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Research Article

Prediction of Refractive Indices of Binary Mixtures of Ionic Liquids and Water

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In the present work, we have successfully predicted the refractive indices of binary mixtures containing ionic liquids (ILs) and water as a component following the approach of Gladstone–Dale and Newton. These approaches were proposed as adequate to predict the refractive indices of mixtures containing imidazolium-based ILs with water or ethanol. It requires just the knowledge of the density and refractive index of pure components to obtain the value of refractive index of binary mixtures at any composition. We have selected 30 binary systems that contain ILs from imidazolium, ammonium, pyrrolidinium, and pyridinium families for validation. The tested data comprise of more than 400 mixture points. The results are presented in terms of average percentage deviations (APDs) from the experimental values. The Gladstone–Dale method gives better results compared to the Newton approach. The reasons why these approaches give large APDs for some mixtures are discussed in the manuscript.

1. Introduction

Ionic liquids (ILs) are compounds that are entirely composed of ions [1]. The mismatch of cation and anion sizes and insufficient crystal packing are the reasons that many of these salts exist in liquid form at or around room temperature [2, 3]. Due to their peculiar properties such as low melting point compared to conventional salts, large liquid range, wide electrochemical window, solvent properties, and designer nature, their use is widespread across many disciplines [4–8].

ILs are being used in many applications where due to their high viscosity, they are most often mixed with other molecular solvents [9–11]. These mixtures differ from the mixtures of molecular solvents where only neutral molecular entities are present [12]. For many biological applications, ILs are dissolved in water to form aqueous solutions [13]. Prior to their use in a given application, the physicochemical properties (e.g., density, viscosity, surface tension, and refractive index) of such mixtures and solutions are often required.

Refractive index is an important property that is often used to characterize materials, which indicates the ratio of the velocity of light in vacuum to the velocity of light in the material [14]. It is a thermodynamic property, which depends on temperature, pressure, and wavelength [15]. Compared to other bulk properties, the measurement of refractive index is relatively fast and convenient; therefore, it is often used to gain molecular insights between the component molecules.

A survey of the literature indicates that several workers have proposed the strategies to predict the refractive indices of pure ILs. Deetlefs et al. [16] presented a simple strategy to calculate refractive indices of pure ILs using parachor and molar refraction data obtained through group contribution method. Wang et al. [17] collected a large amount of data to construct a database which was used to propose a group contribution method to predict the refractive indices of ILs. Sattari et al. [18] developed a linear quantity structure property relation (QSPR) model using genetic function approximation (GFA) to predict the refractive indices of ILs.

More recently, extreme learning machine (ELM) intelligence algorithm was also used to predict the refractive indices of ILs [19].

As far as refractive indices of binary liquid mixtures containing ILs + water/molecular solvents are concerned, only a handful studies are available. Iglesias-Otero et al. [20] proposed correlations between the volumetric properties and refractive indices of binary mixtures of ILs with organic solvents. Similarly, Rilo et al. [21] presented an easy method for the prediction of refractive indices of mixtures of ILs with water/ethanol based on the approach of Gladstone–Dale and Newton. Soriano et al. [22] presented a model based on artificial neural network (ANN) to predict the refractive indices of binary solutions of ILs + alcohols. Goodarzi et al. [23] presented a predictive tool based on the least square support vector machine optimization to determine the refractive indices of mixture of IL + alcohols.

Tremendous progress has been made, in recent years, on the front of predicting refractive indices of ILs and binary mixture containing ILs; however, simple models are still unavailable. Some of the proposed sophisticated predictive models require special skills for their usage. Therefore, in this work, following the suggestions proposed by Rilo et al. [21], we have chosen the Gladstone–Dale and Newton method to determine the refractive indices of aqueous mixtures containing ILs as one of the component. Both these approaches are extremely user friendly and require just information about the refractive indices and density of pure components.

In order to validate these approaches, we have collected data for 30 IL + H_2O mixtures from the literature containing ILs from different families at 298.15 K [24–34]. One of the objectives of this work is to see whether the models work for functionalized imidazolium ILs and can it be applied successfully to other families as well?

