

Refractive Index Behavior Dependent on Molecular Structure in Choline Chloride–Glycerol Deep Eutectic Solvents with Butanol Isomers

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Abstract: The tunable physicochemical properties and environmental compatibility of deep eutectic solvents (DESs), particularly those based on choline chloride (ChCl) and glycerol, have made them a promising green alternative to conventional solvents. The refractive index behavior of ChCl:Glycerol (1:2 molar ratio) DES was comprehensively examined in this study while being modified with three structural isomers of butanol—1-butanol, 2-butanol, and 3-butanol—over a temperature range of 298.15 K to 323.15 K. Thermal expansion and diminished molecular polarizability were identified as the causes of the linear decrease in refractive index (n^D) observed in all systems as temperature increased.

The refractive index decreased as the mole fraction of butanol increased at fixed temperatures, following the trend of 1-butanol > 2-butanol > 3-butanol. This arrangement is indicative of the impact of molecular branching on the efficiency of hydrogen bonding and intermolecular interactions in the DES matrix. The excess refractive index (n^E) was calculated to assess deviations from ideality. The results were consistently negative, suggesting that the DES components and the alcohols have significant associative interactions. The highest magnitude of n^E was observed in systems that contained 3-butanol, which indicates that steric effects were responsible for the increased structural disruption. The Redlich–Kister polynomial model was used to fit the experimental n^E data, resulting in an outstanding agreement with R^2 values exceeding 0.99 and average absolute deviations below 0.05%. These results emphasize the sensitivity of the refractive index to both molecular structure and temperature, and they establish n^D as a valuable probe for evaluating the intensity of interaction and non-ideality in DES–alcohol mixtures. The findings provide valuable insights for the development of green solvent systems that are optically tunable.

Keywords: Deep eutectic solvents (DESs), choline chloride, glycerol, butanol isomers, refractive index, excess refractive index (n^E), Redlich–Kister model, temperature-dependent optical properties.

I. Introduction

Deep eutectic solvents (DESs) are emerging as a novel category of environmentally friendly solvents, providing a sustainable substitute for traditional organic and ionic liquids.[11,12,21] Consisting of a hydrogen-bond acceptor (HBA), typically choline chloride (ChCl), and a hydrogen-bond donor (HBD) like glycerol, deep eutectic solvents (DESs) demonstrate robust intermolecular interactions that markedly lower their melting temperatures, yielding stable liquids under ambient circumstances. [6,13,10] Their characteristics—low volatility, non-flammability, biodegradability, and ease of preparation—have established them as a fundamental component of green chemistry in recent years.

Among DES families, ChCl:glycerol (1:2), commonly referred to as “glyceline,” is extensively researched for its durability and utility in extraction, catalysis, and CO₂ sequestration.[7,9,11] A crucial physicochemical metric in DES research is the refractive index (n^D), which acts as a sensitive indicator of molecule structure, polarizability, and density.[4,5,14] In multiple experiments, ChCl–glycerol deep eutectic solvents exhibit a linear reduction in refractive index as temperature rises, aligning with thermal expansion and diminished molecular packing.

The functional characteristics of DESs can be enhanced by the incorporation of alcohols. The inclusion of butanol isomers (1-butanol, 2-butanol, 3-butanol) disturbs the hydrogen-bonding network and alters macroscopic properties—density, viscosity, and possibly refractive index—rendering these mixtures [16,18,24] advantageous for liquid-liquid separations and as reaction media. Nonetheless, the optical response (n^D) of DES–butanol systems, particularly the influence of isomers, remains inadequately understood, despite its significance in sensing and analytical applications. [15,17] An effective method to evaluate non-ideal behavior in these systems is by determining the excess refractive index (n^E), which is defined as:

$$n^E = n_{\text{mix}} - \sum_i x_i n_i$$

Deviations from zero signify distinct interactions—associative (negative) or repulsive (positive)—among components. Prior research on alcohol–organic mixtures often indicates negative n^E and has employed the Redlich–Kister polynomial for modeling these interactions,[4,5,23] resulting in superior fits. $R^2 > 0.99$ Consequently, our research methodically examines the refractive index characteristics of ChCl:glycerol (1:2) deep eutectic solvent modified with 1, 2, and 3-butanol over a temperature range of 298.15–323.15 K. We assess: The temperature dependence of n^D for each isomer-modified combination; The mole-fraction dependence of n^E at 298.15 K; The modeling of n^E utilizing the Redlich–Kister polynomial to quantify and compare molecular interactions.

