | 0.8048 | 1.5051 | 0.0017 | 1.5000 | 0.0019 | 1.4951 | 0.0022 |
| 0.9275 | 1.5136 | 0.0018 | 1.5087 | 0.0021 | 1.5040 | 0.0025 | 0.9117 | 1.5147 | 0.0007 | 1.5097 | 0.0009 | 1.5047 | 0.0010 |

| 2-Chlorotoluene (1) + 1,4-dioxane (2) | | | | | | | 4-Chlorotoluene (1) + *n*-hexane (2) | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| 0.0612 | 1.4276 | 0.0017 | 1.4229 | 0.0017 | 1.4187 | 0.0022 | 0.0878 | 1.3881 | 0.0026 | 1.3843 | 0.0043 | 1.3813 | 0.0068 |
| 0.1151 | 1.4346 | 0.0032 | 1.4301 | 0.0033 | 1.4259 | 0.0039 | 0.1581 | 1.4075 | 0.0118 | 1.4021 | 0.0118 | 1.3948 | 0.0100 |
| 0.2025 | 1.4465 | 0.0061 | 1.4425 | 0.0068 | 1.4377 | 0.0068 | 0.2445 | 1.4249 | 0.0166 | 1.4241 | 0.0212 | 1.4191 | 0.0216 |
| 0.2969 | 1.4583 | 0.0081 | 1.4541 | 0.0087 | 1.4495 | 0.0089 | 0.2988 | 1.4397 | 0.0235 | 1.4359 | 0.0251 | 1.4316 | 0.0261 |
| 0.4171 | 1.4719 | 0.0093 | 1.4674 | 0.0097 | 1.4630 | 0.0101 | 0.4003 | 1.4569 | 0.0259 | 1.4528 | 0.0271 | 1.4497 | 0.0294 |
| 0.4675 | 1.4772 | 0.0094 | 1.4727 | 0.0098 | 1.4682 | 0.0102 | 0.4700 | 1.4659 | 0.0247 | 1.4597 | 0.0238 | 1.4557 | 0.0251 |
| 0.5174 | 1.4822 | 0.0093 | 1.4777 | 0.0097 | 1.4731 | 0.0100 | 0.5234 | 1.4707 | 0.0217 | 1.4670 | 0.0233 | 1.4615 | 0.0231 |
| 0.5713 | 1.4871 | 0.0086 | 1.4828 | 0.0092 | 1.4784 | 0.0098 | 0.5719 | 1.4749 | 0.0189 | 1.4710 | 0.0202 | 1.4652 | 0.0197 |
| 0.6260 | 1.4921 | 0.0079 | 1.4875 | 0.0083 | 1.4830 | 0.0088 | 0.6219 | 1.4811 | 0.0178 | 1.4760 | 0.0179 | 1.4701 | 0.0173 |
| 0.6840 | 1.4971 | 0.0070 | 1.4927 | 0.0076 | 1.4882 | 0.0080 | 0.6809 | 1.4854 | 0.0135 | 1.4803 | 0.0136 | 1.4751 | 0.0136 |
| 0.7187 | 1.5006 | 0.0069 | 1.4952 | 0.0065 | 1.4906 | 0.0069 | 0.7856 | 1.4959 | 0.0087 | 1.4903 | 0.0083 | 1.4855 | 0.0087 |
| 0.8028 | 1.5074 | 0.0050 | 1.5023 | 0.0050 | 1.4974 | 0.0051 | 0.8602 | 1.5041 | 0.0060 | 1.4987 | 0.0058 | 1.4937 | 0.0059 |
| 0.9289 | 1.5175 | 0.0021 | 1.5125 | 0.0022 | 1.5075 | 0.0023 | 0.9366 | 1.5123 | 0.0031 | 1.5068 | 0.0027 | 1.5019 | 0.0029 |
| 4-Chlorotoluene (1) + cyclohexane (2) | | | | | | | 4-Chlorotoluene (1) + 1,4-dioxane (2) | | | | | | |
| 0.0734 | 1.4307 | 0.0001 | 1.4254 | 0.0003 | 1.4201 | 0.0005 | 0.0743 | 1.4292 | 0.0023 | 1.4246 | 0.0023 | 1.4201 | 0.0026 |
