

# Density and excess properties of *N*-methyldiethanolamine (MDEA) with 1-hexyl-3-methylimidazolium tris(pentafluoroethyl)trifluorophosphate [hmim][FAP]

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**Abstract** In the present work, new data on the densities and refractive indices for 1-hexyl-3-methylimidazolium tris(pentafluoroethyl)trifluorophosphate [hmim][FAP] with *N*-methyldiethanolamine are reported for various concentrations and at temperatures (303.15–328.15) K. Excess molar volumes  $V^E$  and excess refractive indices  $n_D^E$  were calculated from the experimental data. Refractive index values for the binary mixtures were predicted by using Lorentz–Lorenz, Gladstone–Dale and Eykman equations. Excess molar volumes showed positive trend, whereas excess refractive indices showed negative trend over the entire range of temperatures and concentrations studied in the present research.

**Keywords** Density · Excess molar volume · Refractive index · 1-hexyl-3-methylimidazolium tris(pentafluoroethyl)trifluorophosphate · Methyldiethanolamine

## Introduction

Ionic liquids (ILs) are organic salts having melting points less than 100 °C. Ionic liquids consist of large heterocyclic organic cations and inorganic or organic anions. The most

commonly available/used cations are: *N*-alkylpyridinium, tetraalkylammonium, tetraalkylphosphonium and imidazolium. The anionic part could be halides, acetate, nitrate, tetrafluoroborate ([BF<sub>4</sub>]), hexafluorophosphate ([PF<sub>6</sub>]), trifluoromethylsulfonate ([OTf]), bis(trifluoromethylsulfonyl)imide ([Tf<sub>2</sub>N]), tris(pentafluoroethyl)trifluorophosphate ([FAP]) and the others [1, 2]. ILs have garnered much attention over the past decade due to their unique properties, e.g., low vapour pressure, non-flammability, non-volatility, easily modified structure, good solvency power for organic and inorganic compounds and very high thermal stability [3, 4]. These properties make ILs as good candidates and promising green alternative solvents for a number of industrial applications. Some examples of commercial applications of ILs include usage as operating fluid in several electrochemical applications (batteries, photovoltaic cell, capacitors, fuel cells etc.), as lubricants, as biomass-processing fluids, as heat transfer fluids, as reaction solvents, as catalysts, as separating agents in azeotropic and extractive distillation and as absorption media for gasses [5]. Application of ionic liquids towards carbon dioxide capture has recently been studied and reported by many researchers [6–9]. Currently, aqueous amine solutions are being used extensively and effectively (due to their reactivity) in industry for CO<sub>2</sub> removal. However, usage of amine solutions is an energy-extensive and unfriendly process towards environment; furthermore, amine solutions cause corrosiveness to the process equipments [10]. Hence, development of environmentally benign and energy-intensive solvents which are capable of removing CO<sub>2</sub> efficiently is the current focus of the researchers. ILs are envisaged as potential green alternative solvents for CO<sub>2</sub> capture. However, large-scale industrial usage of ILs for the said purpose is not viable due to their high cost of synthesis and high viscosity. Therefore, scientists' pursuit for environment-

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friendly, energy-efficient solvents (to capture CO<sub>2</sub>) which are industrially applicable also is still going on. Recently, hybrid solvents comprising of ILs and amines have been explored by many researchers as alternative solvents for CO<sub>2</sub> capture [11, 12]. These hybrid solvents are expected to possess the environmental-friendly characteristics of ILs coupled with the reactivity of amines. Therefore, highly viscous and highly priced ILs could be mixed with cheap and low-viscous amines to form different combinations of usable binary mixtures for CO<sub>2</sub> capture. Camper et al. [13] put forward the idea of mixing amines with ILs to form mixtures, used for efficient and reversible CO<sub>2</sub> capture. It was reported that attempted mixtures exhibited better CO<sub>2</sub> capture ability as compared to amine functionalized ILs. In the same manner, Feng et al. [14] studied the solubility of CO<sub>2</sub> in mixtures of four ILs with *N*-methyldiethanolamine (MDEA). It was observed that mixtures of ILs with amines showed superior performance (in terms of CO<sub>2</sub> solubility) than the aqueous amine solutions. It has been reported that knowledge of physical properties data pertaining to the solvent used for CO<sub>2</sub> capture is of utmost importance as it aids in designing and scaling up of process equipments. Physical properties data also help to envision the use of certain solvents for a particular application.