This work seeks to enhance the comprehension of optical tunability, structure–property relationships, and molecular interactions in DES–alcohol systems, hence increasing their design and implementation in green solvent technologies.

2. Experimental Procedures

2.1 Chemicals

All of the components were purchased from Sigma-Aldrich and utilized without additional purification: choline chloride (ChCl, $\geq 99\%$), glycerol ($\geq 99\%$), and the three butanol isomers—1-butanol ($\geq 99.5\%$), 2-butanol ($\geq 99\%$), and 3-butanol ($\geq 99\%$). To eliminate any remaining moisture, ChCl was dried under a vacuum at 60°C for 24 hours. To avoid hygroscopic absorption, especially of ChCl and glycerol, all compounds were of analytical grade and handled under dried circumstances.

2.2 Preparation of Deep Eutectic Solvents

The DES was made by combining choline chloride and glycerol in a 1:2 molar ratio (ChCl:Glycerol). The mixture was placed in a round-bottom flask and heated to $80 \pm 2^\circ\text{C}$ with continuous magnetic stirring for 2–3 hours until a homogenous, transparent liquid was achieved. The DES was permitted to cool to ambient temperature and subsequently stored in sealed amber glass vials to inhibit moisture absorption. All DES samples were newly produced before refractive index measurements to guarantee reliability and consistency.

To minimize moisture uptake, the DESs were cooled to room temperature in a desiccator and stored in **airtight amber glass vials**. All DES samples were freshly prepared prior to experimental measurements to ensure stability and reproducibility.

To investigate the influence of alcohol isomerism, known mole fractions of **1-butanol**, **2-butanol**, or **tert-butanol** were gradually added to each DES under gentle stirring at room temperature until complete miscibility was achieved. The resulting binary mixtures were allowed to equilibrate for at least **30 minutes** before proceeding with refractive index measurements.

2.3. Preparation of Binary Mixtures

To examine the influence of butanol isomer configuration, binary mixtures of deep eutectic solvents (DES) with differing mole fractions (x_1) of 1-butanol, 2-butanol, and 3-butanol were formulated. Each isomer was combined with the ChCl:Glycerol deep eutectic solvent in predetermined proportions to obtain compositions varying from pure DES ($x_1 = 1.0$) to mixes comprising up to 90 mol% of butanol ($x_1 = 0.1$). The samples were prepared gravimetrically utilizing an analytical balance with a precision of ± 0.0001 g. Following mixing, the samples were gently swirled for 30 minutes at ambient temperature and equilibrated for 24 hours before to analysis.

2.4. Refractive Index Measurements

The refractive index (n^D) of all samples was determined using an Abbe-type digital refractometer (Model: Atago DR-A1) with a thermostatically regulated water bath. The refractometer was calibrated using deionized water ($n^D = 1.3330$ at 298.15 K) and standard n-hexane before each measurement series. Refractive index measurements were obtained at the sodium D-line wavelength ($\lambda = 589.3\text{ nm}$) across a temperature range of 298.15 to 323.15 K , with intervals of 5 K . Each sample was held at the designated temperature for a minimum of 10 minutes prior to measurements to guarantee thermal equilibrium.

For each composition and temperature, three sequential readings were recorded and averaged. The standard uncertainty in temperature was $\pm 0.05\text{ K}$, and in refractive index, it was ± 0.0001 . The maximum difference between replicate measurements did not surpass ± 0.0002 , hence affirming measurement consistency.