| 0.1401 | 1.4379 | 0.0010 | 1.4329 | 0.0014 | 1.4279 | 0.0019 | 0.1380 | 1.4370 | 0.0038 | 1.4327 | 0.0042 | 1.4281 | 0.0043 |
| 0.2138 | 1.4470 | 0.0031 | 1.4419 | 0.0034 | 1.4371 | 0.0041 | 0.2520 | 1.4503 | 0.0058 | 1.4460 | 0.0063 | 1.4416 | 0.0067 |
| 0.2594 | 1.4521 | 0.0039 | 1.4472 | 0.0043 | 1.4427 | 0.0053 | 0.2987 | 1.4559 | 0.0068 | 1.4512 | 0.0069 | 1.4466 | 0.0071 |
| 0.3561 | 1.4626 | 0.0052 | 1.4579 | 0.0058 | 1.4533 | 0.0067 | 0.3512 | 1.4612 | 0.0069 | 1.4571 | 0.0076 | 1.4525 | 0.0078 |
| 0.4243 | 1.4699 | 0.0060 | 1.4650 | 0.0065 | 1.4604 | 0.0072 | 0.4169 | 1.4686 | 0.0078 | 1.4639 | 0.0079 | 1.4592 | 0.0081 |
| 0.4747 | 1.4748 | 0.0061 | 1.4699 | 0.0066 | 1.4646 | 0.0066 | 0.4663 | 1.4734 | 0.0077 | 1.4686 | 0.0078 | 1.4640 | 0.0080 |
| 0.5238 | 1.4795 | 0.0062 | 1.4747 | 0.0067 | 1.4700 | 0.0073 | 0.5185 | 1.4786 | 0.0078 | 1.4740 | 0.0080 | 1.4692 | 0.0081 |
| 0.5744 | 1.4841 | 0.0060 | 1.4794 | 0.0066 | 1.4746 | 0.0071 | 0.5712 | 1.4837 | 0.0077 | 1.4789 | 0.0077 | 1.4744 | 0.0082 |
| 0.6354 | 1.4895 | 0.0056 | 1.4849 | 0.0063 | 1.4803 | 0.0069 | 0.6250 | 1.4889 | 0.0075 | 1.4842 | 0.0077 | 1.4794 | 0.0079 |
| 0.7515 | 1.4994 | 0.0045 | 1.4949 | 0.0052 | 1.4901 | 0.0056 | 0.7059 | 1.4959 | 0.0065 | 1.4911 | 0.0067 | 1.4862 | 0.0068 |
| 0.8350 | 1.5061 | 0.0033 | 1.5017 | 0.0041 | 1.4970 | 0.0045 | 0.8013 | 1.5036 | 0.0048 | 1.4987 | 0.0049 | 1.4938 | 0.0050 |
| 0.9230 | 1.5126 | 0.0014 | 1.5076 | 0.0016 | 1.5035 | 0.0026 | 0.9033 | 1.5113 | 0.0024 | 1.5063 | 0.0025 | 1.5014 | 0.0026 |
| 1,3-Dichlorobenzene (1) + *n*-hexane (2) | | | | | | | 1,3-Dichlorobenzene (1) + cyclohexane (2) | | | | | | |
| 0.0593 | 1.3866 | 0.0038 | 1.3822 | 0.0049 | 1.3788 | 0.0070 | 0.0207 | 1.4267 | 0.0006 | 1.4212 | 0.0006 | 1.4159 | 0.0008 |
| 0.1575 | 1.4101 | 0.0105 | 1.4059 | 0.0118 | 1.4037 | 0.0150 | 0.0968 | 1.4383 | 0.0030 | 1.4330 | 0.0032 | 1.4277 | 0.0034 |
| 0.2549 | 1.4329 | 0.0167 | 1.4281 | 0.0173 | 1.4246 | 0.0192 | 0.1547 | 1.4460 | 0.0038 | 1.4406 | 0.0038 | 1.4353 | 0.0040 |
| 0.3472 | 1.4506 | 0.0186 | 1.4468 | 0.0202 | 1.4431 | 0.0218 | 0.1824 | 1.4495 | 0.0040 | 1.4441 | 0.0040 | 1.4394 | 0.0048 |