It has been reported that imidazolium-based ILs are much lucrative for CO<sub>2</sub> capture [7]. On the other hand, among the available amines, *N*-methyldiethanolamine (MDEA) is noticeable due to some of its unique characteristics like high thermal and chemical stability, low vapour pressure and less corrosive behaviour [15]. Therefore, in the present work, binary mixtures of 1-hexyl-3-methylimidazolium tris(pentafluoroethyl)trifluorophosphate [hmim][FAP] with *N*-methyldiethanolamine (MDEA) were prepared at different mole fractions. These binary mixtures could be potential solvents for CO<sub>2</sub> capture, and in order to characterize the prepared mixtures, their physical properties have been studied. Density  $\rho$  and refractive indices  $n_D$  for the binary mixtures were measured at different mole fractions and at temperature range of (303.15–328.15) K. Based on the experimental data, excess molar volumes  $V^E$  and excess refractive indices  $n_D^E$  were calculated. Refractive indices values for the binary mixtures were predicted theoretically by using the equations proposed by Lorentz–Lorenz, Gladstone–Dale and Eykman.

## Experimental

### Materials

All the chemicals were purchased from Merck Chemicals. Ionic liquid, 1-hexyl-3-methylimidazolium tris(pentafluoroethyl)trifluorophosphate [hmim][FAP], was supplied with a purity of  $\geq 99\%$  (determined by HPLC). The

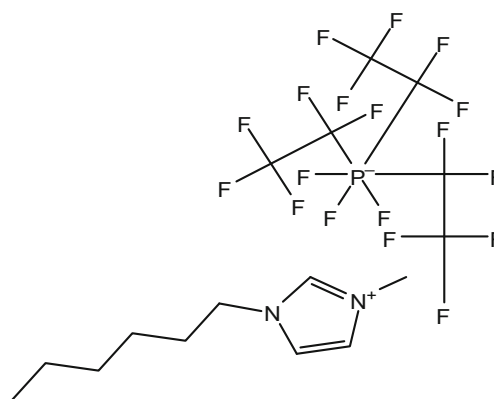
structure of [hmim][FAP] is shown in Fig. 1. [hmim][FAP] was dried under vacuum at 80 °C for 48 h before use. *N*-methyldiethanolamine (MDEA) was supplied with a stated purity  $\geq 99.5\%$  (GC, area %) and used as received.

### Water and chloride content

The water content of [hmim][FAP] was determined by using Karl Fischer coulometric titrator (Mettler-Toledo, DL39) with Hydranal coulomat AG reagent (Riedel-de-Haen). The water content of [hmim][FAP] used in this study was 298 ppm. The chloride content of dried [hmim][BF<sub>4</sub>] was determined by using DL-55 autotitrator (Mettler-Toledo) with 0.005 M, AgNO<sub>3</sub> as the titrant. The estimated chloride content was 34 ppm. The water content of MDEA used in this study was determined by Karl Fischer titrator (Mettler-Toledo, DL-39) using Hydranal coulomat E reagent (Riedel-de-Haen) with benzoic acid (90 mL anolyte + 5 g benzoic acid) as standard procedure described for amines in Mettler-Toledo (DL-39) operating manual. The water content thus estimated was 2768 ppm.

### Apparatus and procedure

All the samples were freshly prepared and retained at desired temperature for 24 h to ensure complete solubility. The samples were prepared on mass basis by using analytical balance (Mettler-Toledo, model AS 120S) and later converted to mole fractions. Uncertainty in the mole fraction calculations was estimated to be around  $\pm 0.0001$ . Binary mixtures were prepared in glass vials and closed with screw caps fitted with PTFE septum. The samples were taken out with a syringe and immediately placed into



1-hexyl-3-methylimidazolium tris(pentafluoroethyl)trifluorophosphate

**Fig. 1** Structure of the ionic liquid studied in this work

the apparatus for each measurement to avoid the humid effects.

