Analysis Of Viscosities And Optical Properties Of Binary System Containing Ester And Cyclic Alcohols At Various Temperatures (298.15-318.15K).

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Abstract
The research examines the excess viscosity and excess refractive index of binary mixtures of triacetin with cyclohexanol / cyclopentanol within a temperature range of 298.15 K to 318.15 K. The study examines the molecular interactions and thermodynamic properties of these mixtures by the measurement of viscosities, and refractive indices. The excess properties, including excess viscosities, and excess refractive indices, are derived from experimental data and examined using several theoretical models, such as the Redlich-Kister equation and mixing rules equations. The findings demonstrate the impact of temperature and composition on the excess characteristics, offering insights into the intermolecular interactions within the mixtures. These discoveries enhance the comprehensive knowledge of the thermophysical characteristics of alcohol mixtures, which are essential for applications in the chemical and petrochemical industries.

Keywords: viscosity, refractive index, excess viscosity and excess refractive index

Introduction

Triacetin, a molecule produced from glycerol, has emerged as a viable biofuel additive because of its ability to improve fuel characteristics and decrease emissions. Glycerol, a byproduct of biodiesel manufacturing, may be effectively transformed into triacetin by esterification methods, so enhancing the value of the biodiesel production chain and mitigating the surplus glycerol problem[1,2]. Triacetin is acknowledged for its elevated energy density and oxygen content, rendering it an efficient addition for enhancing combustion efficiency and mitigating hazardous emissions in diesel engines[3,4]. Research indicates that the use of triacetin in biodiesel blends might reduce the rise in NOx emissions commonly linked to biodiesel, thereby improving the environmental sustainability of biodiesel as a renewable fuel[2,4]. Moreover, the synthesis of triacetin from renewable sources such as glycerol adheres to the tenets of circular bioeconomy, enhancing the sustainability of biofuel production systems. The economic viability of triacetin manufacturing has been established, with evaluations revealing substantial profitability and a brief payback period for dedicated production facilities. Triacetin is a significant biofuel additive that enhances fuel performance while advancing the economic and environmental objectives of the biofuel sector.

Cyclopentanol, a chemical generated from lignocellulose, presents a sustainable alternative to conventional fossil fuels, supporting worldwide initiatives to diminish carbon emissions and dependence on non-renewable energy sources. Its elevated resistance to auto-ignition renders it a desirable addition for gasoline, potentially enhancing engine efficiency and diminishing pollutants[5,6]. The combustion properties of cyclopentanol have been thoroughly investigated, demonstrating its capacity to extend ignition delay and enhance thermal efficiency when mixed with diesel[5]. This feature effectively diminishes soot emissions, a prevalent problem in diesel engines, thereby enhancing combustion cleanliness.

Cyclohexanol, a cyclic alcohol sourced from lignin, has emerged as a viable fuel addition owing to its capacity to improve combustion efficiency and diminish emissions across diverse engine types. Recent research have examined its employment in diesel and gasoline engines, emphasizing its contribution to enhancing fuel attributes and emission characteristics. Cyclohexanol, when combined with biodiesel, enhances combustion efficiency and lowers emissions of hydrocarbons (HC), carbon monoxide (CO), and smoke, but with a little increase in carbon dioxide (CO2) emissions[7]. Cyclohexanol blends in agricultural diesel engines have shown decreased smoke opacity and NOx emissions under specified conditions, while they may lead to increased ignition delay and specific fuel consumption[8,9].

The many advantages of cyclohexanol, cyclopentanol and triacetin as a fuel additive, such as emission reduction, increased combustion properties, and improved blend stability, render it a significant element in the advancement of more sustainable and efficient fuel systems. In the presented research we have studied excess

viscosities and excess refractive indices of the binary mixtures involving cyclohexanol, cyclopentanol and triacetin at a temperature range of 298.15 to 318.15K and 0.1 MPa pressure.