2. Results and Discussion

2.1. Collection of Refractive Index Data for IL + H_2O Mixtures. Table 1 presents the density and refractive index data of neat ILs that were used in this work for the prediction of refractive indices of their binary mixtures with water at 298.15 K [24–34]. The data were used to calculate the molar volume and molar refraction of respective ILs. It must be stressed that the data for the common component (water) were slightly differing in magnitude from one source to another; therefore, an average value of its density and refractive index is reported in Table 1. However, in the calculations, the respective ρ and n_D data were used.

The data are presented in a chronological order of the year of its publication, and each mixture has been assigned a number denoted with Mn, where $n = 1$–30. The density and refractive index data for IL, 1-allyl-3-methylimidazolium chloride were not reported; therefore, the refractive index values for the binary system M4 cannot be calculated.

The density and refractive index variations for all the collected systems as a function of mixtures composition are plotted in Figure 1. It is clear from Figure 1 that the density-composition profile of IL + H_2O mixtures is system specific. In some cases, when the IL is mixed in water, the density increases up to a certain composition and afterwards reaches a plateau whereas, in some cases, the density increases linearly towards the density of IL. In a few cases, where the density of IL is lower than water, the density decreases almost linearly towards the density of ILs. In case of refractive indices, except for a few systems, on mixing the IL in water, the refractive index value increases rapidly up to a certain extent and then reaches almost linearly towards the IL refractive index.

2.2. Calculation of Refractive Index of Binary Mixtures. To calculate the refractive indices of mixtures, the following relation from Gladstone–Dale was used [35]:

$$n_D = \phi_{IL} n_{IL} + \phi_w n_w, \tag{1}$$

where n_D, n_{IL}, and n_w are the refractive index of binary mixture, IL, and water, respectively, whereas ϕ_{IL} and ϕ_w represent the volume fraction of IL and water.

The volume fractions can be defined as follows:

$$\phi_i = \frac{x_i M_i}{x_{IL} M_{IL} + x_w M_w} \frac{\rho}{\rho_i}, \tag{2}$$

where i represents one of the component (water or IL) of binary mixtures. The density ρ at any given composition of the mixtures can be calculated from the values of the density of pure components using the following expression:

$$\rho = \rho_{\mathrm{IL}} \rho_w \frac{(x_{IL} M_{IL} + x_w M_w)}{x_{IL} M_{IL} \rho_w + x_w M_w \rho_{\mathrm{IL}}}. \tag{3}$$

Therefore, the volume fraction of IL can be calculated as follows:

$$\phi_{\mathrm{IL}} = \frac{x_{\mathrm{IL}} M_{\mathrm{IL}} \rho_w}{x_{\mathrm{IL}} M_{\mathrm{IL}} \rho_w + x_w M_w \rho_{\mathrm{IL}}}. \tag{4}$$

Since, for a binary mixture, $\phi_w + \phi_{\mathrm{IL}} = 1$, the volume fraction of water for a given system can simply be calculated as follows:

$$\phi_w = 1 - \phi_{\mathrm{IL}}. \tag{5}$$

Similarly, the Newton relation to obtain the refractive indices of binary liquid mixtures using the values of pure components is indicated as follows [36]:

$$n_D = \sqrt{\left(\phi_{\mathrm{IL}} n_{\mathrm{IL}}^2 + \phi_w n_w^2\right)}. \tag{6}$$

The results of the prediction of both approaches for some selected systems are presented in Figure 2 whereas the same for all systems in the chronological order (according to Table 1) are given in the various subsections of Figure S1 of supplementary material. It is evident that both the methods are predicting the refractive index behavior of liquid mixtures quite accurately. There does not seem any effect of the family of IL (imidazolium, pyrrolidinium, pyridinium, and ammonium) on the predictive efficiency of both the approaches.

Table 1: Details of the properties, viz., molar mass (Mw/g·mol^{-1}), density (ρ/g·cm^{-3}), molar volume (Vm/cm^3·mol^{-1}), refractive index (n_D), and molar refraction (Rm/cm^3·mol^{-1}) of pure ionic liquids (ILs) belong to the binary IL + H_2O mixtures (Mn) used in this work to predict the refractive indices at 298.15 K [24–34].

