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New Method Based on the UNIFAC–VISCO Model for the Estimation of Ionic Liquids Viscosity Using the Experimental Data Recommended by Mathematical Gnostics

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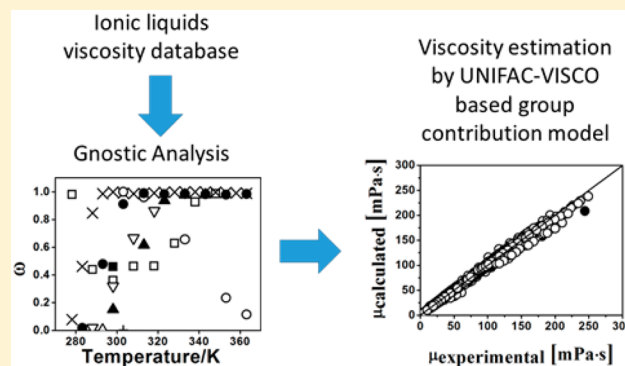
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Supporting Information

ABSTRACT: The viscosity of ionic liquids (ILs) has been modeled as a function of temperature and at atmospheric pressure using a new method based on the UNIFAC–VISCO method. This model extends the calculations previously reported by our group (see Zhao et al. *J. Chem. Eng. Data* 2016, 61, 2160–2169) which used 154 experimental viscosity data points of 25 ionic liquids for regression of a set of binary interaction parameters and ion Vogel–Fulcher–Tammann (VFT) parameters. Discrepancies in the experimental data of the same IL affect the quality of the correlation and thus the development of the predictive method. In this work, mathematical gnostics was used to analyze the experimental data from different sources and recommend one set of reliable data for each IL. These recommended data (totally 819 data points) for 70 ILs were correlated using this model to obtain an extended set of binary interaction parameters and ion VFT parameters, with a regression accuracy of 1.4%. In addition, 966 experimental viscosity data points for 11 binary mixtures of ILs were collected from literature to establish this model. All the binary data consist of 128 training data points used for the optimization of binary interaction parameters and 838 test data points used for the comparison of the pure evaluated values. The relative average absolute deviation (RAAD) for training and test is 2.9% and 3.9%, respectively.



1. INTRODUCTION

Recently, ionic liquids (ILs) have received significant attention due to their unique properties, such as nonvolatility, high chemical stability, and easy operation at the liquid state, and thus have become promising alternatives to traditional liquid solvents.^{1–7} However, the viscosities of ILs are relatively high compared to those of traditional solvents and water.⁸ Accordingly, the viscosity of ILs is a very important parameter in assessing various aspects of chemical processes; high viscosity can negatively affect mass transfer and power requirements for mixing in liquid–liquid systems. A low viscosity IL is desirable as a solvent in order to minimize pumping costs and increase mass transfer rates; while higher viscosity ILs could be favorable for other applications, such as lubrication, membranes,⁹ etc.

To develop new ILs with a tailored viscosity, the conventional experimental trial-and-error method is not practical due to the large quantity of potential cation–anion combinations. Therefore, a variety of methods have been reported for the

correlation/evaluation/prediction of IL viscosity.^{9–29} Methods based on the different correlation equations which provide the correlation for viscosity usually describe the exponential behavior of the temperature dependency; such as the Vogel–Fulcher–Tammann (VFT) equation,^{10–12} fluidity equation,¹² Litovitz equation,^{12,13} Arrhenius equation,^{12,14} power law equation,¹⁵ and the Daubert and Danner correlation.¹⁴

Predictive methods for the viscosity can be categorized as (i) group contribution method (GCM)^{9,16–19} and (ii) quantitative structure–property relationship (QSPR).^{20–27} Gardas et al.⁹ and Daniel et al.¹⁷ developed the group contribution method based on the Orrick–Erbar model³⁰ to predict IL viscosity. However, the Orrick–Erbar model also requires density data for the prediction of the viscosity. To overcome this limitation

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and to attempt the development of an improved viscosity model with lower deviations in estimated viscosities, Gardas et al.¹⁹ proposed a new group contribution method based on the Vogel–Fulcher–Tammann (VFT) equation. Gharagheizi et al.¹⁸ employed a total of 46 substructures in addition to the temperature to predict IL viscosity. Lazzús and Pulgar–Villarreal¹⁶ estimated the viscosity of a number of ILs using an improved Andrade-type approach that is a linear combination of the contribution of groups in the cation and the anion.

Other approaches reported in the literature to describe the IL viscosity are based on the statistical association fluid theory (SAFT) equation of state (EoS).^{28,29} Polishuk²⁸ modeled the viscosity of ILs using SAFT-cubic equation of state coupled with two models, free volume theory (FVT) and modified Yarranton–Satyro (MYS) correlation. Despite unsatisfactory results of free volume theory, the modified Yarranton–Satyro correlation predicted the viscosity of two families of imidazolium-based ILs ($[C_n\text{mim}][\text{PF}_6]$ and $[C_n\text{mim}][\text{BF}_4]$) with appropriate accuracy. Furthermore, Abolala et al.²⁹ modeled the viscosity of pure imidazolium-based ILs ($[C_n\text{mim}][\text{PF}_6]$, $[C_n\text{mim}][\text{BF}_4]$, and $[C_n\text{mim}][\text{NTf}_2]$) using the SAFT-VR-Mie equation of state coupled with three models, namely frictional theory (FT), free-volume theory (FVT), and modified Yarranton–Satyro (MYS) correlation. The result showed that the MYS model calculated the viscosity more precisely than the FVT and FT models. A comparative result from the SAFT-cubic EoS proposed by Polishuk²⁸ showed that the performance of SAFT-VR-Mie EoS was better than the SAFT-cubic EoS. All the equations mentioned are semiempirical, regardless of the basis of their development.

Viscosity measurement is highly sensitive to the presence of impurities (water and halides),³¹ the temperature control,³² and the different experimental techniques,³³ such as the Anton Paar automated microviscometer (AMVn) and the Ubbelohde viscometers, for example. In other words, experimental data of ILs reported in the literature were obtained using different techniques and samples with different impurity levels. Thus, discrepancies in the experimental data occur and affect the quality of the correlation and thus the development of the predictive method. Therefore, one of the main purposes of this work is to do a critical analysis of the viscosity data and to recommend a data set for each IL.

In the present work, we present an extension of the previously developed method³³ based on the UNIFAC–VISCO model to evaluate the viscosity of 70 pure ILs and 11 binary ionic liquid mixtures. Herein, the mathematical gnostics was first employed for the data analysis and data set recommendation. One recommended data set for each IL was used to calculate the binary interaction parameters and ion VFT parameters.

2. DATA ANALYSIS

The impurities (such as halides, water, and metals) in the ionic liquids and the different measurement techniques have a significant effect on the accuracy of the measured viscosity, as discussed elsewhere³³ and above. Discrepancies in the experimental data of the same IL affect the quality of the correlation and thus the development of the predictive method. Therefore, in the present work, mathematical gnostics^{34,35} was used to analyze the experimental data, and a data set for each IL is recommended.

Mathematical gnostics is a novel paradigm of uncertainty.³⁴ This axiomatic theory is built upon the theory of measurement

and based on the fundamental laws of nature such as the special theory of relativity and thermodynamics. The properties of each individual measurement are analyzed and properties of a data sample are obtained by their aggregation. The results are thus valid even for small data samples because, unlike in statistics, the extrapolation from an infinite to a finite data sample is not used. In addition, the data are allowed to speak for themselves; there is no prior assumption on the distribution function of errors, but it is estimated during data analysis. It is possible to describe both unimodal and multimodal distributions using the same approach. In this work, nonlinear regression along with an influence function is utilized. This stems from the methodology of robust statistics³⁶ in which the data errors are assumed to have normal distribution with zero mean but the sample is contaminated by a few values having a gross error. Knowing a distribution function of these gross errors, an influence function is derived. During regressions, the influence function is used for iterative reweighting of the residuals. The procedure is stopped when the values of residuals do not change. The influence function derived from mathematical gnostics does not require any assumption on the distribution function of measurement errors, both the precise data and outliers can be described by the same distribution function estimated during regression. In addition, these *a posteriori* weights obtained by application of the influence function are directly related to the distribution density where the weight equal to one coincides with the location of its maximum, that is, the most probable value. Owing to these properties the values of the *a posteriori* weights together with results of marginal analysis of residuals can successfully be applied to a critical evaluation of the scattered data.

For example, several authors^{8,37–44} have reported viscosity values of the IL 1-ethyl-3-methylimidazolium triflate, $[C_2\text{mim}][\text{OTf}]$. The temperature range, number of data points, purity, and measurement techniques in each reference are summarized in Table 1.

Figure 1 shows the weight of experimental viscosities for $[C_2\text{mim}][\text{OTf}]$. Only two data points^{39,41} were measured at 278.15 K. The weight of one point is close to zero and the other one is close to one. However, it could not be concluded that the viscosity measured by Freire et al.³⁹ at 278.15 K causes a large difference from the estimated location. Only a qualitative judgment can be made by comparison between the data weights at the same temperature. It is seen from Figure 1 that the viscosity data reported by Freire et al.³⁹ manifests a better performance than other data at each reported temperature, with the exception of at 278.15 K.