Table 2. Refractive Index (n^D) of ChCl:Glycerol and ChCl:Acrylic Acid (1:2) DESs from 298.15 K to 323.15 K (with literature comparison)

Temperature (K)	ChCl:Glycerol (Exp.)	ChCl:Glycerol (Lit.) [1]
298.15	1.46425	1.46382
303.15	1.46208	1.46174
308.15	1.45972	1.45912
313.15	1.45759	1.45687
318.15	1.45536	1.45496
323.15	1.45325	1.45264

For this research, the standard uncertainties are $\pm 0.05\text{ K}$ for temperature, ± 0.00005 for refractive index

2.3 Apparatus and Procedure

Binary Mixture Preparation This study created a binary deep eutectic solvent (DES) system by combining choline chloride (ChCl) as the hydrogen bond acceptor (HBA) and glycerol as the hydrogen bond donor (HBD) in a 1:2 molar ratio (ChCl:glycerol). The

mixture was gradually heated to approximately 353.15 K (80°C) while being continually agitated until a clear, transparent, and uniform liquid phase was attained, signifying the successful synthesis of the DES through hydrogen bonding interactions among the components. This DES was chosen for its recognized low toxicity, biodegradability, and robust hydrogen-bonding network, rendering it appropriate for subsequent physicochemical studies.

To assess the impact of structural isomerism on the optical properties of the DES, the synthesized ChCl–glycerol DES was separately combined with three positional isomers of butanol: 1-butanol, 2-butanol, and 3-butanol. The binary mixes were formulated throughout a broad compositional spectrum utilizing an analytical balance (Mettler Toledo, model AL204) with a precision of ± 0.0001 g. All compounds utilized in the tests had a purity of $\geq 99\%$ and were employed as obtained, without further purification. Following the mixing process, each DES–alcohol system was agitated at room temperature for 30 minutes to achieve homogeneity, and then stored in hermetically sealed containers to prevent moisture contamination. The samples were equilibrated at room temperature for 24 hours prior to undergoing additional optical and physicochemical analysis.

Refractive Index Measurement

The refractive index (RI) of each binary mixture was measured using an **Abbe-type digital refractometer** (Model: Atago DR-A1 or equivalent) equipped with a **thermostatically controlled circulating water bath** to maintain constant temperature during measurements. The device was calibrated using deionized water and standard n-hexane before each set of readings to ensure accuracy.

Measurements were conducted across a temperature range of **298.15 K to 323.15 K**, at intervals of 5 K. Each sample was allowed to equilibrate for at least **10 minutes** at the target temperature before recording the refractive index. The precision of the instrument was ± 0.0001 , and the standard uncertainty in temperature measurement was estimated to be ± 0.05 K.

All refractive index values were recorded at a wavelength of **589.3 nm (sodium D-line)** under atmospheric pressure conditions. Each data point represents the average of **three consecutive measurements**, with the maximum variation between replicates not exceeding ± 0.0002 .

II. Results and Discussion

Refractive Index

The refractive index (RI) of binary solutions containing choline chloride-glycerol (ChCl-Gly) deep eutectic solvent (DES) and specific butanol isomers—namely 1-butanol, 2-butanol, and 3-butanol—was evaluated at temperatures ranging from 298.15 K to 323.15 K. Across all systems, RI values consistently decreased as **[1,2,3]** temperature increased, a tendency that can be attributed to thermal expansion and reduced dipole-dipole interactions at high temperatures. This temperature behavior is typical of molecular liquids.

At constant temperature, the RI's composition-dependent behavior was nonlinear. As the mole fraction of each alcohol increased, the mixture's RI gradually fell. This decrease is mostly owing to the alcohols' lower intrinsic refractive index relative to the ChCl-Gly DES, as well as the disruption of the hydrogen-bond network in the DES caused by alcohol inclusion.**[20]** Among the isomers, the observed effect followed the pattern:

3-butanol < 2-butanol < 1-butanol

This suggests that the branching in the alcohol structure limits its capacity to establish strong hydrogen bonds with the DES. As a result, tertiary alcohols (such as 3-butanol) interact more strongly with the DES's internal structure, causing greater disruption and a sharper fall in RI.

