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VISCOSITY MODELING FOR IONIC LIQUID SOLUTIONS BY EYRING-WILSON EQUATION

A semi-theoretical model based on the classical Eyring's mixture viscosity equation and the Wilson activity coefficient equation is presented for correlating the viscosity of ionic liquids with solvent systems. The accuracy of the proposed model was verified by comparing calculated and experimental viscosity values from the literature for 49 mixtures with a total of 1560 data points. The results show that the equation, similar to the Wilson activity coefficient equation, can be well applied to describe the non-ideal term in the Eyring's mixture viscosity equation. The model has a relatively simple mathematical form and can be easily incorporated into process simulation software.

Keywords: ionic liquids; viscosity; Wilson Equation; green solvent.

Ionic liquids (ILs) are often fluid at room temperature, and consist entirely of cations and anions. In recent years, ILs known as environmentally benign solvents are attracting a great deal of attention as possible a replacement for traditional volatile organic compounds (VOCs) [1-3]. ILs can be used in a variety of industrial applications involving catalytic reactions [4], synthesis [5] and phase separations [6,7]; more detailed applications can be found in recent review articles [1-3]. On the other hand, the practical application of ILs to industrial processes is still limited because of the scarcity of available experimental thermodynamic and transport properties. However, the thermodynamic and transport modeling of ILs, in particular for mixtures containing ILs, have lagged behind growth [8,9].

Knowledge of the viscosity for mixtures is essential for the engineering design and modeling of equipments, such as flow behavior through pipes, pumps and heat exchangers. Viscosities of pure ILs and their mixtures with traditional solvents provide an invaluable type of data in the development and engineering application of ILs [10]. Recently, some empirical [11], semi-theoretical [12-14] and QSAR [15,16] models have been proposed for the viscosity model-

ing of ILs and their mixtures. The model of Fang *et al.* [12] for IL mixtures requires the ionic liquid surface area as a starting point. Of course, the equation of Fang *et al.* [12] focuses on the developing of one-parameter model and has its own advantages. On the other hand, the Eyring-UNIQUAC equation [14] with two parameters can always give satisfactory results for mixture viscosity modeling. However, the Eyring-UNIQUAC equation [14] with the complex mathematical formulation requires two additional structure parameters for ILs, namely the surface and volume parameters, which are not easy to obtain. So in the viscosity modeling of ILs mixtures, the Eyring-UNIQUAC model cannot be easily applied.

Recently, a segment-based Eyring-Wilson equation was proposed by Sadeghi [17] to calculate the viscosity of mixtures containing polymers. Many studies have revealed that ILs have some similar properties to polymers, such as the high viscous character [2,15]. It is well known that the Wilson activity coefficient equation [18] based on the local composition concept in thermodynamics has a simple mathematical function than the NRTL [19], UNIQUAC [20,21] and COSMO-RS [22] models. It is the purpose of the present study to extend the Eyring's mixture viscosity model and the Wilson activity coefficient equation to the viscosity modeling of ILs mixtures. The utility of the proposed model is illustrated with the successful representation of viscosity data for ILs mixtures covering a range of temperature and the entire range of ILs composition.

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Theoretical background

Viscous flow of liquids can be regarded as an activated process according to the absolute rate theory approach of Eyring [23]. By the application of the Eyring's absolute rate theory [17,19–26], the viscosity of binary mixtures containing ILs can be expressed as:

$$\ln(\eta V)_{\text{mix}} = x_{\text{ILs}} \ln(\eta_{\text{ILs}} V_{\text{ILs}}) + x_{\text{sol}} \ln(\eta_{\text{sol}} V_{\text{sol}}) + \frac{\Delta E}{RT} \quad (1)$$

where η , V and x are the dynamic viscosity, the molar volume, and the molar concentration of compounds, respectively; subscripts mix, ILs and sol stand for the mixture, ionic liquids and solvent respectively; ΔE is the excess molar energy of activation for moving the sliding layer molecules from one energy minimum to the next layer by one flow unit.