### Density measurement

The densities of all the binary mixture as well as pure MDEA and [hmim][BF<sub>4</sub>] were measured by using oscillating U-tube density meter (Anton Paar, DMA-5000) at temperatures (303.15–328.15) K with an uncertainty of  $\pm 0.01$  K. The density meter was calibrated by using Millipore water and dry air [16]. For the validation of calibrated density meter, pure liquids of known density, namely [bis(2-hydroxyethyl)ammonium acetate, water, monoethanolamine and bis(2-hydroxyethyl)ammonium acetate], were used [16, 17]. The uncertainty of all measurements was better than  $3 \times 10^{-5}$  g cm<sup>-3</sup>, and the overall precision in experimental density measurements was better than  $\pm 1 \times 10^{-5}$  g cm<sup>-3</sup>.

### Refractive index measurement

Refractive indices of all the binary mixture samples as well as pure liquids were measured at temperatures (303.15–328.15) K by using ATAGO programmable digital refractometer (RX-5000 alpha) with a temperature control accuracy of  $\pm 0.05$  °C. The apparatus was precise to within  $2 \times 10^{-5}$  and the uncertainty of the measurement was better than  $\pm 4 \times 10^{-5}$ . The apparatus was calibrated by using Millipore water and the measurements were validated by measuring the refractive index of pure liquids (methanol, 1-butyl-3-methylimidazolium tetrafluoroborate) whose values have been already reported [17, 18].

All the densities and refractive indices measurements for each sample were performed in triplicate, and the average values are reported and considered for further analysis.

### Results and discussions

In this present work, new data on the densities and refractive indices for 1-hexyl-3-methylimidazolium tris(pentafluoroethyl)trifluorophosphate [hmim][FAP] with *N*-methyldiethanolamine (MDEA) are reported for various concentrations and at temperatures (303.15–328.15) K. Excess molar volumes  $V^E$  and excess refractive indices  $n_D^E$  were calculated from the experimental data. Refractive index values for the binary mixtures were predicted by using Lorentz–Lorenz, Gladstone–Dale and Eykman equations. Table 1 compare the measured densities and refractive indices values of pure 1-hexyl-3-

methylimidazolium tris(pentafluoroethyl)trifluorophosphate [hmim][FAP] and *N*-methyldiethanolamine (MDEA) with the available literature data. The measured values were found to be in good agreement with the literature values [19–23]. The present measured values of densities and refractive indices for [hmim][FAP] with

**Table 1** Comparison of present experimental values of densities ( $\rho$ ) and refractive indices ( $n_D$ ) at  $T = (303.15\text{--}313.15)$  K, with literature values

<i>T</i> /K	MDEA		[hmim][FAP]	
	This work	Literature	This work	Literature
$\rho/\text{g cm}^{-3}$				
303.15	1.03401	1.03370 <sup>a</sup> 1.03410 <sup>b</sup> 1.03435 <sup>c</sup>	1.54416	1.54428 <sup>e,k</sup>
308.15	1.03030	1.03020 <sup>a</sup> 1.03032 <sup>b</sup> 1.03042 <sup>c</sup>	1.53869	1.53857 <sup>f,k</sup>
313.15	1.02660	1.02670 <sup>a</sup> 1.02652 <sup>b</sup> 1.02648 <sup>c</sup>	1.53327	1.53310 <sup>g,k</sup>
318.15	1.02295	1.02310 <sup>a</sup> 1.02270 <sup>b</sup> 1.02253 <sup>c</sup>	1.52785	1.52740 <sup>h,k</sup>
323.15	1.01920	1.01940 <sup>a</sup> 1.01887 <sup>b</sup> 1.01857 <sup>c</sup>	1.52249	1.52260 <sup>i,k</sup>
328.15	1.01566	1.01580 <sup>a</sup> 1.01502 <sup>b</sup>	1.51707	1.51729 <sup>j,k</sup>
$n_D$				
303.15	1.46522	1.46532 <sup>d</sup>	1.38433	N/A
308.15	1.46319	N/A	1.38298	N/A
313.15	1.46111	1.46074 <sup>d</sup>	1.38162	N/A
318.15	1.45901	1.45892 <sup>d</sup>	1.38032	N/A
323.15	1.45702	1.45708 <sup>d</sup>	1.37897	N/A
328.15	1.45490	N/A	1.37764	N/A

