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Viscometric and refractive index study of intermolecular interaction between binary mixtures of terpinolene with *o*-, *m*- and *p*-cresol at 303.15, 308.15 and 313.15 K

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Viscosities and refractive indexes (n_D) were experimentally measured for binary mixtures of terpinolene with o-, m- and p-cresol for entire composition range at 303.15, 308.15 and 313.15 K and at atmospheric pressure. Deviation in viscosity ($\Delta\eta$), excess Gibbs energy of activation for viscous flow (ΔG^{*E}), enthalpy (ΔH^{*}) and entropy (ΔS^{*}) of viscous flow, deviation in refractive index (Δn_D), molar refraction (R_m) and deviation in molar refraction (ΔR_m) were also calculated. Various theoretical viscosity relations such as Dolezalek-Schulze (D_{12}), Grunberg-Nissan (G_{12}), Tamura-Kurata (T_{12}), Katti-Chaudhri (W_{vis}/RT), McAllister relation (M_{12} and M_{21}), Kendall-Munroe relation, Bingham relation and Arrhenius-Eyring relation were calculated. Similarly, nine theoretical refractive index relations such as Arago-Biot (A-B), Dale-Glastone (D-G), Lorentz-Lorentz (L-L), Eykman (Eyk), Weiner (WR), Heller (Hr), Newton (Nw), Oster (Os) and Eyring-John (E-J) were also calculated with their standard deviation (σ) values. Results were discussed in terms of the presence of intermolecular interactions between the components of binary mixtures.

Keywords: Viscosity, Refractive index, Theoretical models, Intermolecular interaction, Deviation Properties.

The study of deviation properties of viscosity and refractive index gives us an idea about the presence of intermolecular interactions between these components of mixtures. We can understand the interaction patterns of binary mixtures at different temperatures by studying these properties in terms of using theoretical models. The data from this study can help us choose an appropriate solvent and its concentration for various processes, such as organic synthesis, ion extraction systems, mass transfer phenomena, solvent extraction, separations, and many analytical techniques like HPLC and HPTLC, *etc.*[1]

Terpinolene is a mono terpene hydrocarbon compound that is found as a major constituent component in many essential oils obtained from different herbal plants species[2-4]. In our earlier study, we have examined the intermolecular interactions behavior of terpinolene with halobenzenes[5]. In this present study, we focus on the possibility of formation of new H-bonding and electrostatic type interaction between terpinolene with cresols. Terpinolene molecules have a tendency of electron accepting, while cresol molecules have lone pair electrons on oxygen atoms of the –OH function group, which has an electron donating tendency[6].

Experimental Section

Materials

The chemicals used in this study were of AR grade purity and were purified using standard methods[4,7]. Some other details, such as suppliers, purification method, CAS number and final purity analysis method, are given in Table 1. A comparison of experimental and literature values of viscosities and refractive indices of pure components is given in Table 2. The values are found to be in close proximity.

Apparatus and Procedure

Sample Preparation

All the binary mixtures were prepared by mixing the appropriate masses of components using an electronic balance (Reptech RA-2012) having a precision of ± 0.0001g. All the mixtures were prepared in glass stopper flasks to avoid contamination and losses of solvent due to evaporation, and they were kept in a dark place to avoid any photolytic effect on them. The estimated uncertainty in the mole fraction was ± 0.0001.

Density measurement

Densities of pure components and binary mixtures were measured using Anton Paar's density

Table 1 — List of Chemicals with Details of Supplier, CAS Number, Purity, Purification Method and Applied Method for Final Purity Analysis

Compound/ Grade	Supplier	CAS number	Initial mass-fraction purity	Purification method	Final mass-fraction purity	Analysis method
Terpineol	Sigma-Aldrich Chemical, USA	586-62-9	90%	None	-	-
o-Cresol	S.D.Fine Chemicals Ltd.	95-48-7	99%	Fractional distillation	99.5%	GC[a]
m-Cresol	S.D.Fine Chemicals Ltd.	108-39-4	99%	Fractional distillation	99.5%	GC[a]
p-Cresol	S.D.Fine Chemicals Ltd.	106-44-5	99.%	Fractional distillation	99.5%	GC[a]

[a]GC = Gas-liquid chromatography

Table 2 — Experimental and Literature Values of Viscosity (η) and Refractive index (n_D)of pure components at 303.15, 308.15 and 313.15K.

Pure compound	T/K	η /(mPa·s)		n_D	
		Exp.	Lit.	Exp.	Lit.
Terpinolene	303.15	1.007	--	1.4860	--
	308.15	0.945	--	1.4843	--
	313.15	0.884	--	1.4821	--
o-Cresol	303.15	5.935	--	1.5412	1.5410[39]
	308.15	4.849	--	1.5390	1.5386[40] 1.5370[39]
	313.15	4.032	4.243[41]	1.5364	--
m-Cresol	303.15	9.922	9.806[42]	1.5357	1.5350[39]
	308.15	7.824	7.701[44]	1.5342	1.5320[39]
	313.15	6.349	6.252[41] 6.120[44]	1.5319	--
p-Cresol	303.15	10.927	--	1.5353	1.5340[39]
	308.15	8.646	8.444[43]	1.5338	1.5310[39]
	313.15	6.949	6.661[41] 6.745[43]	1.5305	--

Standard uncertainties u, $u(T)$ = ±0.01 K, $u(\eta)$ = ±0.001 mPa·s and $u(n_D)$ = ±0.0001. All physical quantities are measured at atmospheric pressure.

measurement instrument (Model: 5000M) with an accuracy of ± 0.000005 g cm^{-3}. The temperature was controlled by an in–built temperature-maintenance system having an accuracy of ± 0.001°C. The calibration of this instrument was carried out using highly pure deionized water provided by its manufacturer, Anton Paar.

Viscosity measurement

The viscosity values for pure components and their binary mixtures were measured using an Ubbelohde suspended level viscometer (Agarwal Scientific Glass Company, Mumbai, India). For calibration purposes, the A and B constants of the viscometer were calculated by measuring the flow times of double distilled water at 303.15, 308.15, and 313.15 K. During the measurement of viscosity, the temperature was controlled by a thermostatic water bath (Model No.: 14L-SS, Equiptron Water bath Company, India) having an accuracy of ± 0.01°C. The estimated precision in the viscosity value was ± 0.001 mPa s.