The concepts of classical thermodynamics have been extended to study the viscous flow behavior of ILs mixtures. In the last decade, many studies [17,19–26] have revealed that the approach for the derivation of G^E used in phase equilibrium calculations can be used to derive the ΔE in Eq. (1). Eyring's absolute reaction rate theory seems to provide a good framework for modeling liquid-mixture viscosities, since any excess Gibbs energy function can be plugged into Eq. (1) to account for the ΔE term [26]. In this study, the Wilson equation based on the local composition concept is used to represent the ΔE term. The Wilson model for binary mixtures can be written as [18]:

$$\frac{G^E}{RT} = -x_1 \ln(x_1 + A_{12}x_2) - x_2 \ln(x_2 + A_{21}x_1) \quad (2)$$

where:

$$A_{12} = \frac{V_2}{V_1} \exp\left(-\frac{\lambda_{12}}{RT}\right) \quad (3)$$

$$A_{21} = \frac{V_1}{V_2} \exp\left(-\frac{\lambda_{21}}{RT}\right) \quad (4)$$

Our preliminary calculations show that better results can be achieved for all the ILs systems by coupling the Eyring's Equation with the minus form of the Wilson Equation than with the original form of the Wilson Equation. Therefore, in this study the minus form of the Wilson Equation, *i.e.*, Eq. (2), is utilized. The Eyring-Wilson Equation thus obtained for calculation of the mixture viscosity can be expressed as:

$$\begin{aligned} \ln(\eta_{\text{mix}} V_{\text{mix}}) = & x_1 \ln(\eta_1 V_1) + x_2 \ln(\eta_2 V_2) + \\ & + x_1 \ln\left(x_1 + x_2 \frac{V_2}{V_1} \exp\left(-\frac{\lambda_{12}}{RT}\right)\right) + \\ & + x_2 \ln\left(x_2 + x_1 \frac{V_1}{V_2} \exp\left(-\frac{\lambda_{21}}{RT}\right)\right) \end{aligned} \quad (5)$$

where V_i is the molar volume of pure component i , λ_{12} and λ_{21} are interaction parameters characterized by viscosities. It must be pointed out that parameters of the Wilson activity coefficient model in phase thermodynamics are obtained by fitting phase equilibrium data, while parameters of the Eyring-Wilson viscosity model are obtained by fitting the viscosity data only.

RESULTS AND DISCUSSION

A variety of mixtures containing ILs were used to evaluate the correlative and predictive capability of the Eyring-Wilson viscosity model at different temperatures and ambient pressure. The binary mixtures and temperature range evaluated in this study are listed in Table 1. It contains 49 types of binary mixtures with ILs and a total of 1560 points of experimental viscosity data from literatures. The Eyring-Wilson binary interaction parameters, λ_{12} and λ_{21} , were determined by an optimization algorithm incorporated in Matlab's optimization toolbox.

The interaction parameters and fitting results of the Eyring-Wilson equation are summarized in Table 1. It can be seen that this model can reproduce the viscosities of ILs mixtures satisfactorily. The average ADD% of Eq. (5) was about 2.59% for all 49 ILs systems. Only five systems of ILs mixtures have ADD% larger than 5%. Careful inspection of these systems with relatively large errors, it was found that some points of composition have quite larger errors than others. Because of the large value of viscosity of ILs and the evaporation of solvent during experiment, it was always the case that the viscosity determination for ILs mixtures had big errors for some points. Many studies [46–48] have pointed out that the high viscosity of ILs may be a limiting factor in the industrial application of ILs, because under these conditions the pumping and energy costs will become prohibitive. For real process applications, ILs can be diluted with an organic liquid component, which can confer primarily a suitable density and viscosity to the liquid phase. The well performance of the Eyring-Wilson equation for ILs mixtures presented in this study will be helpful for engineering and optimization calculations of mixtures containing ILs.

For comparison, the results of the Eyring-UNIQUAC [14] and Eyring-NRTL Equation are also shown in the Table 1. Other models such as the Seddon Equation [11] and the Fang and He Equation [12], that focus on the developing of one-parameter models are not compared here. It can be seen that the Eyring-Wilson Equation can give almost the same results as the Eyring-UNIQUAC Equation, but lower than that of the Eyring-NRTL Equation. As pointed by

Table 1. ILs mixtures with the corresponding deviations by the Eyring-Wilson viscosity model

