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Thermophysical properties of ionic liquids and their mixtures from a new equation of state

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Abstract In our previous work, a perturbed hard-trimer-sphere equation of state (PHTS EOS) was developed for modeling the phase equilibria of pure ionic liquids (ILs) (M.M. Alavianmehr et al., Ionics 22 (2016) 2447–2459). In this work, we have successfully extended the model to the mixtures of IL + IL and IL + solvent. Two temperature-dependent parameters appearing in the EOS are correlated with two microscopic scaling constants σ, the effective hard-sphere diameter, and ε, the non-bonded interaction energy. The overall average absolute deviation (AAD) of the estimated densities from the literature data using the proposed model with and without non-additivity parameter (λ_{ij}) was found to be 0.44 and 0.79%, respectively. A modified Enskog equation and rough hard-sphere (RHS) theory are combined with our proposed equation of state to calculate the viscosity coefficient of ionic liquids and their mixtures. Finally, from the results obtained, a linear relation between logarithm of surface tension and viscosity property of ionic liquid was developed.

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Introduction

As we know, ionic liquids (ILs) are organic salts that are liquid near room temperature. They have a negligible vapor pressure under normal operating conditions and have a large liquid working temperature range. In general, ILs have many favorable properties which make them attractive for scientific community. Understanding thermophysical properties of pure ILs and their mixtures is essential to determine potential applications [1].

In recent years, some computational methods have been proposed for predicting densities of ionic liquids. Palomar et al. [2] used COSMO-RS predictive model which was based on the quantum chemistry. Group contribution (GC) and the related methods such as GC-EOS have also been widely used by numerous researchers to predict the density of ILs. Gardas and Coutinho [3] extended group contribution model to predict the density of ILs as a function of the temperature and pressure for varieties of ionic liquids. Valderama et al. [4] presented an artificial neural network and group contribution method to correlate the density of ILs. Aparicio et al. [5] reviewed the available predictive models as well as experimental data reported in literature for the thermophysical properties and especially density and PVT behavior of ionic liquids. Further, a perturbed hard-sphere equation of state has been employed to predict the pressure–volume–temperature properties of some pure and mixture of ionic liquids [6–9]. Also, they predicted the volumetric properties of ILs using Ihm–Song–Mason (ISM) [10] and Tao–Mason [11] equations of state. Hosseini et al. [12] have assessed the role of hard-dimers to present an alternative perturbed hard chain (PHC)

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EOS for ILs. The performance of the perturbed hard-dimer chain (PHDC) model has also been checked for density of ILs. The Tao–Mason (TM) EOS has been employed to model the pressure–volume–temperature (PVT) properties of ILs by the help of an alternative corresponding states correlation based on the measurable scaling constants, i.e., surface tension and liquid density [13]. Alavianmehr and co-workers have revised an attractive part of the preceding EOS by re-evaluating the abovementioned variable parameter as well as the repulsive term. Two temperature-dependent parameters appearing in the revisited EOS have been determined from the corresponding states correlations using the interfacial properties of ILs, i.e., surface tension and liquid density, both at room temperature [14].

Recently, Alavianmehr et al. [15] developed a new version of the PHC EOS, by employing a trimer expression from the statistical associating fluid theory-trimer (SAFT-T) as the reference physical model. The perturbation part of the proposed EOS was taken from the van der Waals dispersion forces. The proposed model was employed to predict the volumetric properties of polymers. In our previous work [16], we examined the predictive power of this EOS for the prediction of the liquid density, isothermal compressibilities, and thermal expansion coefficients as well as the heat capacities and vapor pressure of pure ILs. For this purpose, 39 ILs with various anions were chosen. Our calculation results were summarized as average absolute deviation percent (AAD%) from the literature data. From 6331 data points examined for the studied pure ILs over a broad pressures ranging from 0.1 to 200 MPa and temperatures ranging from 273 to 472.6 K, the AAD was found to be 0.18%.

In the present study, this model is successfully applied to predict thermophysical properties of mixtures containing ionic liquids. Thermophysical properties consist of densities, surface tensions, and viscosity coefficients.

Liquid viscosity is an important transport property in chemical process design. Therefore, prediction of the viscosity coefficient of ionic liquids is a very useful task. Starting from the pioneering work of Hsu and Eyring [17] on the application of the significant structure theory to the calculation of the viscosity of liquid alkali metals, other theoretical prediction methods have been developed so far for this purpose. The temperature dependence of transport properties of fluids is also essential for the most industrial applications, and intensive research has been done for their measurement [18–20]. Fluid viscosity, among other transport properties, can be measured basically with high accuracy. Experimental viscosity of dilute gases and its temperature dependence are used as essential properties for accurate determination of molecular information such as pair interaction potential function. These properties accurately furnish the basic information of fluids so that their correlations provide a good insight into different fluid classes. In this work, we propose a correlation for the temperature and density dependence of the viscosity coefficient of ionic liquids. The correlation has a similar mathematical expression as the modified Enskog theory expression [21] for the viscosity of dense fluids. It is shown that the present approach can well predict the viscosity coefficient of ionic liquids within a few percent of experimental measurements. In the present work, the liquid viscosity for ILs is predicted using the PHTS model in conjunction with the modified Enskog method and rough hard-sphere (RHS) theory [22].

Liquid surface tension as an equilibrium thermodynamic property is also as important as viscosity in the design of industrial applications. Surface tension of a liquid is related to the intermolecular interaction potential energy and the liquid interfacial microstructure; it decreases linearly with temperature in the range of freezing to the boiling temperature and vanishes non-linearly close to the critical point [23–26]. Like viscosity, surface tension can be measured with high accuracy. The temperature dependence of surface tension, the negative surface entropy, can be used to characterize fluids in terms of the molecular energetics and surface microstructure. Contrary to viscosity, the surface tension decreases very smoothly with temperature; its dependence on temperature is constant and close to one another for most normal liquids [27, 28].

In the present study, we have evaluated PHTS EOS by predicting the surface tension for ionic liquids. The thermophysical properties of mixtures are important because they depend on the composition and/or temperature and are of great importance in understanding the nature of molecular aggregation that exists in the binary mixtures.

