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**New Method Based on the UNIFAC-VISCO Model for the
Estimation of **Dynamic** Viscosity of (Ionic Liquid + Molecular Solvent)
Binary Mixtures**

Nan Zhao¹, Johan Jacquemin^{1,2,*}

¹ School of Chemistry and Chemical Engineering, Queen's University Belfast, Belfast,
BT9 5AG, U.K.

² Université Francois Rabelais, Laboratoire PCM2E, Parc de Grandmont 37200 Tours,
France

Corresponding Author: *E-mail: jj@univ-tours.fr or johan.jacquemin@gub.ac.uk

ABSTRACT

The modified UNIFAC-VISCO model was applied to evaluate the viscosity of (ionic liquid + molecular solvent) mixtures as the function of the composition and temperature at atmospheric pressure. The values of interaction parameters between ionic groups, $\alpha_{ion/ion}$ were collected from our previous paper (Zhao et al. J. Chem. Eng. Data 61 (2016) 3908-3921), while the parameters between the common organic groups, $\alpha_{organic/organic}$ were taken from Chevalier et al. (Chem. Eng. Sci. 49 (1994) 1799–1806). Another 376 unknown interaction parameters ($\alpha_{organic/organic}$ and $\alpha_{ion/organic}$) were calculated by regression of 3365 experimental binary viscosity data for 119 different binary systems as a function of temperature and component composition. Then, this model was assessed through the evaluation of 781 viscosity data points for 24 different binary systems not originally included in the correlation set. The relative absolute average deviation (RAAD%) of the correlation and evaluation for the viscosity of the investigated binary mixtures is close to 5.0% and 7.7%, respectively, which proves that the UNIFAC-VISCO based method proposed in this work is reliable for the estimation of the viscosity of ionic liquid-based binary mixtures.

Keywords: UNIFAC-VISCO; Ionic Liquids; Molecular Solvents; Viscosity; Group Contribution Model

1. INTRODUCTION

In recent years, ionic liquids (ILs) have attracted the attention of scientific communities due to their unique properties, such as chemical and thermal stability, high ionic conductivity, low vapor pressure, and easy tuning of their physicochemical properties by altering the cation, anion, or substituent [1]. Among the various properties, viscosity property is of considerable importance in the design of heat transfer equipment, liquid-liquid extractors, process piping, and other units [2]. For industrial applications, IL-cosolvent mixtures are promising alternatives to traditional organic liquids, since pure IL mixing with the organic liquid can confer a suitable viscosity to the mixture [1]. Consequently, many modeling efforts have been reported in the literature [1–13] regarding the viscosity of IL-cosolvent mixtures, used as electrolyte solutions.

In this regard, literature models can be categorized as (i) simple correlation equations, such as the Seddon [3], Grunberg-Nissan [9], and McAllister models [10]; (ii) Eyring's theory based methods, like the Eyring-UNIQUAC [11], Eyring-NRTL [12], Eyring-MTSM [8], Eyring-Flory-Huggins [7], and Eyring-Wilson models [13]; and (iii) predictive methods, for example, UNIFAC-VISCO [6] and ASOG-VISCO models [6].

Wang et al. [2] compared the Eyring-UNIQUAC model and the Eyring-NRTL model (α fixed at 0.25) in the case of 35 binary mixtures containing ILs with a total of 1014 data points. The authors highlighted that a better evaluation of the viscosity could be achieved by using the Eyring-UNIQUAC (RAAD% of 2.6%) than by using Eyring-NRTL

(RAAD% of 3.5%). Bajić et al. [6] used the Seddon, Grunberg-Nissan, McAlister, Eyring-UNIQUAC and Eyring-NRTL models to correlate the viscosity of 11 (IL + solvent) binary systems covering a wide temperature range. Within, McAlister and the three-parameter Eyring-NRTL models showed the best results with average percentage deviations (PD_{max}) below 1% in all cases. It should be noted that the reference in the denominator of the percentage deviation (PD_{max}) calculation is the maximum of experimental values. The Seddon model gives the highest deviation above 1% in all cases when compared with other correlative models, except for three cases; (2-butanone + [C₄mim][BF₄]), (H₂O + [C₃mmim][BF₄]), and (methyl methacrylate + [C₄mim][PF₆]). Fang et al. [7] proposed a method based on the Eyring's absolute reaction rate theory, which was coupled with the Flory-Huggins equation, to estimate the viscosity of 527 binary mixtures including 63 (IL + solvent) mixtures. The RAAD% of the proposed method assessed using 2554 data points of the 63 ionic liquid containing systems is close to 3.73% which shows a better viscosity predictive capability than the Grunberg-Nissan equation (7.42%), for example. Furthermore, Ciocirlan et al. [1] tested 6 methods (*i.e.* G-N, McAllister, Eyring-Flory-Huggins, Eyring-UNIQUAC, Eyring-NRTL, and Eyring-Wilson) in terms of four binary systems ([C₂mim][BF₄] + dimethyl sulfoxide / ethylene glycol / acetonitrile / 1,4-dioxane) at (293.15, 298.15, 303.15, 313.15, 323.15, 333.15, 343.15, and 353.15) K. The McAllister model gives the best viscosity evaluation below 1.34% quantified by RAAD% at each temperature for three systems ([C₂mim][BF₄] + dimethyl sulfoxide / acetonitrile / 1,4-

dioxane). For the other system, $[\text{C}_2\text{mim}][\text{BF}_4]$ + ethylene glycol, the best result was reported using the Eyring-Flory-Huggins based method with a RAAD% of 1.54%, while the McAllister method also showed a good result with RAAD% of 1.60%. The poorest result estimated for these four systems is with the G-N model, ranging from 1.93% up to 6.01% calculated by RAAD% at the eight temperatures. For the series of Eyring-based models, Eyring-Wilson gives the poorest results with a RAAD% of 2.1% and 2.9% in two systems containing dimethyl sulfoxide, and acetonitrile, respectively. Eyring-Flory-Huggins shows the poorest estimation with a 1.5% RAAD% in the system with ethylene glycol and also a poor result with 1.1% of RAAD% in the 1,4-dioxane containing system. Generally, the other two Eyring-based models (i.e. UNIQUAC and NRTL) show a better evaluation, the RAAD% are lower than 1.9% at each temperature point for all the four investigated systems. Atashrouz et al. [8] reported an Eyring's theory method coupled with a modified two-suffix-margules model (Eyring-MTSM). To validate the capability of the Eyring-MTSM model, the authors also made a clear comparison with the Eyring-NRTL model using 5512 experimental data points for 122 IL-based mixtures. Based on this work, these authors demonstrated the higher accuracy of the Eyring-MTSM method, with a RAAD% close to 2% compared to 2.8% observed using the Eyring-NRTL model.