ILs (1)	Solvent (2)	n	λ_{12}^c	λ_{21}^c	T / K	ADD / %			Reference ^b
						This work	Eyring-UNIQUAC	Eyring-NRTL	
[Bmim]BF ₄	Dimethylformamide	15	2420.2	-1122.2	298.15	2.55	2.65	2.70	[27]
[Bmim]BF ₄	Butanone	15	1219.6	-2750.9	298.15	1.99	1.83	1.68	[27]
[Bmim]BF ₄	Acetonitrile	28	-3436.6	-1636.2	298.15	1.31	1.31	1.29	[28,29]
[Bmim]BF ₄	Dichloromethane	15	-2520.5	-2897.5	298.15	3.97	3.52	5.43	[28,29]
[Bmim]BF ₄	Ethanol	13	1340.8	-1204.9	298.15	4.30	3.41	2.26	[28,29]
[Omim]BF ₄	Dichloromethane	15	-1265.3	-2208.7	298.15	5.01	4.40	12.38	[28,29]
[Omim]BF ₄	Butanone	15	-140.4	-2830.2	298.15	2.87	2.52	2.68	[28,29]
[Bmim]BF ₄	Acetone	15	39.7	-2601.6	298.15	2.34	2.18	2.16	[30]
[Bmim]BF ₄	Ethyl formate	15	-1898.3	-2814.6	298.15	2.12	1.85	1.87	[30]
[Bmim]BF ₄	Methyl acetate	15	-4717.9	-2742.0	298.15	4.17	3.89	3.74	[30]
[Bmim]BF ₄	Methyl formate	15	-1922.1	-2078.7	298.15	1.79	1.69	3.11	[30]
[Omim]BF ₄	1-Propanol	15	1918.5	-1928.6	298.15	0.69	0.65	0.86	[31]
[Omim]BF ₄	2-Propanol	15	1983.0	-1987.5	298.15	1.10	1.40	1.66	[31]
[Omim]BF ₄	Ethanol	15	-2287.4	-1558.6	298.15	0.67	0.65	0.63	[31]
[Omim]BF ₄	Methanol	15	-4610.0	-1836.1	298.15	1.91	2.09	8.14	[31]
[Bmim]PF ₆	2-Butanone	15	2268.7	-2223.4	298.15	1.70	1.50	1.29	[32]
[Bmim]PF ₆	Acetone	15	-1342.4	-1663.7	298.15	1.08	0.99	0.87	[32]
[Bmim]PF ₆	Cyclopentanone	15	-2711.1	-995.4	298.15	1.18	1.10	1.11	[32]
[Bmim]PF ₆	Ethyl acetate	15	3875.0	-3060.4	298.15	3.24	3.00	2.54	[32]
[Bmim]PF ₆	Pentanone	15	1929.0	-1977.4	298.15	2.81	2.15	1.61	[32]
[Bmim]PF ₆	Dimethyl sulfoxide	15	-5842.1	580.1	298.15	1.86	2.03	1.93	[33]
[Bmim]PF ₆	Acetonitrile	15	-4471.6	340.9	298.15	2.95	3.03	3.06	[33]
[Bmim]PF ₆	Methanol	15	1396.4	-1457.9	298.15	6.72	5.45	5.29	[33]
[Bmim]PF ₆	Thf	15	-3984.3	-1469.6	298.15	2.40	2.38	2.57	[33]
[Emim]EtSO ₄	Ethanol	26	-2935.3	-1350.8	298.15	0.98	1.22	1.15	[34]
[MTEOA]MeSO ₄	Ethanol	18	-5512.6	-3671.0	298.15	2.44	1.20	16.37	[35]
[Hmim]BF ₄	Butanone	15	-2164.3	-1791.4	298.15	1.50	1.28	1.30	[36]
[Hmim]BF ₄	Butylamine	15	-1193.1	-3630.9	298.15	1.11	1.63	1.56	[36]
[Hmim]BF ₄	Ethyl acetate	15	-1407.8	-2904.1	298.15	0.81	0.86	0.74	[36]
[Hmim]BF ₄	Thf	15	-4354.9	-3148.7	298.15	1.06	2.92	5.82	[36]
[Et ₂ NH]HSO ₄	Dimethyl sulfoxide	75	-2225.7	-2475.8	298.15-328.15	2.46	- ^a	3.82	[37]
[Bmim]Cl	Dimethyl sulfoxide	75	-2321.1	-2897.8	298.15-328.16	2.34	2.52	2.66	[37]
[Bmpy]BF ₄	Methanol	39	482.8	-420.3	298.15-323.15	2.64	0.96	0.93	[38]
[Bmim]ClO ₄	Ethanol	69	2375.9	-1879.1	283.15-343.15	5.80	3.73	3.84	[39]
[Omim]BF ₄	Ethanol	78	-4717.0	-527.7	283.15-343.15	6.53	6.85	6.77	[39]
[Omim]Cl	1-Propanol	39	-316.7	1884.6	298.15-328.15	3.87	3.86	3.90	[40]
[Omim]Cl	Ethanol	33	-1135.2	459.2	298.15-328.15	3.41	3.32	3.31	[40]
[Omim]Cl	Methanol	45	-1924.9	-292.3	298.15-328.15	4.64	4.14	4.47	[40]
[Emim]EtSO ₄	1-Propanol	33	-3014.3	-336.4	298.15-328.15	1.06	1.25	1.23	[41]
[Emim]EtSO ₄	2-Propanol	33	-3089.5	-695.2	298.15-328.15	0.89	0.89	1.00	[41]
[Emim]EtSO ₄	Methanol	39	-4374.3	-2059.6	298.15-328.15	5.85	5.93	8.54	[41]
[Bmim]MeSO ₄	Ethanol	39	-4612.2	-793.7	298.15-328.15	1.43	1.72	1.54	[42]
[Empy]EtSO ₄	Ethanol	36	-1274.6	-1901.1	298.15-328.15	3.25	3.11	3.33	[43]
[Bmim]PF ₆	Monoethanolamine	88	-2313.1	2186.8	288.15-323.15	4.30	3.94	3.49	[44]
[Bmim]PF ₆	<i>N,N</i> -Dimethyl-ethanolamine	88	3256.1	-3173.6	288.15-323.15	3.54	3.05	2.74	[44]
[Bmim]SCN	1-Heptanol	78	1724.4	-1655.5	298.15-328.15	1.56	- ^a	1.50	[45]
[Bmim]SCN	1-Octanol	66	1799.9	-1733.7	298.15-328.15	1.70	- ^a	1.76	[45]