| 0.4204 | 1.4625 | 0.0180 | 1.4585 | 0.0194 | 1.4544 | 0.0205 | 0.2714 | 1.4615 | 0.0053 | 1.4563 | 0.0055 | 1.4510 | 0.0056 |
| 0.4799 | 1.4715 | 0.0168 | 1.4676 | 0.0183 | 1.4641 | 0.0200 | 0.3684 | 1.4735 | 0.0056 | 1.4685 | 0.0061 | 1.4635 | 0.0064 |
| 0.5322 | 1.4787 | 0.0150 | 1.4745 | 0.0162 | 1.4713 | 0.0182 | 0.4677 | 1.4861 | 0.0063 | 1.4811 | 0.0067 | 1.4761 | 0.0070 |
| 0.5704 | 1.4830 | 0.0128 | 1.4789 | 0.0141 | 1.4753 | 0.0157 | 0.5183 | 1.4922 | 0.0063 | 1.4871 | 0.0066 | 1.4819 | 0.0067 |
| 0.6294 | 1.4914 | 0.0111 | 1.4870 | 0.0121 | 1.4831 | 0.0133 | 0.5686 | 1.4983 | 0.0064 | 1.4931 | 0.0066 | 1.4879 | 0.0066 |
| 0.6704 | 1.4959 | 0.0086 | 1.4919 | 0.0100 | 1.4885 | 0.0117 | 0.6684 | 1.5098 | 0.0059 | 1.5047 | 0.0062 | 1.4999 | 0.0065 |
| 0.7263 | 1.5044 | 0.0076 | 1.5006 | 0.0091 | 1.4961 | 0.0097 | 0.7801 | 1.5217 | 0.0044 | 1.5164 | 0.0045 | 1.5115 | 0.0046 |
| 0.8186 | 1.5179 | 0.0053 | 1.5137 | 0.0064 | 1.5087 | 0.0064 | 0.8958 | 1.5333 | 0.0022 | 1.5281 | 0.0023 | 1.5234 | 0.0025 |
| 0.9465 | 1.5360 | 0.0015 | 1.5313 | 0.0021 | 1.5263 | 0.0020 | 0.9346 | 1.5368 | 0.0010 | 1.5318 | 0.0013 | 1.5270 | 0.0015 |
| 1,3-Dichlorobenzene (1) +1,4-dioxane (2) | | | | | | | | | | | | | |
| 0.0625 | 1.4296 | 0.0023 | 1.4251 | 0.0024 | 1.4205 | 0.0026 | | | | | | | |
| 0.1172 | 1.4387 | 0.0046 | 1.4342 | 0.0048 | 1.4298 | 0.0051 | | | | | | | |
| 0.2076 | 1.4526 | 0.0073 | 1.4480 | 0.0074 | 1.4440 | 0.0082 | | | | | | | |
| 0.3032 | 1.4665 | 0.0093 | 1.4619 | 0.0095 | 1.4575 | 0.0099 | | | | | | | |
| 0.3953 | 1.4792 | 0.0106 | 1.4745 | 0.0108 | 1.4699 | 0.0109 | | | | | | | |
| 0.4460 | 1.4855 | 0.0106 | 1.4808 | 0.0108 | 1.4762 | 0.0110 | | | | | | | |
| 0.5372 | 1.4969 | 0.0107 | 1.4919 | 0.0107 | 1.4875 | 0.0111 | | | | | | | |
| 0.5793 | 1.5022 | 0.0108 | 1.4969 | 0.0105 | 1.4921 | 0.0105 | | | | | | | |
| 0.6232 | 1.5066 | 0.0097 | 1.5018 | 0.0099 | 1.4970 | 0.0100 | | | | | | | |
| 0.6676 | 1.5116 | 0.0092 | 1.5067 | 0.0094 | 1.5020 | 0.0095 | | | | | | | |
| 0.7240 | 1.5175 | 0.0081 | 1.5126 | 0.0083 | 1.5080 | 0.0085 | | | | | | | |
| 0.8076 | 1.5261 | 0.0063 | 1.5211 | 0.0065 | 1.5164 | 0.0067 | | | | | | | |
| 0.9081 | 1.5352 | 0.0030 | 1.5305 | 0.0035 | 1.5259 | 0.0038 | | | | | | | |