The comparison between the estimated locations and the experimental values is demonstrated in Figure 2. As shown in Figure 2, large differences are observed especially with the temperature increasing. For example, Yusoff et al.⁴² measured a viscosity value of 11.14 mPa·s at 363.15 K. A difference of 30% was found from 7.75 mPa·s reported by Seddon et al.⁸ and 26.7% deviation from 8.169 mPa·s reported by Freire et al.³⁹ These large deviations can be attributed to different IL batches, measurement techniques, or poor temperature control/calibration. The performance of the data set over a large temperature range measured by Freire et al.³⁹ is better than the other five data sets;^{8,41–44} ignoring three individual data points reported by three different references.^{37,38,40} The data reported by Seddon et al.⁸ shows a good agreement at a temperature range from 303.15 to 343.15 K, while deviations increase at higher and lower temperatures. The average relative deviation (ARD, eq 1)

Table 1. Information for Experimental Viscosities of 1-Ethyl-3-methylimidazolium Triflate [C₂mim][OTf] Collected from Corresponding References^a

ref	N _{data}	T _{range} (K)	source	purity	water (ppm)	chloride (ppm)	experimental technique
37	1	293	synthesized	NA	NA	NA	NA
38	1	298.15	Fluka	98%	NA	NA	HP ^{3D} CE
41	8	278.15–348.15	synthesized	NA	96	NA	cone–plate rheometer
39	18	278.15–363.15	Iolitec	99%	20	NA	Stabinger viscometer
42	5	303.15–363.15	Sigma-Aldrich	98%	NA	NA	concentric cylinders viscometry
8	9	283.15–363.15	NA	NA	237	1300	cone–plate viscometer
40	1	303.15	EMD Chemicals	NA	NA	<2000	cone–plate viscometer
43	5	298.15–343.15	solvionic	99.5%	50–100	NA	Anton Paar viscosimeter
44	4	288.15–318.15	Iolitec	99%	130	NA	Stabinger viscometer
total	52	278.15–363.15					

^aN_{data} number of data points; T_{range} temperature range.

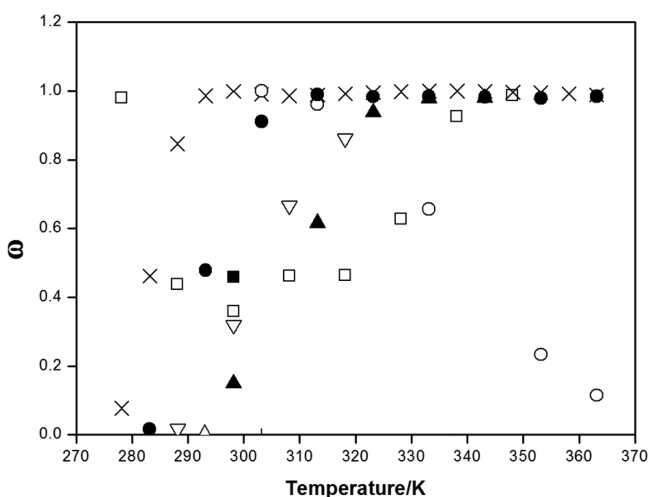


Figure 1. Data weight of the literature values for [C₂mim][OTf]: Δ , Abbott et al.;³⁷ \blacksquare , François et al.;³⁸ \square , Rodríguez et al.;⁴¹ \times , Freire et al.;³⁹ \circ , Yusoff et al.;⁴² \bullet , Seddon et al.;⁸ $+$, Morgan et al.;⁴⁰ \blacktriangle , Tsamba et al.;⁴³ ∇ , Vuksanovic et al.⁴⁴

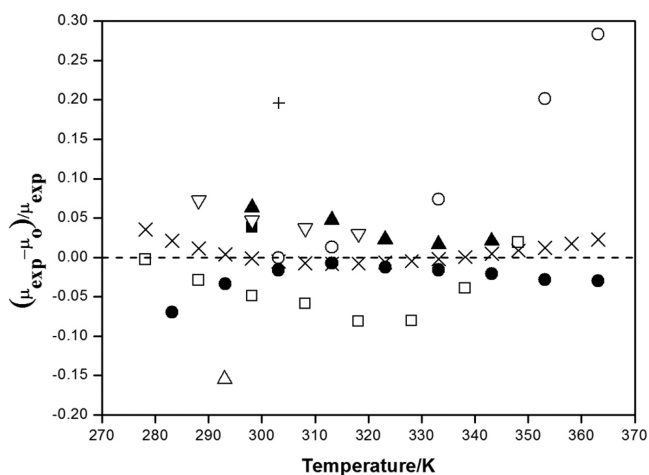


Figure 2. Deviations of the literature values from the estimated locations for [C₂mim][OTf]: Δ , Abbott et al.;³⁷ \blacksquare , François et al.;³⁸ \square , Rodríguez et al.;⁴¹ \times , Freire et al.;³⁹ \circ , Yusoff et al.;⁴² \bullet , Seddon et al.;⁸ $+$, Morgan et al.;⁴⁰ \blacktriangle , Tsamba et al.;⁴³ ∇ , Vuksanovic et al.⁴⁴

of the data set reported by Freire et al.³⁹ is -0.7% compared with the estimated locations, lower than -4.1% of ARD from Seddon et al.⁸ and -4.5% of ARD from Rodríguez et al.⁴¹

While the ARD is up to 9.7% for the data set measured by Yusoff et al.⁴² in a comparison from the estimated locations. Therefore, in general we recommend the data set published by Freire et al.³⁹ to be used for the optimization of the UNIFAC–VISCO fitting parameters.

$$100 \times \text{ARD} = \frac{100}{M} \sum_{i=0}^M \frac{\mu_{\text{exp}} - \mu_0}{\mu_0} \quad (1)$$

where M is the number of data points in the data set; μ_{exp} is the experimental viscosity; and μ_0 is the estimated location of the maximum distribution density.

Following this methodology, we recommended then one data set for each selected ionic liquid. The gnostic analysis of experimental data for each ionic liquid is illustrated in Figures S1–S86 of the Supporting Information. The cations and anions used are among the most commonly studied structures. The cations used include imidazolium, ammonium, phosphonium, pyridinium, and pyrrolidinium. Fifteen different anions are used including the tetrafluoroborate ($[\text{BF}_4]^-$), methylsulfate ($[\text{C}_1\text{SO}_4]^-$), and thiocyanate ($[\text{SCN}]^-$), for example. Selected cations and anions are listed in Table 2 along with the number of data points containing this ion, temperature range, and the references from which the experimental viscosity data were taken. The selected 70 different ILs, the number of data points in the recommended reference, and the temperature range are summarized in Table 3. For some ILs, such as [C₆mim]Br, only one viscosity data set is available in the literature, to date. Recommended data could thus not be obtained by the gnostic analysis and the given set was then considered as the recommended data set.

3. UNIFAC–VISCO BASED METHOD FOR PREDICTION OF VISCOSITY OF IONIC LIQUIDS

3.1. Calculation of Ionic Liquid Viscosity by the UNIFAC–VISCO Method. A new method for the prediction of viscosities of pure and mixtures of ionic liquids has been recently developed by our group.³³ For the calculation of pure IL viscosity based on the UNIFAC–VISCO model, the cation and anion, constituting the ionic liquid, are regarded as individual components and each component is made up of a single group. Below is the summary of the calculations using this method. Full details of the UNIFAC–VISCO model have been described previously.³³

The UNIFAC–VISCO model is a group contribution model developed by Chevalier et al.^{89,90} to predict the viscosities of

Table 2. Selected Cations and Anions Constituting the Selected ILs, and References from Which Experimental Data Were Collected