Generally, correlative models often yield lower deviations, but require enough experimental data for the determination of parameters in the correlation equations. However, a significant advantage of the predictive models is that a given property, such

as viscosity could be evaluated for unknown mixtures by using a set of interaction parameters regressed using well-defined training datasets. Two predictive methods, based on the UNIFAC-VISCO and ASOG-VISCO models, respectively were applied for modeling the viscosity of IL-based mixtures [6]. Bajić et al. [6] calculated the interaction parameters (α_{mn}) between the group ([C₂mim][C₂SO₄], [-mim][BF₄], [-mim][PF₆], and [C₄mim][NO₃]) and the organic functional groups by correlating experimental data of 11 binary mixtures. However, the determination of the groups in the ionic liquid **components** is not clear and uniform, sometimes on the basis of the whole IL (such as [C₂mim][C₂SO₄] and [C₄mim][NO₃]) and sometimes on the basis of the core structure of the IL ([-mim][BF₄] and [-mim][PF₆]). In fact more work is requested to assess properly the quality of the proposed method using novel and original viscosity datasets.

In this work, we aim to develop the UNIFAC-VISCO model for correlation, and then **evaluation of** the viscosity of (IL + molecular solvent) binary mixtures. The method used for molecules' cleavage is different from the division method reported by Bajić et al. [6]. Each cation or anion, constituting the IL, is defined as an individual group, **which is coincident with the method of ionic groups determination for the viscosity estimation of pure ILs and binary ILs mixtures reported in our previous work [14,15].**

As a result, 119 binary systems with a total of 3365 experimental data points were regressed altogether to obtain the interaction parameters between functional groups ($\alpha_{ion/organic}$ and $\alpha_{organic/organic}$). Then other 24 binary mixtures, not involved in

the training set, were compared from the pure evaluated values to test the validity of the interaction parameters and thus the model developed in the current study.

2. DEVELOPMENT OF THE PROPOSED MODEL

2.1 Database. In this work, 4146 experimental viscosity data at 0.1 MPa for 143 binary mixtures were collected from the NIST database, as a function of the composition over a broad temperature range (from 273.15 K to 363.15 K). As discrepancies between the literature datasets for a given system could be observed, the experimental viscosities for each binary system, used to establish the UNIFAC-VISCO model, were taken only from one reference. The UNIFAC-VISCO model calculates the binary viscosities on the basis of the pure components' properties. The references that also report the pure components' densities and viscosities were preferred, thus identical measurement uncertainties for the properties of pure ILs and mixtures containing ILs increase the accuracy of the optimized binary interaction parameters. If the values of the pure component's properties are not provided in the selected reference, the molar volume and viscosity of pure ILs were calculated by using the methods previously proposed by our group [14–17]. In the database, 3365 data points of 119 binary mixtures from literature [18-86], as a function of temperature and component composition, were used for optimization of interaction parameters and 781 data points of another 24 binary mixtures were used to validate the performance of the proposed model. Table 1 shows the list of the selected binary systems along with their corresponding temperature range and composition.

Table 1. List of the selected (ionic liquid + molecular solvent) binary mixtures

No.	Ionic Component (1)	Organic Component (2)	Temperature Range (K)	Range of x_1 (mol%)	No. of Data Points	Measurement Uncertainty	RAAD (%)	Ref. (μ)	Ref. (ρ_{IL})
Correlation Set									
1	[C ₂ mim][C ₂ SO ₄]	1-propanol	298.15 to 328.15	0.0273 to 0.8381	27	± 0.006 (mPa •s)	3.62	[18]	[18]
2	[C ₂ mim][C ₂ SO ₄]	methanol	298.15 to 328.15	0.0083 to 0.8654	33	± 0.006 (mPa •s)	6.35	[18]	[18]
3	[C ₂ mim][C ₂ SO ₄]	ethanol	298.15 to 328.15	0.0137 to 0.9377	30	± 0.006 (mPa •s)	4.34	[19]	[19]
4	[C ₂ mim][BF ₄]	methanol	278.15 to 318.15	0.0499 to 0.9476	65	2%	2.73	[20]	[20]
5	[C ₂ mim][BF ₄]	ethanol	288.15 to 318.15	0.5447 to 0.9054	24	0.3%	3.57	[21]	[20]
6	[C ₂ mim][C ₂ SO ₄]	water	298.15 to 328.15	0.0494 to 0.9285	24	± 0.006 (mPa •s)	2.54	[19]	[19]
7	[C ₂ mim][BF ₄]	dimethyl sulfoxide	303.15 to 333.15	0.0998 to 0.8999	36	<0.6%	2.04	[22]	[22]
8	[C ₂ mim][BF ₄]	N,N-dimethylformamide	303.15 to 333.15	0.1001 to 0.8984	36	<0.6%	0.97	[22]	[22]
9	[C ₂ mim][BF ₄]	dimethylacetamide	303.15 to 333.15	0.1006 to 0.9004	36	<0.6%	1.47	[22]	[22]
10	[C ₄ mim][BF ₄]	water	303.15 to 353.15	0.1001 to 0.8998	72	± 0.01 (mPa •s)	7.41	[23]	[23]
11	[C ₄ mim][BF ₄]	methanol	303.15 to 333.15	0.0999 to 0.8997	36	$\pm 0.35\%$	3.50	[24]	[24]
12	[C ₄ mim][BF ₄]	dimethyl sulfoxide	293.15 to 353.15	0.0722 to 0.9155	63	$\pm 0.1\%$	3.11	[25]	[25]
13	[C ₄ mim][BF ₄]	2-butanone	298.15	0.0498 to 0.8995	13	$\pm 0.3\%$	2.36	[26]	[26]
14	[C ₄ mim][BF ₄]	dimethylacetamide	303.15 to 333.15	0.1001 to 0.8994	36	$\pm 0.35\%$	2.56	[24]	[24]
15	[C ₄ mim][BF ₄]	N,N-dimethylformamide	298.15	0.053 to 0.8976	13	$\pm 0.3\%$	2.46	[26]	[26]
16	[C ₄ mim][BF ₄]	acetonitrile	298.15	0.0487 to 0.9018	13	$\pm 0.3\%$	4.73	[26]	[26]
17	[C ₄ mim][BF ₄]	ethyl formate	298.15	0.05 to 0.8996	13	$\pm 0.3\%$	4.56	[27]	[27]
18	[C ₄ mim][BF ₄]	triethanolamine	303.15 to 343.15	0.1301 to 0.7054	25	<0.1%	11.61	[28]	[28]