Table 1. Continued

ILs (1)	Solvent (2)	n	λ_{12}^c	λ_{21}^c	T/K	ADDI %			Reference ^b
						This work	Eyring-UNIQUAC	Eyring-NRTL	
[Bmim]SCN	1-Nonanol	60	1414.0	-1923.7	298.15-328.15	1.84	- ^a	1.75	[45]
[Bmim]SCN	1-Decanol	72	444.6	-1841.2	298.15-328.15	1.03	- ^a	0.99	[45]
Total		1560	-	-	-	2.59	2.50	3.17	-

^aNot available because of the lack of volume and surface parameters of corresponding ills in the UNIQUAC model; ^bthe density value of mixture and pure substance were taken from the some references; ^cparameters at 298.15 K.

Wang *et al.* [14], the larger error of the Eyring-NRTL Equation may attribute to the fact that the fixed value of a in that equation is not a globally optimized value. It means that the Eyring-NRTL Equation actually needs three parameters to get better results. The Eyring-UNIQUAC Equation needs two parameters and give little better results, however, it requires for each pure substance i , a relative area parameter q_i and a relative volume parameter r_i [14,20]. These parameters can be obtained by group contribution approach for traditional compounds such as alkanes and aromatics. However, due to the lack of pure component volume and area parameters for complex compounds, such as for the ILs, the UNIQUAC model cannot be easily applied for these systems. As shown in the Table 1, we can hardly get the group contribution values for the structure parameters calculations of 1-butyl-3-methylimidazolium thiocyanate ([Bmim]SCN). The Wilson Equation has also well founded theoretical basis as compared to the UNIQUAC and NRTL Equations

but with more simple mathematical forms. The result shows that the Eyring-Wilson Equation is a good compromise between complication and accuracy for the viscosity calculation of ILs mixtures.