code	cation	anion	N_{data}	T_{range} (K)	ref
1	[C ₂ mim] ⁺	[BF ₄] ⁻ , [C ₁ SO ₄] ⁻ , [NTf ₂] ⁻ , [SCN] ⁻ , [OAc] ⁻ , [OTf] ⁻ , [eFAP] ⁻ , [DCA] ⁻ , [C ₂ SO ₄] ⁻ , [C ₈ SO ₄] ⁻ , [C ₁ SO ₃] ⁻ , [Tos] ⁻ , Cl ⁻	172	263.15–373.15	39, 45–51
2	[C ₄ mim] ⁺	[BF ₄] ⁻ , [C ₁ SO ₄] ⁻ , [NTf ₂] ⁻ , [PF ₆] ⁻ , [SCN] ⁻ , [OAc] ⁻ , [OTf] ⁻ , [eFAP] ⁻ , [DCA] ⁻ , [C ₈ SO ₄] ⁻	112	273.15–373.15	8, 49, 52–59
3	[C ₃ mim] ⁺	[NTf ₂] ⁻ , [PF ₆] ⁻	20	298.15–363.15	60, 61
4	[C ₄ m(3)py] ⁺	[BF ₄] ⁻ , [NTf ₂] ⁻ , [DCA] ⁻	36	278.15–363.15	62–64
5	[P ₆₆₆₁₄] ⁺	[eFAP] ⁻ , [DCA] ⁻ , [NTf ₂] ⁻ , [C ₁ SO ₃] ⁻ , Br ⁻	86	268.15–373.15	65–67
6	[C ₄ mmim] ⁺	[BF ₄] ⁻ , [NTf ₂] ⁻ , [eFAP] ⁻	50	273.15–373.15	67, 68
7	[C ₄ py] ⁺	[BF ₄] ⁻ , [NTf ₂] ⁻	22	278.15–363.15	63, 69
8	[C ₆ mim] ⁺	[BF ₄] ⁻ , [PF ₆] ⁻ , [eFAP] ⁻ , [NTf ₂] ⁻ , Br ⁻ , Cl ⁻	54	283.15–363.15	8, 52, 70–72
9	[C ₁₀ mim] ⁺	[BF ₄] ⁻ , [PF ₆] ⁻ , [NTf ₂] ⁻ , [OTf] ⁻	28	283.15–363.15	8, 73
10	[C ₁ mim] ⁺	[NTf ₂] ⁻ , [C ₁ SO ₄] ⁻	15	283.15–353.15	74, 75
11	[C ₄ mpyrro] ⁺	[C ₁ SO ₄] ⁻ , [NTf ₂] ⁻ , [SCN] ⁻ , [OAc] ⁻ , [OTf] ⁻ , [eFAP] ⁻ , [DCA] ⁻	90	283.15–373.15	65, 67, 76–79
12	[C ₄ m(4)py] ⁺	[BF ₄] ⁻ , [NTf ₂] ⁻ , [SCN] ⁻	32	278.15–363.15	63, 77, 80
13	[C ₈ mim] ⁺	[BF ₄] ⁻ , [PF ₆] ⁻ , [NTf ₂] ⁻ , [OTf] ⁻ , Cl ⁻	40	273.15–363.15	8, 81–84
14	[C ₈ py] ⁺	[BF ₄] ⁻ , [NTf ₂] ⁻	33	278.15–363.15	63, 85
15	[N ₁₁₁₄] ⁺	[NTf ₂] ⁻	15	283.15–353.15	86
16	[N ₈₈₈₁] ⁺	[NTf ₂] ⁻	8	273.15–333.15	87
17	[C ₆ py] ⁺	[NTf ₂] ⁻	9	298.15–338.15	88
code	anion	cation	N_{data}	T_{range} (K)	ref
1	[BF ₄] ⁻	[C ₂ mim] ⁺ , [C ₄ mim] ⁺ , [C ₄ mmim] ⁺ , [C ₄ py] ⁺ , [C ₆ mim] ⁺ , [C ₁₀ mim] ⁺ , [C ₄ m(4)py] ⁺ , [C ₈ mim] ⁺ , [C ₈ py] ⁺ , [C ₄ m(3)py] ⁺	87	273.15–363.15	8, 45, 52, 64, 68, 69, 80, 82, 85
2	[C ₁ SO ₄] ⁻	[C ₂ mim] ⁺ , [C ₄ mim] ⁺ , [C ₄ mpyrro] ⁺ , [C ₁ mim] ⁺	45	283.15–373.15	46, 53, 75, 76
3	[NTf ₂] ⁻	[C ₄ mim] ⁺ , [C ₃ mim] ⁺ , [P ₆₆₆₁₄] ⁺ , [C ₄ py] ⁺ , [C ₄ m(3)py] ⁺ , [C ₄ mmim] ⁺ , [C ₁ mim] ⁺ , [C ₂ mim] ⁺ , [C ₄ mpyrro] ⁺ , [C ₄ m(4)py] ⁺ , [C ₈ mim] ⁺ , [C ₈ py] ⁺ , [C ₆ mim] ⁺ , [N ₁₁₁₄] ⁺ , [N ₈₈₈₁] ⁺ , [C ₆ py] ⁺	223	263.15–373.15	47, 54, 60, 63, 65–67, 72–74, 81, 86–88
4	[PF ₆] ⁻	[C ₄ mim] ⁺ , [C ₃ mim] ⁺ , [C ₈ mim] ⁺ , [C ₆ mim] ⁺ , [C ₁₀ mim] ⁺	38	293.15–363.15	8, 55, 61, 70, 83
5	[DCA] ⁻	[P ₆₆₆₁₄] ⁺ , [C ₄ m(3)py] ⁺ , [C ₂ mim] ⁺ , [C ₄ mpyrro] ⁺ , [C ₄ mim] ⁺	68	278.15–363.15	39, 58, 62, 65, 79
6	[SCN] ⁻	[C ₂ mim] ⁺ , [C ₄ mim] ⁺ , [C ₄ m(4)py] ⁺ , [C ₄ mpyrro] ⁺	48	278.15–368.15	39, 56, 77
7	[OAc] ⁻	[C ₂ mim] ⁺ , [C ₄ mim] ⁺ , [C ₄ mpyrro] ⁺	29	298.15–363.15	48, 57, 78
8	[OTf] ⁻	[C ₂ mim] ⁺ , [C ₄ mim] ⁺ , [C ₄ mpyrro] ⁺ , [C ₈ mim] ⁺ , [C ₁₀ mim] ⁺	64	278.15–373.15	8, 39, 67
9	[eFAP] ⁻	[C ₂ mim] ⁺ , [C ₄ mim] ⁺ , [C ₄ mpyrro] ⁺ , [C ₆ mim] ⁺ , [P ₆₆₆₁₄] ⁺ , [C ₄ mmim] ⁺	89	268.15–373.15	49, 67, 70
10	[C ₂ SO ₄] ⁻	[C ₂ mim] ⁺	10	298.15–343.15	50
11	Br ⁻	[C ₆ mim] ⁺ , [P ₆₆₆₁₄] ⁺	27	278.15–363.15	66, 71
12	[C ₈ SO ₄] ⁻	[C ₂ mim] ⁺ , [C ₄ mim] ⁺	28	283.15–373.15	46, 59
13	[C ₁ SO ₃] ⁻	[C ₂ mim] ⁺ , [P ₆₆₆₁₄] ⁺	36	278.15–363.15	39, 66
14	[Tos] ⁻	[C ₂ mim] ⁺	13	303.15–363.15	39
15	Cl ⁻	[C ₂ mim] ⁺ , [C ₈ mim] ⁺ , [C ₆ mim] ⁺	17	283.15–363.15	8, 51, 84

^a N_{data} number of data points; T_{range} temperature range.

liquid mixtures. The main equation of the UNIFAC–VISCO model is

$$\ln(\mu) = \sum_{i=1}^C x_i \ln\left(\mu_i \cdot \frac{V_i}{V_m}\right) + \frac{g_c^E}{RT} - \frac{g_r^E}{RT} \quad (2)$$

Herein, μ is the viscosity of the ionic liquid; subscript i represents the component in the ionic liquid (i.e., the cation and anion); C is the number of components in the ionic liquid ($C = 2$); x_i is the mole fraction of the cation/anion in the ionic liquid, which is equal to 0.5; μ_i is the effective viscosity of the ion and represented by the Vogel–Fulcher–Tamman (VFT) equation (eq 3); V_i is the effective molar volume of the ion and calculated using the volumetric parameters (eq 4), which were developed based on a group contribution method developed by our group previously;^{91,92} V_m is the molar volume of the ionic liquid, and is calculated based on the assumption that the molar volume of the ionic liquid is

the sum of the effective molar volumes of the cation and anion.^{91,92}

$$\mu_{\text{ion}} = A \cdot \exp\left[\frac{B}{T - T_0}\right] \quad (3)$$

$$V_{\text{ion}}^*(T^*, p_{\text{ref}}) = \sum_{i=0}^2 (a_i \cdot (T^*)_{p_{\text{ref}}}^i) \quad (4)$$

where $T^* = T - 298.15$, $p_{\text{ref}} = 0.1$ MPa, and a_i is the volumetric parameters as described by Jacquemin et al.^{91,92} The values of the volumetric parameters used in this paper are given in Table S1 of the Supporting Information.

The residual term and the combinatorial term in eq 2 are defined as follows:

$$\frac{g_r^E}{RT} = \sum_{i=1}^C x_i \left[\sum_{m=1}^N n_{m,i} (\ln \gamma_m - \ln \gamma_{m,i}) \right] \quad (5)$$

Table 3. Ionic Liquids of the Recommended References Including Number of Data Points and Temperature Ranges (see Figures S1–S86 of the Supporting Information)

