ESTIMATING THE REFRACTIVE INDEX OF BINARY LIQUID MIXTURES FOR TUNEABLE OPTICAL PROPERTIES: A THEORETICAL INVESTIGATION OF ACETOPHENONE WITH SELECTED ACETATES

Abstract

For a variety of scientific and industrial purposes, it is crucial to accurately determine the refractive index of liquid mixtures. In this study, theoretical modelling is used to forecast the refractive index behaviour of binary liquid mixes containing acetophenone and certain acetates. The refractive index is a crucial optical characteristic with many uses in medications, colognes, solvents, and coatings. This research gives important insights into the molecular interactions and intermolecular forces influencing the optical characteristics of these liquid mixes through a thorough examination of pertinent literature and computer techniques. The results emphasise the importance of theoretical modelling in understanding binary liquid mixture behaviour and its possible applications.

Keywords: Liquid Mixtures, Mixing Rules, Refractive Index.

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I. INTRODUCTION

Refractive index, a fundamental optical property, plays a pivotal role in understanding the behaviour of light as it interacts with matter. The accurate determination of refractive index in liquid mixtures is of paramount importance in various scientific and industrial applications, such as pharmaceuticals, food processing, and chemical engineering. By studying the refractive index of liquid mixtures, researchers gain valuable insights into molecular interactions, solution behaviour, and composition changes. Binary liquid mixtures, comprised of two components, exhibit complex behaviour due to their diverse chemical compositions. Understanding the refractive index behaviour in binary liquid mixtures is crucial to comprehend their physicochemical properties and potential applications. Acetophenone, a colourless liquid, is widely utilized in the synthesis of pharmaceuticals, dyes, and perfumes, while acetates, a class of organic compounds, find applications in solvents and coatings. In recent years, theoretical approaches have emerged as powerful tools for predicting the refractive index of liquid mixtures with reasonable accuracy, offering insights that complement experimental data. Theoretical estimation allows researchers to explore a wide range of compositions and temperatures, providing a cost-effective and time-efficient alternative to experimental measurements.

Many researchers have made an effort to predict the thermophysical characteristics of liquid mixes from their pure constituents. Dnyaneshwar S Wankhede measured the refractive indices(n), molar volume (Vm) and molar refraction(R) of the binary mixtures of propylene carbonate with tetrahydrofuran, 1,4-dioxane, acetylacetone and acetone at 298.15K, 303.15K and 308.15K. These values were used to measure the excess refractive indices(Δn) and excess molar refraction(ΔR) of these binary liquid mixtures[1]. At a temperature of 298.15 K, Maharolkar et al. studied the density, refractive index, viscosity, and dielectric constant of the binary liquid mixture of allyl chloride and n-butanol. The excess molar volume, excess permittivity, excess refractive index, excess viscosity, excess molar refraction, excess molar polarisation, and Dunsten's constant were all calculated using these values. These parameters' changes with composition and the impact of bonding in binary mixtures were discussed. In addition, they noted that the breaking of H-bonded related species produced between dissimilar molecules at 298 K is what causes the positive value of excess molar volume.[2]. Meenachi et.al. theoretically predicted the refractive index(n) of phenol with hydrocarbons at 303K. Anil Kumar K. et al. measured the reafractive indices(n) of the binary liquid mixture of 1,4-dioxane and 1-butanol at five different temperatures. The measurement was used to compute the values of excess refractive indices of this binary liquid mixture and related coefficients were calculated by using Redlich Kister polynomial equation[3].At atmospheric pressure, Emila M. Zivkovic et al. evaluated the refractive indices, densities, and viscosities of binary liquid solutions containing ethyl methyl ketone in the temperature range of 288.15K to 333.15K. Excess molar volumes, viscosity variances, and refractive index deviations were calculated using these measurements. Redlich-Kister equation was fitted to the calculated data.[4]. At a temperature of 298.15K, I.Y. Jeong et al. evaluated the densities and refractive indices of mixes of dimethyl carbonate, anisole, methanol, phenol, and water. The excess molar volumes and molar refractivity of these combinations were calculated using these measurements. In order to separate the reaction intermediates produced during the synthesis of non-phosgene diphenyl carbonate, they also evaluated the phase equilibria and mixture characteristics. The intermediates were based on anisole, phenol, and dimethyl carbonate. Additionally, they analytically established ternary liquid-liquid equilibria for the mixtures