Standard uncertainties u: u(T) = 0.01 K; u(p) = 0.04 MPa; u(x) = 0.0001, and the standard uncertainty $U_c(n_D)$ = 0.0002.

TABLE-4
ADJUSTABLE PARAMETERS OF REDLICH-KISTER EQUATION ($A^{(n)}$) AND STANDARD DEVIATION (σ)

| Temp. (K) | $A^{(1)}$ | $A^{(2)}$ | $A^{(3)}$ | $A^{(4)}$ | σ | $A^{(1)}$ | $A^{(2)}$ | $A^{(3)}$ | $A^{(4)}$ | σ |
|---|---|---|---|---|---|---|---|---|---|---|
| | 2-Chlorotoluene (1) + *n*-hexane (2) | | | | | 2-Chlorotoluene (1) + cyclohexane (2) | | | | |
| 298.15 | 0.0298 | 0.0692 | 0.0139 | 0.1068 | 0.0000 | 0.0151 | -0.0067 | 0.0048 | -0.0072 | 0.0001 |
| 308.15 | 0.0371 | 0.0602 | 0.0114 | -0.1040 | 0.0000 | 0.0166 | -0.0111 | 0.0065 | -0.0001 | 0.0001 |
| 318.15 | 0.0462 | 0.0562 | 0.0041 | -0.0954 | 0.0000 | 0.0182 | -0.0112 | 0.0107 | -0.0049 | 0.0002 |
| | 2-Chlorotoluene (1) + 1,4-dioxane (2) | | | | | 4-Chlorotoluene (1) + *n*-hexane (2) | | | | |
| 298.15 | 0.0372 | -0.0102 | -0.0074 | 0.0165 | 0.0001 | 0.0937 | -0.0806 | -0.0648 | 0.1287 | 0.0000 |
| 308.15 | 0.0392 | -0.0130 | -0.0084 | 0.0188 | 0.0002 | 0.0974 | -0.0921 | -0.0598 | 0.1277 | 0.0007 |
| 318.15 | 0.0407 | -0.0102 | -0.0079 | 0.0096 | 0.0002 | 0.1010 | -0.1135 | -0.0780 | 0.1946 | 0.0000 |
| | 4-Chlorotoluene (1) + cyclohexane (2) | | | | | 4-Chlorotoluene (1) + 1,4-dioxane (2) | | | | |
| 298.15 | 0.0252 | 0.0002 | -0.0155 | 0.0171 | 0.0002 | 0.0317 | 0.0011 | -0.0011 | -0.0063 | 0.0001 |
| 308.15 | 0.0273 | 0.0016 | -0.0122 | 0.0162 | 0.0002 | 0.0325 | -0.0002 | 0.0005 | -0.0057 | 0.0001 |
| 318.15 | 0.0294 | -0.0023 | -0.0056 | 0.0267 | 0.0003 | 0.0333 | -0.0008 | 0.0017 | -0.0059 | 0.0001 |
| | 1,3-Dichlorobenzene (1) + *n*-hexane (2) | | | | | 1,3-Dichlorobenzene (1) + cyclohexane (2) | | | | |
| 298.15 | 0.0642 | -0.0688 | -0.0135 | 0.0701 | 0.0004 | 0.0257 | 0.0049 | 0.0026 | -0.0183 | 0.0001 |
| 308.15 | 0.0692 | -0.0680 | -0.0056 | 0.0670 | 0.0004 | 0.0269 | 0.0032 | 0.0023 | -0.0140 | 0.0002 |
| 318.15 | 0.0751 | -0.0661 | 0.0040 | 0.0291 | 0.0005 | 0.0275 | 0.0026 | 0.0063 | -0.0143 | 0.0002 |
| | 1,3-Dichlorobenzene (1) + 1,4-dioxane(2) | | | | | | | | | |
| 298.15 | 0.0436 | -0.0036 | -0.0043 | -0.0001 | 0.0002 | | | | | |
| 308.15 | 0.0444 | -0.0073 | 0.0045 | 0.0080 | 0.0001 | | | | | |
| 318.15 | 0.0447 | -0.0067 | 0.0036 | 0.0066 | 0.0002 | | | | | |

Weiner:

$$\frac{n^2 - n_1^2}{n^2 + 2n_2^2} = \left(\frac{n_2^2 - n_1^2}{n_2^2 + 2n_1^2}\right)\phi_2 \quad (8)$$

Newton:

$$n^2 - 1 = (n_1^2 - 1)\phi_1 + (n_2^2 - 1)\phi_2 \quad (9)$$

Eyring and John:

$$n = n_1\phi_1^2 + 2(n_1 n_2)^{1/2}\phi_1\phi_2 + n_2\phi_2^2 \quad (10)$$

In all above equations, $n$, $n_1$ and $n_2$ are the refractive indices of the binary mixture and its pure state values, respectively. The $\phi_1$ and $\phi_2$ used shows the volume fraction of component 1 and 2 in mixtures:

$$\phi_1 = \frac{x_1 V}{\sum x_i V_i} \quad \text{and} \quad \phi_2 = 1 - \phi_1 \quad (11)$$