code	ionic liquid	N_{data}	T_{range} (K)	ref	code	ionic liquid	N_{data}	T_{range} (K)	ref
1	[C ₂ mim][BF ₄]	4	282.55–323.65	45	36	[C ₄ mim][OTf]	9	283.15–363.15	8
2	[C ₄ mim][BF ₄]	7	293.15–323.15	52	37	[C ₈ mim][OTf]	9	283.15–363.15	8
3	[C ₄ mim][C ₁ SO ₄]	9	293.15–343.15	53	38	[C ₄ mpyrro][OTf]	19	283.15–373.15	67
4	[C ₂ mim][C ₁ SO ₄]	19	283.15–373.15	46	39	[C ₁₀ mim][OTf]	9	283.15–363.15	8
5	[C ₄ mim][NTf ₂]	21	273.15–353.15	54	40	[C ₈ mim][NTf ₂]	9	283.15–353.15	81
6	[C ₃ mim][NTf ₂]	10	298.15–343.15	60	41	[C ₈ mim][BF ₄]	14	273.15–353.15	82
7	[C ₃ mim][PF ₆]	10	318.15–363.15	61	42	[C ₈ mim][PF ₆]	4	293.15–353.15	83
8	[C ₄ mim][PF ₆]	7	293.15–323.15	55	43	[C ₂ mim][eFAP]	9	293.15–373.15	49
9	[C ₄ m(3)py][DCA]	10	283.15–328.15	62	44	[C ₄ mim][eFAP]	9	293.15–373.15	49
10	[P ₆₆₆₁₄][DCA]	11	298.15–343.15	65	45	[C ₆ mim][eFAP]	11	293.15–343.15	70
11	[P ₆₆₆₁₄][NTf ₂]	17	278.15–358.15	66	46	[C ₄ mpyrro][eFAP]	19	283.15–373.15	67
12	[C ₄ m(3)py][NTf ₂]	18	278.15–363.15	63	47	[C ₄ mmim][eFAP]	19	283.15–373.15	67
13	[C ₄ mmim][BF ₄]	7	313.15–373.15	68	48	[P ₆₆₆₁₄][eFAP]	22	268.15–373.15	67
14	[C ₄ py][BF ₄]	4	293.15–323.15	69	49	[C ₂ mim][DCA]	18	278.15–363.15	39
15	[C ₄ py][NTf ₂]	18	278.15–363.15	63	50	[C ₂ mim][C ₂ SO ₄]	10	298.15–343.15	50
16	[C ₄ mmim][NTf ₂]	21	273.15–373.15	67	51	[C ₆ mim]Br	9	293.15–333.15	71
17	[C ₆ mim][BF ₄]	8	288.15–323.15	52	52	[P ₆₆₆₁₄]Br	18	278.15–363.15	66
18	[C ₁₀ mim][BF ₄]	9	283.15–363.15	8	53	[C ₄ mim][DCA]	18	278.15–363.15	58
19	[C ₁₀ mim][PF ₆]	6	313.15–363.15	8	54	[C ₄ mpyrro][DCA]	11	293.15–343.15	79
20	[C ₆ mim][PF ₆]	11	293.15–343.15	70	55	[C ₈ py][BF ₄]	15	283.15–353.15	85
21	[C ₁ mim][NTf ₂]	8	283.15–353.15	74	56	[C ₈ py][NTf ₂]	18	278.15–363.15	63
22	[C ₄ mpyrro][NTf ₂]	11	293.15–343.15	65	57	[C ₄ m(3)py][BF ₄]	8	283–343	64
23	[C ₄ mpyrro][C ₁ SO ₄]	10	298.15–343.15	76	58	[C ₂ mim][C ₈ SO ₄]	19	283.15–373.15	46
24	[C ₁ mim][C ₁ SO ₄]	7	293.15–343.15	75	59	[C ₄ mim][C ₈ SO ₄]	9	293.15–373.15	59
25	[C ₂ mim][NTf ₂]	12	263.15–353.15	47	60	[C ₂ mim][C ₁ SO ₃]	18	278.15–363.15	39
26	[C ₄ m(4)py][BF ₄]	8	298.15–348.15	80	61	[P ₆₆₆₁₄][C ₁ SO ₃]	18	278.15–363.15	66
27	[C ₄ m(4)py][NTf ₂]	18	278.15–363.15	63	62	[C ₂ mim][Tos]	13	303.15–363.15	39
28	[C ₂ mim][SCN]	18	278.15–363.15	39	63	[C ₆ mim][NTf ₂]	6	298.15–343.15	72
29	[C ₄ mim][SCN]	18	283.15–368.15	56	64	[C ₁₀ mim][NTf ₂]	4	293.15–323.15	73
30	[C ₄ m(4)py][SCN]	6	298.15–348.15	77	65	[N ₁₁₁₄][NTf ₂]	15	283.15–353.15	86
31	[C ₄ mpyrro][SCN]	6	298.15–348.15	77	66	[N ₈₈₈₁][NTf ₂]	8	273.15–333.15	87
32	[C ₂ mim][OAc]	10	298.15–343.15	48	67	[C ₆ py][NTf ₂]	9	298.15–338.15	88
33	[C ₄ mim][OAc]	5	303.15–343.15	57	68	[C ₂ mim]Cl	4	298.15–313.15	51
34	[C ₄ mpyrro][OAc]	14	298.15–363.15	78	69	[C ₆ mim]Cl	9	283.15–363.15	8
35	[C ₂ mim][OTf]	18	278.15–363.15	39	70	[C ₈ mim]Cl	4	298.15–343.15	84

Table 4. Availability of the Experimental Data for the Selected ILs

combination	[BF ₄] ⁻	[SCN] ⁻	[NTf ₂] ⁻
[C ₂ mim] ⁺	✓	✓	✓
[C ₄ mim] ⁺	✓	✓	✓
[C ₄ m(4)py] ⁺	✓	✓	✓
[C ₄ mpyrro] ⁺		✓	✓

$$\frac{g_c^E}{RT} = \sum_i x_i \ln \frac{\phi_i}{x_i} + \frac{z}{2} \sum_i x_i q_i \ln \frac{\theta_i}{\phi_i} \quad (6)$$

where z is a coordinate value equal to 10, and

$$\theta_i = \frac{x_i q_i}{\sum_j x_j q_j} \quad (7)$$

$$\phi_i = \frac{x_i r_i}{\sum_j x_j r_j} \quad (8)$$

$$q_i = \sum_{k=1}^N n_{i,k} Q_k \quad (9)$$

$$r_i = \sum_{k=1}^N n_{i,k} R_k \quad (10)$$

Herein, the volume R and surface area Q parameters of ions used were obtained by using the COSMOthermX software (version C30_1601) following the methodology previously described by our group⁹³ and are given in Table S2 of the Supporting Information. R and Q values reported herein are higher but in good agreement (i.e., RAAD close to 7.3% and 12% for R and Q values, respectively) with values reported in the case of the [C_{*n*}mim][BF₄] with $n = 2, 4, 6$ or 8 , [C_{*n*}mim][PF₆] with $n = 2$ or 4 , [C_{*n*}mim][OTf] with $n = 4$ or 6 , and [C₈mim]Cl by Santiago et al.⁹⁴ using a DFT-PCM computational approach, for example. Similarly, good agreements are observed between our set of R and Q values with data determined by Banerjee et al.⁹⁵ using a DFT-PCM computational approach. However, by using the empirical methodology proposed originally by Domańska,⁹⁶ we observed that calculated R and Q values of investigated ionic liquids are systematically lower than values reported herein and in the literature with RAAD close to (52% and 48%) with this work, (41% and 29%) and (82% and 33%) with data reported by Santiago et al.⁹⁴ and by Banerjee et al.,⁹⁵ respectively.

Table 5. Calculated Binary Interaction Parameters of the Selected Cations and Anions