(water + DMC + anisole) at 313.15K, (water + methanol + anisole) at 308.15K, and (water + methanol + phenol) at 328.15K, all at atmospheric pressure and constant temperature[5]. At 295.5 K, Khan et al. studied the electronic polarisation, density, and refractive indices of binary liquid mixes of ethanol with water and benzene. These observations were used to calculate the binary liquid mixes' molar refraction. They did this to confirm that the molar refraction is an additive and constitutive feature by comparing the theoretical and experimental values of the molar fraction. Additionally, they came to the conclusion that molar refraction was not influenced by intermolecular interactions[6].In binary liquid mixes of diethyl malonate with dimethylformamide, hexane, tetrahydrofuran, and 1,4-dioxane at 303.15K, Baluja et al. studied the refractive indices and densities. They contrasted these experimentally obtained values with those calculated theoretically using the Lorentz-Lorenz, Heller, Newton, and Gladstone-Dale mixing rules.[7]. At temperatures of 303.15K, 308.15K, and 313.15K over the whole range of ester composition, Rathnam et al. examined density, refractive index, and viscosity of the binary liquid mixes of diethyl maleate with ketones (acetophenone, cyclopentanone, cyclohexanone, and 3-pentnone). These data were used to determine the excess volume, viscosity deviation, and molar refraction deviation. Additionally, the theoretical values of viscosity and refractive index determined using the appropriate mixing principles were compared with the experimental viscosity and refractive index data[8]. For binary liquid mixes of butyl amine with 1-butanol and tert-butanol at 293K, 303K, and 313K over the whole composition range, S. Singh et al. examined the refractive index, density, viscosity, and ultrasonic velocity. These observations were used to determine the excess molar volume, molar refraction deviation, deviation in ultrasonic velocity, and viscosity deviation of these binary liquid mixes, which were then fitted to the Redlich-Kister polynomial graph. The intermolecular interactions present in these combinations are what they used to explain the positive and negative values of excess parameters that they found. Additionally, they contrasted the theoretically calculated refractive indices with those that were observed experimentally.[9]. The refractive indices of pure and binary liquid mixes of benzene and toluene at 293.15 K, heptane and hexane at 313.15 K, and heptane and acetic acid at 293.15 K were measured by Isehunwa S.O. et al. In the range of 293.15K to 308.15K, Janina Nowakowska tested the refractive indices of ethyl alcohol in water. These measured values of this binary liquid mixture's refractive index were contrasted with the experimental values obtained at the same temperature range[10]. The refractive index of a binary liquid mixture of benzene and carbon tetrachloride was measured by K. P. Damor et al. The researchers created a new DDJ equation, computed the result, and compared it to the result of the Lorentz-Lorenz mixing rule[11]. At standard atmospheric pressure at 303.15 K, N.H. Ansari et al. measured the refractive indices of six binary liquid mixes of N-butyl bromide with aniline, benzene, xylene, carbon tetrachloride, n-heptane, and toluene. They determined the molar refraction and deviation in molar refraction from ideal values of these mixtures, as well as the refractive index divergence from ideal values of these combinations. Additionally, they discussed the findings in terms of the interactions of molecules in the binary liquid mixtures[12].Ionic liquids (ILs), 1-butyl-3-methylimidazolium tetraflouroborate, 1-butyl-3-methylimidazolium bis (triflouromethylsulfonyl) imide, and 1-butyl-3-methylimidazolium methylsulfate with 1,2-porpanediol were evaluated using density and speed of sound tests over the full range of composition. Calculating extra molar volume required the measurement of density data. The binary coefficients and standard deviations were obtained by fitting the excess properties to the Redlich-Kister polynomial equation[13].For the binary liquid mixes of benzyl alcohol with chloro and nitro toluene over the complete composition range, L. Venkatramana et al. examined the density in the

temperature range 298.15K - 313.15K and the sound speed at 303.15K and 313.15K. The excess molar volume, excess speed of sound, isentropic compressibility, and excess isentropic compressibility were calculated using the measured density and speed of sound data. Additionally, they used theoretical models like Schaaff's collision theory and Jacobson's free length theory to examine the speed of sound data[14].The density of pure liquids and their mixtures were measured by L. Venkatramana et al. for binary mixtures of benzyl alcohol with 1-heptanol, 1-octanol, 1-nonanol, and 1-decanol at 298.15K to 313.15K spanning the full composition range. Additionally, they measured the binary liquid combinations' sound speeds[15].