M. no	Ionic liquid	Mw (g·mol^{-1})	ρ (g·cm^{-3})	Vm (cm^3·mol^{-1})	n_D	Rm (cm^3·mol^{-1})	Reference
	Water	18.015	0.9971	18.07	1.3324	3.71	Average*
M1	1-Ethyl-3-methylimidazolium ethylsulfate	236.29	1.2376	190.9	1.47940	54.18	Gómez et al. [24]
M2	Pyrrolidinium nitrate	134.13	1.1675	114.9	1.3955	27.57	Anouti et al. [25]
M3	Pyrrolidinium octanoate	215.33	0.9452	227.8	1.4581	62.17	Anouti et al. [26]
M4	1-Allyl-3-methyl-imidazolium chloride	158.63	na	na	na	na	Wu et al. [27]
M5	1-(2-Hydroxyethyl)-3-methylimidazolium tetrafluoroborate	213.97	1.3633	157.0	1.4252	40.15	Rives et al. [28]
M6	1-Ethyl-3-methylimidazolium trifluoromethanesulfonate	260.23	1.38358	188.1	1.4332	48.90	Vercher et al. [29]
M7	1-Butyl-1-ethylimidazolium trifluoromethanesulfonate	302.32	1.26313	239.3	1.43944	63.01	Vercher et al. [29]
M8	1-Butyl-3-methylimidazolium trifluoromethanesulfonat	288.29	1.29743	222.2	1.43729	58.25	Vercher et al. [29]
M9	1-Butyl-1-methylpyrrolidinium trifluoromethanesulfonate	291.33	1.25201	232.7	1.43267	60.44	Vercher et al. [29]
M10	1-Butyl-3-methylimidazolium dicyanamide	205.26	1.06017	193.6	1.50893	57.80	Gonzalez et al. [30]
M11	1-Butyl-3-methylimidazolium trifuoromethanesulfonate	288.29	1.29955	221.8	1.43755	58.18	Gonzalez et al. [30]
M12	1-Butyl-1-methylpyrrolidinium dicyanamide	208.30	1.01353	205.5	1.49681	60.12	Gonzalez et al. [30]
M13	1-Butyl-1-methylpyrrolidinium trifluoromethanesulfonate	291.33	1.25298	232.5	1.43289	60.42	Gonzalez et al. [30]
M14	1-Butyl-3-methylpyridinium trifluoromethanesulfonate	237.05	1.27941	185.3	1.46155	50.89	Gonzalez et al. [30]
M15	1,2- Diethylpyridinium ethylsulfate	261.34	1.21926	214.3	1.51350	64.48	Gonzalez et al. [30]
M16	1-Hexyl-3-methylimidazolium dicyanamide	233.31	1.02856	226.8	1.50415	67.18	Gonzalez et al. [30]
M17	1-Methylpyridinium methylsulfate	205.23	1.34612	152.5	1.51340	45.85	Gonzalez et al. [30]
M18	Ethylammonium acetate	105.14	1.01771	103.3	1.4345	26.93	Hou et al. [31]
M19	Propylammonium acetate	119.16	0.96682	123.2	1.4405	32.51	Hou et al. [31]
M20	Diethylammonium acetate	133.11	1.02141	130.3	1.431	33.73	Umapathi et al. [32]
M21	Diethylammonium hydrogen sulfate	171.06	1.28395	133.2	1.422	33.86	Umapathi et al. [32]
M22	Triethylammonium acetate	161.14	1.05186	153.2	1.501	45.13	Umapathi et al. [32]
M23	Triethylammonium hydrogen sulfate (TEAS)	199.09	1.14289	174.2	1.516	52.62	Umapathi et al. [32]
M24	Trimethylammonium acetate (TMAA)	119.05	1.05385	113.0	1.392	26.90	Umapathi et al. [32]
M25	Trimethylammonium hydrogen sulfate (TMAS)	157.03	1.46758	107.0	1.406	26.28	Umapathi et al. [32]
M26	2-Hydroxy ethylammonium butanoate	149.18	1.07259	139.1	1.4661	38.53	Rocha Pinto et al. [33]
M27	2-Hydroxy ethylammonium pentanoate	163.21	1.04354	156.4	1.4645	43.20	Rocha Pinto et al. [33]
M28	2-Hydroxy ethylammonium hexanoate	177.24	1.01995	173.8	1.4623	47.80	Rocha Pinto et al. [33]
M29	2-Hydroxy diethylammonium hexanoate	221.30	1.05871	209.0	1.4714	58.47	Rocha Pinto et al. [33]
M30	1,1,3,3-Tetramethylguanidine imidazolide	183.26	0.9928	184.6	1.4943	53.77	Ye et al. [34]

*The density and refractive index values were taken from all the sources and averaged.