<i>i</i>	<i>j</i>	α_{ij}	α_{ji}	<i>i</i>	<i>j</i>	α_{ij}	α_{ji}
[BF ₄] ⁻	[DCA] ⁻	331.97	-562.66	[C ₄ mim] ⁺	[DCA] ⁻	361.00	-291.30
[BF ₄] ⁻	[C ₂ SO ₄] ⁻	-111.50	41.59	[C ₄ mim] ⁺	[C ₆ SO ₄] ⁻	-179.20	124.94
[BF ₄] ⁻	[PF ₆] ⁻	-20.70	-59.91	[C ₄ mim] ⁺	[C ₄ py] ⁺	272.88	-294.87
[C ₁₀ mim] ⁺	[BF ₄] ⁻	77.85	-410.46	[C ₄ mim] ⁺	[C ₆ mim] ⁺	-79.43	62.39
[C ₁₀ mim] ⁺	[PF ₆] ⁻	1004.9	-452.60	[C ₄ mmim] ⁺	[eFAP] ⁻	-264.20	74.33
[C ₁₀ mim] ⁺	[OTf] ⁻	394.03	-402.07	[C ₄ mmim] ⁺	[BF ₄] ⁻	-105.01	-768.66
[C ₁₀ mim] ⁺	[NTf ₂] ⁻	128.91	97.59	[C ₄ mmim] ⁺	[NTf ₂] ⁻	-193.25	-8.28
[C ₁ mim] ⁺	[NTf ₂] ⁻	-391.35	931.41	[C ₄ mpyrro] ⁺	[NTf ₂] ⁻	-127.47	-29.34
[C ₁ mim] ⁺	[C ₂ SO ₄] ⁻	-158.30	-54.69	[C ₄ mpyrro] ⁺	[C ₂ SO ₄] ⁻	405.62	-609.91
[C ₂ mim] ⁺	[BF ₄] ⁻	-222.19	-130.81	[C ₄ mpyrro] ⁺	[SCN] ⁻	-42.61	-18.69
[C ₂ mim] ⁺	[C ₂ SO ₄] ⁻	566.37	-354.46	[C ₄ mpyrro] ⁺	[OAc] ⁻	-365.86	-53.83
[C ₂ mim] ⁺	[NTf ₂] ⁻	-322.07	1978.2	[C ₄ mpyrro] ⁺	[OTf] ⁻	121.62	-486.62
[C ₂ mim] ⁺	[SCN] ⁻	5717.4	376.59	[C ₄ mpyrro] ⁺	[eFAP] ⁻	-416.08	356.83
[C ₂ mim] ⁺	[OAc] ⁻	-611.17	6394.8	[C ₄ mpyrro] ⁺	[DCA] ⁻	-208.95	-53.51
[C ₂ mim] ⁺	[OTf] ⁻	1717.39	-465.92	[C ₄ py] ⁺	[BF ₄] ⁻	-404.47	7644.6
[C ₂ mim] ⁺	[eFAP] ⁻	-376.69	807.02	[C ₄ py] ⁺	[NTf ₂] ⁻	86.82	417.64
[C ₂ mim] ⁺	[DCA] ⁻	9765.3	-260.54	[C ₆ mim] ⁺	[BF ₄] ⁻	-155.98	-414.47
[C ₂ mim] ⁺	[C ₆ SO ₄] ⁻	355.51	423.56	[C ₆ mim] ⁺	[PF ₆] ⁻	2912.7	-584.72
[C ₂ mim] ⁺	[C ₆ SO ₄] ⁻	-423.54	2156.2	[C ₆ mim] ⁺	[eFAP] ⁻	439.06	-274.24
[C ₂ mim] ⁺	[C ₂ SO ₃] ⁻	1210.8	1163.3	[C ₆ mim] ⁺	Br ⁻	649.67	-652.40
[C ₂ mim] ⁺	[Tos] ⁻	35.38	-13.35	[C ₆ mim] ⁺	[NTf ₂] ⁻	-251.08	415.53
[C ₂ mim] ⁺	Cl ⁻	-2855.0	-0.56	[C ₆ mim] ⁺	Cl ⁻	905.23	-1620.8
[C ₂ mim] ⁺	[C ₆ mim] ⁺	-215.62	199.37	[C ₆ py] ⁺	[NTf ₂] ⁻	10.57	-8.10
[C ₃ mim] ⁺	[NTf ₂] ⁻	67.96	-111.31	[C ₈ mim] ⁺	[OTf] ⁻	304.75	-419.22
[C ₃ mim] ⁺	[PF ₆] ⁻	2723.3	-599.61	[C ₈ mim] ⁺	[NTf ₂] ⁻	80.21	36.46
[C ₄ m ₍₄₎ py] ⁺	[BF ₄] ⁻	-678.30	1116.6	[C ₈ mim] ⁺	[BF ₄] ⁻	-443.32	29.73
[C ₄ m ₍₄₎ py] ⁺	[NTf ₂] ⁻	65.77	-9.79	[C ₈ mim] ⁺	[PF ₆] ⁻	-506.07	1351.0
[C ₄ m ₍₄₎ py] ⁺	[SCN] ⁻	917.22	-113.69	[C ₈ mim] ⁺	Cl ⁻	35183	-1499.7
[C ₄ m ₍₄₎ py] ⁺	[C ₂ mim] ⁺	154.59	21.49	[C ₈ py] ⁺	[BF ₄] ⁻	-504.65	909.79
[C ₄ m ₍₄₎ py] ⁺	[C ₂ SO ₄] ⁻	130.48	30.82	[C ₈ py] ⁺	[NTf ₂] ⁻	-232.89	73830
[C ₄ m ₍₄₎ py] ⁺	[C ₄ py] ⁺	-109.87	41.82	[N ₁₁₁₄] ⁺	[NTf ₂] ⁻	-3.97	-9.63
[C ₄ m ₍₃₎ py] ⁺	[DCA] ⁻	4174.7	-336.84	[N ₈₈₈₁] ⁺	[NTf ₂] ⁻	17.24	27.69
[C ₄ m ₍₃₎ py] ⁺	[NTf ₂] ⁻	702.97	-262.88	[NTf ₂] ⁻	[C ₂ SO ₄] ⁻	132.29	-123.01
[C ₄ m ₍₃₎ py] ⁺	[BF ₄] ⁻	-660.43	1990.6	[NTf ₂] ⁻	[BF ₄] ⁻	-314.98	113.81
[C ₄ mim] ⁺	[BF ₄] ⁻	-0.06	-464.73	[OAc] ⁻	[C ₂ SO ₄] ⁻	-136.52	2380.7
[C ₄ mim] ⁺	[C ₂ SO ₄] ⁻	112.45	-365.32	[P ₆₆₆₁₄] ⁺	[DCA] ⁻	2040.7	-577.60
[C ₄ mim] ⁺	[NTf ₂] ⁻	322.36	-228.33	[P ₆₆₆₁₄] ⁺	[NTf ₂] ⁻	1463.7	-403.99
[C ₄ mim] ⁺	[PF ₆] ⁻	1248.42	-544.06	[P ₆₆₆₁₄] ⁺	[eFAP] ⁻	1001.6	-359.16
[C ₄ mim] ⁺	[SCN] ⁻	57.26	346530	[P ₆₆₆₁₄] ⁺	Br ⁻	-873.04	29571
[C ₄ mim] ⁺	[OAc] ⁻	4669.6	-712.86	[P ₆₆₆₁₄] ⁺	[C ₂ SO ₃] ⁻	29517	-276.78
[C ₄ mim] ⁺	[OTf] ⁻	-280.50	-14.04	[SCN] ⁻	[C ₂ SO ₄] ⁻	-324.50	287.34
[C ₄ mim] ⁺	[eFAP] ⁻	-256.62	270.83	[SCN] ⁻	[DCA] ⁻	-22.96	252.44

For example, in the case of the [C₄mim][PF₆], *R* and *Q* values have been reported in the literature as close to (*R* = 8.5506 and *Q* = 6.5172 using Bondi method and ionic radii,⁹⁷ (*R* = 8.4606 and *Q* = 6.808 determined by using a DFT-PCM computational approach)⁹⁴ and (*R* = 11.034 and *Q* = 6.958 using a DFT-PCM computational approach)⁹⁵ such values match very well with those determined during this work (*R* = 9.2940 and *Q* = 7.8352) while a large difference is observed in each case by using the empirical methodology proposed originally by Domańska⁹⁶ (*R* = 6.0582 and *Q* = 5.2466) and in fact parameters reported in Table S2 of the Supporting Information can be considered as representative.

Then, $\ln \gamma_m$ the residual activity coefficient in the residual term (eq 5), is obtained by using eq 11:

$$\ln \gamma_m = Q_m \left[1 - \ln \left(\sum_{i=1}^N \Theta_i \Psi_{i,m} \right) - \sum_{i=1}^N \frac{\Theta_i \Psi_{m,i}}{\sum_{j=1}^N \Theta_j \Psi_{j,i}} \right] \quad (11)$$

where:

$$\Theta_m = \frac{x_m Q_m}{\sum_{i=1}^N X_i Q_i}, \quad m = 1, 2, 3, \dots, N \quad (12)$$

where the subscripts represent the defined groups, and

$$\Psi_{mn} = \exp \left(-\frac{\alpha_{mn}}{298.15} \right) \quad (13)$$

The unknown parameters, binary interaction parameters (α_{mn} and α_{nm}) and ion VFT fitting parameters (*A*, *B*, and *T*₀) were estimated by minimizing the objective function (eq 14) using the Marquardt⁹⁸ optimization technique:

$$\text{OF} = \frac{1}{M} \sum_{i=1}^M \left(\frac{\mu_{\text{exp}} - \mu_{\text{cal}}}{\mu_{\text{exp}}} \right)^2 \rightarrow \min \quad (14)$$

where *M* is the number of data points; μ_{exp} and μ_{cal} are the experimental and calculated viscosity data, respectively.

This method is faster than the regression along with an influence function used earlier in the paper. It is worth mentioning that the estimators derived from mathematical gnostics converge to the ordinary least-squares method if the errors are sufficiently small as can be proven by a Taylor series expansion. In this stage, regression was applied to the recommended data sets only which ensures that errors are small enough and a faster algorithm can be utilized.

3.2. Extension of the UNIFAC–VISCO Method. In our previous study,³³ 154 experimental data of the selected 25 ILs were used during the overall regression. In this work, these data were substituted by the recommended ones to estimate the corresponding binary interaction parameters and the VFT parameters of previously selected ions. Then based on the updated parameters, the unknown parameters of other 45 ionic liquids were optimized using their corresponding recommended data sets.

Table 4 shows the availability of the experimental data in the literature for different combinations of cations ($[\text{C}_2\text{mim}]^+$, $[\text{C}_4\text{mim}]^+$, $[\text{C}_4\text{m}_{(4)}\text{py}]^+$, and $[\text{C}_4\text{mpyrro}]^+$) and anions ($[\text{BF}_4]^-$, $[\text{SCN}]^-$, and $[\text{NTf}_2]^-$). It can be observed that the experimental data of $[\text{SCN}]^-$ -based ionic liquids are only reported in the literature for $[\text{C}_2\text{mim}][\text{SCN}]$,³⁹ $[\text{C}_4\text{mim}][\text{SCN}]$,⁵⁶ $[\text{C}_4\text{m}_{(4)}\text{py}][\text{SCN}]$,⁷⁷ and $[\text{C}_4\text{mpyrro}][\text{SCN}]$.⁷⁷ The experimental data of $[\text{C}_4\text{m}_{(4)}\text{py}]^+$ -based ionic liquids are only available in the literature for $[\text{C}_4\text{m}_{(4)}\text{py}][\text{BF}_4]$,⁸⁰ $[\text{C}_4\text{m}_{(4)}\text{py}][\text{SCN}]$,⁷⁷ and $[\text{C}_4\text{m}_{(4)}\text{py}][\text{NTf}_2]$.⁶³ In other words, the experimental data of the ionic liquids consisting of each ion denoted with bold letters (see Table 4) having been reported in literature are all listed in Table 4. For the other ions the VFT fitting parameters have been obtained from the regression analysis of the first 25 ILs. Therefore, the VFT parameters of only $[\text{C}_4\text{m}_{(4)}\text{py}]^+$, and $[\text{SCN}]^-$ are unknown. The VFT parameters of $[\text{C}_4\text{m}_{(4)}\text{py}]^+$ only depend on the available experimental values of $[\text{C}_4\text{m}_{(4)}\text{py}][\text{BF}_4]$, $[\text{C}_4\text{m}_{(4)}\text{py}][\text{SCN}]$, and $[\text{C}_4\text{m}_{(4)}\text{py}][\text{NTf}_2]$. The VFT parameters of $[\text{SCN}]^-$ only depend on the available experimental values of $[\text{C}_2\text{mim}][\text{SCN}]$, $[\text{C}_4\text{mim}][\text{SCN}]$, $[\text{C}_4\text{m}_{(4)}\text{py}][\text{SCN}]$, and $[\text{C}_4\text{mpyrro}][\text{SCN}]$. Herein, the experimental data of these six ionic liquids were regressed together to get the proper VFT fitting parameters of $[\text{C}_4\text{m}_{(4)}\text{py}]^+$, and $[\text{SCN}]^-$, and the binary interaction parameters of these six ionic liquids as well. Following this methodology and based on these parameters having been obtained,

more binary interaction parameters and VFT parameters for other ILs were optimized.