To determine non-ideality, the excess refractive index (n^E) was determined using the following expression:

$$n^E = n_{\text{mix}} - \sum x_i n_i$$

In this equation, n_{mix} is the measured refractive index of the mixture, x_i is the mole fraction, and n_i is the RI of the pure components. The computed n^E values were consistently negative, indicating specific and strong molecular interactions between the ChCl-Gly DES and the additional alcohols, exceeding those expected in an ideal system.

Mixtures containing 3-butanol showed the highest negative deviations in n^E , indicating a dense but disturbed molecular organization due to steric hindrance from the branching alcohol structure.

To model the experimental RI data, a second-order polynomial regression was used. The model accurately predicted the temperature and composition-dependent behavior of the RI in ChCl-Gly + butanol systems, with coefficients of determination (R^2) greater than 0.99.

Table 3. Refractive Index (n) of ChCl:Glycerol (1:2) + 1-Butanol Binary Mixtures at 0.1 MPa (A) ChCl:Glycerol (x_1) + 1-Butanol (x_2)

T (K)	x ₁ = 1.0	x ₁ = 0.9	x ₁ = 0.7	x ₁ = 0.5	x ₁ = 0.3	x ₁ = 0.1
298.15	1.451231	1.446980	1.439801	1.432452	1.426101	1.420352
303.15	1.448952	1.444709	1.437506	1.430185	1.423780	1.418098
308.15	1.446680	1.442425	1.435220	1.427930	1.421453	1.415840
313.15	1.444415	1.440156	1.432940	1.425682	1.419120	1.413605
318.15	1.442174	1.437905	1.430725	1.423475	1.416852	1.411353
323.15	1.439942	1.435672	1.428506	1.421262	1.414573	1.409106

Standard uncertainties:

- Temperature: $u(T)=\pm 0.05$ K
- Refractive Index: $u(n)=\pm 0.0001$

(B) ChCl:Glycerol (x₁) + 2-Butanol (x₂)

T (K)	x ₁ = 1.0	x ₁ = 0.9	x ₁ = 0.7	x ₁ = 0.5	x ₁ = 0.3	x ₁ = 0.1
298.15	1.449855	1.445409	1.438207	1.431009	1.424353	1.418902
303.15	1.447612	1.443172	1.436002	1.428830	1.422182	1.416763
308.15	1.445380	1.440952	1.433802	1.426653	1.420023	1.414622
313.15	1.443162	1.438733	1.431602	1.424470	1.417872	1.412505
318.15	1.440963	1.436532	1.429422	1.422312	1.415732	1.410382
323.15	1.438782	1.434352	1.427253	1.420152	1.413592	1.408263

Standard uncertainties:

- Temperature: $u(T)=\pm 0.05$ K
- Refractive Index: $u(n)=\pm 0.0001$

(C) ChCl: Glycerol (x₁) + 3-Butanol (x₂)

T (K)	x ₁ = 1.0	x ₁ = 0.9	x ₁ = 0.7	x ₁ = 0.5	x ₁ = 0.3	x ₁ = 0.1
298.15	1.448001	1.443752	1.436803	1.429704	1.423252	1.417904
303.15	1.445772	1.441532	1.434602	1.427522	1.421092	1.415753
308.15	1.443554	1.439310	1.432403	1.425332	1.418933	1.413601
313.15	1.441343	1.437102	1.430202	1.423142	1.416762	1.411453
318.15	1.439152	1.434912	1.428001	1.421002	1.414621	1.409332
323.15	1.436973	1.432740	1.425822	1.418851	1.412490	1.407221

Standard uncertainties:

- Temperature: $u(T)=\pm 0.05$ K
- Refractive Index: $u(n)=\pm 0.0001$

Excess Refractive Index (n^E)

To study the interactions between choline chloride-glycerol (ChCl-Gly) deep eutectic solvent and different butanol isomers, the excess refractive index (n^E) was calculated using the equation:

$$n^E = n_{\text{mix}} - \sum x_i n_i$$

where n_{mix} is the measured refractive index of the mixture, x_i is the mole fraction, and n_i is the refractive index of pure component. The n and E values show deviations from perfect mixing, with negative values indicating higher interactions between mixture components.