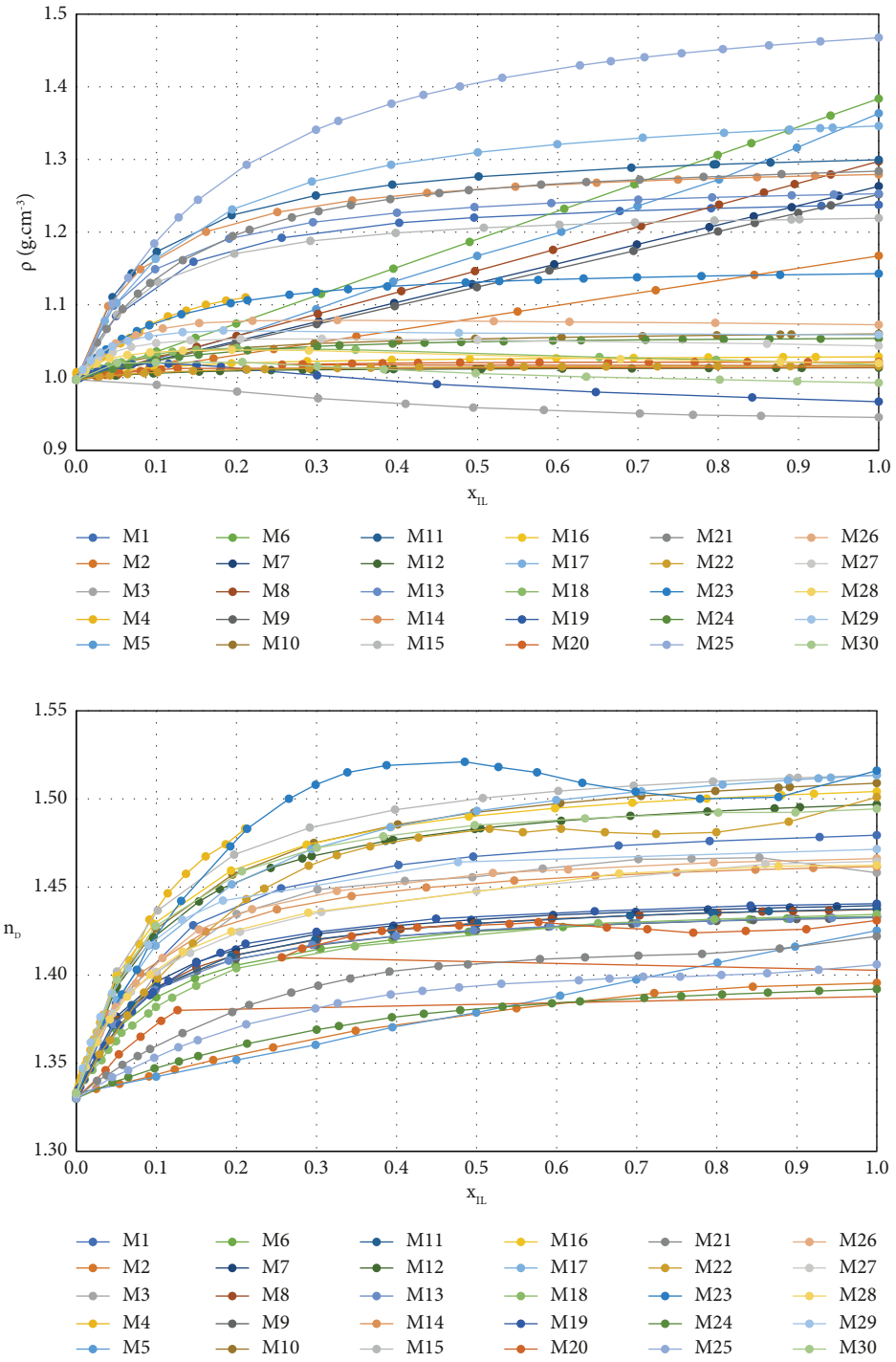

FIGURE 1: Density-composition (ρ-X_{IL}) and refractive index-composition (n_D-X_{IL}) profiles of the binary ionic liquid + H_2O systems used in this study reported at 298.15 K [24–34].