19	[C ₄ mim][BF ₄]	methyl acetate	298.15	0.0501 to 0.8999	13	±0.3%	4.58	[27]	[27]
20	[C ₄ mim][BF ₄]	pyridine	298.15	0.193 to 0.8941	8	±1%	0.28	[29]	[29]
21	[C ₄ mim][BF ₄]	chloroform	298.15	0.1509 to 0.8921	9	±1%	1.52	[29]	[29]
22	[C ₄ mim][BF ₄]	dichloromethane	298.15	0.1605 to 0.9	9	±1%	3.02	[29]	[29]
23	[C ₄ mim][BF ₄]	N-methyl-2-pyrrolidone	298.15 to 318.15	0.3049 to 0.7945	15	^b	1.59	[30]	[30]
24	[C ₂ mim][NTf ₂]	ethanol	278.15 to 338.15	0.051 to 0.907	40	0.1%	1.96	[31]	[31]
25	[C ₂ mim][NTf ₂]	N-methyl-2-pyrrolidone	293.15 to 323.15	0.0978 to 0.9002	63	<0.1%	3.24	[32]	[32]
26	[C ₂ mim][NTf ₂]	benzene	298.15 to 328.15	0.508 to 0.865	16	±0.35%	1.67	[33]	[33]
27	[C ₄ mim][NTf ₂]	thiophene	298.15 to 328.15	0.23 to 0.911	20	±0.35%	4.17	[33]	[33]
28	[C ₄ mim][NTf ₂]	γ-butyrolactone	293.15 to 323.15	0.1005 to 0.8995	63	1%	3.06	[34]	[34]
29	[C ₄ mim][NTf ₂]	2,2,2-trifluoroethanol	278.15 to 333.15	0.1252 to 0.9123	72	2%	4.59	[35]	[35]
30	[C ₄ mim][NTf ₂]	isopropyl acetate	298.15	0.1082 to 0.9486	11	±0.5%	4.65	[36]	[36]
31	[C ₆ mim][NTf ₂]	methyl diethanolamine	303.15 to 323.15	0.101 to 0.9011	45	^b	6.97	[37]	[37]
32	[C ₆ mim][NTf ₂]	1-propanol	278.15 to 338.15	0.048 to 0.8999	40	^c	3.70	[38]	[38]
33	[C ₆ mim][NTf ₂]	chloroform	288.2 to 318.2	0.074 to 0.906	40	2~3%	4.18	[39]	^a
34	[C ₆ mim][NTf ₂]	1-octene	283.15 to 348.15	0.72 to 0.95	16	±1%	3.07	[40]	[41]
35	[C ₆ mim][BF ₄]	ethanol	288.15 to 318.15	0.098 to 0.9	36	0.3%	5.30	[21]	[42]
36	[C ₆ mim][BF ₄]	2-methyl-2-propanol	298.15	0.1017 to 0.9	9	±0.01 (mPa •s)	7.98	[43]	[43]
37	[C ₆ mim][BF ₄]	3-amino-1-propanol	303.15 to 308.15	0.0536 to 0.9204	18	±0.01 (mPa •s)	4.86	[43]	[43]
38	[C ₆ mim][BF ₄]	water	288.15 to 318.15	0.28 to 0.8899	32	±0.35%	3.79	[44]	^a
39	[C ₆ mim][BF ₄]	propylamine	298.15	0.0988 to 0.8475	9	±0.01 (mPa •s)	33.98	[43]	[43]
40	[C ₆ mim][BF ₄]	2-butanone	298.15	0.0492 to 0.8931	13	±1%	1.83	[45]	[45]
41	[C ₆ mim][BF ₄]	ethyl acetate	298.15	0.0498 to 0.8908	13	±1%	0.88	[45]	[45]
42	[C ₆ mim][BF ₄]	tetrahydrofuran	298.15	0.0498 to 0.8925	13	±1%	1.45	[45]	[45]