Theory

Equation of state for single substance

The general form of PHTS EOS is as follows:

$$Z = Z^{H3} + Z^{pert} \tag{1}$$

In Eq. (1), the reference equation takes the form proposed by Srivastava and Khanna [29]. The formalism mentioned here is based on the assumption that the chain formed by the pairs of trimers:

$$Z^{H3} = 3Z^{HS}_{MV} - 2\left\{1 + y\frac{\partial\ln\left((Z^{HS}_{MV}-1)/4y\right)}{\partial y}\right\} \tag{2}$$

The reference system is the new version of the hard-sphere (HS) model expressed by Malijevsky and Veverka (MV) equation [30]:

$$Z^{HS}_{MV} = \frac{1 + 1.056y + 1.6539y^2 + 0.3262y^3}{(1 + 0.056y + 0.5979y^2 + 0.3076y^3)(1-y)^3} \tag{3}$$

where y is the packing fraction of hard-core which is defined as:

$$y = \frac{b(T)\rho}{4} \tag{4}$$

The perturbed term used in this work is van der Waals attractive form:

$$Z^{pert} = -\frac{a(T)\rho}{k_B T} \tag{5}$$

The two temperature-dependent parameters $a(T)$ and $b(T)$ can be expressed in terms of the universal function of reduced temperature according to the law of corresponding states principle. They are correlated with the hard-sphere diameter, σ, and the non-bonded interaction energy between two trimer spheres, ε, according to the following equations:

$$a(T) = \frac{2\pi}{3}\sigma^3 \varepsilon F_a\left(\frac{k_B T}{\varepsilon}\right) \tag{6}$$

$$b(T) = \frac{2\pi}{3}\sigma^3 F_b\left(\frac{k_B T}{\varepsilon}\right) \tag{7}$$

where F_a and F_b are the universal functions of the reduced temperature ($k_B T/\varepsilon$), which can be written as the following formula:

$$F_a(k_B T/\varepsilon) = 1.058393\exp[-0.879380(k_B T/\varepsilon)]$$
$$+ 0.609445\exp\left[-0.314038(k_B T/\varepsilon)^{3/2}\right] \tag{8}$$

$$F_b(k_B T/\varepsilon) = 0.798019\exp[0.908086(k_B T/\varepsilon)]$$
$$+ 0.521304\exp\left[-0.407266(k_B T/\varepsilon)^{3/2}\right] \tag{9}$$

Mixture version of the model

The mixture version of the proposed equation of state can be written as below:

$$Z_m = 3Z_{MV,m}^{HS} - 2\left\{1 + y_m \frac{\partial\ln\left(\left(Z_{MV,m}^{HS}-1\right)/4y_m\right)}{\partial y_m}\right\}$$
$$- \frac{\rho}{RT}\sum_{ij}^{m} x_i x_j a(T)_{ij} \tag{10}$$

where x_i and x_j are the mole fractions of ith and jth components, respectively. η_m is the packing fraction of the

mixtures of hard-spheres [31]. This parameter is defined by the following expression:

$$y_m = \frac{\rho}{4}\sum_i x_i b(T)_i \tag{11}$$

The compressibility factor of hard-sphere mixtures takes the form:

$$Z_{MV,m}^{HS} = \frac{1 + 1.056 y_m + 1.6539 y_m^2 + 0.3262 y_m^3}{\left(1 + 0.056 y_m + 0.5979 y_m^2 + 0.3076 y_m^3\right)\left(1-y_m\right)^3} \tag{12}$$

The attractive forces between two hard-sphere species of a mixture including i and j components can be written as follows:

$$a(T)_{ij} = \frac{2\pi}{3}\sigma_{ij}^3 \varepsilon_{ij} F_a\left(\frac{k_B T}{\varepsilon}\right)_{ij} \tag{13}$$

The present method for calculating two temperature-dependent parameters can be extended to mixtures by using simple geometric and arithmetic mean for the adjustable scaling constants, i.e.,

$$\varepsilon_{ij} = \left(1-\lambda_{ij}\right)\sqrt{\left(\varepsilon_i \varepsilon_j\right)} \tag{14}$$

and

$$\sigma_{ij} = \frac{1}{2}\left(\sigma_i + \sigma_j\right) \tag{15}$$

where λ_{ij} is a binary interaction parameter which shows non-additivity of the mixture. Further, the universal function F for ij mixture is proposed to be obtained by the following geometric mean from its corresponding ones for pure substances, i.e., Eqs. (8) and (9):

$$F_a\left(\frac{k_B T}{\varepsilon}\right)_{ij} = \sqrt{F_a(k_B T/\varepsilon)_i F_a(k_B T/\varepsilon)_j} \tag{16}$$

Results and discussion

In previous section, a new equation of state was developed to model thermophysical properties of ILs and their binary mixtures. To utilize the proposed PHTS EOS, the values of two pure component parameters, σ and ε, of studied ILs and solvents must be characterized. Physically, theses parameters reflect the hard-core

diameter and non-bonded interaction energy between pair-wise trimers, respectively. In this work, these parameters were optimized for each ILs and solvents by fitting the proposed model with experimental densities data. To achieve this goal, the following objective function was minimized using the least-squares method:

$$OF = \min \frac{1}{n} \sum_{i=1}^{n} \frac{|M_i^{\text{Lit.}} - M_i^{\text{Cal.}}|}{M_i^{\text{Lit.}}} \qquad (17)$$

where M and n represent thermodynamic properties and the number of relevant density data points for a given ILs and/or solvents, respectively. The numerical values of optimized pure component parameters of studied fluids are reported in Table 1. Generally, from 1927 literature data points applied to nine studied ILs (cited in Table 1), the minimum value of OF was found to be 0.080.

In the present study, the proposed model is extended to various binary mixtures formed by ILs and molecular

solvents including water, ethanol, methanol, 1-propanol, THF, and acetonitrile using some mixing and combining rules. Table 2 tabulates the AAD of the predicted densities of several binary of IL + IL and IL + solvent mixtures at various temperatures and mole fractions from literature values [32–39]. Besides, the maximum deviations (MD) of the calculations have also been included in Table 2. It should be mentioned that the maximum deviation of the calculated volumetric properties of all binary mixtures studied in this work was found to be 3.46%. It is noticeable that the uncertainty of the calculated density for all binary mixtures studied in this work was of the order of ± 1.55 in temperature range 278–358 K and atmospheric pressure.