The results are also presented in terms of average percentage deviations (APDs), which were calculated as follows:

$$\text{APD} = \left(\frac{\sum_{i=1}^{N} \left(n_{\text{exp}} - n_{\text{cal}} \right)}{N} \times 100 \right), \tag{7}$$

where n_{exp} is the experimentally measured refractive index, n_{cal} is the calculated ones, and N is the number of data points. Table 2 lists the APDs obtained for both the approaches for each system.

It must be clarified that to understand the efficiency of the G-D and Newton approach, in Table 2, the systems are divided according to the families of ILs. Afterwards, the

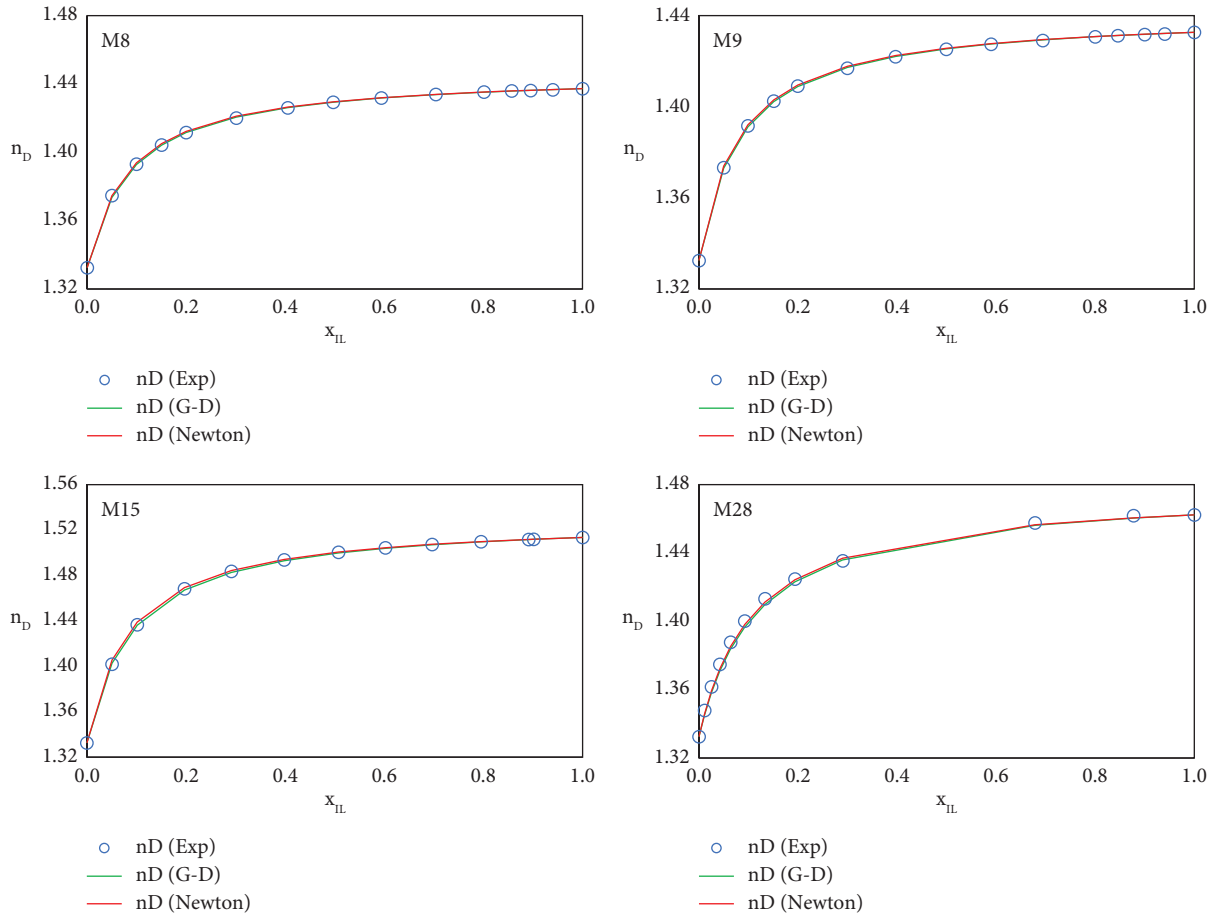

Figure 2: Experimental refractive index values (circular symbols) as a function of ionic liquid (IL) mole fraction (x_{IL}) for the four selected IL + H_2O systems belong to imidazolium (M8), pyrrolidinium (M9), pyridinium (M15), and ammonium (M28) families along with the predicted values using Gladstone–Dale (green line) and Newton (red line) approach at 298.15 K. The best results (lowest average percentage deviations) were obtained for these systems in respective families.