43	[C ₄ py][NTf ₂]	water	298.15 to 363.15	0.752	14	±0.35%	2.46	[46]	[46]
44	[C ₄ py][BF ₄]	dichloromethane	298.15	0.08071 to 0.94508	8	1%	7.72	[47]	- ^a
45	[C ₄ py][BF ₄]	water	298.15	0.024 to 0.82817	9	1%	4.43	[47]	- ^a
46	[C ₄ py][BF ₄]	methanol	298.15	0.05255 to 0.91576	9	1%	3.87	[47]	- ^a
47	[C ₈ mim][NTf ₂]	2-propanol	298.15	0.1034 to 0.9669	12	±0.5%	1.76	[48]	[48]
48	[C ₈ mim][NTf ₂]	isopropyl acetate	298.15	0.1009 to 0.9585	11	±0.5%	1.20	[48]	[48]
49	[C ₈ mim][NTf ₂]	methanol	298.15	0.098 to 0.9346	10	±0.5%	3.09	[49]	[49]
50	[C ₈ mim][BF ₄]	1-propanol	298.15	0.0473 to 0.9118	13	- ^b	7.26	[50]	[50]
51	[C ₈ mim][BF ₄]	2-butanone	298.15	0.0482 to 0.899	13	±0.01 (mPa •s)	1.64	[51]	[51]
52	[C ₈ mim][BF ₄]	methanol	298.15	0.0495 to 0.9049	13	- ^b	1.79	[50]	[50]
53	[C ₈ mim][BF ₄]	ethyl acetate	298.15	0.047 to 0.8908	13	±0.01 (mPa •s)	3.88	[51]	[51]
54	[C ₄ mim][PF ₆]	2-butanone	298.15	0.0513 to 0.9005	13	±0.2%	5.81	[52]	[52]
55	[C ₄ mim][PF ₆]	acetonitrile	298.15	0.1082 to 0.901	10	±1%	1.87	[29]	[29]
56	[C ₄ mim][PF ₆]	dichloromethane	298.15	0.1322 to 0.9247	10	±1%	1.99	[29]	[29]
57	[C ₄ mim][PF ₆]	chloroform	298.15	0.2674 to 0.9143	8	±1%	2.84	[29]	[29]
58	[C ₄ mim][PF ₆]	cyclopentanone	298.15	0.0482 to 0.9064	13	±0.2%	1.19	[52]	[52]
59	[C ₄ mim][PF ₆]	N,N-dimethylethanolamine	288.15 to 323.15	0.0872 to 0.8779	72	±0.1%	5.01	[53]	[53]
60	[C ₄ mim][PF ₆]	N,N-dimethylformamide	303.15 to 323.15	0.0276 to 0.0988	9	0.35%	2.08	[54]	[54]
61	[C ₄ mim][PF ₆]	dimethyl sulfoxide	298.15	0.05 to 0.893	13	±0.5%	2.28	[55]	[55]
62	[C ₄ mim][PF ₆]	water	298.15	0.7854 to 0.9482	5	±1%	2.13	[29]	[29]
63	[C ₄ mim][PF ₆]	methanol	298.15	0.0503 to 0.8889	13	±0.5%	6.42	[55]	[55]
64	[C ₄ mim][PF ₆]	methyl methacrylate	283.15 to 353.15	0.0993 to 0.9014	117	<2.612%	4.72	[56]	[56]
65	[C ₄ mim][PF ₆]	monoethanolamine	288.15 to 323.15	0.0972 to 0.8815	72	±0.1%	2.43	[53]	[53]
66	[C ₄ mim][PF ₆]	tetrahydrofuran	298.15	0.0502 to 0.9595	13	±0.5%	2.73	[55]	[55]

67	[C ₄ mim][PF ₆]	2,2,2-trifluoroethanol	278.15 to 333.15	0.0583 to 0.5368	60	2%	11.03	[35]	[35]
68	[C ₄ mim][DCA]	water	278.15 to 358.15	0.5169 to 0.941	54	±0.35%	6.88	[57]	[57]
69	[C ₄ mim][DCA]	γ-butyrolactone	273.15 to 323.15	0.0504 to 0.8871	56	<0.5%	2.17	[58]	[58]
70	[C ₂ mim][DCA]	ethanol	298.15 to 343.15	0.0673 to 0.898	40	0.65%	4.01	[59]	[59]
71	[C ₄ mpyrro][NTf ₂]	acetonitrile	288.15 to 308.15	0.1924 to 0.8159	12	±0.2%	2.32	[60]	[60]
72	[C ₄ mpyrro][NTf ₂]	N,N-dimethylformamide	295.2	0.26 to 0.87	7	±1%	7.00	[61]	^a
73	[C ₄ mpyrro][NTf ₂]	γ-butyrolactone	298.15 to 323.15	0.1 to 0.8925	54	1%	2.07	[62]	[62]
74	[C ₄ mpyrro][NTf ₂]	methanol	288.15 to 308.15	0.2054 to 0.7067	12	±0.2%	1.86	[60]	[60]
75	[C ₄ mpyrro][NTf ₂]	propylene carbonate	293.15 to 318.15	0.05 to 0.95	108	0.01 (mPa •s)	3.20	[63]	[63]
76	[C ₄ mpyrro][SCN]	water	298.15 to 348.15	0.0343 to 0.9147	48	±0.1%	7.50	[64]	[64]
77	[C ₄ mim][SCN]	ethanol	298.15 to 348.15	0.0473 to 0.9174	54	<1%	8.38	[65]	[65]
78	[C ₄ mim][SCN]	1-hexanol	298.15 to 348.15	0.0898 to 0.9576	60	<0.05%	4.82	[66]	[66]
79	[C ₄ mim][SCN]	1-decanol	298.15 to 348.15	0.0747 to 0.9587	60	<1%	3.31	[67]	[67]
80	[C ₄ mim][SCN]	methanol	298.15 to 328.15	0.0154 to 0.9046	44	<1%	3.42	[65]	[65]
81	[C ₄ mim][SCN]	thiophene	298.15 to 328.15	0.299 to 0.897	20	±0.35%	3.73	[33]	[33]
82	[C ₄ mim][SCN]	benzene	298.15 to 328.15	0.4906 to 0.7666	16	±0.35%	1.45	[33]	[33]
83	[C ₁ mim][C ₁ SO ₄]	water	298.15 to 328.15	0.0515 to 0.9503	33	<±0.2 (mPa •s)	5.72	[68]	[68]
84	[C ₁ mim][C ₁ SO ₄]	ethanol	298.15 to 328.15	0.055 to 0.9331	33	<±0.2 (mPa •s)	3.35	[68]	[68]
85	[C ₄ mim][OTf]	benzene	298.15 to 328.15	0.427 to 0.878	20	±0.35%	2.18	[33]	[33]
86	[C ₂ mim][OTf]	water	278.15 to 348.15	0.0648 to 0.7714	40	±2%	12.88	[69]	[69]
87	[C ₂ mim][OAc]	ethanol	298.15 to 343.15	0.1334 to 0.8941	40	0.65%	6.57	[59]	[59]
88	[C ₂ mim][OAc]	water	298.15 to 343.15	0.0933 to 0.9008	40	0.65%	8.96	[59]	[59]
89	[C ₄ mim][OAc]	methyl diethanolamine	293.15 to 343.15	0.2 to 0.8	24	±0.5%	9.52	[70]	[70]
90	[C ₄ mim][OAc]	diethanolamine	303.15 to 343.15	0.2 to 0.8	20	±0.5%	10.24	[70]	[70]