Refractive index measurement

The refractive index values for pure components and their binary mixtures were measured by a thermostated Abbe's refractometer (SER No. 995033). The refractive index of 1-bromonaphthalene, methanol, and double distilled water were used to calibrate the Refractometer. During the measurement of refractive index, the temperature was controlled by circulating water using a pump with a thermostatic water bath (Model No. 14L-SS, Equiptron Water Bath Company, India) with an accuracy of ± 0.01°C. The estimated precision measurement of refractive index was ± 0.0001.

Results and Discussion

Viscosity (η), Deviation in Viscosity (Δη) and Excess Gibbs' Free Energy of Activation of Viscous Flow (ΔG^{*E})

The values of experimentally measured viscosities (η) of pure components and their binary mixtures, the calculated values of deviation in viscosities ($\Delta\eta$) and excess Gibbs' free energy of activation of viscous

Table 3 — Viscosity (η), Deviation in Viscosity ($\Delta\eta$), Excess Free Energy of Activation of Viscous Flow (ΔG^{*E}), Enthalpy (ΔH^{*E}) and Entropy (ΔS^{*E}) of activation of viscous flow *vs*. Mole Fraction (x_1)Terpinolene (1) + *o*-, *m*- and *p*-Cresol (2) at 303.15, 308.15 and 313.15K.

x_1	ρ (g·cm^3)			η (mPa·s)			$\Delta\eta$ (mPa·s)			ΔG^{*E} (J·mol^{-1})			ΔH^* (kJ·mol^{-1})	ΔS^* (J·K^{-1}·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	303.15 K	308.15 K	313.15 K		
Terpinolene (1) + *o*-Cresol (2)														
0.0000	1.037029	1.032655	1.028261	5.935	4.849	4.032	0.000	0.000	0.000	0.00	0.00	0.00	29.85	-134.91
0.0677	1.019331	1.014881	1.010434	5.538	4.269	3.478	-0.063	-0.316	-0.341	142.86	-27.32	-101.79	36.05	-114.10
0.1404	1.001488	0.997051	0.992583	5.043	3.724	2.951	-0.200	-0.577	-0.639	245.02	-59.56	-228.62	41.63	-95.20
0.2188	0.983525	0.979095	0.974682	4.524	3.251	2.493	-0.333	-0.744	-0.850	332.14	-68.04	-346.58	46.34	-79.07
0.3035	0.965436	0.961057	0.956698	4.044	2.830	2.088	-0.396	-0.834	-0.988	435.66	-60.27	-464.88	51.45	-61.63
0.3953	0.947226	0.942921	0.938634	3.591	2.447	1.748	-0.397	-0.859	-1.040	551.37	-43.44	-560.98	56.13	-45.57
0.4950	0.928884	0.924644	0.920409	3.210	2.104	1.474	-0.286	-0.812	-1.000	715.74	-10.91	-609.50	60.73	-29.77
0.6040	0.910381	0.906192	0.902005	2.809	1.819	1.278	-0.151	-0.672	-0.853	862.11	68.41	-555.02	61.47	-26.60
0.7233	0.891733	0.887586	0.883458	2.380	1.565	1.134	0.009	-0.460	-0.621	968.85	173.96	-404.18	57.83	-37.60
0.8547	0.872911	0.868847	0.864801	1.860	1.279	0.996	0.137	-0.233	-0.345	919.96	191.56	-237.74	48.60	-66.35
1.0000	0.853890	0.849991	0.846075	1.007	0.945	0.884	0.000	0.000	0.000	0.00	0.00	0.00	9.59	-190.54
Terpinolene (1) + *m*-Cresol (2)														
0.0000	1.026135	1.022187	1.018215	9.922	7.824	6.349	0.000	0.000	0.000	0.00	0.00	0.00	34.64	-123.45
0.0684	1.009415	1.005379	1.001334	8.811	6.637	5.199	-0.501	-0.717	-0.776	109.22	-36.49	-153.71	41.01	-101.71
0.1417	0.992572	0.988521	0.984458	7.713	5.586	4.233	-0.945	-1.263	-1.341	209.17	-68.06	-299.27	46.72	-82.06
0.2206	0.975649	0.971601	0.967515	6.648	4.694	3.418	-1.307	-1.613	-1.725	299.85	-76.11	-440.39	51.85	-64.21
0.3057	0.958614	0.954564	0.950497	5.674	3.921	2.732	-1.522	-1.800	-1.946	398.84	-68.19	-578.77	57.01	-46.24
0.3978	0.941462	0.937442	0.933392	4.788	3.244	2.173	-1.587	-1.844	-2.002	506.14	-50.94	-698.38	61.68	-29.77
0.4977	0.924217	0.920196	0.916159	4.067	2.660	1.732	-1.418	-1.740	-1.897	671.65	-17.28	-774.36	66.67	-12.30
0.6065	0.906817	0.902814	0.898796	3.366	2.182	1.420	-1.149	-1.470	-1.614	818.14	60.76	-737.40	67.42	-8.61
0.7254	0.889317	0.885328	0.881323	2.671	1.775	1.194	-0.784	-1.058	-1.190	912.54	167.54	-587.05	62.84	-22.16
0.8560	0.871671	0.867702	0.863726	1.944	1.364	1.015	-0.347	-0.571	-0.656	848.91	185.17	-355.14	50.57	-60.33
1.0000	0.853890	0.849991	0.846075	1.007	0.945	0.884	0.000	0.000	0.000	0.00	0.00	0.00	9.59	-190.54
Terpinolene (1) + *p*-Cresol (2)														
0.0000	1.025991	1.022115	1.018204	10.927	8.646	6.949	0.000	0.000	0.000	0.00	0.00	0.00	35.13	-122.63
0.0684	1.009142	1.005191	1.001249	9.533	7.256	5.571	-0.716	-0.864	-0.963	81.49	-46.19	-192.83	41.78	-99.86
0.1417	0.992253	0.988257	0.984315	8.236	6.062	4.445	-1.285	-1.492	-1.644	166.40	-77.42	-373.57	48.04	-78.31
0.2207	0.975286	0.971270	0.967318	7.052	5.055	3.555	-1.686	-1.892	-2.055	259.62	-84.65	-520.64	53.40	-59.68
0.3058	0.958238	0.954204	0.950258	5.963	4.191	2.818	-1.932	-2.100	-2.276	355.72	-74.14	-660.54	58.46	-41.93
0.3978	0.941094	0.937042	0.933118	5.002	3.437	2.231	-1.979	-2.145	-2.305	470.70	-55.37	-769.60	63.01	-25.81
0.4977	0.923846	0.919783	0.915881	4.189	2.790	1.766	-1.801	-2.023	-2.164	624.44	-22.55	-841.62	67.46	-10.00
0.6065	0.906467	0.902394	0.898519	3.426	2.261	1.426	-1.484	-1.714	-1.844	768.45	52.74	-817.87	68.48	-5.34
0.7255	0.889001	0.884912	0.881054	2.693	1.820	1.178	-1.038	-1.239	-1.371	866.89	162.97	-685.68	64.51	-16.78
0.8560	0.871448	0.867369	0.863548	1.918	1.381	0.995	-0.518	-0.672	-0.762	780.83	181.24	-439.20	51.03	-58.77
1.0000	0.853890	0.849851	0.846075	1.007	0.944	0.884	0.000	0.000	0.000	0.00	0.00	0.00	9.59	-190.54