This article aims to investigate the refractive index of binary liquid mixtures of acetophenone with selected acetates using theoretical mixing rules. By employing various mixing rules, we intend to predict the refractive index behaviour across various compositions and temperatures. This study will contribute to a deeper understanding of the intermolecular forces and molecular interactions governing the optical properties of these liquid mixtures.

II. METHODOLOGY

Mixtures of Acetophenone with all other three chemicals were considered for the theoretical estimation of refractive Index at three different temperatures (T= 303.15K, 313.15K and 323.15K) with increasing molar fraction of acetophenone. The three mixtures are as follows:

Acetophenone – Methyl Acetate
Acetophenone – Ethyl Acetate
Acetophenone – Propyl Acetate

The Refractive index of considered liquid mixtures were obtained using four different relations which are given below:

1. **Gladstone Dale Equation:** It is given as

$$\frac{(n_{12} - 1)}{r_{12}} = \frac{n_1 - 1}{r_1} w_1 + \frac{n_2 - 1}{r_2} w_2$$

where r_{12} is the density of liquid mixture, r_1, w_1 and r_2, w_2 are the density and weight fraction of pure components 1 and 2 respectively.

2. **Heller's Equation:** It is based on light scattering equation of Debye and Rayleigh and is given by

$$\frac{(n_{12} - n_1)}{n_1} = \frac{3}{2} f_2 \left[\left(\frac{n_2}{n_1}\right)^2 - 1 \right] / \left[\left(\frac{n_2}{n_1}\right)^2 + 2 \right]$$

where n_{12} is refractive index of the mixture, n_1 and n_2 are refractive indices of pure components 1 and 2 respectively. f_1 and f_2 are volume fractions of components 1 and 2 respectively and is given by:

$$f_i = \frac{x_i V_i}{\sum x_i V_i}$$

where x_i and V_i are the mole fraction and molar volume of ith constituent of binary mixture.

3. **Newton's Equation:** It applies to isotropic bodies of spherically symmetrical shape and proposes volume additivity and is given by:

$$(n_{12}^2 - 1) = (n_1^2 - 1)\phi_1 + (n_2^2 - 1)\phi_2$$

4. **Lorentz- Lorentz Equation:** It is given by

$$\frac{(n_{12}^2 - 1)}{(n_{12}^2 + 2)} = \phi_1 \frac{(n_1^2 - 1)}{(n_1^2 + 2)} + \phi_2 \frac{(n_2^2 - 1)}{(n_2^2 + 2)}$$

III. RESULTS AND DISCUSSION

Tables 1 to 3 shows the refractive index of binary liquid mixture of Acetophenone with Acetates (Methyl acetate, Ethyl acetate and Propyl acetate) at temperature 303.15K, 313.15K and 323.15K with respect to the mole fraction (x_1) of Acetophenone.

Figures 1 to 3 further illustrate the trends observed in the refractive index of the binary liquid mixtures of Acetophenone with Acetates (Methyl acetate, Ethyl acetate and Propyl acetate) at temperature 303.15K, 313.15K and 323.15K with respect to the mole fraction (x_1) of Acetophenone. These graphical representations provide a visual insight into the variations of the refractive index with changing mole fractions of Acetophenone. The graphs clearly depict how the refractive index responds to different concentrations of Acetophenone at each temperature, emphasizing the role of molecular interactions and composition in influencing optical behaviour. Comparing the results obtained from the four mixing rules reveals intriguing insights into the accuracy and reliability of each approach in predicting the refractive index behaviour. The variations in refractive index values across different temperatures and compositions underscore the complex nature of these binary liquid mixtures and the need for robust models to describe their optical properties accurately. The observed trends and patterns in the refractive index data shed light on the molecular interactions and structural changes occurring within the binary liquid mixtures. These findings deepen our understanding of the optical behaviour of Acetophenone-Acetate systems and pave the way for their potential applications in diverse fields, including pharmaceuticals, cosmetics, and materials science.

Table 1: Refractive Index of (Acetophenone + Methyl acetate) mixture at 303.15K, 313.15K and 323.15K by Gladstone- Dale, Hellar, Newton's and Lorentz- Lorentz Mixing Rules.