In the current work, the non-additivity effect on our proposed model has also been investigated. Table 2 lists the results with and without the non-additivity effect. The AADs of the predicted mixture densities for 1927 data points using the present model were found to be 0.44 and 0.79% with and without adjustable parameter (λ_{ij}), respectively. As Table 2 shows, the numerical values of

Table 1 The scaling pure component parameters to be used in Eqs. (4), (5) and (18)

	M_{W} (g mol^{-1})	ε/k_{B} (K)	σ (nm)	α (mPas)	β	ω
Ionic liquid						
[C$_2$mim][Triflat]	260.23	4833.7	0.6382	1271.530	− 7.907	247.511
[C$_4$mim][Triflat]	288.29	7106.3	0.6917	1544.080	− 11.832	554.110
[C$_2$mim][EtSO$_4$]	236.29	7465.0	0.6595	5162.113	− 13.470	720.392
[C$_4$mim][C(CN)$_3$]	229.28	5010.5	0.6716	883.100	− 8.080	259.506
[C$_4$mim][N(CN)$_2$]	205.20	7950.0	0.6652	940.500	− 13.609	735.645
[C$_4$mim][MeSO$_4$]	250.32	6355.0	0.6703	18,526.398	− 11.450	522.002
[C$_2$mim][NTf$_2$]	391.31	5539.7	0.7144	656.641	− 9.014	320.230
[C$_4$mim][NTf$_2$]	419.36	5919.0	0.7486	1313.646	− 9.817	381.816
[C$_6$mim][NTf$_2$]	447.42	5735.8	0.7747	1991.562	− 9.498	358.139
[C$_8$mim][NTf$_2$]	475.47	5335.2	0.7975	2882.353	− 8.766	305.382
[C$_{10}$mim][NTf$_2$]	503.52	5627.2	0.8249	2491.448	− 9.096	328.596
[C$_2$mim][BF$_4$]	197.97	2938.8	0.6052	1147.998	− 9.091	327.337
[C$_3$mim][BF$_4$]	211.97	3940.0	0.6069	2047.131	− 6.167	150.845
[C$_4$mim][BF$_4$]	226.02	5391.4	0.6410	2000.958	− 8.659	297.645
[C$_6$mim][BF$_4$]	254.08	5220.0	0.6766	7796.281	− 8.640	297.215
[C$_8$mim][BF$_4$]	282.13	5598.0	0.7132	5275.015	− 8.785	307.319
[C$_4$mim][PF$_6$]	284.18	5005.0	0.6605	3922.981	− 7.657	233.157
[C$_6$mim][PF$_6$]	312.24	5406.0	0.6973	8438.436	− 8.466	285.631
[C$_8$mim][PF$_6$]	340.29	7292.0	0.7731	61,055.050	− 13.210	696.093
Solvent						
Ethanol	46	1690.0	0.38605	4.123	− 1.966	14.002
Water	18	6720.0	0.30055	4.971	− 10.308	400.453
Methanol	32	1650.0	0.33955	–	–	–
1-Propanol	60	2680.0	0.44405	–	–	–
Acetonitrile	41.05	1765.0	0.37555	–	–	–
THF	72.11	1550.0	0.42550	–	–	–

M_{W} molar weight

Table 2 AAD of the predicted densities of binary mixtures including IL + solvent and IL + IL using the proposed model from the literature ones

Mixture	T_{min}–T_{max} (K)	NP	λ_{ij}	AAD[a] %	AAD[b]%	MD %	Ref.
[C$_2$mim][BF$_4$]+[C$_6$mim][BF$_4$]	298–308	135	0.06	0.33	0.77	0.96	[32]
	293–343	121		0.39	0.60	1.19	[39]
[C$_2$mim][BF$_4$]+[C$_3$mim][BF$_4$]	293–343	121	0.04	0.35	0.45	0.98	[39]
[C$_3$mim][BF$_4$]+[C$_6$mim][BF$_4$]	293–343	121	0.04	0.41	0.56	1.19	[39]
[C$_4$mim][BF$_4$]+[C$_4$mim][MeSO$_4$]	298–308	144	0.04	0.18	0.46	0.62	[32]
[C$_4$mim][PF$_6$]+[C$_4$mim][BF$_4$]	298–308	126	0.011	0.23	0.27	0.62	[32]
[C$_4$mim][N(CN)$_2$]+water	278–358	63	− 0.05	0.084	0.29	0.37	[37]
[C$_4$mim][BF$_4$]+water	308–353	88	0.11	0.42	1.35	− 2.81	[33]
[C$_4$mim][Triflate]+water	303–343	77	− 0.05	0.36	0.71	− 2.81	[34]
[C$_2$mim][EtSO$_4$]+water	278–308	112	− 0.09	0.83	1.01	− 2.26	[35]
[C$_2$mim][Triflate]+water	278–338	133	0.08	0.38	1.03	− 1.91	[36]
[C$_4$mim][C(CN)$_3$]+water	278–358	81	0.12	0.20	1.08	− 0.51	[37]
[C$_2$mim][EtSO$_4$]+methanol	278–308	147	0.01	0.95	0.95	3.46	[35]
[C$_2$mim][EtSO$_4$]+ethanol	278–308	126	0.13	0.47	0.99	1.43	[35]
[C$_2$mim][Triflate]+ethanol	278–328	60	0.16	0.73	2.00	3.12	[36]
[C$_2$mim][Triflate]+1-propanol	278–338	91	0.01	0.29	0.29	0.59	[36]
[C$_2$mim][EtSO$_4$]+acetonitrile	278–308	168	0.05	0.67	0.74	− 1.89	[35]
[C$_4$mim][PF$_6$]+THF	298	13	0.01	0.62	0.62	1.10	[38]
Overall		1927		0.44	0.79		

$$AAD = 100/NP \sum_{i=1}^{NP} | \rho_i^{Calc.} - \rho_i^{Lit.} | / \rho_i^{Lit.}$$

*NP* number of literature data points examined, *MD* maximum deviation of the calculated ρ_{mix}

[a] The present model with adjustable parameter

[b] The present model without adjustable parameter

the non-additivity parameter (λ_{ij}) for the studied mixtures are nearly low. A nonzero non-additivity parameter indicates some interaction between the unlike components. The low values of this parameter are in favor of this fact that the volumes of ionic liquids are nearly additive. However, considering non-additivity effect improves slightly the results of the calculated densities of IL mixtures.

In general, a wide space of experimental data points regarding the binary mixtures has been taken to check the predictive power of the proposed EOS. To show how the mixture version of our model passes through the experimental points, Fig. 1 has been provided. Figure 1 shows the calculated and literature values of densities of binary mixtures [C$_2$mim][Triflate] + 1-Propanol at different temperatures.