The estimation capability of above mentioned mixing rules have been analyzed in terms of standard deviation depicted in Table-5. Figs. 1-3 show the deviation in refractive index data *versus* mole fraction of the first component in the mixtures formed. Light requires a medium to travel through. The medium is solely responsible for so many properties corresponding to the speed of light in any medium. If we talk about refractive index, which we all know is $n = c/v$ where 'c' represents speed of light in vacuum and 'v' represents the speed of light in any medium. From the proven facts, light travel at its actual speed or maximum speed only in the vacuum. Therefore, refractive index is always greater than one. Deviation in the refractive index is directly proportional to the density of any medium. Light gets scattered/absorbed/transmitted through particles present in any medium, which is responsible for its different speeds in different medium. If molecules in any medium are close to each other or any associative binding force act between them, the medium becomes dense, then the speed of light will be less in that medium due to obstruction caused by the binding of molecules. The values are positive for the present binary system as shown in Figs. 1-3. The $\Delta n$ values of the mixture may be due to two reasons (i) due to intermolecular bonding and (ii) due to molecular packing of different components. If the intermolecular interactions in binary components are stronger than in the pure species, the outcome will be revealed as reduction or contraction and *vice-versa*. The positive $\Delta n$ values in present binary system is due to the fact that the interactions among different molecules are less than between similar molecules. Cyclohexane is a non-polar hydrocarbon whereas chloro-substituted benzene derivatives are polar in nature. The addition of cyclohexane break/disrupt the orientation order and intermolecular interaction in chlorobenzene resulting the positive deviation. As n-π intermolecular interactions between 1,4-dioxane and chlorobenzene increase, due to the lone pair on chlorine and oxygen atom, the density of medium increases, which affects the refractive index and shows deviation. The order of deviation with cyclohexane is 1,3-dichlorobenzene > 4-chlorotoluene > 2-chlorotoluene as shown in Fig. 4. This is because two sites are available for interactions in 1,3-dichlorobenzene, whereas 1 site available for interactions in 2-chlorotoluene and 4-chlorotoluene. Cyclohexane can change its structure to the chair form to attain stability [34]. This order may also be due to the symmetrical effect of 1,3-dichlorobenzene and 4-chlorotoluene. The *n*-hexane molecule exists in a rigid zigzag shape [35], it can interact effectively along its length, which is most in 4-chlorotoluene, then in 1,3-dichlorobenzene and the least in 2-chlorotoluene. Therefore, it follows the order 4-chlorotoluene > 1,3-dichlorobenzene > 2-chlorotoluene as shown in Fig. 5. 1,4-Dioxane is a conformationally flexible molecule. It has a tendency to attain both boat or chair form to attain its stability according to the requirement of the system. It has two interactional sites at oxygen atom. The oxygen atom

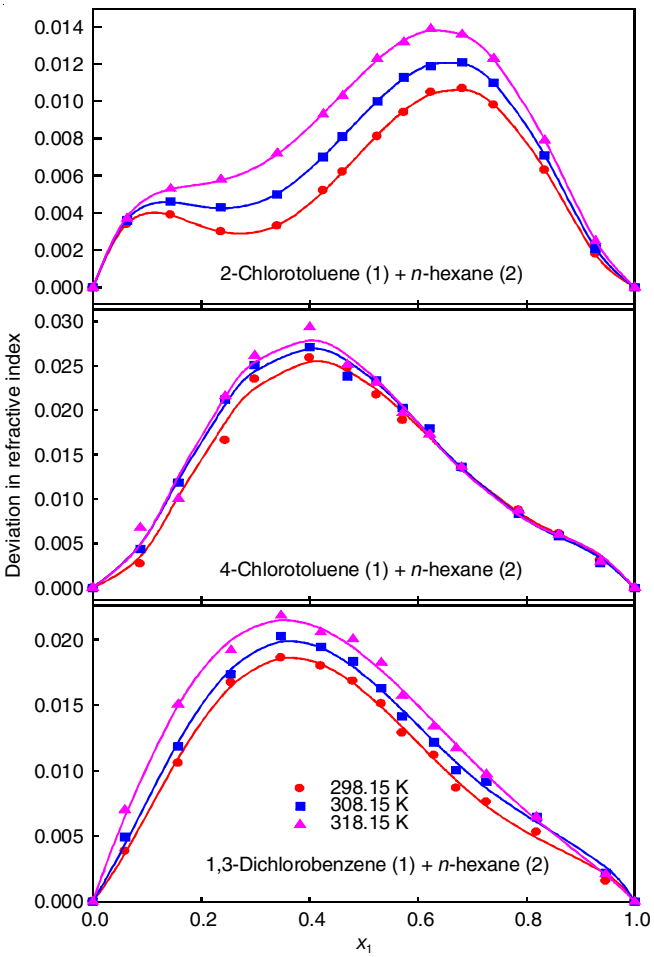


Fig. 1. Deviation in refractive index ($\Delta n$) for the binary mixture (2-chlorotoluene or 4-chlorotoluene or 1,3-dichlorobenzene (1) + *n*-hexane (2)) mixtures as a function of mole fraction ($x_1$) at 298.15-318.15 K. Symbols represent experimental values and lines represent values calculated from Redlich-Kister equation