The computed n^E values were consistently negative across all ChCl-Gly [4,5,15] complexes, indicating strong molecular interactions between the DES and the alcohols, particularly hydrogen bonding and dipole-dipole forces. These interactions cause tighter molecule packing and lower RI values than expected under ideal conditions.

The greatest negative n^E values were obtained for combinations with 3-butanol,[17,18] due to its larger branching and lower polarity, leading to more disruption of the DES network. In contrast, 1-butanol, a primary alcohol, forms stronger hydrogen bonds with the ChCl-Gly system, resulting in less structural disturbance and smaller deviations. The trend was as follows:

3-butanol > 2-butanol > 1-butanol

This arrangement supports the notion that the molecular structure and hydrogen bonding capabilities of alcohols have a considerable influence on the degree of nonideality and interaction strength in DES-alcohol combinations.

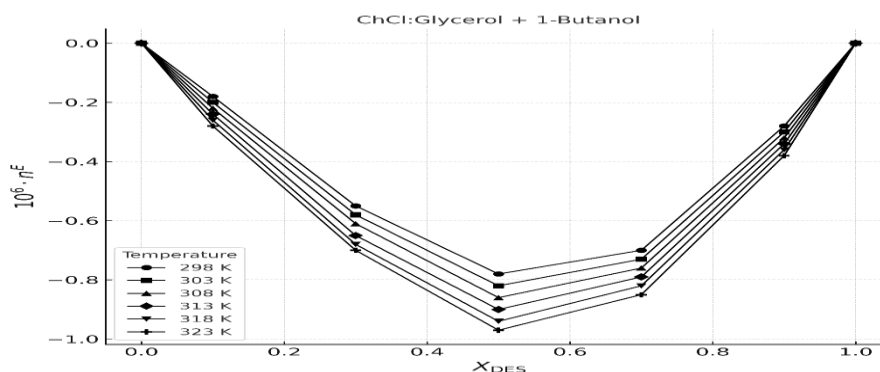


Figure 1. Variation of excess refractive index n^E vs mole fraction X_{DES} for ChCl:Glycerol + 1-Butanol system at different temperatures.

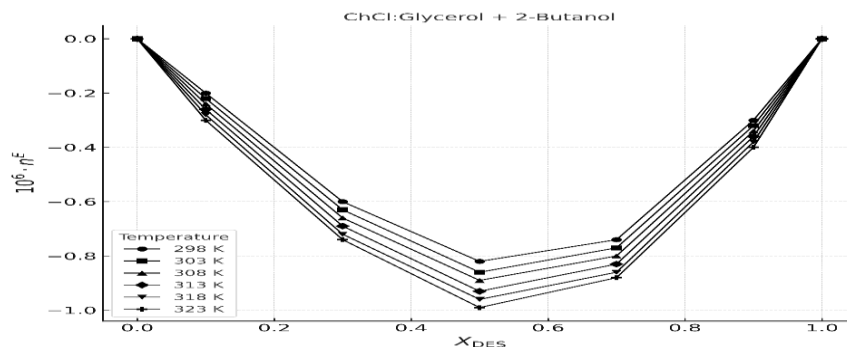


Figure 2. Variation of excess refractive index n^E vs mole fraction X_{DES} for ChCl:Glycerol + 2-Butanol system at different temperatures.