Experimental

Triacetin, cyclohexanol, cyclopentanol were purchased with high purity. The binary mixtures were prepared gravimetrically and stored in sealed bottles immediately to avoid moisture absorption. The Abbemat 550 digital refractometer, provided by Anton Paar, was used to assess the refractive indices of liquid mixes used in experiments. It operates at 589 nm wavelength, which is the same as the sodium D line. With an uncertainty in temperature of 0.03K, this refractometer operates in the temperature range of 277.15 to 358.15K. With distilled water, the instrument was promptly calibrated. The refractive index can be measured, with an uncertainty of 0.00002 units within the range of 1.26-1.72[10].

The viscosities of mixtures as well as pure liquids were assessed by using Lovis 2000 M/ME Micro Viscometer. It operates on the premise of a rolling ball. It is very practical for assessing the kinematic, dynamic, relative, and intrinsic viscosities of liquids with a very low viscosity range. The micro viscometer is named after the little sample volume required for operation (<1mL). The glass capillary containing the sample is placed into a temperature-regulated capillary block. The rolling time of the ball in the capillary correlates directly with the sample's viscosity; higher viscosity results in an extended rolling time. This device quantifies the duration of rolling.

The accuracy of instrument can be predicted by experimental refractive indices and viscosities values that are very well compared with values available in literature presented in **Table 1.**

Table 1. presents the experimental viscosities, η and refractive index, n_D values and values available in literature at various temperatures.

Liquid	T/K	Dynamic Viscosity, η, (m Pas)		Refractive index, nD	
		Exptl.	literature	Exptl.	literature
Triacetin	298.15K	16.42	Not available	1.429727	Not available
	303.15K	12.92	Not available	1.428116	Not available
	308.15K	10.28	Not available	1.426051	Not available
	313.15K	8.34	Not available	1.423940	Not available
	318.15K	6.92	Not available	1.421519	Not available
Cyclohexanol	298.15K	57.08	58.966[11]	1.464385	1.46449[12]
	303.15K	42.04	41.072[13] , 43.42[11]	1.46287	Not available
	308.15K	32.378	32.387[11]	1.460355	Not available
	313.15K	25.784	25.847[11]	1.458835	Not available
	318.15K	20.132	20.138[11]	1.456508	Not available
Cyclopentanol	298.15K	9.68	9.72, 10.413[11], 9.6[14]	1.450673	1.45159[15]
	303.15K	7.82	8.068, 8.088[11], 7.891[14]	1.448837	Not available
	308.15K	6.52	7.077, 6.528[11], 6.564[14]	1.44694	Not available
	313.15K	5.51	6.04, 5.442[11], 5.509[14]	1.444937	Not available
	318.15K	4.62	5.19, 4.742[11], 4.657[14]	1.442901	Not available

Source: Authors and literature

Result & Discussion

Excess viscosities, η^E of all mixtures at different temperatures were calculated by using experimental data on viscosities, η. Equation 1 was used to calculate excess viscosities.

$$\eta^E=\eta_m - \exp(x_i \ln\eta_i + x_j \ln\eta_j) \quad (1)$$

Where, x_i, x_j = mole fractions and η_i, η_j are viscosities of component i and component j, η_m is the viscosity of mixture. The excess viscosities calculated were given in **Table 1** and graphically presented in **figure a and b.**

Table 2. **shows excess viscosities, η^E of triacetin(i) and cyclohexanol, cyclopentanol(j), mixture at 0.1 MPa of pressure and a range of temperature, T= 298.15- 318.15K.**

Mole fraction	Excess viscosities, η^E				
x_i	*298.15K*	*303.15K*	*308.15K*	*315.15K*	*318.15K*
Triacetin(i) + cyclohexanol(j)					
0.0613	-3.02291	-2.68091	-2.38887	-2.04035	-1.68636
0.0968	-4.53177	-4.04626	-3.61503	-3.11131	-2.61393