91	[C ₄ mim][OAc]	acetonitrile	298.15	0.1244 to 0.6794	4	±2%	7.59	[71]	- ^a
92	[C ₄ mim][OAc]	N,N-dimethylformamide	298.15	0.1933 to 0.848	4	±2%	6.48	[71]	- ^a
93	[P ₆₆₆₁₄][NTf ₂]	water	298.15 to 358.15	0.912	13	±0.35%	4.91	[72]	[72]
94	[N ₁₁₁₄][NTf ₂]	1,2-butanediol	323.15 to 353.15	0.0969 to 0.7709	42	±0.1%	4.36	[73]	[73]
95	[N ₁₁₁₄][NTf ₂]	2,3-butanediol	323.15 to 343.15	0.1858 to 0.9121	30	±0.1%	5.69	[73]	[73]
96	[C ₄ mmim][BF ₄]	dimethyl sulfoxide	298.15 to 353.15	0.138 to 0.8785	56	±0.1%	2.40	[74]	[74]
97	[C ₄ mmim][BF ₄]	acetonitrile	298.15 to 343.15	0.1118 to 0.8423	42	±0.1%	4.58	[74]	[74]
98	[C ₈ mim]Cl	methanol	298.15 to 328.15	0.0197 to 0.9435	39	<1 (mPa •s)	9.98	[75]	[75]
99	[C ₈ mim]Cl	ethanol	298.15 to 328.15	0.0206 to 0.9551	27	<1 (mPa •s)	9.11	[75]	[75]
100	[C ₂ mim][BF ₄]	water	288.15 to 318.15	0.1009 to 0.9015	36	±0.35%	6.65	[44]	- ^a
101	[C ₄ mim][BF ₄]	ethanol	288.15 to 318.15	0.0986 to 0.9235	36	0.3%	10.65	[21]	- ^a
102	[C ₄ mim][BF ₄]	methyl formate	298.15	0.05 to 0.8993	13	±0.3%	3.81	[27]	[27]
103	[C ₄ mim][NTf ₂]	butyl acetate	298.15 to 323.15	0.11 to 0.9056	54	3%	12.69	[76]	[76]
104	[C ₄ mim][NTf ₂]	ethanol	298.15	0.1016 to 0.9207	12	±0.5%	2.24	[77]	[77]
105	[C ₆ mim][BF ₄]	butylamine	298.15	0.0492 to 0.8787	13	±1%	8.47	[45]	[45]
106	[C ₆ mim][BF ₄]	methyl diethanolamine	303.15 to 323.15	0.1176 to 0.903	45	±3%	6.03	[78]	[78]
107	[C ₈ mim][NTf ₂]	ethanol	298.15	0.1033 to 0.9041	11	±0.5%	11.16	[79]	[79]
108	[C ₈ mim][NTf ₂]	methyl acetate	298.15	0.1016 to 0.9301	11	±0.5%	1.96	[49]	[49]
109	[C ₈ mim][BF ₄]	ethanol	298.15	0.0517 to 0.9286	13	- ^b	1.39	[50]	[50]
110	[C ₈ mim][BF ₄]	butyl acetate	298.15	0.05001 to 0.886	13	±0.01 (mPa •s)	1.76	[51]	[51]
111	[C ₈ mim][BF ₄]	methyl acetate	298.15	0.0485 to 0.8827	13	±0.01 (mPa •s)	5.24	[51]	[51]
112	[C ₄ mim][PF ₆]	ethanol	288.15 to 308.15	0.52 to 0.8	10	±3%	10.08	[80]	- ^a
113	[C ₂ mim][DCA]	water	298.15 to 343.15	0.047 to 0.8701	40	0.65%	4.92	[59]	[59]
114	[C ₄ mpyrro][DCA]	γ-butyrolactone	273.15 to 323.15	0.0987 to 0.9013	42	1%	1.59	[81]	[81]

115	[C ₄ mim][C ₁ SO ₄]	water	298.15 to 328.15	0.0534 to 0.9675	30	<±0.2 (mPa •s)	8.82	[82]	[82]
116	[C ₄ mim][C ₁ SO ₄]	ethanol	298.15 to 328.15	0.0555 to 0.9349	33	<±0.2 (mPa •s)	10.20	[82]	[82]
117	[C ₄ mim][OTf]	water	298.15	0.0805 to 0.8907	10	±1%	13.36	[29]	[29]
118	[C ₄ mim][OAc]	water	278.15	0.03236 to 0.75268	5	±2%	25.84	[71]	- ^a
119	[C ₄ mim][OAc]	1,2-ethanediol	298.15	0.12944 to 0.78032	4	±2%	14.40	[71]	- ^a