The regression procedure was completed by developing scripts in MATLAB. MATLAB program (available in the Supporting Information; filename: Calculation_Single_Ionic_Liquid) contains all parameters (binary interaction and VFT parameters) estimated during this study. This program can read experimental data of pure ionic liquids from 'Recommended data sets.xlsx', determine the evaluated viscosity data of selected ionic liquids, and calculate the relative average absolute deviation (RAAD) between the calculated and the experimental viscosity data for each ionic liquid, for each series of ionic liquids (like imidazolium, pyridinium, and ammonium-based ionic liquids), and for all the 70 ionic liquids. Plotting for the comparison between the predicted values and the experimental data can be shown as well.

The MATLAB program (Filename: Calculation_Binary_Ionic_Liquids) contains all parameters for the binary mixtures of ionic liquids estimated during this study. The program can read experimental data of binary mixtures from 'Binary data.xlsx', determine the evaluated viscosity of selected mixtures, and calculate the RAAD between the calculated and the experimental viscosity data for each binary system, and also for the 11 types of binary mixtures. Plotting for the comparison can be shown as well.

4. RESULTS AND DISCUSSION

Binary interaction parameters (see Table 5) for 70 pure ILs and VFT parameters (see Table 6) for 17 cations and 15 anions were estimated by following the regression analysis (eq 14) of 819 experimental data points.

The quality of the regression was then determined by calculating the relative average absolute deviations (RAAD, eq 15) between the calculated and the experimental viscosity data:

$$\text{RAAD} = 100 \times \frac{1}{M} \sum_i^M \left| \frac{\mu_{\text{exp}} - \mu_{\text{cal}}}{\mu_{\text{exp}}} \right| \quad (15)$$

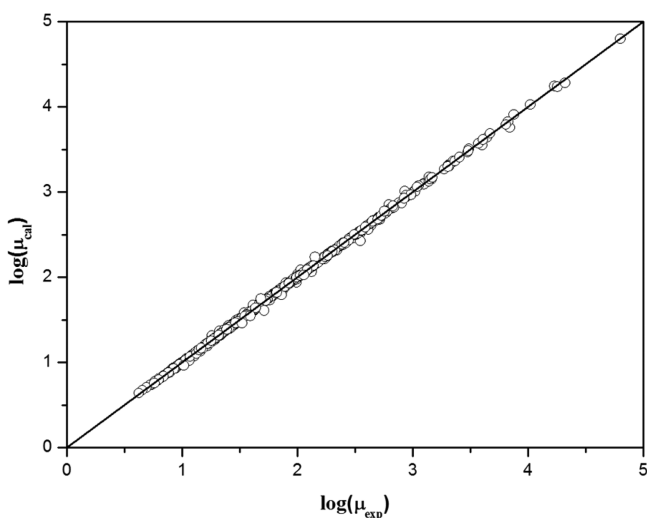
The RAAD values for the regression of each pure IL are presented in Table 7. The model results show a global relative average absolute deviation of 1.4% from the 819 experimental data, which demonstrate an excellent correlation ability of this

Table 6. VFT Fitting Parameters of the Selected Cations and Anions

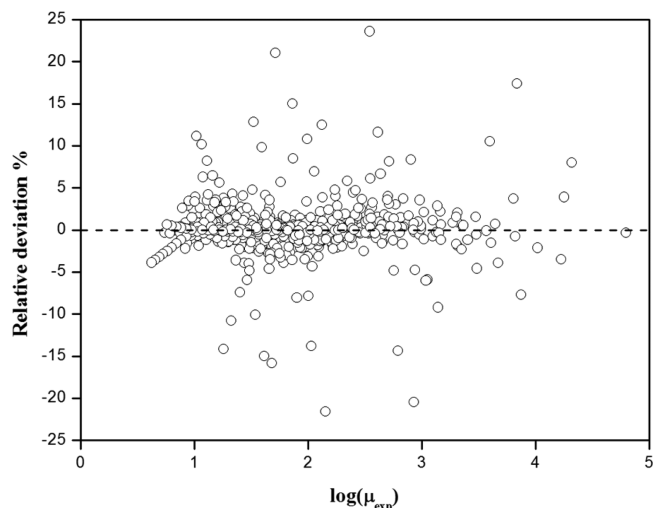
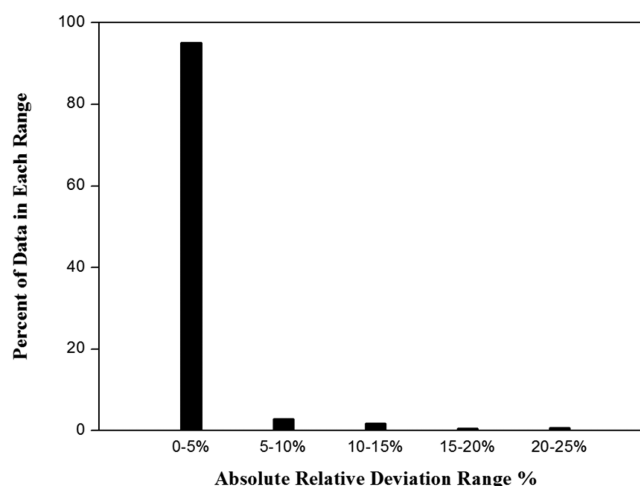
cation/anion	A (mPa·s)	B (K)	T ₀ (K)	cation/anion	A (mPa·s)	B (K)	T ₀ (K)
$[\text{C}_1\text{mim}]^+$	0.521	385.2	182.9	$[\text{C}_6\text{py}]^+$	0.034	1066.3	171.6
$[\text{C}_2\text{mim}]^+$	0.436	792.8	110.1	$[\text{BF}_4]^-$	0.403	532.9	182.7
$[\text{C}_3\text{mim}]^+$	0.091	664.4	180.4	$[\text{C}_1\text{SO}_4]^-$	0.517	756.9	186.0
$[\text{C}_4\text{mim}]^+$	0.062	926.9	155.8	$[\text{NTf}_2]^-$	2.007	581.0	170.7
$[\text{C}_6\text{mim}]^+$	0.032	1192.9	145.7	$[\text{PF}_6]^-$	6.206	378.1	215.4
$[\text{C}_{10}\text{mim}]^+$	0.013	1965.2	142.1	$[\text{DCA}]^-$	4.760	282.5	197.9
$[\text{C}_4\text{mmim}]^+$	0.010	1075.2	161.1	$[\text{SCN}]^-$	8.425	690.3	177.9
$[\text{C}_4\text{py}]^+$	0.587	1031.8	171.6	$[\text{OAc}]^-$	3.497	254.1	244.2
$[\text{C}_4\text{m}_{(3)}\text{py}]^+$	0.250	766.9	174.8	$[\text{OTf}]^-$	1.492	412.2	188.0
$[\text{C}_4\text{mpyrro}]^+$	0.022	966.9	160.7	$[\text{eFAP}]^-$	1.371	587.6	183.6
$[\text{P}_{66614}]^+$	2.520	1029.4	130.0	$[\text{C}_2\text{SO}_4]^-$	1.049	1285.1	174.4
$[\text{C}_4\text{m}_{(4)}\text{py}]^+$	0.063	944.8	171.2	Br^-	0.036	1958.5	161.8
$[\text{C}_8\text{mim}]^+$	0.021	1577.3	143.6	$[\text{C}_8\text{SO}_4]^-$	0.611	1159.5	184.4
$[\text{C}_8\text{py}]^+$	1.781	772.0	182.7	$[\text{C}_1\text{SO}_3]^-$	2.190	1344.0	183.1
$[\text{N}_{1114}]^+$	0.060	989.1	176.8	$[\text{Tos}]^-$	0.226	1006.4	232.7
$[\text{N}_{8881}]^+$	0.022	1828.4	161.8	Cl^-	0.006	71.7	258.6

Table 7. Relative Absolute Average Deviation (RAAD) of the Calculated Viscosity from the Experimental Viscosity Data for Selected ILs