To ascertain the degree of reliability of the present model, we compare the correlated and predicted results from the proposed model with other models given in literature. In this respect, we compare the outcomes of our calculations with those obtained from Fadaei-Nobandegani et al [40]. Table 3 contains the AAD of the calculated densities of binary mixtures using the present model and other equations of state [40]
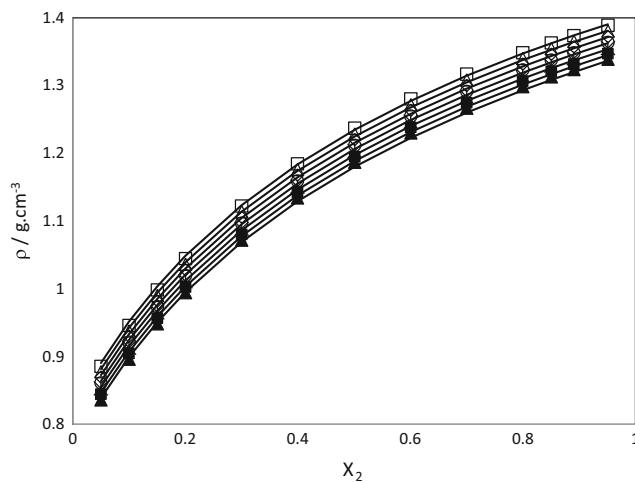
Fig. 1 Density of [C$_2$mim][Triflate](x_1) + 1-Propanol (x_2) as a function of mole fraction (*x*) of 1-Propanol at different temperatures and pressures (with adjustable parameter). The markers represent the literature data [36], 278.15 K (white square); 288.15 K (white up-pointing triangle), 298.15 K (white diamond), 308.15 K (white circle), 318.15 K (asterisk), 328.15 K (black square), and 338.15 K (black up-pointing triangle), while the solid curves represent the results calculated with the proposed model

Table 3 Comparison of the AAD of calculated density of binary mixtures of ILs (IL + IL and IL + solvent) in this work and those obtained by PHS EOS [40] compared with the literature data [32–36]

	T_{min}–T_{max} (K)	NP	This work	PHS	Ref.
[C$_2$mim][BF$_4$]+[C$_6$mim][BF$_4$]	298–308	135	0.33	1.20	[32]
[C$_4$mim][PF$_6$]+[C$_4$mim][BF$_4$]	298–308	126	0.23	1.14	[32]
[C$_4$mim][BF$_4$]+water	308–353	88	0.42	0.89	[33]
[C$_4$mim][Triflate]+water	303–343	77	0.36	0.68	[34]
[C$_2$mim][EtSO$_4$]+water	278–308	112	0.83	0.65	[35]
[C$_2$mim][Triflate]+water	278–338	133	0.38	0.48	[36]
[C$_2$mim][EtSO$_4$]+methanol	278–308	147	0.95	0.39	[35]
Overall		818	0.50	0.78	

$$AAD = 100/NP \sum_{i=1}^{NP} |\rho_i^{Calc.} - \rho_i^{Lit.}|/\rho_i^{Lit.}$$

NP number of literature data points examined

from the literature data [32–36]. Close inspection of the results shows the ability of the present model to describe liquid density data for IL mixtures. Interestingly, the results are in favor of superiority of the present model over the one proposed in literature [40].

The proposed model was further evaluated by prediction of other thermophysical properties including viscosity and surface tension.

Viscosity

Modified Enskog theory

According to the modified Enskog theory (MET) [21], the viscosity coefficient of a dense gas of rigid spherical molecules is related to pair distribution function, density, and dilute gas viscosity. As we know, MET is applicable to dense gases and

Table 4 The coefficients c_1–c_3 for each IL and solvent to be used in Eq. (22) obtained from the viscosity data at pressure 0.1 MPa

	c_1	c_2	c_3
Ionic liquid			
[C$_2$mim][Triflat]	3.4167×10^{-05}	$- 9.9008 \times 10^{-04}$	7.2170×10^{-03}
[C$_4$mim][Triflat]	2.4070×10^{-05}	$- 9.8118 \times 10^{-04}$	1.0069×10^{-02}
[C$_2$mim][EtSO$_4$]	6.8953×10^{-05}	$- 3.1636 \times 10^{-03}$	3.6476×10^{-02}
[C$_4$mim][C(CN)$_3$]	1.8434×10^{-05}	$- 5.4093 \times 10^{-04}$	3.9811×10^{-03}
[C$_4$mim][N(CN)$_2$]	1.2583×10^{-05}	$- 5.8308 \times 10^{-04}$	6.7837×10^{-03}
[C$_4$mim][MeSO$_4$]	2.6713×10^{-04}	$- 1.0623 \times 10^{-02}$	1.0596×10^{-01}
[C$_2$mim][NTf$_2$]	1.3882×10^{-05}	$- 4.4431 \times 10^{-04}$	3.5898×10^{-03}
[C$_4$mim][NTf$_2$]	2.3726×10^{-05}	$- 8.1998 \times 10^{-04}$	7.1295×10^{-03}
[C$_6$mim][NTf$_2$]	3.5197×10^{-05}	$- 1.1835 \times 10^{-03}$	9.9993×10^{-03}
[C$_8$mim][NTf$_2$]	5.2316×10^{-05}	$- 1.6415 \times 10^{-03}$	1.2931×10^{-02}
[C$_{10}$mim][NTf$_2$]	4.4995×10^{-05}	$- 1.4625 \times 10^{-03}$	1.1938×10^{-02}
[C$_2$mim][BF$_4$]	1.5683×10^{-05}	$- 4.3050 \times 10^{-04}$	2.9816×10^{-03}
[C$_3$mim][BF$_4$]	5.8411×10^{-05}	$- 1.3501 \times 10^{-03}$	7.8424×10^{-03}
[C$_4$mim][BF$_4$]	4.2444×10^{-05}	$- 1.3238 \times 10^{-03}$	1.0374×10^{-02}
[C$_6$mim][BF$_4$]	1.4511×10^{-04}	$- 4.6621 \times 10^{-03}$	3.7552×10^{-02}
[C$_4$mim][PF$_6$]	1.1141×10^{-04}	$- 3.1130 \times 10^{-03}$	2.1824×10^{-02}
[C$_6$mim][PF$_6$]	1.6786×10^{-04}	$- 5.1185 \times 10^{-03}$	3.9110×10^{-02}
[C$_8$mim][PF$_6$]	6.7529×10^{-04}	$- 3.0489 \times 10^{-02}$	3.4482×10^{-01}
Solvent			
Ethanol	5.7766×10^{-07}	$- 5.1388 \times 10^{-06}$	1.1890×10^{-05}
Water	1.4248×10^{-07}	$- 4.9829 \times 10^{-06}$	4.5083×10^{-05}

therefore cannot be applied to liquids. Therefore, we take the simple form of MET and propose the following equation for the viscosity of liquids:

$$\eta = \alpha\left\{1 + \beta\big(b(T)\rho\big(Z_{MV}^{HS}-1\big)/4y\big) + \omega\big(b(T)\rho\big(Z_{MV}^{HS}-1\big)/4y\big)^2\right\} \quad (18)$$

where α, β, and ω are adjustable parameters. The experimental viscosities over wide range of temperatures and pressures were used to fix aforementioned adjustable parameters. The values of α, β, and ω are reported in Table 1.