Table 2: Results of the predictive refractive index values at 298.15 K for binary mixtures of ionic liquids + H_2O systems using Gladstone–Dale (G-D) and Newton approach in terms of average percentage deviations (APD) from respective experimental values. The systems are divided in different cation families and arranged from lowest APD to highest APD magnitude in each group.

M. no	Ionic liquid	Data points	G-D	Newton	Reference
Imidazolium (8)					
M8	1-Butyl-3-methylimidazolium trifluoromethanesulfonat	15	0.0044	−0.0200	Vercher et al. [29]
M11	1-Butyl-3-methylimidazolium trifuoromethanesulfonate	13	−0.0047	−0.0330	Gonzalez et al. [30]
M7	1-Butyl-1-ethylimidazolium trifluoromethanesulfonate	15	−0.0118	−0.0364	Vercher et al. [29]
M6	1-Ethyl-3-methylimidazolium trifluoromethanesulfonate	15	0.0183	−0.0058	Vercher et al. [29]
M1	1-Ethyl-3-methylimidazolium ethylsulfate	10	0.0291	−0.0177	Gómez et al. [24]
M16	1-Hexyl-3-methylimidazolium dicyanamide	13	−0.0626	−0.1216	Gonzalez et al. [30]
M10	1-Butyl-3-methylimidazolium dicyanamide	13	−0.0737	−0.1391	Gonzalez et al. [30]
M5	1-(2-Hydroxyethyl)-3-methylimidazolium tetrafluoroborate	11	−1.8608	−1.8812	Rives et al. [28]
Pyrrolidinium (5)					
M9	1-Butyl-1-methylpyrrolidinium trifluoromethanesulfonate	15	−0.0010	−0.0230	Vercher et al. [29]
M13	1-Butyl-1-methylpyrrolidinium trifluoromethanesulfonate	13	0.0023	−0.0188	Gonzalez et al. [30]
M12	1-Butyl-1-methylpyrrolidinium dicyanamide	21	−0.0445	−0.1246	Gonzalez et al. [30]
M3	Pyrrolidinium octanoate	11	0.4253	0.3964	Anouti et al. [26]
M2	Pyrrolidinium nitrate	12	−0.6393	−0.6537	Anouti et al. [25]
Pyridinium (3)					
M15	1,2-Diethylpyridinium ethylsulfate	13	0.0214	−0.0444	Gonzalez et al. [30]
M17	1-Methylpyridinium methylsulfate	15	−0.0772	−0.1526	Gonzalez et al. [30]
M14	1-Butyl-3-methylpyridinium trifluoromethanesulfonate	13	0.1961	0.1573	Gonzalez et al. [30]

Table 2: Continued.

M. no	Ionic liquid	Data points	G-D	Newton	Reference
Ammonium (13)					
M28	2-Hydroxy ethylammonium hexanoate	12	0.1484	0.0903	Rocha Pinto et al. [33]
M27	2-Hydroxy ethylammonium pentanoate	12	0.1549	0.0930	Rocha Pinto et al. [33]
M20	Diethylammonium acetate	21	0.1989	0.1655	Umapathi et al. [32]
M25	Trimethylammonium hydrogen sulfate (TMAS)	21	−0.2157	−0.2349	Umapathi et al. [32]
M24	Trimethylammonium acetate (TMAA)	21	−0.2686	−0.2815	Umapathi et al. [32]
M30	1,1,3,3-Tetramethylguanidine imidazolide	11	0.3059	0.2429	Ye et al. [34]
M18	Ethylammonium acetate	18	0.3981	0.3594	Hou et al. [31]
M29	2-Hydroxy diethylammonium hexanoate	11	0.4337	0.3599	Rocha Pinto et al. [33]
M23	Triethylammonium hydrogen sulfate (TEAS)	21	0.4358	0.3293	Umapathi et al. [32]
M21	Diethylammonium hydrogen sulfate	21	−0.4621	−0.4926	Umapathi et al. [32]
M19	Propylammonium acetate	20	0.5106	0.4685	Hou et al. [31]
M26	2-Hydroxy ethylammonium butanoate	12	0.5630	0.5007	Rocha Pinto et al. [33]
M22	Triethylammonium acetate	21	−0.5690	−0.6603	Umapathi et al. [32]