0.1466	-6.37004	-5.73778	-5.14344	-4.46941	-3.83076
0.1981	-7.95045	-7.22030	-6.49187	-5.68948	-4.96186
0.2431	-9.08167	-8.30044	-7.47977	-6.59683	-5.82610
0.3123	-10.3978	-9.58251	-8.65862	-7.69518	-6.89895
0.3953	-11.3491	-10.5364	-9.54027	-8.52866	-7.73425
0.4523	-11.6328	-10.8355	-9.81749	-8.79339	-8.00516
0.5114	-11.6252	-10.8487	-9.82933	-8.80532	-8.02069
0.5867	-11.1854	-10.4382	-9.44751	-8.44136	-7.65524
0.6538	-10.3943	-9.67770	-8.74222	-7.77199	-6.98455
0.7422	-8.77521	-8.11830	-7.30412	-6.42418	-5.65853
0.8212	-6.75593	-6.19477	-5.54547	-4.81064	-4.12479
0.9141	-3.64601	-3.29583	-2.92771	-2.48527	-2.03749
Triacetin(i) + cyclopentanol(j)					
0.0613	-1.60583	-1.35995	-1.07454	-0.81771	-0.63131
0.0968	-2.37513	-2.03967	-1.6548	-1.30464	-1.05304
0.1466	-3.28351	-2.87247	-2.41007	-1.98182	-1.67796
0.1981	-4.04107	-3.59794	-3.11190	-2.65185	-2.32840
0.2431	-4.57333	-4.12842	-3.65336	-3.19375	-2.87126
0.3123	-5.19254	-4.77179	-4.34455	-3.91523	-3.60914
0.3953	-5.66932	-5.28958	-4.92961	-4.55144	-4.26318
0.4523	-5.85220	-5.49355	-5.16715	-4.81779	-4.52921
0.5114	-5.92650	-5.57681	-5.26552	-4.93302	-4.62903
0.5867	-5.84894	-5.48956	-5.16550	-4.83218	-4.48752
0.6538	-5.60123	-5.21832	-4.85861	-4.50758	-4.11485
0.7422	-4.96593	-4.55196	-4.13683	-3.75789	-3.31072
0.8212	-4.02334	-3.61610	-3.18643	-2.81139	-2.36259
0.9141	-2.32023	-2.02827	-1.70605	-1.43451	-1.11084

Source: Authors

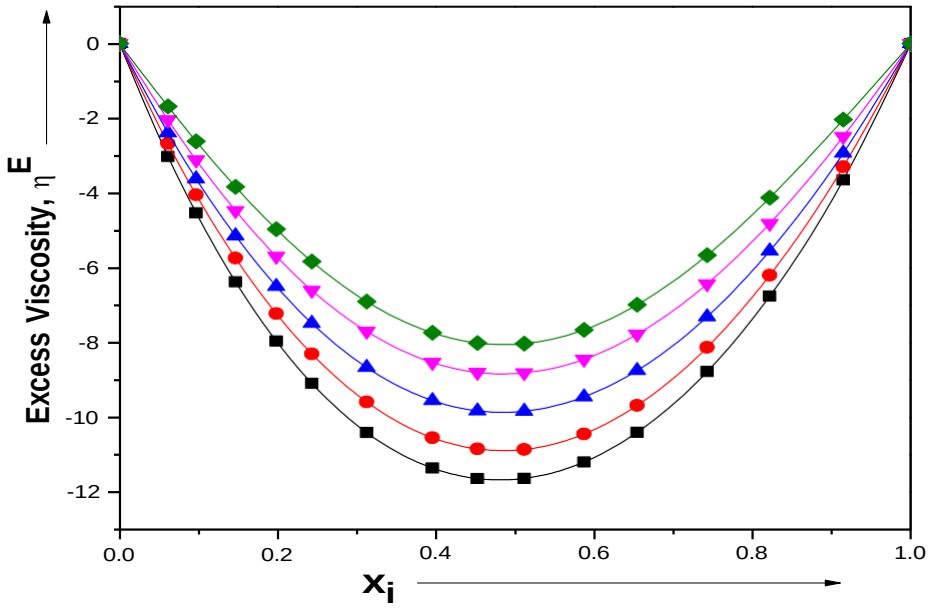

Figure a. show the change in Excess viscosities, η^E with increasing concentration of Triacetin in a binary mixture of triacetin(i) and cyclohexanol(j) at temperatures 298.15 (■), 303.15(●), 308.15(▲),

313.15 (▼) and 318.15 K (♦); lines are for R.K. equation values and symbols present experimental values.