Test Set									
1	[C ₂ mim][C ₂ SO ₄]	2-propanol	298.15 to 328.15	0.0294 to 0.8772	27	±0.006 (mPa •s)	6.44	[18]	[18]
2	[C ₂ mim][NTf ₂]	1-propanol	278.15 to 338.15	0.0519 to 0.9025	35	- ^c	11.28	[38]	[38]
3	[C ₄ mim][BF ₄]	acetone	298.15	0.0508 to 0.8949	13	±0.3%	11.19	[27]	[27]
4	[C ₄ mim][NTf ₂]	1-butanol	298.15	0.1382 to 0.8952	7	1.5%	23.11	[83]	- ^a
5	[C ₄ mim][NTf ₂]	1-propanol	278.15 to 338.15	0.0508 to 0.9030	40	- ^c	8.58	[38]	[38]
6	[C ₄ mim][NTf ₂]	ethyl acetate	298.15	0.1067 to 0.9478	11	±0.5%	21.42	[77]	[77]
7	[C ₆ mim][NTf ₂]	ethanol	278.15 to 338.15	0.047 to 0.903	40	0.1%	5.91	[31]	[31]
8	[C ₆ mim][BF ₄]	2-methyl-1-propanol	303.15 to 308.15	0.1004 to 0.8327	16	- ^b	7.82	[84]	[84]
9	[C ₆ mim][BF ₄]	1-propanol	293.15 to 333.15	0.1006 to 0.8894	35	- ^b	9.80	[85]	[85]
10	[C ₆ mim][BF ₄]	2-propanol	293.15 to 333.15	0.08354 to 0.85073	45	- ^b	14.55	[86]	[86]
11	[C ₈ mim][NTf ₂]	ethyl acetate	298.15	0.0984 to 0.9507	11	±0.5%	4.04	[79]	[79]
12	[C ₈ mim][BF ₄]	2-propanol	298.15	0.0459 to 0.8697	13	- ^b	7.11	[50]	[50]
13	[C ₄ mim][PF ₆]	acetone	298.15	0.0519 to 0.8853	13	±0.2%	7.55	[52]	[52]
14	[C ₄ mim][PF ₆]	3-pentanone	298.15	0.052 to 0.8912	13	±0.2%	7.93	[52]	[52]
15	[C ₄ mim][PF ₆]	ethyl acetate	298.15	0.0496 to 0.9063	13	±0.2%	15.84	[52]	[52]
16	[C ₄ mim][SCN]	1-heptanol	298.15 to 348.15	0.0806 to 0.9793	66	<1%	4.71	[67]	[67]

2.2 Functional Groups. In our previous studies, the UNIFAC-VISCO model was developed to estimate the viscosity of pure ILs where each ion was regarded as an individual functional group [14,15]. In the present study, a similar approach was used to define the IL-viscosity contribution for investigated mixtures. In terms of the organic component in the mixture, the original cleavage reported by Chevalier et al. [87] was also used in the current study. The cleavages are identical for the branched and linear hydrocarbons with the same number of carbon atoms, which limits the number of organic functional groups [87]. Although isomers of organic components are not characterized by using this definition, ILs with different alkyl chain shapes (branched and linear) could be evaluated thanks to the 3D structure of ions contributing to the volume and surface area values (R and Q) [14].

Chevalier et al. [88] reported that methanol could not be split into the groups CH_3 and OH like other alcohols, due to a peculiarity. Several molecular solvents also show a similar characteristic and are defined as the single groups, such as water, chloroform, dichloromethane, pyridine, and thiophene. Table 2 shows the list of organic components and their corresponding functional groups used during this work.

2.3 Proposed Model. The UNIFAC-VISCO model is a group contribution model developed by Chevalier et al. [87,88] to predict the viscosity of the liquid mixtures.

Briefly, the viscosity of a given mixture is calculated as follows:

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

where, μ is the viscosity of the mixture; subscript i represents the pure component in the mixture (i.e., the IL and the solvent); C is the total number of the pure components existing in the mixture, and equals 2 because only binary mixtures were investigated in this paper. x_i is the mole fraction of component i ; μ_i is the pure-component viscosity of component i ; V_i is the pure-component molar volume of component i ; V_m is the ideal molar volume of the mixture calculated by using the following equation:

$$V_m = \sum_{i=1}^C x_i V_i \quad (2)$$

The UNIFAC-VISCO combinatorial term reported in the eq. 1 is defined by:

$$\frac{g_c^E}{RT} = \sum_{i=1}^C x_i \ln \frac{\phi_i}{x_i} + 5 \sum_{i=1}^C x_i q_i \ln \frac{\theta_i}{\phi_i} \quad (3)$$

where

$$\theta_i = \frac{x_i q_i}{\sum_j x_j q_j} \quad (4)$$

$$\phi_i = \frac{x_i r_i}{\sum_j x_j r_j} \quad (5)$$

$$q_i = \sum_{k=1}^N n_{i,k} Q_k \quad (6)$$

$$r_i = \sum_{k=1}^N n_{i,k} R_k \quad (7)$$

The volume R and surface area Q of ionic groups were obtained by using the COSMOthermX software (version C30_1601) as reported by our group previously [14,15,89]. R and Q values of organic groups are collected from values reported by

Fredenslund et al. [90,91]. R and Q values of all the functional groups are presented in the Table S2 of the Supporting Information.

The UNIFAC-VISCO residual term reported in the eq. 1 is defined by:

$$\frac{g_r^E}{RT} = \sum_{i=1}^C x_i [\sum_{m=1}^N n_{m,i} (\ln \gamma_m - \ln \gamma_{m,i})] \quad (8)$$

where N is the total number of the functional groups existing in the binary system; n_m and $\ln \gamma_m$ refer to the group fractions in the mixture, and $n_{m,i}$ and $\ln \gamma_{m,i}$ refer to the group fractions in pure component i . $\ln \gamma_m$ is defined as:

$$\ln \gamma_m = Q_m \left[1 - \ln \left(\sum_{i=1}^N \Theta_i \Psi_{i,m} \right) - \sum_{i=1}^N \frac{\Theta_i \Psi_{m,i}}{\sum_{j=1}^N \Theta_j \Psi_{j,i}} \right] \quad (9)$$

The Θ value is obtained by using eq 10:

$$\Theta_m = \frac{x_m Q_m}{\sum_{i=1}^N x_i Q_i}, m = 1, 2, 3 \dots N \quad (10)$$

The group interaction parameters Ψ_{mn} are calculated as follows:

$$\Psi_{mn} = \exp\left(-\frac{\alpha_{mn}}{298.15}\right) \quad (11)$$

where α_{mn} are the group interaction potential energy parameters between the defined functional groups m and n .