Rough hard-sphere-based model

The proposed RHS model for transport properties of ILs can be expressed as [22]:

$$\eta_{RHS}^* = C_\eta \eta_{SHS\text{-}MET}^* = C_\eta \frac{(M_w\varepsilon/N_a)^{1/2}}{\sigma^2 F_\eta}\eta_{SHS\text{-}MET} \quad (19)$$

Table 5 AAD (%) of the calculated viscosity of ionic liquids using the MET [21] and RHS [22] models from the experimental data [33, 34, 37, 39, 41–48]

	T_{min}–T_{max} (K)	NP	MET	RHS	Ref
Pure					
[C$_2$mim][BF$_4$]	293–343	11	2.14	2.48	[39]
[C$_3$mim][BF$_4$]	293–343	11	4.56	4.90	[39]
[C$_6$mim][BF$_4$]	293–343	11	8.20	9.06	[39]
[C$_4$mim][BF$_4$]	303–343	07	2.13	2.71	[33]
[C$_4$mim][C(CN)$_3$]	278–358	09	17.18	22.20	[37]
[C$_4$mim][N(CN)$_2$]	278–358	09	15.88	17.91	[37]
[C$_4$mim][MeSO$_4$]	293–313	05	0.87	0.88	[42]
[C$_{10}$mim][NTf$_2$]	298–343	03	0.026	0.83	[43]
[C$_8$mim][NTf$_2$]	293–343	06	6.68	7.13	[44]
[C$_6$mim][NTf$_2$]	293–343	06	5.56	6.08	[44]
[C$_4$mim][NTf$_2$]	293–343	06	4.43	4.89	[44]
[C$_2$mim][NTf$_2$]	293–343	06	2.66	3.10	[44]
[C$_2$mim][Triflat]	278–338	07	4.55	6.85	[48]
[C$_4$mim][Triflat]	303–343	07	1.38	1.57	[34]
[C$_4$mim][PF$_6$]	303–363	07	8.15	8.86	[41]
[C$_6$mim][PF$_6$]	303–363	07	14.71	15.42	[41]
[C$_8$mim][PF$_6$]	293–323	07	4.66	4.80	[45]
[C$_2$mim][EtSO$_4$]	298–313	04	0.29	0.35	[46]
Ethanol	283–343	13	0.41	0.49	[46]
Water	303–353	08	0.54	0.61	[33]
Overall		150	5.25	6.06	

$$AAD = 100/NP \sum_{i=1}^{NP} |\eta_i^{Calc.} - \eta_i^{Lit.}|/\eta_i^{Lit.}$$

NP number of literature data points examined, *MET* modified Enskog theory [21], *RHS* rough hard-sphere [22]
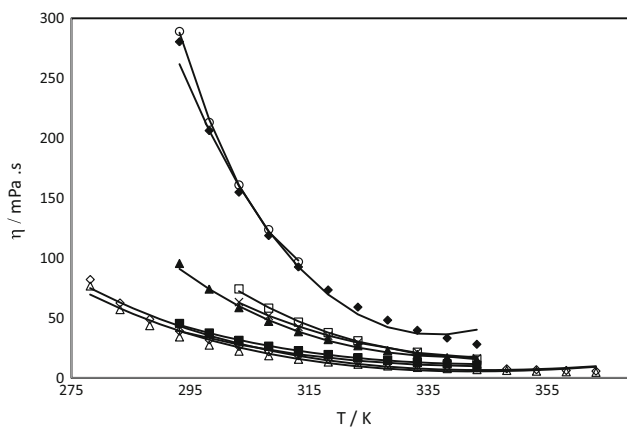

Fig. 2 Linear dependency viscosity of some selected ionic liquids on the temperature curve. The markers represent the literature data [33, 34, 37, 39, 42, 44], [C$_4$mim][BF$_4$] (white square), [C$_4$mim][C(CN)$_3$] (white up-pointing triangle), [C$_4$mim][N(CN)$_2$] (white diamond), [C$_6$mim][MeSO$_4$] (white circle), [C$_2$mim][BF$_4$] (black square), [C$_3$mim][BF$_4$] (black up-pointing triangle), [C$_2$mim][NTf$_2$] (plus sign), [C$_6$mim][BF$_4$] (black diamond), and [C$_4$mim][Triflate] (multiplication sign), and solid lines are those predicted by the proposed model

where η^* is reduced viscosity and η is dynamic viscosity in Pas units. $\eta_{SHS\text{-}MET}^*$ is the relevant expression to viscosity from smooth hard sphere-modified Enskog's theory, viz.:

$$\eta_{SHS\text{-}MET}^* = 4y\left[\frac{1}{(Z_{MV}^{HS}-1)} + 0.8 + 0.761\big(Z_{MV}^{HS}-1\big)\right] \quad (20)$$

In this study, C_η is proposed to be expressed in terms of universal function of reduced pressure ($P_r = P / 0.1$ MPa)
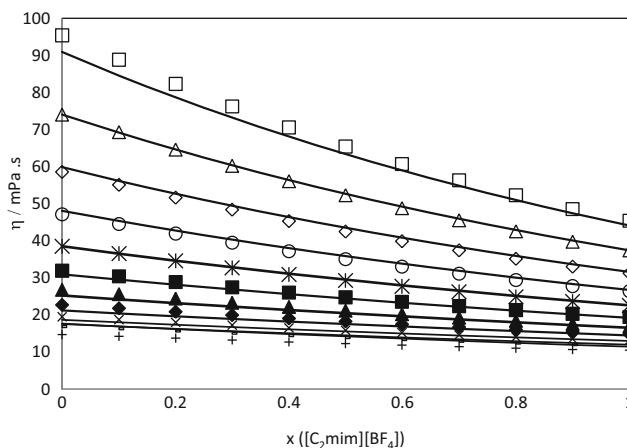
Fig. 3 Viscosity of mixtures of x [C$_2$mim][BF$_4$] + $(1 - x)$ [C$_3$min][BF$_4$] in terms of mole fraction. Symbols refer to experimental values [39] at 293.15 K (white square), 298.15 K (white up-pointing triangle), 303.15 K (white diamond), 308.15 K (white circle), 313.15 K (asterisk), 318.15 K (black square), 323.15 K (black up-pointing triangle), 328.15 K (black diamond), 333.15 K (multiplication sign), 338.15 K (white square), and 343.15 K (plus sign), while the solid curves represent the results calculated with the proposed modeling atmospheric pressure
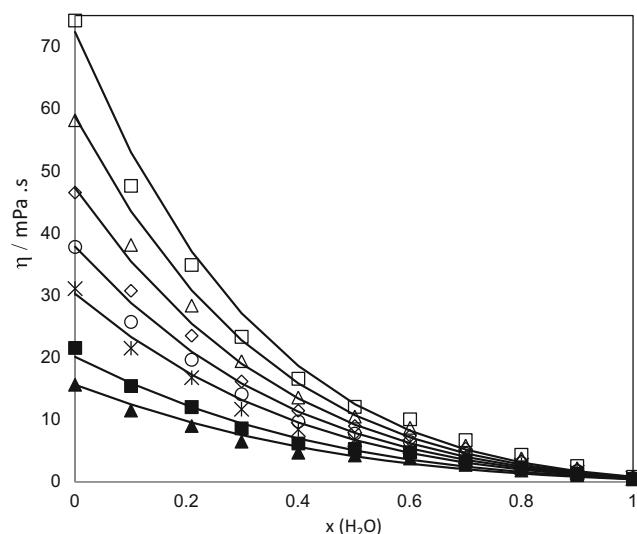
Fig. 4 Viscosity of x [H$_2$O] + $(1 - x)$ [C$_4$min][BF4]. Symbols refer to experimental values [33] at 303.15 K (white square), 308.15 K (white up-pointing triangle), 313.15 K (white diamond), 318.15 K (white circle), 323.15 K (asterisk), 333.15 K (black square), and 343.15 K (black up-pointing triangle), while the solid curves represent the results calculated with the proposed modeling atmospheric pressure