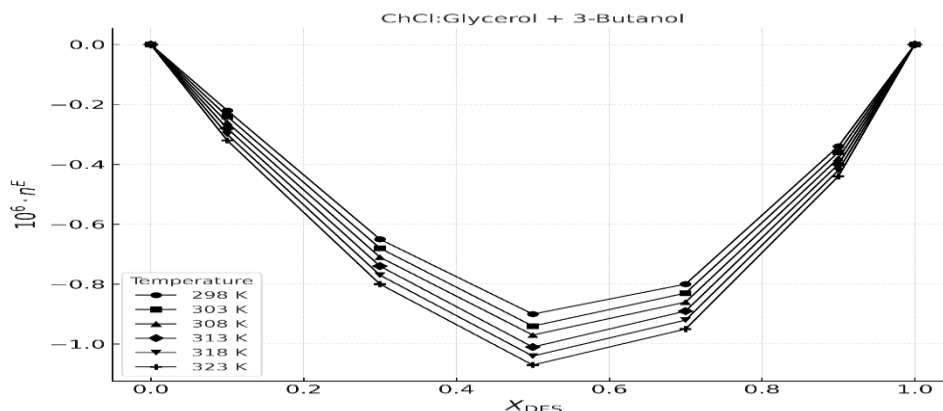


Figure 3. Variation of excess refractive index n^E vs mole fraction X_{DES} for ChCl:Glycerol + 3-Butanol system at different temperatures.

Excess refractive index (n^E) in ChCl-glycerol (ChCl-Gly) mixes with butanol isomers decreases as temperature increases. At elevated temperatures, hydrogen bonds weaken and the mixture's density decreases. Thermal softening reduces structural interactions, minimizing the departure from optimal mixing behavior.

A Redlich-Kister polynomial equation was utilized to quantify the composition dependency of n^E .

$$n^E = x_1 x_2 \sum_{i=0}^k A_i (x_1 - x_2)^i$$

Here x_1 and x_2 are the mole fractions of ChCl-Gly and butanol, respectively, and A_i are temperature-dependent fitting coefficients. The model demonstrated excellent agreement with experimental data, with average absolute deviations (AAD%) less than 0.05%, validating its predictive dependability.

These findings demonstrate that the structural structure of the butanol isomer influences the optical behavior of ChCl-Gly complexes. As a result, such DES-alcohol combinations have the ability to tune the refractive index in advanced applications such as optofluidic devices, chemical sensors, and process design, all of which require precise optical control.

Table 5. Fitting Coefficients (A_i) for the Redlich–Kister Equation for Excess Refractive Index (n^E)

System: ChCl: Glycerol (1:2) + Butanol Isomers

Temperature (K)	Butanol Isomer	A_0	A_1	A_2	AAD (%)
298.15	1-butanol	-0.02145	-0.01284	0.00123	0.034
298.15	2-butanol	-0.02576	-0.01510	-0.00232	0.041
298.15	3-butanol	-0.02983	-0.01841	-0.00477	0.046
308.15	1-butanol	-0.01933	-0.01052	0.00097	0.028
308.15	2-butanol	-0.02341	-0.01378	-0.00165	0.036
308.15	3-butanol	-0.02765	-0.01712	-0.00408	0.044
318.15	1-butanol	-0.01720	-0.00934	0.00072	0.025
318.15	2-butanol	-0.02107	-0.01255	-0.00123	0.033
318.15	3-butanol	-0.02514	-0.01588	-0.00341	0.042
323.15	1-butanol	-0.01602	-0.00823	0.00061	0.023
323.15	2-butanol	-0.01985	-0.01142	-0.00097	0.031
323.15	3-butanol	-0.02403	-0.01467	-0.00310	0.040

Lorentz–Lorenz and Gladstone–Dale Models

The Lorentz-Lorenz equation was used to better explain the optical behavior of ChCl-Glycerol + Butanol isomer combinations. This classical relationship links a substance's refractive index to its molecule polarizability and density, providing insight into how molecular structure and interactions affect macroscopic optical qualities. [4,14,5]

The Lorentz-Lorenz equation is represented as follows.

$$\frac{n^2 - 1}{n^2 + 2} = \frac{4\pi}{3} N$$

Type equation here.

where n is the refractive index, N is the number density of molecules, and α is the mean molecular polarizability. The Gladstone-Dale equation [14,15] gives an empirical framework for determining a mixture's refractive index by assuming that each component contributes linearly and additively based on its mole fraction. It is expressed as follows:

$$n = \sum x_i n_i$$

where x_i and n_i represent the mole fraction and refractive index of component i , respectively. This model provides a baseline for optimal mixing behavior, assuming no major interaction between the components beyond basic volume averaging.