2.4 Determination of The Interaction Parameters α_{mn} . A set of interaction parameters α_{mn} is required for the calculation of the viscosity of binary mixtures using the UNIFAC-VISCO model. The values of interaction parameters between ionic groups ($\alpha_{ion/ion}$) were collected from our previous paper [14]. The values of interaction parameters between some organic groups ($\alpha_{organic/organic}$) were collected from Chevalier et al. [88]. The other unknown interaction parameters between organic and ionic groups ($\alpha_{ion/organic}$ and $\alpha_{organic/organic}$) were regressed by using the experimental training datasets. To obtain the values of α_{mn} , we

performed the Marquardt [92] optimization of the following objective function:

$$OF = \frac{1}{M} \sum_{i=1}^M \left(\frac{\mu_{exp} - \mu_{cal}}{\mu_{exp}} \right)^2 \rightarrow \min \quad (12)$$

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

To carry out the regression, the 'lsqnonlin' function of the MATLAB Optimization Toolbox was used. In this work, 3365 training data were regressed altogether to obtain the global optimal values of interaction parameters.

3. RESULTS AND DISCUSSION

All the binary interaction parameters used in this work are presented in the Table S3 of the Supporting Information. The regression result was characterized by calculating the relative absolute average deviation (RAAD%, eq. 13) between the calculated (μ_{cal}) and experimental viscosity (μ_{exp}) data.

$$RAAD\% = 100 \times \frac{1}{M} \sum_{i=0}^M \left| \frac{\mu_{exp} - \mu_{cal}}{\mu_{exp}} \right| \quad (13)$$

The deviation between each experimental viscosity dataset and correlated (training set; overall RAAD% close to 5.0%) for each investigated binary system used to determine the missing UNIFAC-VISCO parameters is reported in Table 1. As shown in Table 1 and in Figure 1, the quality of the developed UNIFAC-VISCO model was then assessed by comparing experimental data with those calculated for wide ranges of temperature, composition, and chemical structures leading to large values of viscosity up to 14000 mPa.s. As illustrated in Figures 1 and 2, the correlated results show, generally, a good agreement with the experimental data, except in two cases: i.e. the ([C₆mim][BF₄] + propylamine) viscosity dataset reported by Kermanpour et al. [43] and the ([C₄mim][OAc] + water) dataset published by Fendt et al. [71] for which a deviation higher than 20% is observed in both cases.

To further assess the UNIFAC-VISCO model, 781 test data points for 24 binary mixtures, not originally used during the training set, were then compared to the evaluated viscosity data by using the interaction parameters estimated by using the training set.

An arbitrary binary mixture with unknown viscosity could also be estimated thanks to the optimized values of the binary interaction parameters between the functional groups, constituting the given mixture. The evaluation result for each binary system is presented in Table 1 and is also shown in Figures 1-2. The overall RAAD% for the test set is close to 7.7% (see Table 1 and Figure 2), which demonstrates that the interaction parameters estimated during this work are accurate enough to evaluate then the viscosity data for original mixtures.

Nevertheless, as shown in Figure 2, a correlated data point for the ([C₄mim][OAc] + water) binary system presents a relative deviation higher than 60%. However, only one research group reported the experimental viscosity data for the system ([C₄mim][OAc] + water), to date [71]. The viscosity of the pure [C₄mim][OAc] measured by Fendt et al. [71] is close to 485 mPa·s at 298.15 K; while by looking at the literature, it is obvious to highlight a clear discrepancy between experimental viscosity values reported for the [C₄mim][OAc] at 298.15 K; e.g. 297 mPa·s [93], 440 mPa·s [94], 485.1 mPa·s [95], and 485 mPa·s [71]. As reported in our previous paper [14], a viscosity dataset [96] was recommended through the gnostic analysis of five viscosity datasets [71,93–96] for [C₄mim][OAc] over a temperature range from (283.15 to 393.15) K. The interaction parameters between ions [C₄mim]⁺ and [OAc]⁻ were then calculated by regressing the recommended dataset [96], leading to an estimated viscosity value close to 424.6 mPa·s at 298.15 K for the pure component [C₄mim][OAc] by using the UNIFAC-VISCO method [14]. Therefore, such a discrepancy on the pure component viscosity values definitely contributes to increasing errors during the estimation of the viscosity for the

([C₄mim][OAc] + water) binary system as shown in Figure 2.

Table 3 shows the estimation results (including correlated and evaluated data) for several main types of molecular solvents to further analyze the effect of the molecular solvent structure on the predictive capability of the UNIFAC-VISCO model. The deviation between the experimental viscosity data and those determined using the UNIFAC-VISCO model follows the order: water > amines > alcohols > esters > ketones > amides. The binary systems with water show the poorest estimation result. This trend could be attributed to the complexity of (IL + H₂O) systems, driven by strong solvent-ion interactions, like H-bonds, between water and IL, which may induce experimental (hydrolysis reaction [97], trace of water originally present in the pure IL, etc.) and computational errors. The difference between the experimental viscosity values for pure ILs used in this work for the calculation of the viscosity data of selected mixtures and those used in our previous paper [14] to determine the interaction parameters $\alpha_{ion/ion}$ also contribute to an increase in the observed errors of the UNIFAC-VISCO model for some IL-based mixtures. In other words, the quality of the proposed method is strongly affected by the overall accuracy of data reported in the literature as expected for a such complex property like the viscosity.