<sup>a</sup> Data from Ref. [19]

<sup>b</sup> Data from Ref. [20]

<sup>c</sup> Data from Ref. [21]

<sup>d</sup> Data from Ref. [22]

<sup>e</sup> At 303.01 K

<sup>f</sup> At 307.90 K

<sup>g</sup> At 313.05 K

<sup>h</sup> At 318.38 K

<sup>i</sup> At 322.78 K

<sup>j</sup> At 327.53 K

<sup>k</sup> Data from Ref. [23]

**Table 2** Experimental values of densities  $\rho$ , refractive indices  $n_D$  and excess molar volumes  $V^E$ , for the binary system [hmim][FAP] (1) + MDEA (2)

$x_1$	$\rho/\text{g cm}^{-3}$	$V^E/\text{cm}^3 \text{ mol}^{-1}$	$n_D$	$\rho/\text{g cm}^{-3}$	$V^E/\text{cm}^3 \text{ mol}^{-1}$	$n_D$
	$T = 303.15 \text{ K}$			$T = 308.15 \text{ K}$		
0.0000	1.03401	0.0000	1.46522	1.03030	0.0000	1.46319
0.1007	1.17216	0.4577	1.44235	1.16778	0.4827	1.44052
0.1989	1.26254	0.8816	1.42644	1.25784	0.9117	1.42485
0.2995	1.32985	1.1798	1.41508	1.32488	1.2197	1.41354
0.4026	1.38206	1.3864	1.40683	1.37690	1.4324	1.40536
0.5001	1.42079	1.5360	1.40086	1.41548	1.5893	1.39948
0.5999	1.45323	1.5830	1.39602	1.44785	1.6324	1.39467
0.7039	1.48264	1.2561	1.39197	1.47719	1.3013	1.39061
0.8003	1.50555	0.9302	1.38891	1.50004	0.9728	1.38756
0.8993	1.52580	0.5505	1.38636	1.52032	0.5702	1.38500
1.0000	1.54416	0.0000	1.38433	1.53869	0.0000	1.38298
	$T = 313.15 \text{ K}$			$T = 318.15 \text{ K}$		
0.0000	1.02660	0.0000	1.46111	1.02295	0.0000	1.45901
0.1007	1.16350	0.4980	1.43865	1.15911	0.5324	1.43679
0.1989	1.25312	0.9485	1.42322	1.24850	0.9765	1.42161
0.2995	1.31997	1.2556	1.41197	1.31509	1.2914	1.41042
0.4026	1.37174	1.4850	1.40387	1.36664	1.5315	1.40241
0.5001	1.41018	1.6487	1.39808	1.40487	1.7136	1.39671
0.5999	1.44248	1.6888	1.39330	1.43709	1.7523	1.39197
0.7039	1.47175	1.3545	1.38924	1.46631	1.4101	1.38792
0.8003	1.49452	1.0290	1.38620	1.48901	1.0847	1.38489
0.8993	1.51485	0.5995	1.38363	1.50938	0.6298	1.38231
1.0000	1.53327	0.0000	1.38162	1.52785	0.0000	1.38032
	$T = 323.15 \text{ K}$			$T = 328.15 \text{ K}$		
0.0000	1.01920	0.0000	1.45702	1.01566	0.0000	1.45490
0.1007	1.15490	0.5359	1.43499	1.15054	0.5786	1.43310
0.1989	1.24385	1.0030	1.42005	1.23914	1.0547	1.41841
0.2995	1.31013	1.3368	1.40890	1.30512	1.4029	1.40733
0.4026	1.36144	1.5953	1.40095	1.35601	1.7079	1.39946
0.5001	1.39954	1.7853	1.39534	1.39422	1.8603	1.39395
0.5999	1.43167	1.8277	1.39063	1.42628	1.8981	1.38927
0.7039	1.46086	1.4764	1.38657	1.45543	1.5351	1.38522
0.8003	1.48349	1.1540	1.38355	1.47798	1.2140	1.38221
0.8993	1.50395	0.6637	1.38095	1.49853	0.6836	1.37961
1.0000	1.52249	0.0000	1.37897	1.51707	0.0000	1.37764