Mole Fraction (x_i)	303.15K					313.15K					323.15K				
	n_m GD	n_m GD	n_m Exp.	n_m H	n_m N	n_m L-L	n_m Exp.	n_m H	n_m N	n_m L-L	n_m Exp.	n_m H	n_m N	n_m L-L	n_m Exp.
0.0637	1.3666	1.3666	1.3716	1.3628	1.3648	1.3509	1.3571	1.3591	1.3490	1.3659	1.3562	1.3524	1.3544	1.3443	1.3612
0.1313	1.3817	1.3817	1.3867	1.3789	1.3799	1.3757	1.3719	1.3739	1.3638	1.3807	1.3606	1.3668	1.3698	1.3587	1.3756
0.2032	1.3968	1.3968	1.4018	1.3930	1.3950	1.3906	1.3868	1.3888	1.3787	1.3956	1.3851	1.3813	1.3833	1.3732	1.3901
0.2799	1.4119	1.4119	1.4169	1.4081	1.4101	1.4054	1.4016	1.4036	1.3936	1.4104	1.3994	1.3976	1.3976	1.3875	1.4044
0.3617	1.4171	1.4171	1.4321	1.4233	1.4253	1.4202	1.4164	1.4174	1.4083	1.4252	1.4138	1.4100	1.4120	1.4019	1.4188
0.4494	1.4422	1.4422	1.4472	1.4384	1.4404	1.4371	1.4313	1.4333	1.4232	1.4401	1.4282	1.4244	1.4264	1.4163	1.4332
0.5434	1.4573	1.4573	1.4623	1.4535	1.4455	1.4498	1.4460	1.4480	1.4379	1.4548	1.4466	1.4388	1.4408	1.4307	1.4476
0.6446	1.4723	1.4723	1.4773	1.4685	1.4705	1.4747	1.4609	1.4629	1.4528	1.4697	1.4571	1.4533	1.4453	1.4452	1.4621
0.7537	1.4875	1.4875	1.4925	1.4837	1.4857	1.4795	1.4757	1.4777	1.4576	1.4845	1.4714	1.4676	1.4696	1.4595	1.4764
0.8718	1.5025	1.5025	1.5075	1.4987	1.5007	1.4944	1.4906	1.4926	1.4825	1.4994	1.4867	1.4829	1.4849	1.4748	1.4917

Table 2: Refractive Index of (Acetophenone + Ethyl acetate) mixture at 303.15K, 313.15K and 323.15K by Gladstone- Dale, Hellar, Newton's and Lorentz- Lorentz Mixing Rules.

Mole Fraction (x_i)	303.15K					313.15K					323.15K				
	n_m GD	n_m GD	n_m Exp.	n_m H	n_m N	n_m L-L	n_m Exp.	n_m H	n_m N	n_m L-L	n_m Exp.	n_m H	n_m N	n_m L-L	n_m Exp.
0.0775	1.3786	1.3728	1.3748	1.3247	1.3816	1.3728	1.3670	1.3690	1.3589	1.3758	1.3248	1.3610	1.3630	1.3529	1.3698
0.1574	1.3908	1.3870	1.3830	1.3789	1.3958	1.3850	1.3712	1.3832	1.3731	1.3900	1.3785	1.3737	1.3787	1.3656	1.3835
0.2396	1.4047	1.4029	1.4029	1.3988	1.4097	1.3991	1.3953	1.3973	1.3872	1.4041	1.3951	1.3883	1.3903	1.3802	1.3971
0.3244	1.4188	1.4150	1.4170	1.4069	1.4238	1.4132	1.4094	1.4134	1.4113	1.4182	1.4057	1.4119	1.4039	1.3938	1.4107
0.4119	1.4319	1.4291	1.4211	1.4210	1.4379	1.4274	1.4236	1.4256	1.4155	1.4324	1.4194	1.4156	1.4176	1.4075	1.4244
0.5021	1.4468	1.4430	1.4450	1.4349	1.4518	1.4435	1.4397	1.4397	1.4296	1.4465	1.4330	1.4292	1.4322	1.4231	1.4380
0.5953	1.4609	1.4581	1.4591	1.4490	1.4659	1.4556	1.4518	1.4538	1.4437	1.4606	1.4436	1.4418	1.4448	1.4347	1.4516
0.6915	1.4750	1.4712	1.4742	1.4621	1.4800	1.4697	1.4659	1.4679	1.4578	1.4747	1.4602	1.4564	1.4574	1.4483	1.4652
0.7909	1.4892	1.4854	1.4874	1.4773	1.4942	1.4839	1.4801	1.4821	1.4710	1.4889	1.4739	1.4731	1.4721	1.4610	1.4789
0.8937	1.5032	1.4994	1.5014	1.4903	1.5082	1.4991	1.4943	1.4963	1.4862	1.5031	1.4865	1.4837	1.4857	1.4756	1.4925