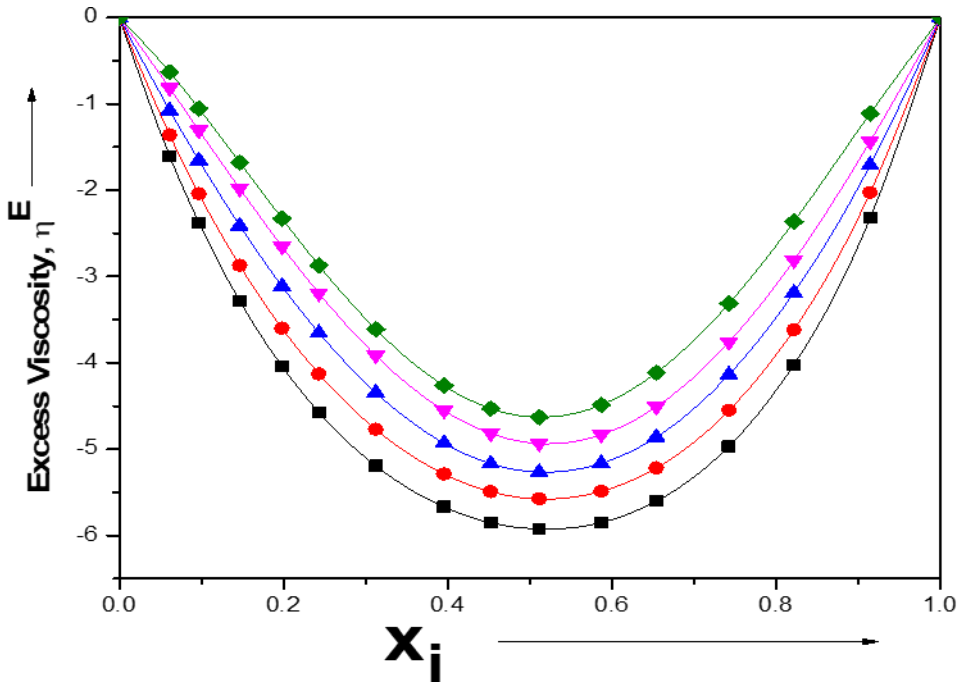

Figure b. show the change in Excess viscosities, η^E with increasing concentration of Triacetin in a binary mixture of triacetin(i) and cyclopentanol(j) at temperatures 298.15 (■), 303.15(●), 308.15(▲), 313.15 (▼) and 318.15 K (♦); lines are for R.K. equation values and symbols present experimental values.

From figure a & b it is visible that excess viscosities of binary mixtures of triacetin with cyclohexanol and cyclopentanol are negative. The excess viscosities of triacetin is more negative with cyclohexanol as compared to cyclopentanol. Negative excess viscosities indicate that the interactions among the molecules in the mixture are more robust than those in the pure components, resulting in a more organized or dense configuration. Cyclohexanol forms stronger hydrogen bonds and exerts a greater influence on the mixture's structure than cyclopentanol.