as follows:

$$C_\eta = \exp\left(6.4 \times 10^{-4}(P_r - 1)\right) \quad (21)$$

In Eq. (19), F_η is a temperature-dependent function which is expressed in terms of the reduced temperature ($T_r = k_B T / \varepsilon$) as follows:

$$F_\eta(k_B T/\varepsilon) = c_1 + c_2 T_r + c_3 T_r^3 \quad (22)$$

where $k_B T$ is thermal energy per one molecule. The coefficients c_1–c_3 were fixed for each IL by correlating the

viscosity data at 0.1 MPa and their numerical values were reported in Table 4.

Table 5 contains the AAD of the calculated viscosity coefficients of studied pure ILs from the experimental ones [33, 34, 37, 39, 41–48] in temperature interval 278–363 K and atmospheric pressure. For 150 data point examined, the overall AADs for MET and RHS models were found to be 5.78 and 6.06, respectively. Further, Fig. 2 displays the predicted viscosity of several pure ILs in terms of temperature in atmospheric pressure using the MET method. Our results are in accord with those given in literature. It should be mentioned that the errors associated with the calculated viscosity using our model are of the order of ± 26.40.

Extension to mixtures

It is interesting to check further the present model by predicting the viscosity of mixtures. In this regard, the ideal Grunberg and Nissan mixing law [49] was employed to predict the mixture viscosity of ionic liquids as follows:

$$\log_{10}(\eta_m) = x_1 \log_{10}(\eta_1) + x_2 \log_{10}(\eta_2)$$
$$+ 2 x_1 x_2 k_{12} \left(\frac{M_{w,1} M_{w,2}}{M_{w,m}}\right)^{1/2} \quad (23)$$

where η_m represents the absolute viscosity of the mixture, x_i is the mole fraction of component i, and η_i is the absolute viscosity of pure component i. k_{12} is an interaction parameter.

Figures 3 and 4 represent the calculated viscosity of binary mixtures formed by ILs in terms of mole fraction and at different temperatures and atmospheric

Table 6 AAD (%) of the calculated viscosity of binary IL-IL and IL-solvents using the present model compared with the experiment [33, 34, 37, 39]

	T_{min}–T_{max} (K)	NP	AAD[a]	AAD[b]	k_{12}	Ref.
C$_2$mim][BF$_4$]+[C$_3$mim][BF$_4$]	293–343	121	2.41	2.40	3×10^{-5}	[39]
[C$_2$mim][BF$_4$]+[C$_6$mim][BF$_4$]	293–343	121	6.39	3.62	0.00497	[39]
[C$_3$mim][BF$_4$]+[C$_6$mim][BF$_4$]	293–343	121	6.39	4.38	0.00416	[39]
[C$_4$mim][BF$_4$]+water	303–343	77	29.32	13.65	0.0760	[33]
[C$_4$mim][C(CN)$_3$]+water	278–358	81	33.04	15.08	0.112	[37]
[C$_4$mim][N(CN)$_2$]+water	278–358	63	36.55	11.74	0.113	[37]
[C$_4$mim][Triflate]+water	303–343	77	43.28	17.64	0.143	[34]
Overall		661	22.48	9.79		

$$\text{AAD} = 100/\text{NP} \sum_{i=1}^{\text{NP}} |\eta_{m,i}^{Calc.} - \eta_{m,i}^{Lit.}|/\eta_{m,i}^{Lit.}$$
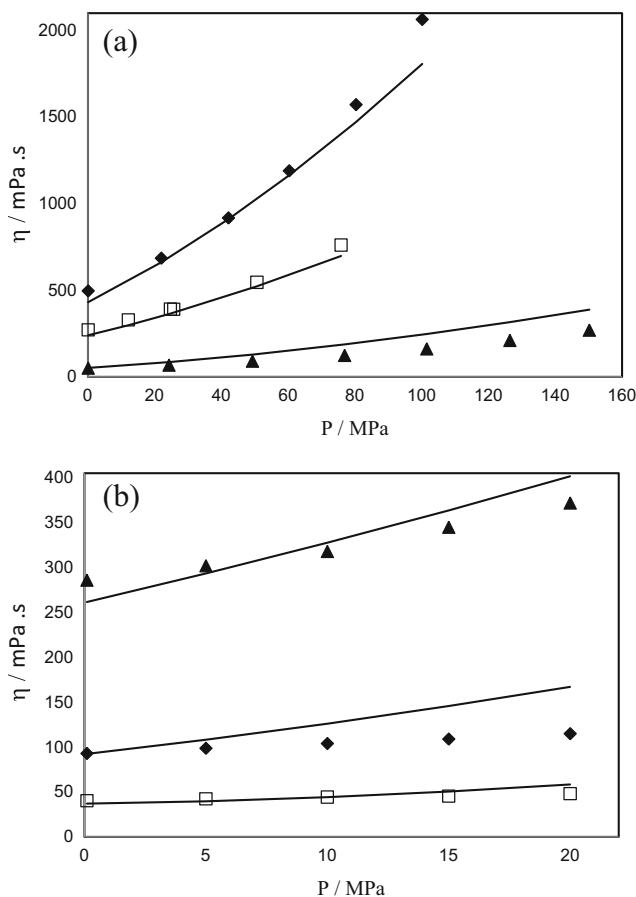
NP number of literature data points examined