Standard uncertainties u, $u(T)$= ±0.01 K, $u(x_1)$ = ±0.0001, and, $u(\eta)$ = ±0.001 mPa·s. Further fitting coefficients of $\Delta\eta$ and ΔG^{*E} with standard error are given in Tables S1 and S2. All physical quantities are measured at atmospheric pressure.

flow (ΔG^{*E}) for all binary mixtures for all studied temperatures are given in Table 3. The graphical representation of $\Delta\eta$ and ΔG^{*E} *vs* mole fraction (x_1) is shown as Fig. 1 and Fig. 2, respectively. The values of the fitting coefficients (A_0, A_1, A_2, A_3) of the Redlich-Kister polynomial equation for $\Delta\eta$ and ΔG^{*E} are listed in Tables S1 and S2 of supplementary data, respectively. The deviation in viscosity ($\Delta\eta$) and excess Gibbs' free energy for activation of viscous flow (ΔG^{*E}) were calculated using the following relations:

$$\Delta\eta\ (mPa \cdot s) = \eta_{12} - (x_1\eta_1 + x_2\eta_2) \qquad \ldots (1)$$

$$\Delta G^{*E}\ (J \cdot mol^{-1}) = RT\,\{\ln(\eta V) - x_1 \ln(\eta_1 V_1) - x_2 \ln(\eta_2 V_2)\} \qquad \ldots (2)$$

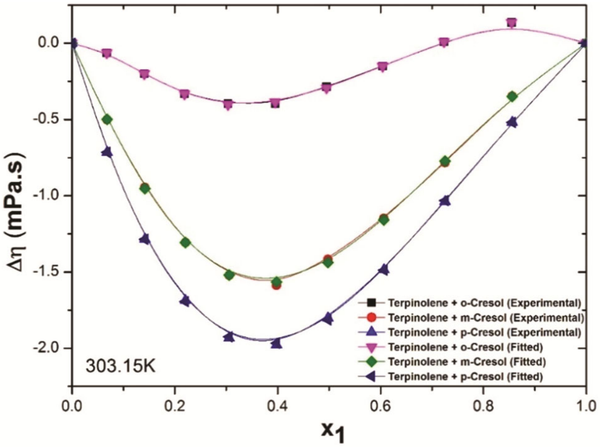

Fig. 1 — Deviation in viscosity ($\Delta\eta$) for the system Terpinolene (1) + *o*-, *m*-, and *p*-cresol (2) as a function of mole fraction (x_1) at *T* = 303.15K

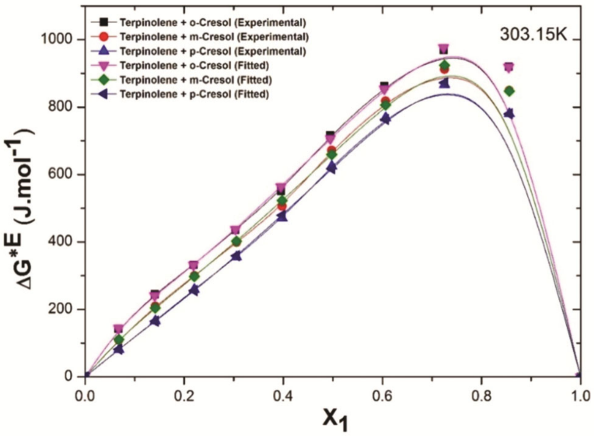

Fig. 2 — The excess Gibb's energy of activation for viscous flow (ΔG^{*E}) for the system Terpinolene (1) + *o*-, *m*-, and *p*-cresol (2) as a function of mole fraction (x_1) at *T* = 303.15K

where, V_1, V_2 and V represent the molar volume of component 1, 2, and mixture, respectively; η_1, η_2, and η represent the viscosity of component 1, 2, and mixture, respectively; and R and T have their usual meaning.

Some observations made from Table III are as follows:

1. All the binary mixtures show the negative values of $\Delta\eta$.
2. The negative values of $\Delta\eta$ increase with an increase in temperature.
3. The ΔG^{*E} values shift from positive values to more negative values with an increase in temperature.
4. Going from an *o*-cresol to a *p*-cresol binary mixture, the negative values of $\Delta\eta$ become more negative, and ΔG^{*E} values become less positive or more negative in some cases.