*N*-methyl-diethanolamine (MDEA) are listed in Table 2. It is evident that [hmim][FAP] has higher density as compared to MDEA. For binary mixtures, the density values increased as the mole fraction of [hmim][FAP] increased. The density values of binary mixtures as well as pure liquids (MDEA, [hmim][FAP]) decreased with increase in temperature as expected (Table 2). Based on this analysis, the measured density values can be expressed as a function

of temperature and concentration simultaneously by using the following form of expression [18]:

$$\rho/\text{g cm}^{-3} \text{ or } n_D = \sum_{i=0}^2 [A_i x^i + B_i x^i (T/K) + C_i x^i (T/K)^2]; \quad (1)$$

where  $x$  is the mole fraction of [hmim][FAP],  $T$  is the temperature in Kelvin and  $A_i$ ,  $B_i$ ,  $C_i$  are the correlation

**Table 3** Fitting parameters of Eq. (1) for [hmim][FAP] (1) + MDEA (2) system, along with the values of the standard deviations

		$A_i$	$B_i$	$C_i$
$\rho/\text{g cm}^{-3}$	$i = 0$	1.4071	$-1.2183 \times 10^{-3}$	$6.7338 \times 10^{-7}$
	$i = 1$	0.66090	$2.0794 \times 10^{-3}$	$-4.5621 \times 10^{-6}$
	$i = 2$	-0.13633	$-2.4892 \times 10^{-3}$	$4.7742 \times 10^{-6}$
	$\sigma$		$7.03 \times 10^{-5}$	
$n_D$	$i = 0$	1.5724	$-3.8384 \times 10^{-4}$	$-1.7146 \times 10^{-8}$
	$i = 1$	-0.24637	$2.7745 \times 10^{-4}$	$1.2524 \times 10^{-7}$
	$i = 2$	0.16087	$-2.5314 \times 10^{-4}$	$1.1523 \times 10^{-8}$
	$\sigma$		$1.35 \times 10^{-4}$	

coefficients obtained by regression. The estimated correlation coefficients are presented in Table 3 along with the standard deviation ( $\sigma$ ) values obtained by using the following relation:

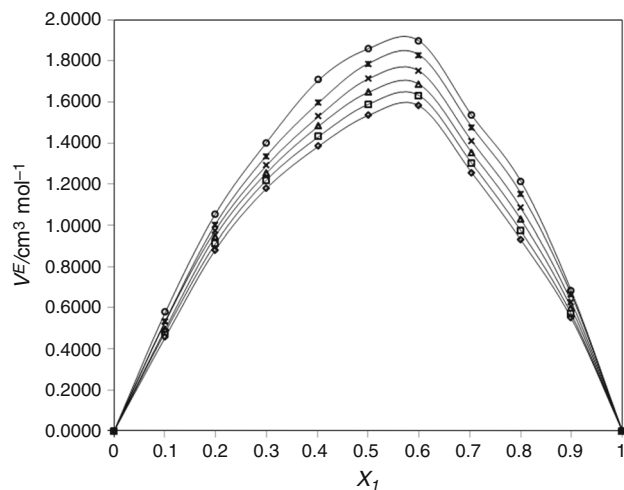
$$\sigma = \left[ \frac{\sum (Z_{\text{exp}} - Z_{\text{cal}})^2}{n} \right]^{\frac{1}{2}} \quad (2)$$

where  $Z_{\text{exp}}$  is the experimental value,  $Z_{\text{cal}}$  is the calculated value and 'n' is the number of experimental data points.