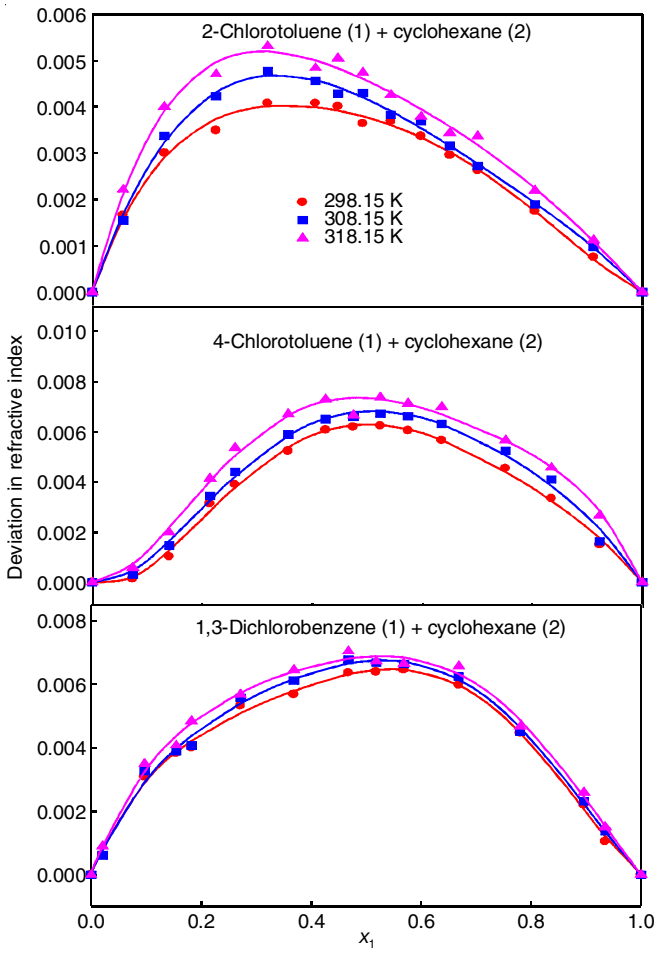


Fig. 2. Deviation in refractive index ($\Delta n$) for the binary mixture (2-chlorotoluene or 4-chlorotoluene or 1,3-dichlorobenzene (1) + cyclohexane (2)) mixtures as a function of mole fraction ($x_1$) at 298.15-318.15 K. Symbols represent experimental values and lines represent values calculated from Redlich-Kister equation

TABLE-5
STANDARD DEVIATION OF CALCULATED VALUES OF REFRACTIVE INDEX (*n*) USING VARIOUS CORRELATIONS AND MIXING RULES

| Binary system | Temp. (K) | AB | GD | LL | Weiner | Heller | Newton | Erying |
|---|---|---|---|---|---|---|---|---|
| 2-Chlorotoluene (1) + *n*-hexane (2) | 298.15 | 0.0018 | 0.0018 | 0.0023 | 0.0012 | 0.0021 | 0.0013 | 0.0020 |
| | 308.15 | 0.0112 | 0.0112 | 0.0128 | 0.0091 | 0.0122 | 0.0098 | 0.0120 |
| | 318.15 | 0.0129 | 0.0129 | 0.0145 | 0.0097 | 0.0139 | 0.0114 | 0.0137 |
| 2-Chlorotoluene (1) + Cyclohexane (2) | 298.15 | 0.0020 | 0.0020 | 0.0027 | 0.0026 | 0.0026 | 0.0013 | 0.0023 |
| | 308.15 | 0.0023 | 0.0023 | 0.0030 | 0.0029 | 0.0029 | 0.0016 | 0.0026 |
| | 318.15 | 0.0028 | 0.0028 | 0.0036 | 0.0038 | 0.0034 | 0.0022 | 0.0032 |
| 2-Chlorotoluene (1) + 1,4-dioxane (2) | 298.15 | 0.0009 | 0.0009 | 0.0017 | 0.0055 | 0.0014 | 0.0004 | 0.0013 |
| | 308.15 | 0.0013 | 0.0013 | 0.0021 | 0.0052 | 0.0018 | 0.0007 | 0.0017 |
| | 318.15 | 0.0015 | 0.0015 | 0.0023 | 0.0050 | 0.0020 | 0.0008 | 0.0018 |
| 4-Chlorotoluene (1) + *n*-hexane (2) | 298.15 | 0.0214 | 0.0214 | 0.0230 | 0.0146 | 0.0225 | 0.0199 | 0.0222 |
| | 308.15 | 0.0226 | 0.0226 | 0.0241 | 0.0152 | 0.0237 | 0.0210 | 0.0233 |
| | 318.15 | 0.0103 | 0.0103 | 0.0118 | 0.0087 | 0.0113 | 0.0089 | 0.0111 |
| 4-Chlorotoluene (1) + cyclohexane (2) | 298.15 | 0.0070 | 0.0070 | 0.0077 | 0.0050 | 0.0074 | 0.0063 | 0.0073 |
| | 308.15 | 0.0038 | 0.0038 | 0.0045 | 0.0043 | 0.0042 | 0.0031 | 0.0041 |
| | 318.15 | 0.0044 | 0.0044 | 0.0051 | 0.0043 | 0.0049 | 0.0038 | 0.0047 |
| 4-Chlorotoluene (1) + 1,4-dioxane (2) | 298.15 | 0.0004 | 0.0004 | 0.0007 | 0.0056 | 0.0005 | 0.0010 | 0.0004 |
| | 308.15 | 0.0003 | 0.0003 | 0.0009 | 0.0053 | 0.0007 | 0.0007 | 0.0006 |
| | 318.15 | 0.0005 | 0.0005 | 0.0012 | 0.0051 | 0.0010 | 0.0004 | 0.0008 |
| 1,3-Dichlorobenzene (1) + *n*-hexane (2) | 298.15 | 0.0186 | 0.0186 | 0.0209 | 0.0119 | 0.0201 | 0.0165 | 0.0197 |
| | 308.15 | 0.0201 | 0.0201 | 0.0223 | 0.0125 | 0.0216 | 0.0179 | 0.0211 |
| | 318.15 | 0.0219 | 0.0219 | 0.0241 | 0.0132 | 0.0234 | 0.0197 | 0.0230 |

| | | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| 1,3-Dichlorobenzene (1) + cyclohexane (2) | 298.15 | 0.0040 | 0.0040 | 0.0050 | 0.0067 | 0.0048 | 0.0031 | 0.0045 |
| | 308.15 | 0.0043 | 0.0043 | 0.0053 | 0.0066 | 0.0051 | 0.0034 | 0.0048 |
| | 318.15 | 0.0046 | 0.0046 | 0.0057 | 0.0066 | 0.0054 | 0.0037 | 0.0051 |
| 1,3-Dichlorobenzene (1) + 1,4-dioxane (2) | 298.15 | 0.0016 | 0.0016 | 0.0028 | 0.0075 | 0.0023 | 0.0006 | 0.0021 |
| | 308.15 | 0.0018 | 0.0018 | 0.0030 | 0.0073 | 0.0025 | 0.0008 | 0.0023 |
| | 318.15 | 0.0021 | 0.0021 | 0.0033 | 0.0070 | 0.0028 | 0.0011 | 0.0026 |

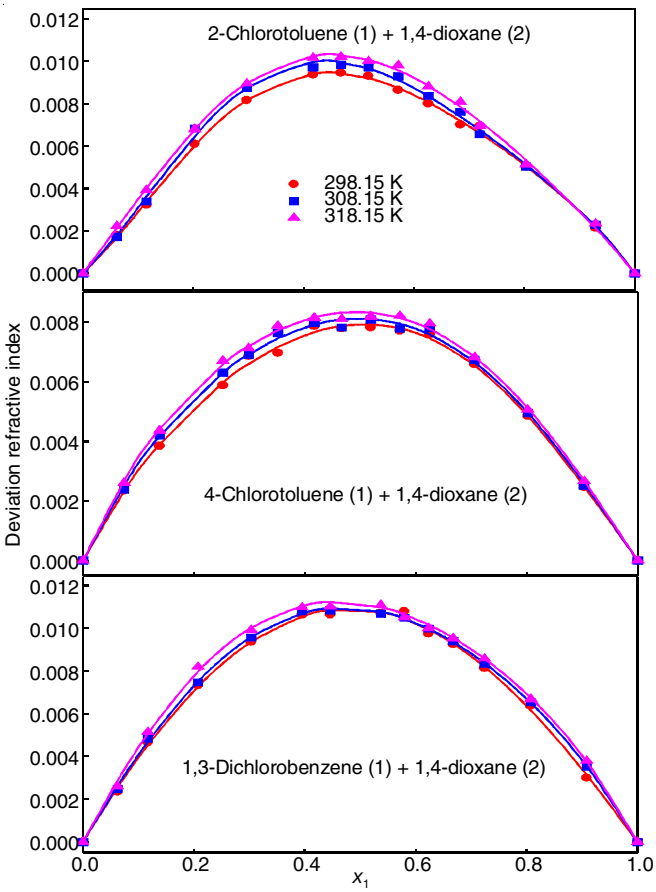


Fig. 3. Deviation in refractive index ($\Delta n$) for the binary mixture (2-chlorotoluene or 4-chlorotoluene or 1,3-dichlorobenzene (1) + 1,4-dioxane (2)) mixtures as a function of mole fraction ($x_1$) at 298.15-318.15 K. Symbols represent experimental values and lines represent values calculated from Redlich-Kister equation

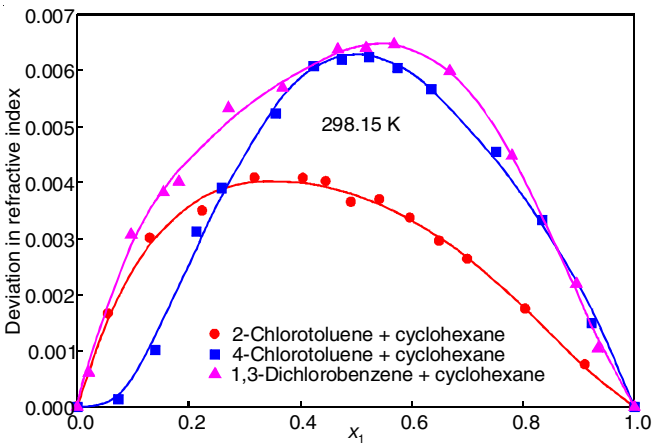


Fig. 4. Deviation in refractive index ($\Delta n$) for the binary mixture (2-chlorotoluene or 4-chlorotoluene or 1,3-dichlorobenzene (1) + cyclohexane (2)) mixtures as a function of mole fraction ($x_1$) at 298.15 K. Symbols represent experimental values and lines represent values calculated from Redlich-Kister equation