By using experimental data on refractive index (n_D), calculation of the excess refractive index (n^E_D). **Equation 2** is employed to calculated excess refractive indices[16].

$$n^E_D = n_D - n^{id}_D = n_D - (\sum_i \varphi_i \, n^2_{D,i})^{\frac{1}{2}} \qquad (2)$$

Where $n_{D,i}$: refractive index of pure component i
n_D : refractive index of the mixture
φ_i : volume fraction pure component i
Calculated excess refractive indices, n^E_D are given in **Table 3** and graphically presented in **figure c & d.**

Table 3. shows excess refractive indices, n^E_D of triacetin(i) and cyclohexanol, cyclopentanol(j), mixture at 0.1 MPa of pressure and a range of temperature, T= 298.15- 318.15K

Mole fraction	**Excess refractive indices, n^E_D**				
x_i	***298.15K***	***303.15K***	***308.15K***	***315.15K***	***318.15K***
Triacetin(i) + Cyclohexanol(j)					
0.0623	2.82E-05	3.25E-05	3.55E-05	4.24E-05	4.70E-05
0.0958	4.39E-05	4.97E-05	5.36E-05	6.28E-05	6.89E-05
0.1426	6.56E-05	7.24E-05	7.72E-05	8.83E-05	9.57E-05
0.1961	8.88E-05	9.59E-05	0.000101	0.000113	0.000121
0.2531	0.000110	0.000117	0.000122	0.000134	0.000142

0.3173	0.000129	0.000135	0.000140	0.000150	0.000158
0.3913	0.000142	0.000147	0.000152	0.000160	0.000167
0.4553	0.000145	0.000150	0.000155	0.000162	0.000168
0.5174	0.000141	0.000145	0.000151	0.000157	0.000163
0.5767	0.000131	0.000135	0.000141	0.000147	0.000153
0.6638	0.000106	0.000112	0.000118	0.000125	0.000132
0.7322	8.16E-05	8.78E-05	9.48E-05	0.000102	0.000109
0.8412	3.92E-05	4.59E-05	5.25E-05	6.00E-05	6.73E-05
0.9241	1.26E-05	1.74E-05	2.18E-05	2.73E-05	3.23E-05
Triacetin(i) + cyclopentanol(j)					
0.0623	-0.00030	-0.00037	-0.00041	-0.00045	-0.00053
0.0958	-0.00055	-0.00064	-0.00070	-0.00077	-0.00088
0.1426	-0.00096	-0.00107	-0.00116	-0.00126	-0.00139
0.1961	-0.00148	-0.00161	-0.00172	-0.00185	-0.00200
0.2531	-0.00203	-0.00217	-0.00230	-0.00247	-0.00263
0.3173	-0.00258	-0.00274	-0.00290	-0.00309	-0.00326
0.3913	-0.00308	-0.00324	-0.00342	-0.00364	-0.00383
0.4553	-0.00333	-0.00350	-0.00371	-0.00394	-0.00414
0.5174	-0.00340	-0.00357	-0.00380	-0.00405	-0.00427
0.5767	-0.00329	-0.00347	-0.00372	-0.00397	-0.00422
0.6638	-0.00286	-0.00304	-0.00331	-0.00356	-0.00383
0.7322	-0.00233	-0.00250	-0.00278	-0.00300	-0.00329
0.8412	-0.00128	-0.00142	-0.00166	-0.00182	-0.00209
0.9241	-0.00051	-0.00059	-0.00074	-0.00083	-0.00100