IL	RAAD (%)	IL	RAAD (%)
[C ₂ mim][BF ₄]	1.71	[C ₄ mim][OTf]	1.09
[C ₄ mim][BF ₄]	0.25	[C ₈ mim][OTf]	1.00
[C ₄ mim][C ₁ SO ₄]	0.17	[C ₄ mpyrro][OTf]	0.72
[C ₂ mim][C ₁ SO ₄]	0.16	[C ₁₀ mim][OTf]	0.99
[C ₄ mim][NTf ₂]	0.38	[C ₈ mim][NTf ₂]	0.88
[C ₃ mim][NTf ₂]	0.13	[C ₈ mim][BF ₄]	0.53
[C ₃ mim][PF ₆]	0.11	[C ₈ mim][PF ₆]	0.88
[C ₄ mim][PF ₆]	0.29	[C ₂ mim][eFAP]	0.39
[C ₄ m(3)py][DCA]	0.23	[C ₄ mim][eFAP]	1.45
[P ₆₆₆₁₄][DCA]	0.42	[C ₆ mim][eFAP]	0.48
[P ₆₆₆₁₄][NTf ₂]	0.22	[C ₄ mpyrro][eFAP]	1.60
[C ₄ m(3)py][NTf ₂]	0.12	[C ₄ mmim][eFAP]	2.16
[C ₄ mmim][BF ₄]	1.63	[P ₆₆₆₁₄][eFAP]	1.23
[C ₄ py][BF ₄]	3.46	[C ₂ mim][DCA]	2.31
[C ₄ py][NTf ₂]	0.83	[C ₂ mim][C ₂ SO ₄]	0.14
[C ₄ mmim][NTf ₂]	1.84	[C ₆ mim]Br	12.33
[C ₆ mim][BF ₄]	0.45	[P ₆₆₆₁₄]Br	1.44
[C ₁₀ mim][BF ₄]	1.73	[C ₄ mim][DCA]	0.40
[C ₁₀ mim][PF ₆]	0.57	[C ₄ mpyrro][DCA]	7.19
[C ₆ mim][PF ₆]	0.54	[C ₈ py][BF ₄]	1.12
[C ₁ mim][NTf ₂]	0.73	[C ₈ py][NTf ₂]	0.24
[C ₄ mpyrro][NTf ₂]	0.32	[C ₄ m(3)py][BF ₄]	1.48
[C ₄ mpyrro][C ₁ SO ₄]	0.66	[C ₂ mim][C ₆ SO ₄]	0.80
[C ₁ mim][C ₁ SO ₄]	0.45	[C ₄ mim][C ₆ SO ₄]	3.37
[C ₂ mim][NTf ₂]	1.78	[C ₂ mim][C ₁ SO ₃]	2.71
[C ₄ m(4)py][BF ₄]	3.32	[P ₆₆₆₁₄][C ₁ SO ₃]	2.60
[C ₄ m(4)py][NTf ₂]	0.20	[C ₂ mim][Tos]	0.25
[C ₂ mim][SCN]	0.15	[C ₆ mim][NTf ₂]	0.23
[C ₄ mim][SCN]	0.68	[C ₁₀ mim][NTf ₂]	14.72
[C ₄ m(4)py][SCN]	0.57	[N ₁₁₁₄][NTf ₂]	0.04
[C ₄ mpyrro][SCN]	2.20	[N ₈₈₈₁][NTf ₂]	0.75
[C ₂ mim][OAc]	7.72	[C ₆ py][NTf ₂]	0.34
[C ₄ mim][OAc]	0.62	[C ₂ mim]Cl	9.84
[C ₄ mpyrro][OAc]	4.31	[C ₆ mim]Cl	3.46
[C ₂ mim][OTf]	0.09	[C ₈ mim]Cl	3.99

**Figure 3.** Comparison of experimental (μ_{exp}) and evaluated viscosity (μ_{cal}) data for pure ionic liquids.

model, except in the case of the [C₆mim]Br (RAAD = 12.3%) and [C₁₀mim][NTf₂] (RAAD = 14.7%).

**Figure 4.** Relative deviation of evaluated viscosity values versus experimental data.**Figure 5.** Distribution of relative deviations of evaluated viscosities from experimental ones.

The evaluated viscosities in comparison with the experimental values are presented in Figure 3. In addition, the relative deviation of the model in comparison with the experimental data is shown in Figure 4. The distribution of absolute relative deviations from experimental data is shown in Figure 5. For this figure, it appears that 94.9% of the viscosities are evaluated within 0–5%; 2.7% within 5–10%; 2.0% within 10–20%; and the deviations of only four data points beyond 20%. The four points with larger deviation are from [C₆mim]Br and [C₁₀mim][NTf₂]. However, due to a large range of viscosity values from bromide-based ILs, it was difficult to regress and obtain the optimal parameters of all ILs combined all together.

Tables 8 and 9 show the results for different cation series of ILs and different anion types of ILs, respectively. This deviation for the estimation follows the order for (1) cations as ammonium < pyridinium < phosphonium < imidazolium < pyrrolidinium, and for (2) anions as [C₂SO₄][−] < [Tos][−] < [C₁SO₄][−] < [PF₆][−] < [SCN][−] < [OTf][−] < [NTf₂][−] < [eFAP][−] < [BF₄][−] < [C₈SO₄][−] < [DCA][−] < [C₁SO₃][−] < [OAc][−] < Br[−] < Cl[−].

By using the group interaction potential energy and VFT parameters of ions obtained by the regression of the viscosity data of pure ILs and by following the same methodology, the

Table 8. RAAD for Different Cation Types of ILs

cation type	no. of structures	no. of data	temp. range (K)	experimental viscosity range (mPa·s)	RAAD (%)
pyridinium	11	132	278.15–363.15	7.673–640.14	0.78
pyrrolidinium	7	90	283.15–373.15	8.98–593.7	2.30
phosphonium	5	86	268.15–373.15	17.567–16955	1.26
imidazolium	45	488	263.15–373.15	4.229–63100	1.53
ammonium	2	23	273.15–353.15	14.7–3729.6	0.29

Table 9. RAAD for Different Anion Types of ILs

anion type	no. of structures	no. of data	temp. range (K)	experimental viscosity range (mPa·s)	RAAD (%)
[PF ₆] ⁻	5	38	293.15–363.15	17.77–1052	0.42
[C ₁ SO ₄] ⁻	4	45	283.15–373.15	9–467.5	0.32
Cl ⁻	3	17	283.15–363.15	33.2–63100	5.09
[C ₁ SO ₃] ⁻	2	36	278.15–363.15	13.33–7526.5	2.65
[NTf ₂] ⁻	17	223	263.15–373.15	7.673–3729.6	0.83
[BF ₄] ⁻	10	84	273.15–373.15	18.4–2291	1.38
[DCA] ⁻	5	68	278.15–363.15	4.229–562.45	1.99
[SCN] ⁻	4	48	278.15–368.15	5.485–112.41	0.66
[eFAP] ⁻	6	89	268.15–373.15	7.45–3067.3	1.35
[OAc] ⁻	3	29	298.15–363.15	8.98–288.8	4.85
[C ₂ SO ₄] ⁻	1	10	298.15–343.15	18.72–97.6	0.14
[C ₈ SO ₄] ⁻	2	28	283.15–373.15	23.3–1879	1.62
Br ⁻	2	27	278.15–363.15	85.436–16955	5.07
[Tos] ⁻	1	13	303.15–363.15	33.22–1417	0.25
[OTf] ⁻	5	64	278.15–373.15	8.169–2059	0.67

Table 10. Experimental Viscosity Data of Binary Mixtures of ILs Collected from Literature

binary system	mole fraction range (<i>x</i>)	temp. range (K)	no. of data points	ref
$x[C_2mim][DCA] + (1-x)[C_2mim][BF_4]$	0.05527–0.9552	298.15	7	101
$x[C_6mim][BF_4] + (1-x)[C_2mim][BF_4]$	0.0978–0.9455	293.15–343.15	216	99, 100
$x[C_2mim][OAc] + (1-x)[C_2mim][C_2SO_4]$	0.0993–0.9007	298.15–358.15	63	102
$x[C_2mim][C_2SO_4] + (1-x)[C_2mim][SCN]$	0.0495–0.801	298.15–353.15	72	103
$x[C_2mim][DCA] + (1-x)[C_2mim][SCN]$	0.2397–0.7415	298.15–353.15	36	103
$x[C_4m(4)py][NTf_2] + (1-x)[C_2mim][C_2SO_4]$	0.0513–0.9525	293.15–353.15	70	104
$x[C_4py][BF_4] + (1-x)[C_4m(4)py][NTf_2]$	0.0575–0.9535	293.15–353.15	70	105
$x[C_4mim][BF_4] + (1-x)[C_4mim][C_1SO_4]$	0.1209–0.9447	298.15–308.15	117	99
$x[C_4mim][PF_6] + (1-x)[C_4mim][BF_4]$	0.0568–0.945	298.15–308.15	117	99
$x[C_4mim][BF_4] + (1-x)[C_4py][BF_4]$	0.1009–0.8998	293.15–343.15	99	106
$x[C_6mim][BF_4] + (1-x)[C_4mim][BF_4]$	0.1083–0.946	298.15–308.15	99	99

viscosity of binary mixtures of ILs were then correlated to obtain missing interaction parameters between cation–cation and/or anion–anion. The viscosity data of 11 binary mixtures of ILs reported in eight references were used. The compositions of the mixtures, temperature range, number of data points, and references are summarized in Table 10. Herein, only one set of data for each binary mixture was used as the training set to determine missing group interaction potential energy parameters. The other data available were then used to evaluate the accuracy of the UNIFAC–VISCO model as the function of temperature and composition for each ILs mixture. The prediction result is highlighted by the RAAD values, presented in Table 11, which is 3.8% for all collected binary data. In addition, the parity plot given in Figure 6 shows good agreement between the experimental data and calculated viscosities by using the model. In the case of the ($[C_2mim][BF_4] + [C_6mim][BF_4]$) binary mixture, two viscosity data sets are available in the literature,^{99,100} and discrepancies between these series of measurements, which were performed in two different laboratories for different compositions and temperatures within an uncertainty close to 1.0%⁹⁹ and 4.0%,¹⁰⁰ do in fact exist.