Excess molar volumes  $V^E$  for the binary mixtures were calculated from the measured density data by employing the following relation [24]:

$$V^E/(\text{cm}^3 \text{mol}^{-1}) = \frac{x_1 M_1 + x_2 M_2}{\rho} - \frac{x_1 M_1}{\rho_1} - \frac{x_2 M_2}{\rho_2} \quad (3)$$

where  $\rho$  is the density of the binary mixture;  $x_1$  and  $x_2$  are the mole fractions of pure component 1 and 2, respectively;  $M_1$  and  $M_2$  are the molar masses of pure components 1 and 2, respectively;  $\rho_1$  and  $\rho_2$  are the densities of the pure components 1 and 2, respectively. The calculated values of excess molar volumes are listed in Table 2. Excess molar volume values showed positive trend over the entire range of temperatures and concentrations (Fig. 2). It has been observed that excess molar volume is the resultant of several opposing effects. These effects can arbitrarily be divided into three types: chemical, physical and structural. Physical contributions which are non-specific forces of interaction, like dispersion forces or weak dipole-dipole interactions (due to hydrogen bond rupture) between unlike molecules in the mixtures, lead to positive values of  $V^E$ . Meanwhile, positive values of excess molar volumes are also attributed to the structural breaking effects [25–29]. Positive values of  $V^E$  increased with increase in mole fraction of [hmim][FAP] and reached a maximum value at  $x_1 = 0.5999$ . After that point with increase in mole fraction of [hmim][FAP], the positive values of  $V^E$  decreased (Fig. 2). The values of  $V^E$  changed predominantly with



**Fig. 2** Excess molar volume  $V^E$  versus mole fraction  $x_1$  for the system [hmim][FAP] (1) + MDEA (2), at several temperatures: diamond 303.15 K; square box 308.15 K; triangle 313.15 K; cross sign 318.15 K; asterisk 323.15 K; circle 328.15 K. The solid lines were calculated by  $\rho_{\text{calc}}$  using Eq. 1

variation in concentration of either pure component ([hmim][FAP] or MDEA). In [hmim][FAP]-rich areas ( $x_1 = 0.8003, 0.8993$ ) or in MDEA-rich areas ( $x_1 = 0.1007, 0.1989$ ), excess molar volume values were less positive indicating the ability of solvents to oppose the structural breaking effects, while one is in high concentration as compared to the other constituent comprising the mixture. Refractive index values were higher for MDEA as compared to [hmim][FAP]. In binary mixtures the refractive index values increased as the mole fraction of [hmim][FAP] decreased. Refractive index values decreased with increasing temperature, as expected (Table 2). Variations in the refractive indices values with change in temperatures and concentrations are appreciable, and hence an attempt has been made to correlate refractive indices as a function of temperature and concentration simultaneously by using Eq. 1. The estimated fitting parameters are presented in Table 3.

Refractive index values of the binary mixtures were predicted theoretically by using the equations proposed by Lorentz–Lorenz (Eq. 4), Gladstone–Dale (Eq. 5) and Eykman (Eq. 6).

$$\frac{n_D^2 - 1}{n_D^2 + 2} = \sum_{i=1}^N \left[ \varphi_i \left( \frac{n_{Di}^2 - 1}{n_{Di}^2 + 2} \right) \right] \quad (4)$$

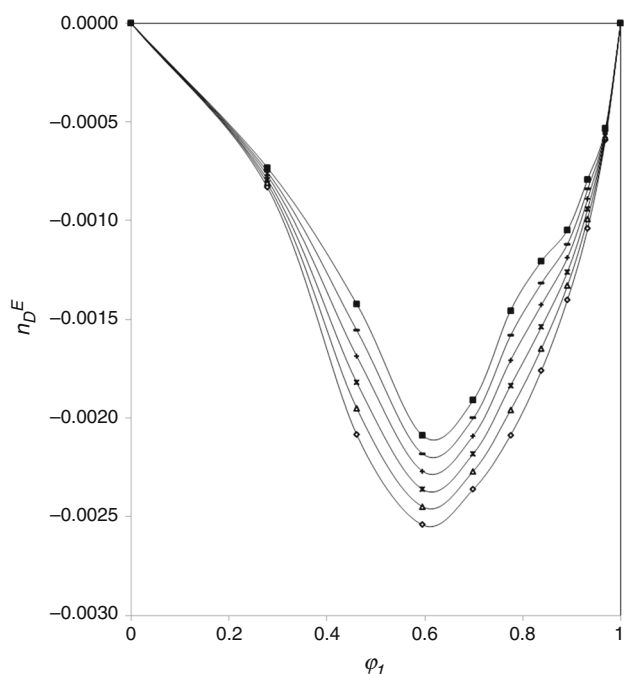
$$n_D^2 - 1 = \sum_{i=1}^N [\varphi_i (n_{Di} - 1)] \quad (5)$$