As an illustration of the performance of the Eyring-Wilson Equation, the calculations of the mixture viscosity by Eq. (5) for ILs solutions determined by Zhu *et al.* [33] were recently given in Figure 1. These mixtures include [Hmim][BF₄] type of ILs with four polar and nonpolar solvents. The line represents the calculation results of the Eyring-Wilson viscosity model, and the symbols represent the experimental data. From the figure and table, it can be seen that the agreement between experimental data and the correlation is excellent. The average absolute mixture viscosity deviation for the data in the Figure 1 is less than 1.5%. In Figure 1, it can also be seen that the addition of organic solvents into ILs dramatically lowers down the viscosity of mixtures. In the solvent rich region ($x_{ILs} < 0.5$) the viscosity of mixtures is only have

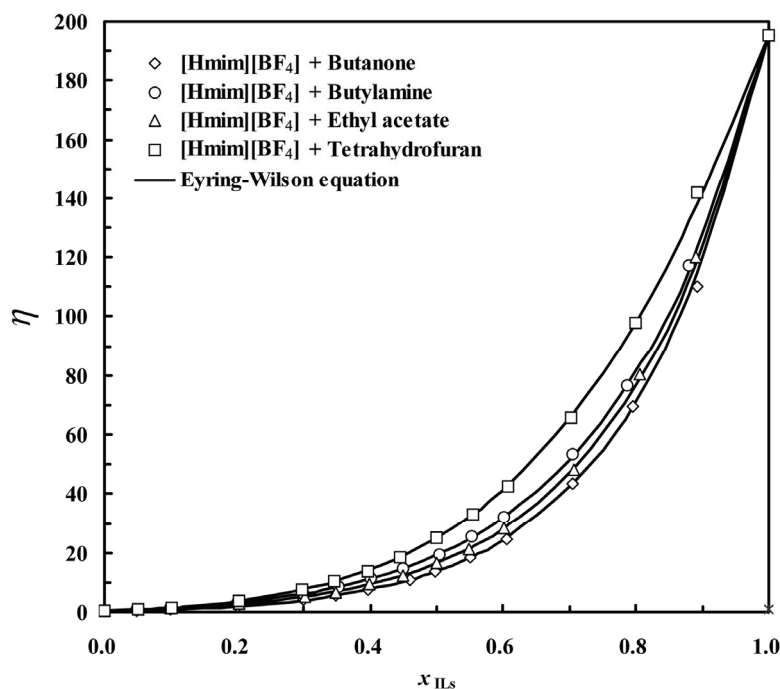


Figure 1. Plot of the experimental viscosity from Zhu *et al.* [36] against the viscosity calculated by the Eq. (5) for some [Hmim][BF₄] + solvents at $T = 298.15$ K.

about 10% of value of relative pure ILs. The Eyring-Wilson equation can well represent the viscosity behavior for both ILs rich and solvent rich regions.

If the viscosities of ILs mixtures cover a wide range of temperatures, it is necessary to consider temperature dependence for the binary parameters of the Eyring-Wilson Equation. The commonly used formulations for the temperature dependence of parameters in activity coefficient models were adopted:

$$\lambda_{ij} = A_{ij} + \frac{B_{ij}}{T} \quad (6)$$

where T is the temperature; A_{ij} and B_{ij} are temperature independent constants for a given ILs mixtures. As shown in Figure 2, a single set of two binary parameters with temperature dependence by the Eq. (5) is sufficient for the correlation of the $[\text{Et}_2\text{NH}][\text{HSO}_4] + \text{DMSO}$ mixtures [37] at different temperatures. Overall, very good agreement between calculated and experimental data at all the temperatures is presented. However, it must be pointed out that the application of the Eyring-Wilson Equation should be limited in which the temperature does not vary largely over its calibrated range.

CONCLUSION

Ionic liquids (ILs) are attractive as green solvents primarily because of their environmental benefits. However, the modeling of transport properties for

mixtures containing ILs has lagged behind growth. In this work, a semi-theoretical equation is presented for the viscosity calculations of ILs mixtures. The model is based on Eyring's mixture viscosity model and assumes that the non-ideal term can be represented by the equation similar to the Wilson activity coefficient equation. The accuracy of the model was tested by comparing calculated and experimentally measured viscosity values from the literature. The results show that the model adequately describes the viscosity relationship with composition and temperature. The proposed Eyring-Wilson Equation has a relatively simple mathematical form and can be easily incorporated into process simulation software for processes containing ILs mixtures.