Generally, the positive values of $\Delta\eta$ and ΔG^{*E} indicate the presence of strong intermolecular interactions between the components of binary mixtures[8,9]. But the difference in viscosity values of pure components is large. So negative $\Delta\eta$ values are expected even for the presence of strong interactions instead of positive $\Delta\eta$ value[10,11]. The enthalpy (ΔH^*) and entropy (ΔS^*) of viscous flow were also calculated using the absolute reaction rate theory of Eyring *et al.*[12], Ali *et al.*[13] and Trabelsi *et al.*[14]. The following is the relationship between kinematic viscosity and the free energy of activation of viscous flow.

$$\Delta G^* = RT \ln\left(\frac{\eta V}{h\, N_A}\right) \quad \dots (3)$$

$$\Delta G^* = \Delta H^* - T\Delta S^* \quad \dots (4)$$

$$V = [x_1 M_1 + x_2 M_2]/\rho \quad \dots (5)$$

where, V, h, N_A and ρ represents molar volume, Plank's constant, Avogadro's number, and the density of the mixture, respectively.

The values of ΔH^* and ΔS^* were calculated by [assuming that activation parameters ΔH^* and $\Delta S^*$are temperature independent] plotting a graph between $R \ln\left(\frac{\eta V}{h\, N_A}\right)$ against $\frac{1}{T}$ using both graphical and least square methods[14]. The values of ΔH^* and ΔS^* of activation of viscous flow are given in Table 3.

A perusal of Table 3 shows that the *o*-cresol molecules show the highest intermolecular interactions with the terpinolene molecule and have the lowest negative $\Delta\eta$ value. Less negative $\Delta\eta$ values reveal that interactions between unlike molecules contribute more positively to this magnitude. These interactions decrease at higher temperatures. Similarly, the positive values of ΔG^{*E} also support the above discussion. When cresol molecules are added to terpinolene molecules, new electrostatic interactions or H-bonding is formed between the components of binary mixtures. This leads to a closer orientation of molecules and an increase in viscosity and Gibbs free energy. The order of interaction is as follows:

o-cresol > *m*-cresol > *p*-cresol

Viscosity Interaction Parameters and Models

The values of viscosity interaction parameters and models such as Bingham, Arrhenius, Kendall-Munroe, Grunberg-Nissan (G_{12}), Dolezalek-Schulze (D_{12}), Tamura-Kurata (T_{12}), Katti-Chaudhri

(W_{vis}/RT), and 3-body McAllister (M_{12} and M_{21}) with standard percentage deviation (σ %) are given in Table 4. The graphical representation of experimental and theoretically calculated viscosities values *vs*. mole fraction (x_1) for three binary mixtures at 303.15 K is shown in Figures S1-S3 of the supplementary data. Their standard equations for various models are given as follows:

I. Dolezalek and Schulze[15]

$$\eta = x_1^2\eta_1 + x_2^2\eta_2 + 2x_1x_2D_{12} \qquad \text{... (6)}$$

II. Grunberg and Nissan[16]

$$\ln\eta = x_1\ln\eta_1 + x_2\ln\eta_2 + x_1x_2G_{12} \qquad \text{... (7)}$$

III. Tamura and Kurata[17]

$$\eta = x_1\phi_1\eta_1 + x_2\phi_2\eta_2 + 2(x_1x_2\phi_1\phi_2)^{0.5}T_{12} \qquad \text{... (8)}$$

IV. Katti and Chaudhri[18]

$$\ln(\eta V) = x_1\ln(\eta_1V_1) + x_2\ln(\eta_2V_2) + x_1x_2\left(\frac{W_{vis}}{RT}\right) \text{... (9)}$$

V. The McAllister three-body interaction model[19]

$$\ln v_{12} = x_1^3\ln v_1 + 3x_1^2x_2\ln M_{12} + 3x_1x_2^2\ln M_{21} + x_2^3\ln v_2 - ln\left(x_1 + \left(\frac{x_2M_2}{M_1}\right)\right) + 3x_1^2x_2\ln\left(\left(\frac{2}{3}\right) + \left(\frac{M_2}{3M_1}\right)\right) + 3x_1x_2^2\ln\left(\left(\frac{1}{3}\right) + \left(\frac{2M_2}{3M_1}\right)\right) + x_2^3\ln\left(\frac{M_2}{M_1}\right) \text{... (10)}$$

VI. Kendall-Munroe relation[20,21]

$$\ln\eta = \textstyle\sum_{i=1}^{2} x_i\, ln\,\eta_i \qquad \text{... (11)}$$

VII. Bingham relation[22]

$$\eta = \textstyle\sum_{i=1}^{2} x_i\,\eta_i \qquad \text{...(12)}$$

VIII. Arrhenius-Eyring relation[20]

$$ln\,\eta V = \textstyle\sum_{i=1}^{2} x_i\ln\eta_i V_i \qquad \text{... (13)}$$

Table 4 — Adjustable Parameters for the Correlation of Mixture Viscosities for Terpinolene (1) + *o*-, *m*- and *p*-Cresol (2) at 303.15, 308.15 and 313.15K.

Parameters	*T* (K)			σ(%)		
	303.15	308.15	313.15	303.15	308.15	313.15
Terpinolene (1) + *o*-Cresol (2)						
Bingham	-	-	-	0.672	2.640	4.558
Arrhenius	-	-	-	2.076	0.353	1.702
Kendall-Munroe	-	-	-	1.972	0.373	1.888
Grunberg-Nissan (G_{12})	1.154	-0.050	-0.939	0.904	0.357	0.130
Dolezalek-Schulze (D_{12})	2.930	1.229	0.422	0.181	0.713	0.874
Tamura-Kurata (T_{12})	3.510	1.688	0.794	0.561	0.483	0.490
Katti-Chaudhri (W_{vis}/RT)	1.237	0.036	-0.854	0.898	0.351	0.131
McAllister (M_{12})	1.542	0.764	0.188	4.545	4.908	7.067
McAllister (M_{21})	0.660	0.347	0.068			
Terpinolene (1) + *m*-Cresol (2)						
Bingham	-	-	-	2.439	4.612	7.508
Arrhenius	-	-	-	1.952	0.353	2.284
Kendall-Munroe	-	-	-	1.850	0.381	2.472
Grunberg-Nissan (G_{12})	1.067	-0.062	-1.193	0.871	0.359	0.133
Dolezalek-Schulze (D_{12})	2.539	0.745	-0.350	0.252	1.178	1.677
Tamura-Kurata (T_{12})	3.544	1.509	0.251	0.608	0.756	1.021
Katti-Chaudhri (W_{vis}/RT)	1.145	0.018	-1.113	0.864	0.353	0.139
McAllister (M_{12})	1.674	0.925	0.210	4.491	4.366	5.747
McAllister (M_{21})	0.988	0.663	0.332			
Terpinolene (1) + *p*-Cresol (2)						
Bingham	-	-	-	3.070	5.153	8.565
Arrhenius	-	-	-	1.827	0.356	2.621
Kendall-Munroe	-	-		1.723	0.394	2.818
Grunberg-Nissan (G_{12})	0.977	-0.075	-1.344	0.843	0.363	0.104
Dolezalek-Schulze (D_{12})	2.198	0.543	-0.695	0.293	1.336	2.043
Tamura-Kurata (T_{12})	3.307	1.392	-0.038	0.676	0.858	1.290
Katti-Chaudhri (W_{vis}/RT)	1.056	0.006	-1.261	0.836	0.357	0.111
McAllister (M_{12})	1.670	0.957	0.186	4.482	4.316	5.798
McAllister (M_{21})	1.029	0.725	0.346			