$$\frac{n_D^2 - 1}{n_D^2 + 0.4} = \sum_{i=1}^N \left[ \varphi_i \left( \frac{n_{Di}^2 - 1}{n_{Di}^2 + 0.4} \right) \right] \quad (6)$$

**Table 4** Standard deviations ( $\sigma$ ) for the predicted refractive indices with respect to the corresponding experimental data for [hmim][FAP] (1) + MDEA (2) system

$T/K$	Lorentz–Lorenz $\times 10^{-3}$	Gladstone–Dale $\times 10^{-3}$	Eykman $\times 10^{-4}$
303.15	2.48	3.45	12.03
308.15	2.30	3.26	10.43
313.15	2.12	3.06	8.86
318.15	1.95	2.87	7.34
323.15	1.76	2.67	5.69
328.15	1.59	2.48	4.16

In the above equations,  $n_D$ , is the refractive index of the mixture;  $\varphi_i$  and  $n_{Di}$  are the volume fraction and refractive index of the component  $i$ , respectively. The deviations between the experimental values of refractive indices and the one calculated with the help of above equations are listed in Table 4. The calculated values of refractive indices with Lorentz–Lorenz, Gladstone–Dale, and Eykman equations agreed well with the experimental values as the deviation was within the allowable limit ( $\leq 0.005$ ). In order to calculate the ideal refractive index ( $n_D^{id}$ ) and excess refractive index ( $n_D^E$ ) of the binary mixtures ([hmim][FAP] with *N*-methyl-diethanolamine (MDEA)) the equations proposed by Reis et al. [30] were used. The ideal refractive index is expressed as:



**Fig. 3** Excess refractive indices  $n_D^E$  for the system [hmim][FAP] (1) + MDEA (2), at several temperatures; *diamond* 303.15 K; *triangle* 308.15 K; *asterisk* 313.15 K; *plus* 318.15 K; *minus* 323.15 K; *filled square*, 328.15 K. The *solid curves* were calculated by  $n_{Dcalc}$  using Eq. 1

$$n_D^{id} = \left[ \varphi_1 (n_{D1})^2 + \varphi_2 (n_{D2})^2 \right]^{\frac{1}{2}} \quad (7)$$

where  $\varphi_1$  and  $\varphi_2$  are the volume fractions of pure component 1 and 2 and  $n_{D1}$  and  $n_{D2}$  are the refractive index of pure component 1 and 2, respectively. The excess refractive index was calculated by using the following expression.

$$n_D^E = n_D - n_D^{id} \quad (8)$$

where  $n_D^E$  is the excess refractive index and  $n_D$  is refractive index of mixtures and  $n_D^{id}$  is the ideal refractive index calculated using Eq. (7). Excess refractive indices values showed negative trend over the entire range of temperatures and compositions (Fig. 3).

## Conclusions

Binary mixtures of 1-hexyl-3-methylimidazolium tris(pentafluoroethyl)trifluorophosphate [hmim][FAP] with *N*-methyl-diethanolamine (MDEA) were prepared at different mole fractions. The densities and refractive indices of the binary mixtures were measured at a temperature range of (303.15–328.15) K. The densities of the present prepared binary mixtures increased as the mole fraction of [hmim][FAP] increased. Refractive index values were higher for MDEA as compared to [hmim][FAP]. In mixtures the refractive index values increased as the mole fraction of [hmim][FAP] decreased. Excess molar volumes showed positive values over the entire range of temperature and concentrations, indicating the presence of weak interaction between [hmim][FAP] and MDEA. Excess refractive index values showed negative trend over the whole compositions and temperatures.

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