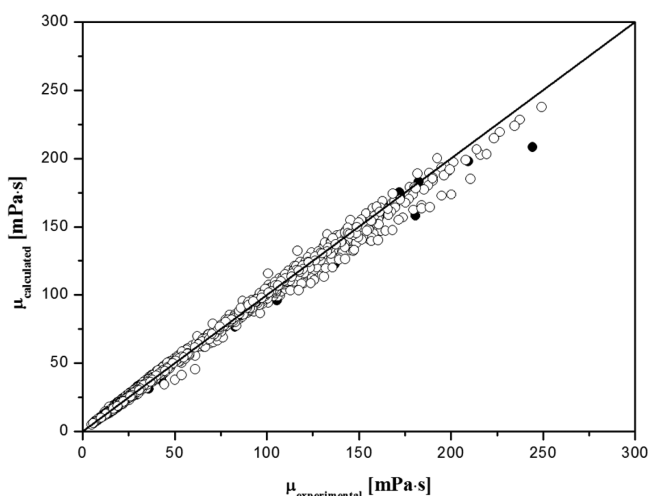
As a consequence, for this particular binary mixture, a training data point, and some predicted data, shown as a filled and empty circles in Figure 6, respectively, seem to be underestimated.

5. CONCLUSIONS

A critical analysis of IL viscosity data has been made for the selection of available data sets for 70 ILs. An extension of a previously reported method based on the UNIFAC–VISCO model has been made using 819 experimental data of 70 ILs in a temperature range from 263.15 to 373.15 K at 0.1 MPa. The binary interaction parameters and the ion VFT parameters were obtained by a regression analysis of experimental viscosity data. The deviation of 1.4% from experimental data shows a good correlation quality of this model. The viscosity of binary mixtures of ionic liquids was then predicted by using this method, with a relative average absolute deviation lower than 3.8%. This proves that the UNIFAC–VISCO-based model can be used to correlate and then evaluate the viscosities of pure ionic liquids with different functionalities (i.e., branched or not alkyl chain shape, for example) thanks to the 3D structure of their ions (effective molar volume, effective viscosity, *R* and *Q*

Table 11. RAADs of Training Data, Test Data, and Experimental Data for Each Binary Mixture of ILs

binary system	no. of training data	RAAD for training	no. of test data	RAAD for test	RAAD for each system
$x[\text{C}_2\text{mim}][\text{DCA}] + (1-x)[\text{C}_2\text{mim}][\text{BF}_4]$	7	2.25	0		2.25
$x[\text{C}_6\text{mim}][\text{BF}_4] + (1-x)[\text{C}_2\text{mim}][\text{BF}_4]$	20	7.88	196	5.58	5.79
$x[\text{C}_2\text{mim}][\text{OAc}] + (1-x)[\text{C}_2\text{mim}][\text{C}_2\text{SO}_4]$	7	5.05	56	4.77	4.80
$x[\text{C}_2\text{mim}][\text{C}_2\text{SO}_4] + (1-x)[\text{C}_2\text{mim}][\text{SCN}]$	12	4.32	60	8.27	7.61
$x[\text{C}_2\text{mim}][\text{DCA}] + (1-x)[\text{C}_2\text{mim}][\text{SCN}]$	12	1.35	24	2.60	2.18
$x[\text{C}_4\text{m}_{(4)}\text{py}][\text{NTf}_2] + (1-x)[\text{C}_2\text{mim}][\text{C}_2\text{SO}_4]$	7	2.19	63	4.54	4.31
$x[\text{C}_4\text{py}][\text{BF}_4] + (1-x)[\text{C}_4\text{m}_{(4)}\text{py}][\text{NTf}_2]$	14	2.39	56	2.57	2.54
$x[\text{C}_4\text{mim}][\text{BF}_4] + (1-x)[\text{C}_4\text{mim}][\text{C}_1\text{SO}_4]$	9	0.56	108	1.46	1.39
$x[\text{C}_4\text{mim}][\text{PF}_6] + (1-x)[\text{C}_4\text{mim}][\text{BF}_4]$	9	0.37	108	2.74	2.56
$x[\text{C}_4\text{mim}][\text{BF}_4] + (1-x)[\text{C}_4\text{py}][\text{BF}_4]$	22	1.48	77	1.76	1.70
$x[\text{C}_6\text{mim}][\text{BF}_4] + (1-x)[\text{C}_4\text{mim}][\text{BF}_4]$	9	0.76	90	3.86	3.57
all mixtures	128	2.92	838	3.92	3.79

**Figure 6.** Comparison of experimental (μ_{exp}) and evaluated viscosity (μ_{cal}) data for binary ionic liquids mixtures where data used for developing correlation (●) and predicted data (○) are shown.

values) and their interaction in solution (binary interaction parameters), as well as mixtures of ionic liquids, including electrolyte blends with great accuracy.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.jced.6b00689.

All the volumetric parameters, R and Q values for each ion estimated in this work are presented in Tables S1–S2. The gnostic analysis figures for all the other ionic liquids were illustrated in Figures S1–S86 (PDF)

Matlab files for the calculation of pure IL and binary mixtures (ZIP)

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Notes

The authors declare no competing financial interest.

■ ABBREVIATIONS

Cations:

$[\text{C}_1\text{mim}]^+$	1,3-dimethylimidazolium
$[\text{C}_2\text{mim}]^+$	1-ethyl-3-methylimidazolium
$[\text{C}_3\text{mim}]^+$	1-propyl-3-methylimidazolium
$[\text{C}_4\text{mim}]^+$	1-butyl-3-methylimidazolium
$[\text{C}_6\text{mim}]^+$	1-hexyl-3-methylimidazolium
$[\text{C}_8\text{mim}]^+$	1-octyl-3-methylimidazolium
$[\text{C}_{10}\text{mim}]^+$	1-decyl-3-methylimidazolium
$[\text{C}_4\text{mmim}]^+$	1-butyl-2,3-dimethylimidazolium
$[\text{C}_4\text{py}]^+$	1-butylpyridinium
$[\text{C}_6\text{py}]^+$	1-hexylpyridinium
$[\text{C}_8\text{py}]^+$	1-octylpyridinium
$[\text{C}_4\text{m}_{(3)}\text{py}]^+$	1-butyl-3-methylpyridinium
$[\text{C}_4\text{m}_{(4)}\text{py}]^+$	1-butyl-4-methylpyridinium
$[\text{C}_4\text{mpyrro}]^+$	1-butyl-1-methylpyrrolidinium
$[\text{N}_{1114}]^+$	butyl-trimethylammonium
$[\text{N}_{8881}]^+$	methyl-trioctyl-ammonium
$[\text{P}_{66614}]^+$	trihexyl(tetradecyl)phosphonium

Anions:

$[\text{BF}_4]^-$	tetrafluoroborate
$[\text{NTf}_2]^-$	bis(trifluoromethylsulfonyl)imide
$[\text{C}_1\text{SO}_4]^-$	methylsulfate
$[\text{C}_2\text{SO}_4]^-$	ethylsulfate
$[\text{C}_8\text{SO}_4]^-$	octylsulfate
$[\text{C}_1\text{SO}_3]^-$	methanesulfonate
$[\text{PF}_6]^-$	hexafluorophosphate
$[\text{DCA}]^-$	dicyanamide
$[\text{SCN}]^-$	thiocyanate
$[\text{OAc}]^-$	acetate
$[\text{OTf}]^-$	triflate
$[\text{eFAP}]^-$	tris(pentafluoroethyl)trifluorophosphate
$[\text{Tos}]^-$	tosylate
Br^-	bromide
Cl^-	chloride

Nomenclature

Roman Letters:

a_i	volumetric parameter
A	VFT equation parameter
B	VFT equation parameter
C	total number of components in UNIFAC–VISCO method
g_c^E	combinatorial contribution term in UNIFAC–VISCO method
g_r^E	residual contribution term in UNIFAC–VISCO method

$n_{i,k}$ total number of k^{th} group present in component i
 M number of data points
 N total number of groups
 p_{ref} reference pressure (0.1 MPa)
 q_i van der Waals' surface area of component i
 Q_k group surface area parameter
 r_i van der Waals' volume of component i
 R_k group volume parameter
 \mathcal{R} gas constant (J/mol·K)
 T temperature (K)
 T_0 VFT equation parameter
 V_i pure-component molar volume (m^3/kmol)
 V_m mixture molar volume (m^3/kmol)
 x_i mole fraction of the component i
 z coordinate value in UNIFAC–VISCO method

Greek Letters:

α_{mn} group interaction potential energy parameter between groups m and n
 γ_m residual activity coefficient
 θ_i molecular surface area fraction of component i
 Θ_i area fraction for group i
 μ viscosity of mixture (mPa·s)
 μ_i viscosity of component i
 μ_{exp} viscosity experimentally measured
 μ_{cal} viscosity calculated by our method
 μ_o viscosity value with the maximum distribution density
 μ_{ion} effective viscosity of ion
 ϕ_i molecular volume fraction of component i
 $\Psi_{m,i}$ group interaction parameter
 ω gnostic a posteriori weight

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