where, η, V represents the viscosity, molar volume of binary mixture. η_i, V_i, x_i, ϕ_i, M_i represents the viscosity, molar volume, mole fraction, volume fraction, and molar mass of i^{th} component, respectively.

Some observations made from the values of Table 4 are as follows:

1. The Dolezalek-Schulze (D_{12}) relation shows the lowest; and the 3-body McAllister relation shows the highest σ % value for all binary mixtures at 303.15K.
2. For 308.15K, the Katti-Chaudhri (W_{vis}/RT) relation for *o*-cresol binary mixtures, and Arrhenius and Katti-Chaudhri (W_{vis}/RT) for *m*-cresol and *p*-cresol binarymixtures, shows the lowest σ % value.
3. At 313.15K, the Grunberg-Nissan (G_{12}) relation shows the lowest values, and the McAllister relation for *o*-cresol binaries, the Bingham relation for *m*-cresol binaries and the *p*-cresol binaries show the highest σ % values.

According to the survey of literature data, the positive values of interaction parameters G_{12}, D_{12} and T_{12} support the presence of strong intermolecular interaction between unlike molecules[23,24]. The positive values of G_{12}, D_{12} and T_{12} become less positive, and in some cases, they become negative, which shows the weakening of intermolecular interactions at higher temperatures.

Refractive index

Deviation in refractive index $(\Delta \mathbf{n_D})$ and deviation in molar refraction $(\Delta \mathbf{R_m})$

Experimentally measured values of refractive index (n_D) and calculated values of deviation in refractive index (Δn_D) and deviation in molar refraction (ΔR_m) for all binary mixtures at 303.15, 308.15, and 313.15K are listed in Tables 5 and Table 6. The graphical representation of Δn_D and (ΔR_m) *vs.* volume fraction (ϕ_1) is shown in Fig. 3 and Fig. 4. The values of the fitting coefficients (A_0, A_1, A_2, A_3) of the Redlich-Kister polynomial equation for Δn_D are listed in Tables S3 and S4 of the supplementary data. The deviation in refractive index (Δn_D) and the deviation in molar refraction (ΔR_m) were calculated using the following equations:

$$\Delta n_D = n_{D_{exp}} - (n_{D_1}\phi_1 + n_{D_2}\phi_2) \qquad \ldots (14)$$

$$\Delta R_m = R_{m_{exp}} - (R_{m_1}\phi_1 + R_{m_2}\phi_2) \qquad \ldots (15)$$

where, $\phi_1, \phi_2; n_{D_1}, n_{D_2}$ and $R_{m_1} R_{m_2}$ indicate the volume fraction, refractive index, and molar refraction of components 1 and 2, respectively. The values of ϕ_i, R_{m_i} and V_i were calculated using the following equations:

$$\phi_i = \frac{x_i V_i}{\sum_{i=1}^{2} x_i V_i} \qquad \ldots (16)$$

$$R_{m_i} = \frac{n_{D_i}^2 - 1}{n_{D_i}^2 + 2} V_i \qquad \ldots (17)$$

$$V_i = \frac{M_i}{\rho_i} \qquad \ldots (18)$$

where, x_i, V_i, M_i represent mole fraction, molar volume and the molecular mass of i^{th} component, respectively.

Some observations made from the values of Tables 5 and Table 6 are as follows:

1. All the binary mixtures show a positive value of deviation in refractive index (Δn_D) over the entire composition range at all studied temperatures.
2. With increase of temperature, the Δn_D values for all binary mixtures shift toward less positive values.
3. Terpinolene + *o*-cresol binary mixtures show the highest positive Δn_D values at all studied temperatures.
4. The positive Δn_D values decrease when binary mixtures change from *o*-cresol to *p*-cresol.
5. All the binary mixtures show the highest Δn_D values at a 0.5 volume fraction value.
6. All the binary mixtures show negative ΔR_m values for the entire composition range at all studied temperatures.
7. Changing *o*-cresol to *p*-cresol binary mixtures, the negative values of ΔR_m decreases.
8. The largest negative values of ΔR_m are observed for the *o*-cresol binary mixture.

A perusal of Table 5 and Fig. 3 shows the positive values of Δn_D for all binary mixtures. The largest positive values of Δn_D is observed in cases of *o*-cresol containing binary mixtures. It indicates that *o*-cresol molecules show the strongest intermolecular interaction with terpinolene molecules. These interactions may be type of a delocalization of charge or hydrogen bonding. In cresols' binary mixtures, oxygen (O) atom have a lone pair of electrons and have more chances for hydrogen bonding or delocalization of charge type interaction to occur. Due to these types of interactions, the polarizability of a

Table 5 — Refractive Index (n_D) and Deviation in Refractive Index (Δn_D) *vs*. Volume Fraction (ϕ_I) for Terpinolene + + *o*-, *m*- and *p*-Cresol Mixtures at 303.15, 308.15 and 313.15 K.