Furthermore, the impact of the non-ideality of the volumetric properties of the solution on the quality of the proposed method was also exemplified by using the ([C₂mim][C₂SO₄] + 1-propanol) binary system. The experimental viscosities, densities, and excess molar volumes of this selected system are collected from González et al. [18] and provided in Table S4 of the Supporting Information, together with the

calculated binary viscosities by implementing the ideal and then the real molar volumes into the UNIFAC-VISCO model, respectively. Figure 3 shows the excess molar volumes of the system ([C₂mim][C₂SO₄] + 1-propanol), along with the fitted curves by using the Redlich-Kister [98] equation (Eq. 14) as a function of the mole fraction of solvent.

$$V^E = x_s(1 - x_s) \sum_{p=0}^M B_p(2x_s - 1)^p \quad (14)$$

where V^E is the excess molar volume; x_s is the mole fraction of solvent; B_p are the Redlich-Kister fitting parameters; and M is the degree of the polynomial expansion. The fitting parameters reported by González et al. [18] are presented in Table S5 of the Supporting Information.

Figure 4 shows the difference on the calculated viscosities induced by using the non-ideality of the system ([C₂mim][C₂SO₄] + 1-propanol). The minimum of V^E presents at approximately $x_s=0.6$, as shown in Figure 3. While the largest difference between the calculated viscosities by two different molar volumes is present at nearly 0.8 mole fraction of 1-propanol. The negative excess molar volume decreases as the temperature increases. Analogous temperature dependence behavior was observed for the calculated viscosities difference in Figure 4. However, the minimum difference between the calculated viscosities is better than -0.6% for the system ([C₂mim][C₂SO₄] + 1-propanol). The RAAD% between the experimental viscosities and the calculated viscosities assuming the system to be ideal is 3.62%, and the RAAD% between the experimental viscosities and the calculated values considering their non-ideality is 3.55%. Hence, only a global 0.07% deviation was caused by the non-ideality

of the binary system. Therefore, it is reasonable and acceptable to assume all binary systems investigated in this work as ideal.

4. CONCLUSIONS

In this work, the experimental viscosity values of (IL + molecular solvent) binary mixtures were estimated by using the modified UNIFAC-VISCO model. During this work, 3365 experimental viscosity data for 119 different binary systems were regressed altogether to determine missing interaction parameters between ions and molecular solvent group parameters. Then, 781 experimental data of 24 binary systems, not originally included in the training set, were used to assess the performance of the established model. The overall estimation result is close to 5.5%, amongst 5% for the training result and 7.7% for the test result, which proves that the UNIFAC-VISCO-based method is able to correlate and then evaluate the viscosity of (ILs + molecular solvent) binary mixtures as the function of temperature and composition at 0.1 MPa.

ASSOCIATED CONTENT

Supporting Information

The volumetric parameters for selected cations and anions are provided in Table S1.

R and Q values of the functional groups, and the interaction parameters α_{mn} used in this work are presented in Tables S2-S3. Experimental density, viscosity, and excess molar volume data, along with calculated viscosity values for the system ([C₂mim][C₂SO₄] + 1-propanol) are given in Table S4. Parameters fitted to the Redlich-Kister equation for the excess molar volume of ([C₂mim][C₂SO₄] and 1-propanol) are provided in Table S5.

AUTHOR INFORMATION

Corresponding Author

*E-mail: jj@univ-tours.fr or johan.jacquemin@qub.ac.uk

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Notes

The authors declare no competing financial interest.

ABBREVIATIONS

UNIQUAC *UN*iversal *QU*asichemical Activity Coefficient

UNIFAC-VISCO *UN*iversal Functional-group Activity Coefficient-*VISCO*sity

NRTL *Non-Random Two-Liquid*

MTSM *Modified Two-Suffix-Margules*

ASOG-VISCO *Analytical Solution Of Groups-VISCO*sity

Cations:

[mim]⁺ methylimidazolium

[C₁mim]⁺ 1,3-dimethylimidazolium

[C₂mim]⁺ 1-ethyl-3-methylimidazolium

[C₃mmim]⁺ 1-propyl-2,3-dimethylimidazolium

[C₄mim]⁺ 1-butyl-3-methylimidazolium

[C₆mim]⁺ 1-hexyl-3-methylimidazolium

[C₈mim]⁺ 1-octyl-3-methylimidazolium

[C₄mmim]⁺ 1-butyl-2,3-dimethylimidazolium

[C₄py]⁺ 1-butylpyridinium

[C₄mpyrro]⁺ 1-butyl-1-methylpyrrolidinium

[N₁₁₁₄]⁺ butyl-trimethyl-ammonium

[P₆₆₆₁₄]⁺ trihexyl(tetradecyl)phosphonium

Anions:

[BF₄]⁻ tetrafluoroborate

[NO₃]⁻ nitrate

$[\text{NTf}_2]^-$ bis(trifluoromethylsulfonyl)imide

$[\text{C}_1\text{SO}_4]^-$ methylsulfate

$[\text{C}_2\text{SO}_4]^-$ ethylsulfate

$[\text{PF}_6]^-$ hexafluorophosphate

$[\text{DCA}]^-$ dicyanamide

$[\text{SCN}]^-$ thiocyanate

$[\text{OAc}]^-$ acetate

$[\text{OTf}]^-$ triflate

Cl^- chloride

Nomenclature

Roman Letters:

a parameter in Seddon equation

C total number of pure components in UNIFAC-VISCO method

g_c^E combinatorial term in UNIFAC-VISCO method

g_r^E residual 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

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)

V_i pure-component molar volume (m³/kmol)

V_m mixture molar volume (m³/kmol)

x_i mole fraction of the component i

x_s mole fraction of the molecular solvent

Greek Letters:

α_{mn} group interaction parameter between groups m and n

γ_m residual activity coefficient

ρ_{IL} density of pure ionic liquid (g·cm⁻³)

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 pure component i

μ_{IL} viscosity of pure ionic liquid

μ_{exp} viscosity experimentally measured

μ_{cal} viscosity calculated by our method

μ_{corre} viscosity correlated by the Seddon equation

ϕ_i molecular volume fraction of component i

$\Psi_{m,i}$ group interaction parameter

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