Source: Authors

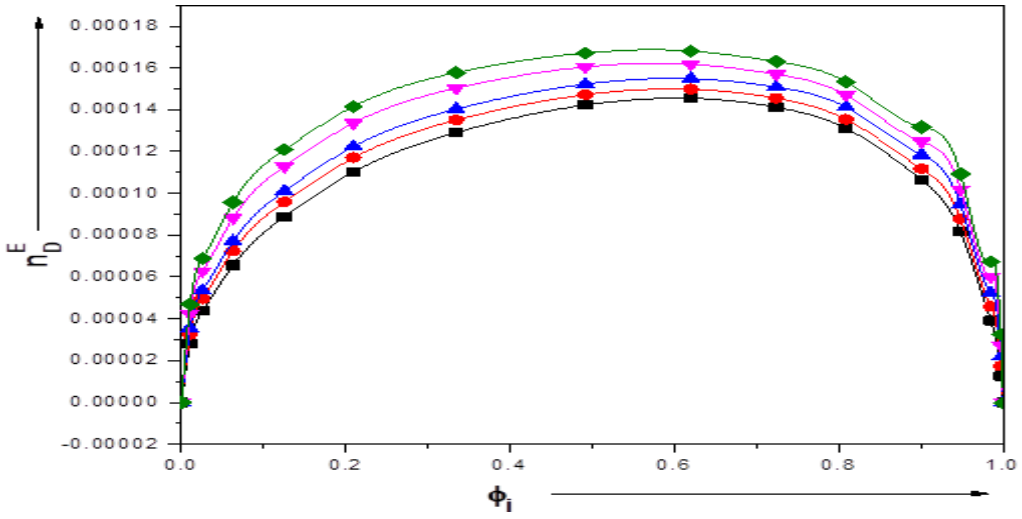

Figure c show the change in Excess Refractive index n_D^E with increasing concentration of Triacetin in a binary mixture of triacetin and cyclohexanol at temperatures 298.15 (■), 303.15(●), 308.15(▲), 313.15 (▼) and 318.15 K (♦); lines are for R.K. equation values and symbols present experimental values.

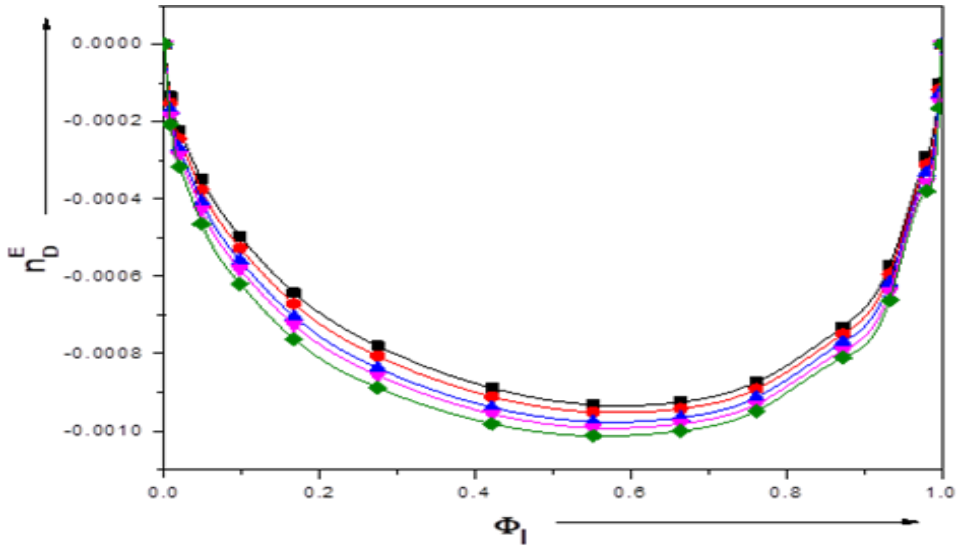

Figure d show the change in Excess Refractive index $n_D^E$with increasing concentration of Triacetin in a binary mixture of triacetin and cyclopentanol at temperatures 298.15 (■), 303.15(●), 308.15(▲), 313.15 (▼) and 318.15 K (♦); lines are for R.K. equation values and symbols present experimental values.

The positive excess refractive index in triacetin mixture with cyclohexanol indicates strong intermolecular interactions, presumably resulting from hydrogen bonding between cyclohexanol and ester molecules. Cyclohexanol, being a bigger and more flexible molecule, may establish stable hydrogen bonds, resulting in denser packing and a positive deviation in refractive index. Cyclopentanol has a negative excess refractive index when combined with triacetin. This signifies less intermolecular interactions or less efficient hydrogen bonding relative to cyclohexanol. The reduced ring size and structural stiffness of cyclopentanol may constrain its capacity to establish robust hydrogen bonds, leading to a more dispersed molecular configuration and a negative deviation.