ϕ_I		n_D			Δn_D	
	303.15 K	308.15 K	313.15 K	303.15 K	308.15 K	313.15 K
Terpinolene (1) + *o*-Cresol (2)						
0.0000	1.5412	1.5390	1.5364	0.00000	0.00000	0.00000
0.1000	1.5362	1.5340	1.5314	0.00052	0.00047	0.00043
0.2000	1.5311	1.5289	1.5263	0.00094	0.00084	0.00076
0.3000	1.5259	1.5237	1.5211	0.00126	0.00111	0.00099
0.4000	1.5205	1.5184	1.5158	0.00138	0.00128	0.00112
0.5000	1.5151	1.5130	1.5105	0.00150	0.00135	0.00125
0.6000	1.5094	1.5074	1.5049	0.00132	0.00122	0.00108
0.7000	1.5037	1.5017	1.4993	0.00114	0.00099	0.00091
0.8000	1.4978	1.4959	1.4935	0.00076	0.00066	0.00054
0.9000	1.4919	1.4901	1.4878	0.00038	0.00033	0.00027
1.0000	1.4860	1.4843	1.4821	0.00000	0.00000	0.00000
Terpinolene (1) + *m*-Cresol (2)						
0.0000	1.5357	1.5342	1.5319	0.00000	0.00000	0.00000
0.1000	1.5312	1.5296	1.5272	0.00047	0.00039	0.00028
0.2000	1.5266	1.5249	1.5225	0.00084	0.00068	0.00056
0.3000	1.5219	1.5201	1.5177	0.00111	0.00087	0.00074
0.4000	1.5170	1.5152	1.5128	0.00118	0.00096	0.00082
0.5000	1.5121	1.5103	1.5079	0.00125	0.00105	0.00090
0.6000	1.5070	1.5052	1.5028	0.00112	0.00094	0.00078
0.7000	1.5018	1.5000	1.4976	0.00089	0.00073	0.00056
0.8000	1.4966	1.4948	1.4924	0.00066	0.00052	0.00034
0.9000	1.4913	1.4895	1.4872	0.00033	0.00021	0.00012
1.0000	1.4860	1.4843	1.4821	0.00000	0.00000	0.00000
Terpinolene (1) + *p*-Cresol (2)						
0.0000	1.5353	1.5338	1.5305	0.00000	0.00000	0.00000
0.1000	1.5308	1.5292	1.5259	0.00043	0.00035	0.00024
0.2000	1.5262	1.5245	1.5213	0.00076	0.00060	0.00048
0.3000	1.5214	1.5197	1.5166	0.00089	0.00075	0.00062
0.4000	1.5166	1.5149	1.5119	0.00102	0.00090	0.00076
0.5000	1.5117	1.5100	1.5071	0.00105	0.00095	0.00080
0.6000	1.5067	1.5049	1.5021	0.00098	0.00080	0.00064
0.7000	1.5016	1.4998	1.4971	0.00081	0.00065	0.00048
0.8000	1.4964	1.4946	1.4920	0.00054	0.00040	0.00022
0.9000	1.4912	1.4894	1.4870	0.00027	0.00015	0.00006
1.0000	1.4860	1.4843	1.4821	0.00000	0.00000	0.00000

Standard uncertainties u are $u(T)$ = ±0.01 K, $u(\phi_I)$ = ±0.0001, $u(n_D)$ = ±0.0001. Further fitting coefficients of Δn_D with standard error are given in Table S3. All physical quantities are measured at atmospheric pressure.

binary mixture increases, which leads to a positive deviation in refractive index Δn_D values[6]. According to Campos *et al*. and Brocos *et al*., the positive values of Δn_D indicate the presence of strong intermolecular interaction between components of binary mixtures[25,26].

The reason for decreasing the intermolecular interactions with increase of temperature can be: (i) increase of kinetic energy of molecules due to increase of temperature leads the molecules to orient at large distances from each other (ii) increase of declustering of components with increase of temperature (iii) also weakening of dipole-dipole interactions and hydrogen bonding leads to decrease in polarizability due to increase of temperature[27].

The negative values of deviation in molar refraction ΔR_msupport the results of Δn_D that *o*-cresol binary mixtures show the strongest possible intermolecular interactions among all binary mixtures[28,29]. From the Tables 5 and Table 6 and Fig. 3 and Fig. 4, the order of Δn_D values at maxima for cresols with terpinolene is as under.

o-cresol > *m*-cresol > *p*-cresol

Table 6 — Molar Refraction (R_m) and Deviation in Molar Refraction (ΔR_m) *vs*. Volume Fraction (ϕ_1) for Terpinolene + *o*-, *m*- and *p*-Cresol Mixtures at 303.15, 308.15 and 313.15 K.

ϕ_1	R_m(cm^3·mol^{-1})			ΔR_m(cm^3·mol^{-1})		
	303.15 K	308.15 K	313.15 K	303.15 K	308.15 K	313.15 K
Terpinolene (1) + *o*-Cresol (2)						
0.0000	32.7775	32.8050	32.8127	0.0000	0.0000	0.0000
0.1000	33.6720	33.7038	33.7145	-0.4084	-0.4085	-0.4082
0.2000	34.6311	34.6647	34.6775	-0.7521	-0.7550	-0.7552
0.3000	35.6617	35.6974	35.7097	-1.0245	-1.0298	-1.0331
0.4000	36.7665	36.8089	36.8206	-1.2225	-1.2258	-1.2321
0.5000	37.9673	38.0097	38.0264	-1.3246	-1.3324	-1.3363
0.6000	39.2576	39.3069	39.3233	-1.3372	-1.3426	-1.3494
0.7000	40.6714	40.7221	40.7454	-1.2262	-1.2347	-1.2374
0.8000	42.2117	42.2716	42.2945	-0.9888	-0.9927	-0.9982
0.9000	43.9142	43.9822	44.0114	-0.5892	-0.5895	-0.5913
1.0000	45.8063	45.8791	45.9128	0.0000	0.0000	0.0000
Terpinolene (1) + *m*-Cresol (2)						
0.0000	32.8449	32.8947	32.9043	0.0000	0.0000	0.0000
0.1000	33.7437	33.7939	33.8017	-0.3974	-0.3993	-0.4035
0.2000	34.7056	34.7535	34.7632	-0.7316	-0.7381	-0.7429
0.3000	35.7361	35.7811	35.7929	-0.9972	-1.0090	-1.0140
0.4000	36.8387	36.8867	36.8992	-1.1907	-1.2018	-1.2085
0.5000	38.0349	38.0848	38.0983	-1.2907	-1.3021	-1.3103
0.6000	39.3227	39.3761	39.3907	-1.2990	-1.3093	-1.3187
0.7000	40.7240	40.7803	40.7959	-1.1939	-1.2035	-1.2143
0.8000	42.2593	42.3191	42.3362	-0.9547	-0.9632	-0.9749
0.9000	43.9432	44.0066	44.0323	-0.5669	-0.5741	-0.5796
1.0000	45.8063	45.8791	45.9128	0.0000	0.0000	0.0000
Terpinolene (1) + *p*-Cresol (2)						
0.0000	32.8291	32.8765	32.8323	0.0000	0.0000	0.0000
0.1000	33.7317	33.7789	33.7348	-0.3951	-0.3986	-0.4055
0.2000	34.6948	34.7408	34.7014	-0.7297	-0.7378	-0.7470
0.3000	35.7209	35.7704	35.7364	-1.0013	-1.0092	-1.0200
0.4000	36.8295	36.8828	36.8541	-1.1905	-1.1978	-1.2104
0.5000	38.0251	38.0825	38.0591	-1.2926	-1.2991	-1.3135
0.6000	39.3192	39.3743	39.3565	-1.2962	-1.3083	-1.3241
0.7000	40.7263	40.7858	40.7740	-1.1868	-1.1979	-1.2146
0.8000	42.2601	42.3247	42.3201	-0.9507	-0.9599	-0.9765
0.9000	43.9470	44.0160	44.0261	-0.5615	-0.5697	-0.5786
1.0000	45.8063	45.8867	45.9128	0.0000	0.0000	0.0000