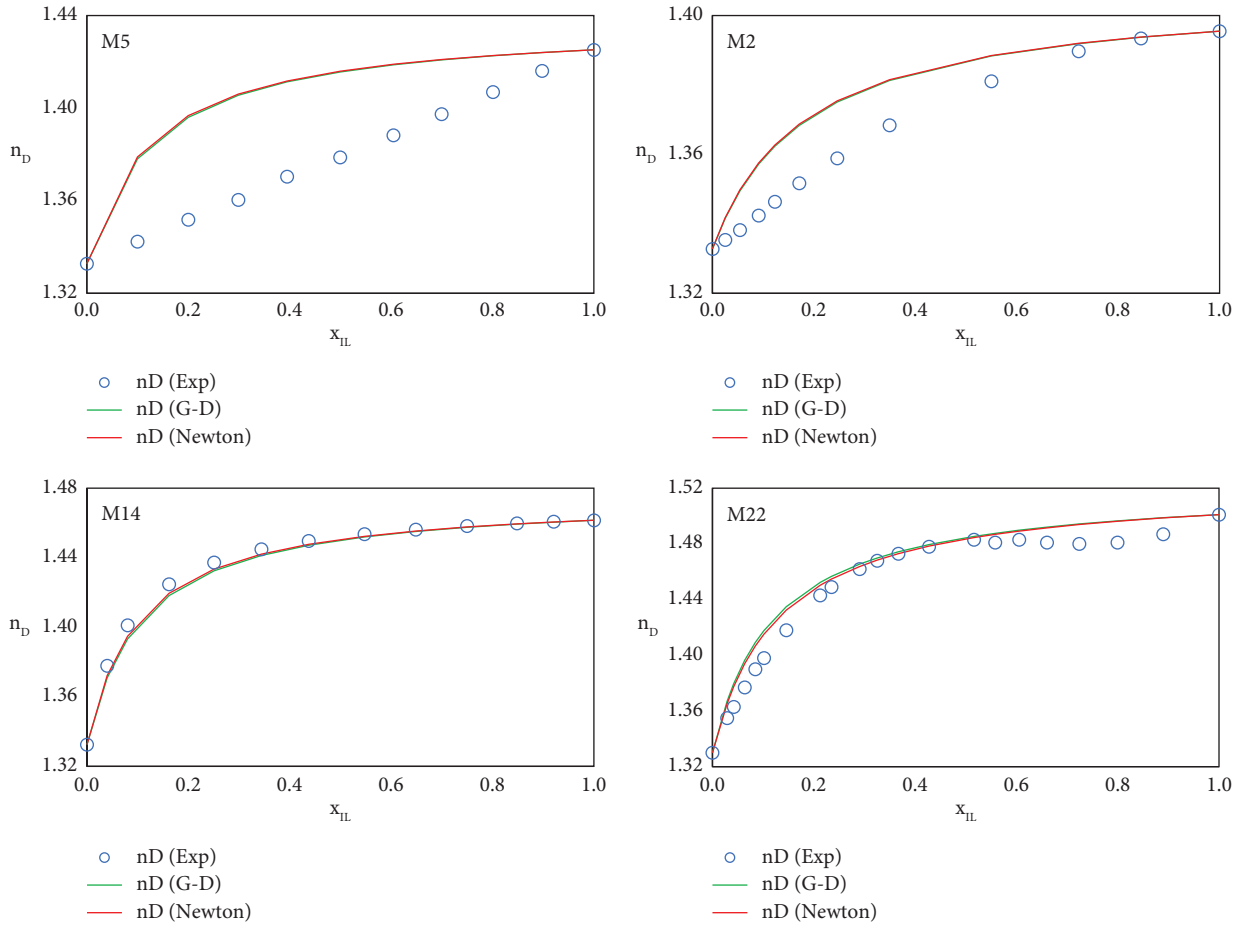

Figure 3: Experimental refractive index values (circular symbols) as a function of ionic liquid (IL) mole fraction (x_{IL}) for the four selected IL + H_2O systems belong to imidazolium (M5), pyrrolidinium (M2), pyridinium (M14), and ammonium (M22) families along with the predicted values using Gladstone–Dale (green line) and Newton (red line) approach at 298.15 K. The poorest results (highest average percentage deviations) were obtained for these systems in respective families.

systems are listed according to the lowest APD obtained to highest APD obtained in case of each family.