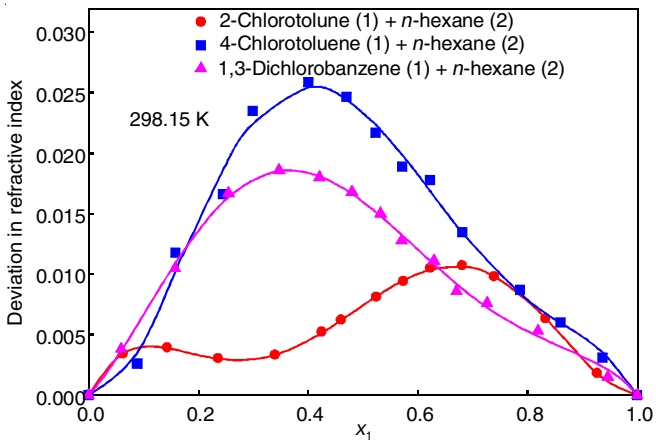


Fig. 5. Deviation in refractive index ($\Delta n$) for the binary mixture (2-chlorotoluene or 4-chlorotoluene or 1,3-dichlorobenzene (1) + *n*-hexane (2)) mixtures as a function of mole fraction ($x_1$) at 298.15 K. Symbols represent experimental values and lines represent values calculated from Redlich-Kister equation

is more electronegative than chlorine atom, can interact easily with 2 chlorine atoms in 1,3-dichlorobenzene then with 2-chlorotoluene and then with 4-chlorotoluene. Here, the polarity factor dominates over symmetrical factor, due to most electronegative oxygen atom present in dioxane molecule. Hereby follows the order 1,3-dichlorobenzene > 2-chlorotoluene > 4-chlorotoluene as shown in Fig. 6.

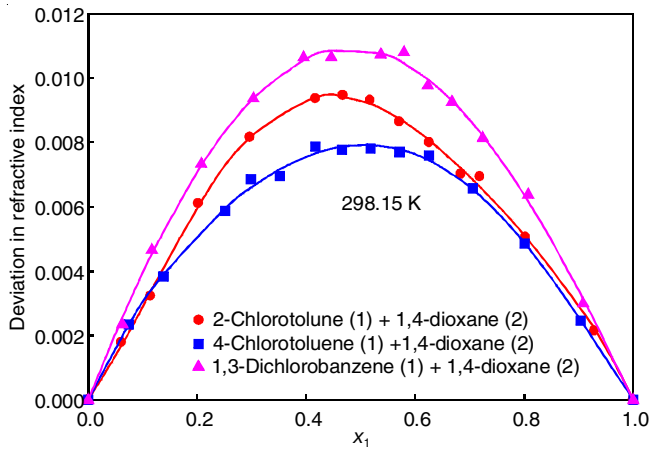


Fig. 6. Deviation in refractive index ($\Delta n$) for the binary mixture (2-chlorotoluene or 4-chlorotoluene or 1,3-dichlorobenzene (1) + 1,4-dioxane (2)) mixtures as a function of mole fraction ($x_1$) at 298.15 K. Symbols represent experimental values and lines represent values calculated from Redlich-Kister equation

## Conclusion

In this work, the refractive indices ($n$) of haloarenes, hydrocarbons, ether, alcohol and respective possible mixtures were experimentally determined over the whole compositions at T

= (298.15-318.15) K. The mixtures selected were 2-chlorotoluene or 4-chlorotoluene or 1,3-dichlorobenzene (1) + *n*-hexane or cyclohexane or 1,4-dioxane with its possible combinations. The $\Delta n$ values were calculated using experimental *n* values. The $\Delta n$ is positive for all binary mixtures at all investigated compositions. The positive values in current binary systems are due to the fact that the interactions between different molecules are less than between similar molecules. Different rules of mixing like Lorentz-Lorentz, Erying-John, Arago-Biot, *etc*. were also used to predict *n* values. Then $\Delta n$ values were discussed in terms of ongoing intermolecular interactions among the components of the selected mixtures.

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## CONFLICT OF INTEREST

The authors declare that there is no conflict of interests regarding the publication of this article.

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