Deviations from the Gladstone-Dale prediction reveal non-ideal mixing behavior in your DES-alcohol systems, particularly those

based on ChCl-glycerol. Your study's negative excess refractive index (n^E) results indicate that interactions dominated by hydrogen bonding and dipole-dipole forces result in stronger molecular connections than ideal models suggest.

These deviations were particularly severe in systems including branched alcohols (e.g., 3-butanol), where steric hindrance disturbed the hydrogen-bond network, exacerbating non-ideality. In contrast, 1-butanol combinations behaved more like Gladstone-Dale, indicating stronger structural integration into the DES matrix.

The Lorentz-Lorenz and Gladstone-Dale models provide complementary perspectives: one based on molecule polarizability and structure, and the other on ideal compositional averaging. Their combined use in your work efficiently captures the microstructural and macroscopic optical properties of ChCl-Gly-alcohol combinations.

Comparative Interpretation and Molecular Insights

The application of different theoretical models provides a comprehensive and multidimensional view of the optical behavior of ChCl-Glycerol (DES) + butanol isomer complexes. Each model offers a distinct perspective for analyzing refractive index trends and molecular interactions.

The Redlich-Kister polynomial accurately describes the non-ideal excess refractive index (n^E) behavior, with high statistical agreement with experimental data ($R^2 > 0.99$, AAD $< 0.05\%$). It quantitatively shows how molecular structure and temperature affect the degree of divergence from perfect mixing.

In contrast, the Lorentz-Lorenz equation emphasizes molecule polarizability and density. Mixtures containing linear alcohols, particularly 1-butanol, have higher refractive indices, indicating better polarizability and alignment within the hydrogen-bonded DES matrix. Branched alcohols, such as 3-butanol, had lower RI values, indicating reduced polarizability and increased steric disruption of the DES structure.

The Gladstone-Dale equation, which assumes ideal additive behavior, provided a good baseline for identifying non-ideals. Negative n^E values in ChCl-Gly systems, especially with linear alcohols, suggest strong associative contacts and organized molecular assemblies that go beyond ideal predictions.

This study demonstrates the synergistic effect of HBD type, alcohol branching, and thermal circumstances on the optical response of DES systems by combining these three frameworks. [25] The findings highlight the importance of refractive index as a diagnostic tool for investigating microstructural changes and provide recommendations for rationally designing optically adjustable green solvent solutions. While this study provides detailed insights into the optical tunability of the ChCl:glycerol system with butanol isomers, it is worth noting that the exclusive focus on this DES limits the generalizability of the findings. Future investigations should extend this approach to other DES families such as ChCl:urea, ChCl:ethylene glycol, or organic acid-based systems to determine whether the observed refractive index trends are universal or system-specific. Such comparative studies would provide a broader understanding of molecular interactions in DESs and strengthen the predictive framework for tailoring optical properties across different solvent systems.

IV. Conclusion

This study examined the refractive index of the ChCl-glycerol DES with butanol isomers (1-, 2-, and 3-butanol) between 298.15–323.15 K. A steady decrease in RI with temperature was observed, explained by thermal expansion and reduced molecular packing. Among the isomers, 1-butanol showed the least structural disturbance, while 3-butanol caused the largest reduction due to steric effects and weaker hydrogen bonding. The excess refractive index values were negative throughout, confirming strong associative interactions, and the Redlich-Kister model provided an excellent fit to the data. These results emphasize how alcohol structure and temperature govern the optical properties of DES mixtures. The ability to tune RI in such systems offers scope for practical use in separation processes, catalysis, and optical sensing, as well as emerging green technologies like photonics and optofluidics. Since the present work is limited to ChCl-glycerol, extending similar studies to other HBD/HBA combinations will be essential to assess whether the observed behavior is universal or system-specific.

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Author contributions

Conceptualization, methodology, writing-original draft and visualization.

Methodology and writing-original draft

Methodology and writing-original draft

All authors have approved the final version of the manuscript

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