[a] Calculations have been performed with k_{12} equal to zero

[b] Calculations have been performed with non-zero k_{12}

pressure. The results show that the model is able to describe liquid viscosity data for binary mixture of ILs. We have calculated the viscosities of binary mixtures of ILs over the whole range of concentration. Further, Table 6 shows the AAD of the calculated viscosities of mixtures of several ionic liquids from the experimental ones [33, 34, 37, 39]. From 661 data points examined for the aforementioned mixtures, the mean AAD of the predicted viscosities from measurement was found to be 22.48% and 9.79 without and with binary interaction parameter. The pleasure harmony between the results and those obtained from literature confirms the reliability of our model. Figure 5 represents the predicted viscosity of three ILs at 298.15 K (a-plot) and viscosity of $[C_6mim][BF_4]$ predicted at three temperatures 293.15, 313.15, and 333.15 K up to 20 MPa (b-plot). The markers represent the experimental viscosity [50–52] and the solid lines are those obtained from the proposed MET model. As Fig. 5

illustrates, the viscosity of IL mixture can be shown well by the present model.

Finally, we assessed the proposed MET model by studying the excess viscosity of IL-IL and IL-solvent systems which is defined as:

$$\Delta\eta = \eta_m - \sum_i^2 x_i\eta_i \tag{24}$$

The excess behaviors of two selected IL-IL and IL-solvent systems were predicted using the proposed MET model, and the results were compared with those obtained from experimental mixture viscosity data [34, 39]. The outcomes of the computations are shown in Fig. 6.
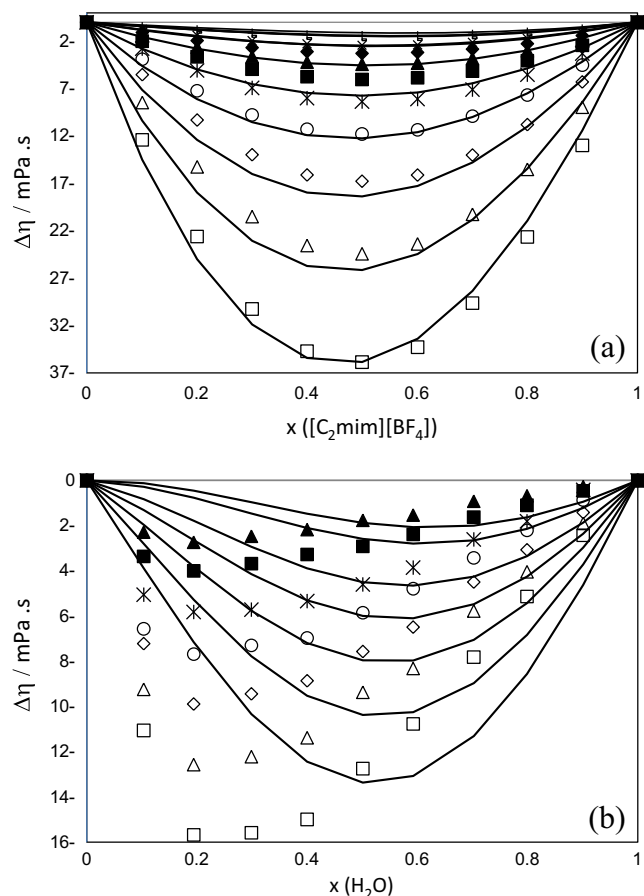
Fig. 6 Excess viscosity for the $x\,[C_2mim][BF_4] + (1-x)\,[C_6min][BF_4]$ in terms of mole fraction. Symbols refer to experimental values [39] for temperatures 293.15 K (white square), 298.15 K (white up-pointing triangle), 303.15 K (white diamond), 308.15 K (white circle), 313.15 K (asterisk), 318.15 K (black square), 323.15 K (black up-pointing triangle), 328.15 K (black diamond), 333.15 K (multiplication sign), 338.15 K (white square), and 343.15 K (plus sign) (a-plot). b-plot represents the excess viscosity of $x\,[H_2O] + (1-x)\,[C_4min][Triflate]$. Symbols refer to experimental values [34] at temperatures 303.15 K (white square), 308.15 K (white up-pointing triangle), 313.15 K (white diamond), 318.15 K (white circle), 323.15 K (asterisk), 333.15 K (black square), and 343.15 K (black up-pointing triangle). The solid curves represent the results calculated with the MET model in atmospheric pressure

Fig. 5 The viscosity of some selected ionic liquids in terms of pressure. The markers represent the literature data [51, 52] for $[C_4mim][PF_4]$ (white square), $[C_4mim][NTf_2]$ (black up-pointing triangle), $[C_6mim][PF_6]$ and (black diamond) at 298.15 K (a-plot). b-plot represents the viscosity of $[C_6mim][BF_4]$ in terms of pressure. The markers represent the literature data [50] at temperatures 293.15 K (black up-pointing triangle), 313.15 K (black diamond), and 333.15 K (white square). Solid lines are those predicted by the MET model
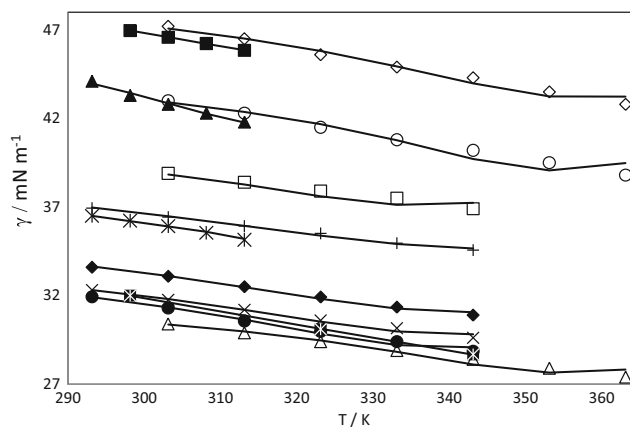
Surface tension

Due to the fact that temperature-dependent surface tension of fluids (below their critical temperatures) are perfectly linear and according to the successful applicability of temperature dependence of viscosity [53], we propose that the surface tension can be related to the fluidity as,

$$\ln\gamma = C + D\left(\frac{1}{\eta}\right)^{\phi} \quad (25)$$

where C and D are substance-dependent constants and, for studied ILs, constants C and D are reported in Table 7. The exponent $\phi = 0.30$ is used in Eq. (25) to fit the imidazolium-based ILs with simple anion content using the experimental viscosity [41] and surface tension [54] data.

In Fig. 7, the surface tension in terms of temperature has been provided and compared with literature data [42, 45, 46, 54, 55]. The results show that the model is able to describe surface tension data for pure ILs.