The calculated n_D^E and η^E data were fitted to the Redlich-Kister equation given below

$$X^E = x_i x_j [A^{(0)} + A^{(1)}(2x_i - 1)^+ A^{(2)})(2x_i - 1)^2 \qquad (2)$$

Where X^E represents excess property and A^n is the fitting parameter.

The standard deviation (σ) and fitting parameter were calculated using the following equation

$$\sigma X^E = [\Sigma(X^E_{expt} - X^E_{calc})^2 / (m - n)]^{1/2} \qquad (3)$$

Table 4 shows Values of adjustable parameters A^1, A^2, A^3 of Redlich–Kister equation and standard deviation, σ_{nD} for refractive indices values at various temperatures

Systems	T/K	A^1	A^2	A^3	σ
Excess refractive indices, n_D^E					
Triacetin(i) + cyclohexanol(j)	298.15	0.000573	-0.00018	-3.29E-04	0.000142
	303.15	0.000589	-0.00019	-2.55E-04	0.000151
	308.15	0.000610	-0.00018	-2.07E-04	0.000158
	313.15	0.000636	-0.00020	-1.09E-04	0.000172
	318.15	0.000660	-0.00020	-4.10E-05	0.000183
Triacetin(i) + cyclopentanol(j)	298.15	-0.013580	-0.00092	0.009868	0.022107
	303.15	-0.014280	-0.00104	0.009285	0.060003
	308.15	-0.015180	-0.00182	0.008640	0.105013
	313.15	-0.016160	-0.00210	0.008521	0.166999
	318.15	-0.017010	-0.00279	0.007094	0.240737
Excess viscosities, η^E					
Triacetin(i) + cyclohexanol(j)	298.15	-46.6003	3.387369	-3.84668	12.86983

	303.15	-43.4774	2.656641	-1.01561	11.81805
	308.15	-39.3944	2.483436	0.075779	10.65165
	313.15	-35.2946	2.353736	2.469973	9.403284
	318.15	-32.1547	2.278751	6.29697	8.32611
Triacetin(i) + cyclopentanol(j)	298.15	-23.6849	-1.30583	-6.97265	6.663337
	303.15	-22.2839	-1.45602	-3.41310	6.061582
	308.15	-21.034	-1.7367	1.086273	5.434306
	313.15	-19.6958	-2.14949	4.675389	4.831313
	318.15	-18.4966	-1.46309	8.10788	4.342738

Source: Authors

The refractive index of pure components can also be used to measure the refractive index of the mixture by using mixing rules[17–19]. Equations 4 to 7 illustrate the most widely used mixing rules, which include the Lorentz-Lorentz, Gladstone-dale, Weiner, and Heller relations. These mixing principles were attempted to be validated in the current study, and **Table 4** shows a strong correlation between experimental and theoretical data.

Gladstone-Dale (G-D) [20]

$$n_D - 1 = (n_{D1}\text{-}1)\,\varphi_1 + (n_{D2}\text{-}1)\,\varphi_2 \qquad (4)$$

Lorentz -Lorentz (L-L) [21]

$$\frac{n_D^2-1}{n_D^2+2} = \left(\frac{n_D^2-1}{n_D^2+2}\right)\varphi_1 + \left(\frac{n_D^2-1}{n_D^2+2}\right)\varphi_2 \qquad (5)$$

Heller (H)[22]

$$\frac{n_D-n_{D_1}}{n_{D_1}} = \frac{3}{2}\left(\frac{(n_{D2}/n_{D_1})^2-1}{(n_{D_2}/n_{D_1})^2+2}\right)\varphi_2 \qquad (6)$$

Weiner[23]

$$\frac{n_D^2-n_{D1}^2}{n_D^2+2n_{D2}^2} = \left(\frac{n_{D2}^2-n_{D1}^2}{n_{D2}^2+2n_{D2}^2}\right)\varphi_2 \qquad (7)$$

Where φ_1, n_{D1}, φ_2, and n_{D2} represents the volume fraction and the refractive index of components 1 and 2 respectively.