Standard uncertainties u are $u(T)$ = ±0.01 K, $u(\phi_1)$ = ±0.0001. Further fitting coefficients of ΔR_m with standard error are given in Table S4.

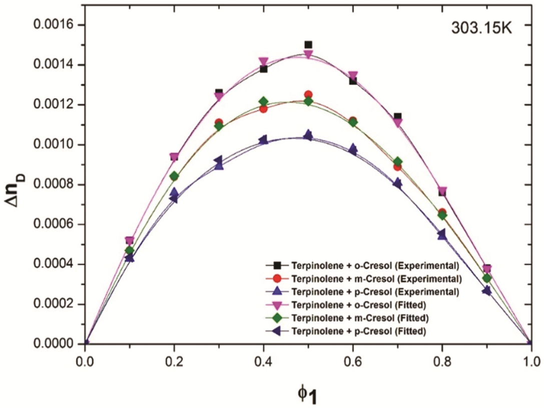

Fig. 3 — Deviation in refractive index (Δn_D) for the system Terpinolene (1) + *o*-, *m*-, and *p*-cresol (2) as a function of volume fraction ϕ_1 at *T* = 303.15 K

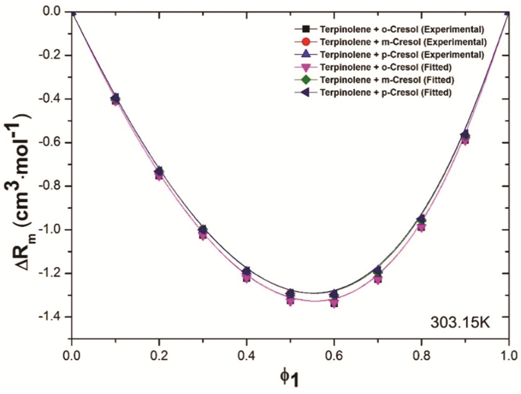

Fig. 4 — Deviation in molar refraction (ΔR_m) for the system Terpinolene (1) + *o*-, *m*-, and *p*-cresol (2) as a function of volume fraction ϕ_1 at *T* = 303.15 K

Table 7 — Average Percentage Deviation ($\sigma\%$) in the Refractive Index from Nine Different Mixing Relations for Terpinolene + *o*-, *m*- and *p*-Cresol Mixtures at 303.15, 308.15 and 313.15 K.

Mixing Relations	*T* (K)		
	303.15	308.15	313.15
Terpinolene (1) + *o*-Cresol (2)			
A–B	0.000836	0.000750	0.000668
D-G	0.000836	0.000750	0.000668
L–L	0.001008	0.000918	0.000834
WR	-0.000480	-0.000544	-0.000609
Heller	0.001015	0.000926	0.000842
Newton	0.000685	0.000602	0.000522
Eyring–John	0.000912	0.000824	0.000741
Eykman	0.001200	0.001107	0.001020
Oster	0.000738	0.000653	0.000573
Terpinolene (1) + *m*-Cresol (2)			
A–B	0.000714	0.000577	0.000464
D-G	0.000714	0.000577	0.000464
L–L	0.000852	0.000717	0.000603
WR	-0.000353	-0.000499	-0.000610
Heller	0.000858	0.000723	0.000609
Newton	0.000591	0.000454	0.000340
Eyring–John	0.000775	0.000639	0.000525
Eykman	0.001008	0.000875	0.000760
Oster	0.000634	0.000497	0.000384
Terpinolene (1) + *p*-Cresol (2)			
A–B	0.000614	0.000505	0.000391
D-G	0.000614	0.000505	0.000391
L–L	0.000750	0.000642	0.000522
WR	-0.000436	-0.000555	-0.000623
Heller	0.000756	0.000648	0.000528
Newton	0.000493	0.000383	0.000274
Eyring–John	0.000674	0.000565	0.000449
Eykman	0.000904	0.000797	0.000671
Oster	0.000535	0.000426	0.000316

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

Theoretical nine mixing relations of refractive index

The theoretical refraction index values of binary mixtures were calculated using the following nine theoretical relations:

Arago-Biot (A-B)[30]

$$n_D = n_{D_1}\phi_1 + n_{D_2}\phi_2 \qquad \dots (19)$$

Dale-Glastone (D-G)[31]

$$n_D - 1 = \left(n_{D_1} - 1\right)\phi_1 + (n_{D_2} - 1)\phi_2 \qquad \dots (20)$$