One can see that the obtained APD values are very small for both G-D and Newton methods. In general, the G-D approach is presenting lower APD values compared to the Newton. It shows the applicability of these approaches to obtain the refractive indices of IL + H_2O binary systems irrespective of the family of ILs. There are systems in each family that are showing APD values that are several orders of magnitude higher compared to the APDs of other systems in

the same group.. Four such systems, from each imidazolium, pyrrolidinium, pyridinium, and ammonium families, show that the highest APDs are presented in Figure 3. These results can be explained considering the applicability of these approaches to systems that show nearly ideal behavior [37].

For instance, in imidazolium family, the system containing IL 1-(2-hydroxyethyl)-3-methylimidazolium tetrafluoroborate + H_2O (M5) shows the highest APD within the family as well as among the systems undertook in this work (Table 2). It is clear from Figure 3 that both the approaches are giving completely different trend compared to the experimentally obtained linear trends of refractive index *versus* composition. It is interesting to note that this IL is very similar to other ILs studied in the imidazolium family except it is slightly functionalized with an additional -OH group attached to the cation. The excess volume for the system shows large negative values which indicate strong molecular interactions between the component molecules [28]. Since the system is not quasi-ideal, both the approaches were not able to consider the large change in volumes and failed to predict the correct trends for this binary system.

The other systems that show relatively high APD values among pyrrolidinium family are pyrrolidinium octanoate + H_2O (M3) and pyrrolidinium nitrate + H_2O (M2). It must be stressed that both of these ILs are protic ILs, and the excess volumes for these systems were large positive [25, 26]. The protic ILs influence the structure of water immensely like polar solvents [38].

In case of pyridinium family, both the approaches were able to satisfactorily predict the refractive index trends for the system M14, 1-butyl-3-methylpyridinium trifluoromethanesulfonate + H_2O, but the APD value was highest among the family. The excess volumes for the system show asymmetrical curves due to large difference in the molar masses of the component molecules [30].

The case for ammonium family is very different, as all the systems are showing highest APD values among the families considered in this work. One of the reason for this behavior is due to the fact that all these systems involve protic ILs that many a times show irregular volumetric trends [31, 32]. The thermophysical properties of protic IL systems behave in a peculiar manner and are therefore difficult to predict [9].

For all ionic liquids discussed in this manuscript, experimental refractive index values (circular symbols) as a function of ionic liquid (IL) mole fraction (x_{IL}) for the IL + H_2O systems used in this work along with the predicted values using Gladstone–Dale (green line) and Newton (red line) approach at 298.15 K are plotted in Figure S1.

3. Conclusions

The refractive indices of 30 binary mixtures containing ionic liquid and water as a common component were predicted using two simple approaches, namely, Gladstone–Dale and Newton. These approaches just require the knowledge of density and refractive indices of pure components. The results were presented in terms of average percentage deviations (APDs), and satisfactory outcomes from both the approaches were obtained. Systems that were not ideal in terms of large or asymmetrical excess volume trends give high APD values. Both these approaches can be applied to any family of ILs and to the systems that show quasi-ideal behavior. However, care must be taken to use it to predict the refractive index–composition trends for systems involving protic ILs.

Data Availability

All data were included in the manuscript.

Conflicts of Interest

The authors declare that there are no conflicts of interest regarding the publication of this paper.

Authors' Contributions

Mohd Sajid Ali investigated the study, proposed the methodology, wrote the original draft, and reviewed and edited the paper; Mohammad Tariq conceptualized the study, investigated the study, and reviewed and edited the paper.

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Supplementary Materials

Figure S1: experimental refractive index values (circular symbols) as a function of ionic liquid (IL) mole fraction (x_{IL}) for the IL + H_2O systems used in this work along with the predicted values using Gladstone–Dale (green line) and Newton (red line) approach at 298.15 K. The systems are presented in chronological order and marked as M1–M30 as per Table 1 of the main text. (*Supplementary Materials*)

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