To assess further the degree of accuracy of the proposed model, we compare the AADs of our calculations with those obtained from other methods. In Table 7, we have listed the AAD of the predicted surface tension for 81 data points using the present model, the results obtained from the Alavianmehr et al. [56], quantitative structure–property relationship (QSPR) strategy [57], and corresponding states group contribution (CS–GC) method [58] from the literature data [42, 45,

Fig. 7 The calculated surface tension of some selected ionic liquids as a function of temperature. The markers represent the literature data [42, 45, 54, 55], [C$_6$mim][BF$_4$] (white square), [C$_8$mim][BF$_4$] (white up-pointing triangle), [C$_4$mim][PF$_6$] (white diamond), [C$_6$mim][PF$_6$] (white circle), [C$_8$mim][PF$_6$] (asterisk), [C$_2$mim][EtSO$_4$] (black square), [C$_4$mim][MeSO$_4$] (black up-pointing triangle), [C$_2$mim][NTf$_2$] (plus sign), [C$_4$mim][NTf$_2$] (black diamond), [C$_6$mim][NTf$_2$] (multiplication sign), [C$_8$mim][NTf$_2$] (black circle), and [C$_{10}$mim][NTf$_2$] (wingdings 181), and solid lines are those predicted by the proposed model in atmospheric pressure

46, 54, 55]. The interesting point of the present study is that the AADs of the calculated surface tension from the present model are remarkably lower than those obtained from other methods. The overall AAD of the predicted surface tension for 81 data points using the present model, Alavianmehr et al., QSPR, and CS–GC methods were found to be 0.29, 1.45,

Table 7 The scaling constants to be used in Eq. (25) and AAD (%) of the calculated surface tension using the present model and those obtained by Alavianmehr et al. [56], QSPR [57] strategy, and CS–GC [58] method

	C	$-D$	T_{min}–T_{max} (K)	NP	This work	Alavianmehr et al.	QSPR	CS–GC	Ref.
[C$_4$mim][BF$_4$]	3.86	0.348	303–343	07	0.12	2.09	9.48	–	[54]
[C$_6$mim][BF$_4$]	3.74	0.375	303–363	05	0.64	–	–	–	[54]
[C$_8$mim][BF$_4$]	3.51	0.491	303–363	07	0.61	3.81	15.69	6.14	[54]
[C$_4$mim][PF$_6$]	3.95	0.464	303–363	07	0.45	2.03	2.45	–	[54]
[C$_6$mim][PF$_6$]	3.85	0.505	303–363	07	0.71	0.23	6.38	–	[54]
[C$_8$mim][PF$_6$]	3.85	0.505	293–313	05	0.087	1.79	11.93	–	[45]
[C$_2$mim][EtSO$_4$]	3.96	0.428	298–313	04	0.021	–	–	–	[45]
[C$_4$mim][MeSO$_4$]	3.92	0.735	293–313	05	0.18	–	–	–	[42]
[C$_{10}$mim][NTf$_2$]	3.62	0.631	298–343	03	0.082	–	–	–	[55]
[C$_2$mim][Triflat]	3.87	0.229	278–338	07	0.062	–	–	–	[55]
[C$_8$mim][NTf$_2$]	3.59	0.532	293–343	06	0.38	–	–	–	[55]
[C$_6$mim][NTf$_2$]	3.59	0.447	293–343	06	0.27	0.21	6.59	5.20	[55]
[C$_4$mim][NTf$_2$]	3.64	0.447	293–343	06	0.23	1.00	3.98	3.23	[55]
[C$_2$mim][NTf$_2$]	3.73	0.376	293–343	06	0.17	0.42	5.33	2.00	[55]
Overall				81	0.29	1.45	7.73	4.14	

$$AAD = 100/NP \sum_{i=1}^{NP} |\gamma_i^{Calc.} - \gamma_i^{Lit.}|/\gamma_i^{Lit.}$$

NP number of literature data points examined

7.73, and 4.14%, respectively. As we can see from Table 7, the present model is the most accurate among other abovementioned methods. It should be mentioned that the error propagation uncertainty of calculated surface tension was of the order of ± 1.73.

Conclusion

We successfully applied the PHTS EOS to the mixture of ILs using alternative corresponding states correlations for two temperature-dependent parameters that appeared in the EOS. When tested against experimental data, the calculated liquid densities for binary mixtures including IL + solvent and IL + IL gave the mean average absolute deviations of 0.44% for 1927 data points. Also, we showed that the liquid densities and molecular parameters taken from previously developed EOS were adequately rational for predicting the densities of mixture involving ILs using some simple mixing rules.

The thermophysical properties such as viscosity and surface tension were predicted by the present model. When compared with other works and also with literature data, the accuracy of our model was further revealed. The predicted viscosity coefficient and surface tension of ionic liquids were in good agreement with experimental data.

Finally, the non-additivity behavior of the studied mixtures was also investigated. The sign of the non-additivity parameter indicates a tendency toward attraction between the dissimilar molecules in the mixture. However, the value of this parameter was not large, which implied that the hard-sphere model is able to model the excess properties of the present mixtures. However, considering the non-additivity effect in our calculations showed that this effect can improve slightly the results of mixture densities.

Nomenclature

AAD; Average absolute deviation (%),g; Pair radial distribution function of hard-spheres at contact (m), $b(T)$; van der Waals covolume, m^3, P; Pressure, Pa, R; Gas constant, J/mol K, T; Absolute temperature, K; k_B; Boltzmann's constant, $J/K M_w$; Molecular weight, c_1–c_3; Coefficients, N_a; Avogadro's number, mol, F_η; Temperature-dependent function, C_η; Coupling parameters, $a(T)$; Strengths of attractive forces, J m^{-3}

F_a and F_b Universal functions

y; Packing fraction, V; Molar volume, C; Substance-dependent constant, D; Substance-dependent constant, M; Thermodynamic properties, Z; Compressibility factor, x; Molar fraction

Greek letters

ε; Non-bonded interaction energy parameter, J, σ; Effective hard-sphere diameter, nm, η; Viscosity, mPa s, η^*; Reduced viscosity, ρ; Molar density, mol/m^3, α; Adjustable parameter, β; Adjustable parameter, ω; Adjustable parameter, ϕ; Exponent, γ; Surface tension, mN m^{-1}, π; pi = 3.14, λ; Adjustable parameter

Superscript

Li; Literature, Calc; Calculated, E; Excess, H^3; Hard-trimer-chain reference system, Pert; Perturbed, HS; Hard-sphere, RHS; Rough hard-sphere

Subscript

MV; Malijevsky–Veverka, m; Mixture

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