Table 3: Refractive Index of (Acetophenone + Propyl acetate) mixture at 303.15K, 313.15K and 323.15K by Gladstone- Dale, Hellar, Newton's and Lorentz- Lorentz Mixing Rules.

| Mole Fraction (xi)	303.15K					313.15K					323.15K				
	n_m GD	n_m GD	n_m Exp.	n_m H	n_m N	n_m L-L	n_m Exp.	n_m H	n_m N	n_m L-L	n_m Exp.	n_m H	n_m N	n_m L-L	n_m Exp.
0.0896	1.3772	1.3834	1.3854	1.3753	1.3922	1.3839	1.3781	1.3801	1.3700	1.3869	1.3761	1.3723	1.3743	1.3642	1.3811
0.1794	1.4002	1.3964	1.3984	1.3883	1.4052	1.3949	1.3911	1.3921	1.3830	1.3999	1.3875	1.3847	1.3867	1.3766	1.3935
0.2695	1.4122	1.4194	1.4144	1.4013	1.4182	1.4070	1.4042	1.4062	1.3961	1.4130	1.4110	1.3972	1.3992	1.3851	1.4060
0.3599	1.4262	1.4224	1.4244	1.4153	1.4312	1.4210	1.4142	1.4192	1.4191	1.4260	1.4136	1.4098	1.4118	1.4017	1.4186
0.4506	1.4392	1.4384	1.4374	1.4273	1.4442	1.4340	1.4302	1.4322	1.4121	1.4390	1.4260	1.4122	1.4342	1.4141	1.4310
0.5415	1.4521	1.4483	1.4503	1.4402	1.4571	1.4480	1.4432	1.4452	1.4351	1.4520	1.4385	1.4347	1.4367	1.4266	1.4435
0.6326	1.4551	1.4613	1.4633	1.4542	1.4701	1.4600	1.4562	1.4542	1.4491	1.4650	1.4510	1.4472	1.4492	1.4391	1.4560
0.7241	1.4781	1.4743	1.4773	1.4662	1.4831	1.4731	1.4593	1.4713	1.4612	1.4781	1.4635	1.4597	1.4617	1.4516	1.4685
0.8158	1.4912	1.4863	1.4893	1.4792	1.4961	1.4861	1.4823	1.4843	1.4752	1.4911	1.4750	1.4722	1.4742	1.4631	1.4810
0.9078	1.5051	1.5003	1.5023	1.4922	1.5091	1.4991	1.4953	1.4973	1.4872	1.5041	1.4885	1.4847	1.4867	1.4766	1.4935
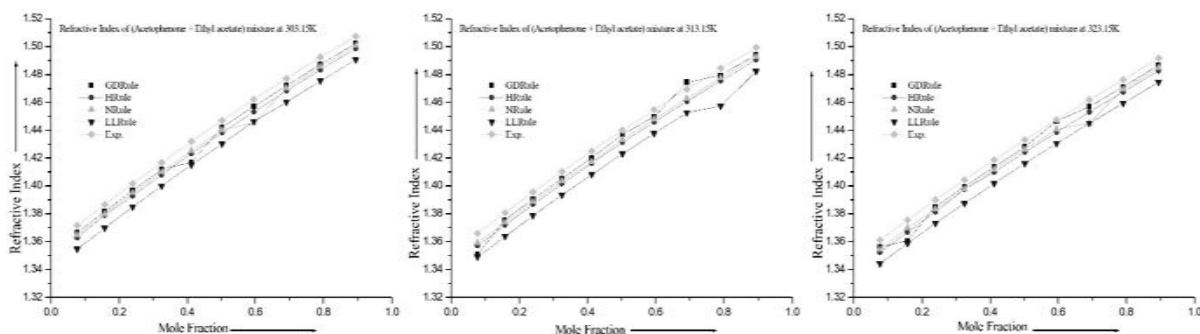
Figure 1: Mole fraction versus Refractive Index of Acetophenone + Methyl Acetate at 303K, 313.15K and 323.15K
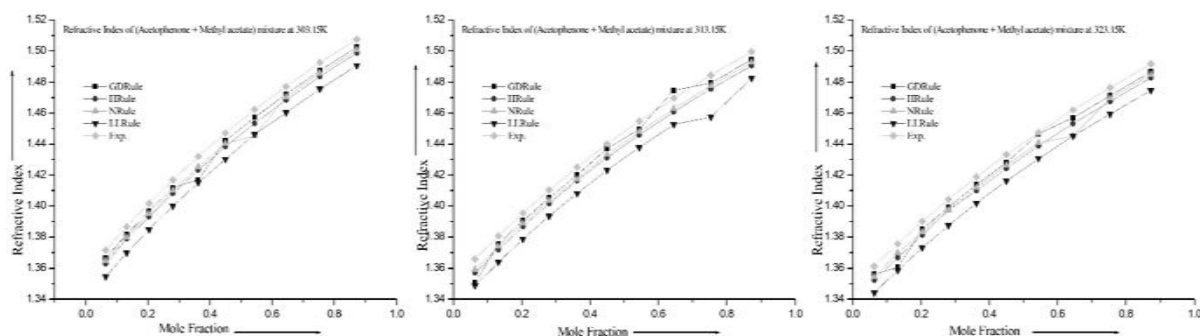