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Abbreviations

[Bmim]BF₄ - 1-butyl-3-methylimidazolium tetrafluoroborate

[Bmim]PF₆ - 1-butyl-3-methylimidazolium hexafluorophosphate

[Bmim]Cl - 1-butyl-3-methylimidazolium chloride

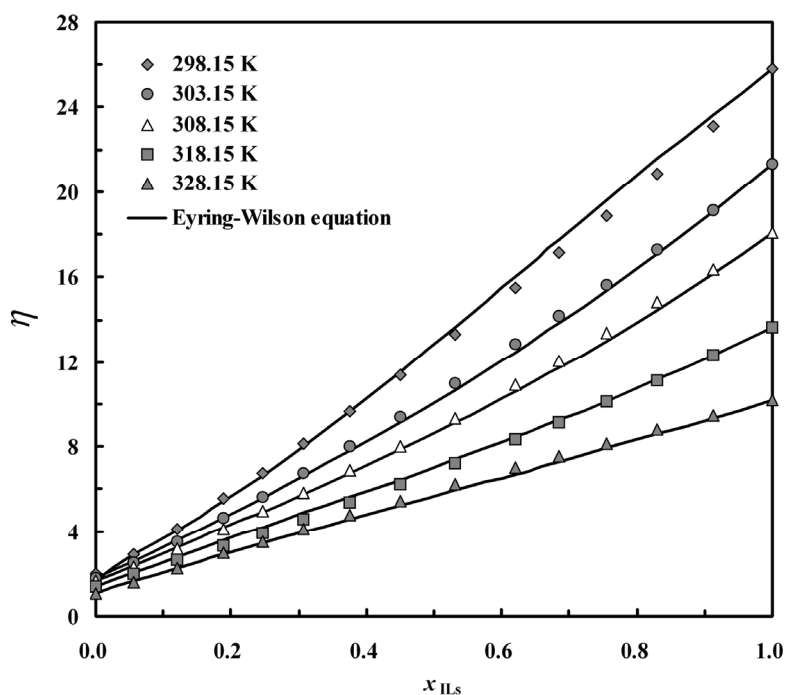


Figure 2. Plot of the experimental viscosity from Govinda *et al.* [37] against the viscosity calculation for $[\text{Et}_2\text{NH}][\text{HSO}_4] + \text{DMSO}$ mixture at different temperatures.

[Bmpy]BF₄ - 1-butyl-3-methylpyridinium tetrafluoroborate
 [Bmim]ClO₄ - 1-butyl-3-methylimidazolium perchlorate
 [Bmim]MeSO₄ - 1-butyl-3-methylimidazolium methylsulfate
 [Bmim]SCN - 1-butyl-3-methylimidazolium thiocyanate
 [Emim]EtSO₄ - 1-ethyl-3-methylimidazolium ethylsulfate
 [Empy]EtSO₄ - 1-ethyl-3-methylpyridinium ethylsulfate
 [Et₂NH]HSO₄ - diethyl ammonium hydrogen sulfate
 [Hmim]BF₄ - 1-hexyl-3-methylimidazolium tetrafluoroborate
 [MTEOA]MeSO₄ - tris-(2-hydroxyethyl)-methylammonium methylsulfate
 [Omim]BF₄ - 1-octyl-3-methylimidazolium tetrafluoroborate
 [Omim]Cl - 1-octyl-3-methylimidazolium chloride.

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NAUČNI RAD

MODELOVANJE VISKOZNOSTI RASTVORA JONSKIH TEČNOSTI POMOĆU EYRING- WILSONOVE JEDNAČINE

Poluteorijski model zasnovan na klasičnoj Eyringovoj jednačini za određivanje viskoznosti smeša i Wilsonovoj jednačini koeficijenata aktivnosti predstavljen je za korelisanje viskoznosti sistema jonska tečnost-rastvarač. Tačnost predloženog modela verifikovana je poređenjem izračunatih i eksperimentalno određenih vrednosti viskoznosti iz literature za 49 smeša sa ukupno 1560 eksperimentalnih tačaka. Rezultati pokazuju da jednačina slična Wilsonovoj jednačini koeficijenata aktivnosti može dobro biti primenjena za opisanje neidealnog člana u Eyringovoj jednačini za određivanje viskoznosti smeša. Model je relativno jednostavnog matematičkog oblika i može se lako inkorporirati u softver za simulaciju procesa.

Ključne reči: jonske tečnosti; viskoznost; Wilsonova jednačina; zeleni rastvarač.