Table 5. Experimental and theoretical n_D values by various mixing rules at temperature 298.15K.

x_i	Exptl.	G.D.	H.R.	L.L	W.R.
Triacetin(i) + cyclohexanol(j)					
0.0706	1.464064	1.464031	1.463889	1.464027	1.465139
0.1077	1.463560	1.463506	1.463365	1.463495	1.464594
0.1621	1.462304	1.462215	1.462079	1.462189	1.463256
0.2204	1.460190	1.460056	1.459930	1.460008	1.461021
0.2778	1.457322	1.457143	1.457029	1.457071	1.458006
0.3461	1.453046	1.452824	1.452728	1.452728	1.453540
0.4234	1.447631	1.447385	1.447311	1.447276	1.447921
0.4882	1.443174	1.442931	1.442876	1.442829	1.443325
0.5475	1.439542	1.439317	1.439277	1.439231	1.439600
0.6055	1.436579	1.436383	1.436355	1.436316	1.436577
0.6923	1.433322	1.433178	1.433163	1.433139	1.433277
0.7617	1.431630	1.431528	1.431520	1.431507	1.431579
0.8577	1.430298	1.430253	1.430251	1.430247	1.430268
0.935057	1.429834	1.429820	1.429820	1.429819	1.429823
Triacetin(i) + cyclopentanol (j)					
0.0613	1.450186	1.450489	1.450438	1.450488	1.450894
0.0941	1.449665	1.450215	1.450164	1.450212	1.450614
0.1426	1.448581	1.449537	1.449487	1.449529	1.449921
0.1956	1.446923	1.448386	1.448340	1.448371	1.448746
0.2485	1.444797	1.446801	1.446758	1.446777	1.447127
0.3128	1.441825	1.444377	1.444341	1.444344	1.444653
0.3870	1.438156	1.441194	1.441165	1.441155	1.441405
0.4506	1.435181	1.438472	1.438451	1.438434	1.438631
0.5099	1.432820	1.436183	1.436167	1.436150	1.436299

0.5690	1.431001	1.434268	1.434257	1.434242	1.434348
0.6593	1.429271	1.432117	1.432111	1.432101	1.432158
0.7333	1.428663	1.430984	1.430981	1.430975	1.431006
0.8383	1.428815	1.430096	1.430095	1.430094	1.430103
0.9253	1.429284	1.429793	1.429793	1.429792	1.429794

Source: Authors

Table 6 shows the values of the standard deviations, σ, of the refractive index that were calculated using different mixing rules for binary systems.

T/K	σ (G.D)	σ(H.R)	σ (L.L)	σ (W.R.)
Triacetin(i) + cyclohexanol(j)				
298.15	0.000515	0.000776	0.000699	0.001451
303.15	0.000536	0.000799	0.000721	0.001442
308.15	0.000552	0.000808	0.000732	0.001378
313.15	0.000588	0.000854	0.000774	0.001411
318.15	0.000515	0.000776	0.000699	0.001451
Triacetin(i) + cyclopentanol(j)				
298.15	0.007415	0.007318	0.007348	0.00816
303.15	0.007940	0.007845	0.007875	0.00867
308.15	0.008574	0.008476	0.008507	0.009316
313.15	0.009209	0.009111	0.009142	0.00996
318.15	0.009914	0.009811	0.009844	0.010694

Source: Authors

Conclusions

Throughout the whole mole fraction range, excess viscosity of the triacetin(i) and cyclopentanol, cyclohexanol(j) binary mixtures were found to be negative while excess refractive indices (n_D^E) of cyclohexanol is positive and negative in case of cyclopentanol. These measurements were made at five different temperatures (298.15, 303.15, 308.15, 313.15, and 318.15K). To verify different mixing rules, experimental values of the refractive index, n_D , were used. It was found that these values correlated well with values predicted from all mixing rules.

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