Figure 2: Mole fraction versus Refractive Index of Acetophenone + Ethyl Acetate at 303K, 313.15K and 323.15K
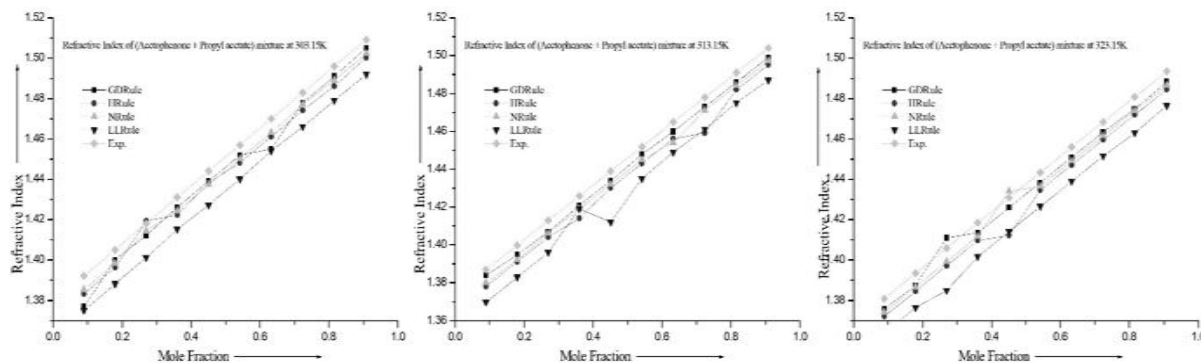
Figure 3: Mole fraction versus Refractive Index of Acetophenone + Propyl Acetate at 303K, 313.15K and 323.15K

IV. CONCLUSION

In conclusion, our investigation into the refractive indices of binary liquid mixtures consisting of Acetophenone and Acetates (Methyl acetate, Ethyl acetate, and Propyl acetate) has provided valuable insights into the optical behavior of these systems. Using four different mixing rules, we have examined the intricate relationship between composition, temperature, and refractive index, offering a comprehensive understanding of the studied mixtures. The data presented in Tables 1 to 3 and Figures 1 to 3 highlight the significant impact of both composition and temperature on the refractive indices of the binary liquid mixtures. The diverse patterns observed underscore the complex interplay between the different components and their interactions, reflecting the intricate nature of these systems. Our findings contribute to the fundamental understanding of these binary liquid mixtures and their optical properties. The four mixing rules employed in this study have allowed us to approach the investigation from multiple perspectives, enhancing the robustness of our conclusions and supporting potential applications across various fields, including pharmaceuticals, perfumes, solvents, and coatings. In essence, the findings presented in this study contribute to the broader understanding of the complex interactions within binary liquid mixtures and their potential significance in diverse industrial sectors. We believe that this research lays a foundation for further investigations and applications in the realm of optical properties and compositional behaviour of liquid mixtures.

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