Lorentz-Lorentz (L-L)[32]

$$\frac{n_D^2-1}{n_D^2+2} = \frac{\left(n_{D_1}^2-1\right)}{\left(n_{D_1}^2+2\right)}\phi_1 + \frac{\left(n_{D_2}^2-1\right)}{\left(n_{D_2}^2+2\right)}\phi_2 \qquad \dots (21)$$

Eykman (Eyk)[33]

$$\frac{n_D^2-1}{n_D^2+0.4} = \frac{\left(n_{D_1}^2-1\right)}{\left(n_{D_1}^2+0.4\right)}\phi_1 + \frac{\left(n_{D_2}^2-1\right)}{\left(n_{D_2}^2+0.4\right)}\phi_2 \qquad \dots (22)$$

Weiner (WR)[34]

$$\left(\frac{n_D^2-n_{D_1}^2}{n_D^2+2n_{D_1}^2}\right) = \phi_2\left(\frac{n_{D_2}^2-n_{D_1}^2}{n_{D_2}^2+2n_{D_1}^2}\right) \qquad \dots (23)$$

Heller (Hr)[35]

$$\frac{n_D-n_{D_1}}{n_{D_1}} = \left(\frac{\left(n_{D_2}-n_{D_1}\right)^2-1}{\left(n_{D_2}-n_{D_1}\right)^2+2}\right)\phi_2 \qquad \dots (24)$$

Newton (Nw)[36]

$$n_{D_1}^2 - 1 = \left(n_{D_1}^2 - 1\right)\phi_1 + (n_{D_2}^2 - 1)\phi_2 \qquad \dots (25)$$

Oster (Os)[37]

$$\left[\frac{\left(n_D^2-1\right)\left(2n_D^2+1\right)}{n_D^2}\right]V = \left[\frac{\left(n_{D_1}^2-1\right)\left(2n_{D_1}^2+1\right)}{n_{D_1}^2}\right]x_1V_1 + \left[\frac{\left(n_{D_2}^2-1\right)\left(2n_{D_2}^2+1\right)}{n_{D_2}^2}\right]x_2V_2 \qquad \dots (26)$$

Eyring-John (E-J)[12]

$$n_D = n_{D_1}\phi_1^2 + 2(n_{D_1}n_{D_2})^{1/2}\phi_1\phi_2 + n_{D_2}\phi_2^2 \qquad \dots (27)$$

where, $\phi_i$and n_{D_i} represents volume fraction and refractive index of pure i^{th} components respectively. The average percentage deviation (σ %) from nine mixing relations is shown in Table 7.

Some observations made from the values of Table 7 are as follows:

1. Arago-Biot (A-B) and Dale-Gladstone (D-G) relations show the same values of σ % for all binary mixtures.
2. The Weiner (WR) relation shows the lowest σ % value for all binary mixtures at 303.15K.
3. The Newton (NW) relation shows the lowest σ % value for all binary mixtures at 308.15 and 313.15 K except for the *o*-cresol binary mixture at 308.15K (Weiner (WR) relation -0.000544).
4. The Eykman (Eyk) relation shows the highest σ % value for all binary mixtures at all studied temperatures.

The calculated values of $\Delta\eta$, ΔG^{*E}, Δn_D, and ΔR_m were fitted with the Redlich-Kister[38] polynomial equation. The calculated values of the fitting coefficient are given in Tables S1 to S4 of supplementary data.

$$Y^E = x_1(1 - x_1)\sum_{i=1}^{n} A_i(2x_1 - 1)^i \quad \dots (28)$$

where, Y^E, x_1 and A_i represent the excess/deviation properties, mole fraction values of component 1, and fitting coefficient, respectively. The standard deviation (σ) was also calculated using the following standard equation:

$$\sigma(Y) = \left[\frac{\sum(Y_{exp}^E - Y_{cal}^E)}{N-P}\right]^{\frac{1}{2}} \quad \dots (29)$$

where, Y_{exp}^E, Y_{cal}^E, N and Pindicate the experimentally calculated values of excess/deviation properties, the number of experimental points, and the number of parameters of the Redlich-Kister equation.

4. Conclusion

The viscosity (η) and refractive index (n_D) were experimentally measured for binary mixtures of terpinolene + *o*-, *m*- and *p*-cresol at 303.15, 308.15, and 313.15 K for the whole composition range. Using the values of primary physical properties, various deviation properties like deviation in viscosity $(\Delta\eta)$, excess Gibbs' free energy for activation of viscous flow (ΔG^{*E}), deviation in refractive index (Δn_D) and deviation in molar refraction (ΔR_m) were calculated. Some theoretical viscosity relations, like Dolezalek-Schulze (D_{12}), Grunberg-Nissan (G_{12}), Tamura-Kurata (T_{12}), Katti-Chaudhri (W_{vis}/RT), McAllister relation $(M_{12}$ and $M_{21})$, Kendall-Munroe relation, Bingham relation and Arrhenius-Eyring relation were used for calculation of theoretical viscosities. Among these relations, Dolezalek-Schulze (D_{12}) shows the lowest deviation $(\sigma\,\%)$ and 3-body McAllister relation shows the highest deviation $(\sigma\,\%)$ for all binary mixtures at 303.15 K. Nine relations like Arago-Biot (A-B), Dale-Glastone (D-G), Lorentz-Lorentz (L-L), Weiner (WR), Heller (Hr), Newton (Nw), Eyring-John (E-J), Eykman (Eyk), and Oster (Os) are used to calculate theoretical refractive indices. Weiner (WR) relation of refractive index showed the lowest deviation $(\sigma\,\%)$ values for all binary mixtures at 303.15 K. Negative values of $\Delta\eta$, ΔR_m and positive values of Δn_D indicated the presence of strong intermolecular interactions between components of binary mixtures. Positive values of interaction parameters like Grunberg-Nissan (G_{12}), Dolezalek-Schulze (D_{12}) and Tamura-Kurata (T_{12}) also indicated the presence of strong intermolecular interactions between components of binary mixtures. On the basis of the studied parameters, the order of molecular interaction strength for these terpinolene + cresol binaries is *o*-cresol > *m*-cresol > *p*-cresol.

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Supplementary Information

Supplementary information is available in the website: http://nopr.niscpr.res.in/ handle/123456789/58776.

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