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Corresponding states theory for the prediction of surface tension of ionic liquids

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Abstract The surface tension of ionic liquids is studied according to phenomenological scaling and the law of corresponding states. The reduced coordinates γ^*–T^*, where γ^* represents the reduced surface tension and T^* is the reduced temperature, are introduced for the prediction of the surface tension from the melting point up to boiling point. It has been shown that the correlation can be expressed as a unique straight-line plot with a linear correlation coefficient of 0.984 that requires only the melting and boiling point parameters and can predict the surface tension accurately.

Keywords Corresponding states · Ionic liquids · Surface tension

Introduction

Ionic liquids (ILs) were typically composed of large organic cations and small organic/inorganic anions and usually considered as a kind of special organic salt. In the past decade, they have received increasing attention due to their promising practice applications in chemistry and engineering such as reaction, separation, and catalyzation [1, 2]. Liquid surface tension as equilibrium thermodynamic property is also important in the design of industrial applications. Surface tension of a liquid is related to the intermolecular interaction potential energy and the liquid interfacial microstructure. Despite numerous research works, investigation of the thermophysical properties of ILs, their temperature dependences, and their correlations are still open fields of research [3–8].

Very few works have systematically studied the qualitative and/or quantitative relationships between the structures of ILs and their fundamental properties, such as melting point, viscosity, density, thermal and electrochemical stability, solvent properties, and surface tension [9–11]. To better understand the nature of ionic liquids and rationally expand their applications, knowledge of their physical properties is required. At present, however, data for many other physicochemical properties of ionic liquids are in short supply or too unreliable to allow similar structure–property relationship studies. Recently, Deetlefs et al. [10] attempted to predict surface tension by using the quantitative structure–property relationship (QSPR) correlations previously proposed by Knotts et al. [11]. They attempted with limited success due to the lack of experimental data. Then, Gardas et al. [12, 13] applied the QSPR correlation of Knotts et al. to predict surface tensions of assembled large database for ionic liquids. Ghatee et al. [3] correlated a linear relation between logarithm of surface tension and fluidity involving the characteristic exponent.

In this work, we propose simple regularities for surface tension of ILs, which has been applied for alkali metals [14] and alkali halides [15]. The reliability and utility of the modification proposed herein are established by performing detailed comparisons of the experimental values from the melting temperature up to the boiling temperature.

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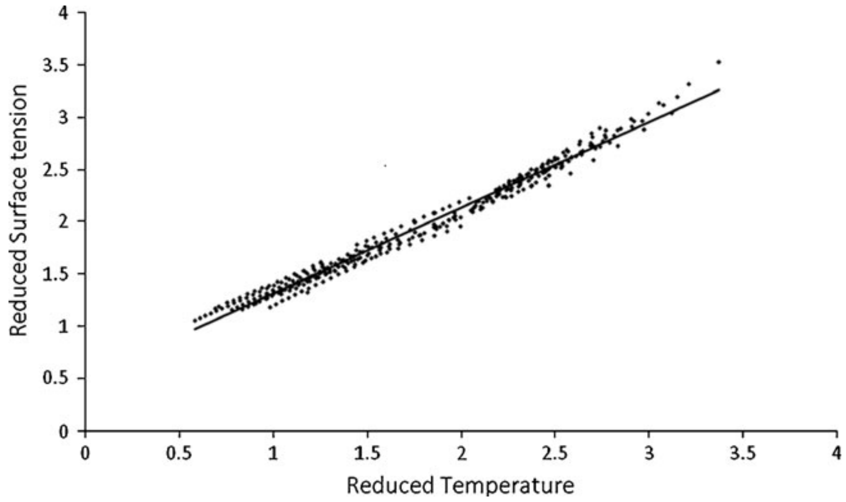

Fig. 1 Correlation results of reduced surface tension, γ^*, vs. reduced temperature, T^*, for ionic liquids

Table 1 List of ionic liquids, boiling [38] and melting temperatures, temperature range, and deviation of experiment surface tension and surface tension predicted by Eq. (4)

Ionic liquid	T_b/K	T_m/K	ΔT/K	AAD(%)	Refs.[a]
$[C_2mim]TFSO_3$	662	262	296–353	4.13	[39, 54]
$[C_4mim]OcSO_4$	895	307	278–328	4.10	[40, 55]
$[C_4mim]dca$	783	267	293–344	1.53	[41, 56]
$[C_4mim]PF_6$	554	280	288–352	2.35	[42, 57]
[N1134] NTf_2	761	293	299–324	1.50	[39, 58]
[N6222] NTf_2	829	293	299–347	2.63	[39, 59]
$[C_2mim]$ $EtSO_4$	712	236	278–328	0.86	[40, 60]
$[C_6mim]$ NTf_2	908	272	283–351	1.70	[43, 61]
$[C_4mim]$ NTf_2	862	270	284–351	0.91	[44, 62]
$[C_3mim]$ PF_6	531	252	318–352	0.90	[45, 63]
$[C_1mim]$ $MeSO_4$	666	308	288–313	3.72	[46, 64]
$[C_2mim]$ NTf_2	816	255	283–351	3.52	[44, 65]
$[C_6mim]Cl$	603	188	298–393	2.67	[42, 65]
$[C_6mim]$ BF_4	541	191	298–393	6.14	[42, 65]
$[C_4mim]BF_4$	495	202	284–351	7.58	[44, 65]
$[C_6mim]$ PF_6	600	199	284–353	2.40	[45, 65]
$[C_8mim]$ PF_6	646	203	283–353	1.60	[45, 65]
$[C_8mim]Cl$	649	218	298–393	6.59	[42, 65]
$[C_8mim]$ BF_4	586	185	298–361	0.85	[41, 65]
$[C_8mim]$ NTf_2	954	265	293–343	3.77	[47, 65]
$[C_4mim]$ $MeSO_4$	735	298	283–313	2.60	[48, 65]
$[C_4mim]$ $TFSO_3$	707	286	293–343	4.30	[49, 56]
$[C_2C_2im]$ NTf_2	839	287	293–343	1.61	[50, 65]
$[C_3mpyr]$ NTf_2	736	281	293–343	2.14	[50, 66]
$[C_4mpyr]$ NTf_2	759	265	293–343	1.61	[50, 66]
$[C_4mim]I$	613	201	298–393	0.75	[42, 67]
$[C_4mim]Cl$	558	314	298–393	4.33	[42, 67]
$[C_2py]$ NTf_2	778	303	288–338	0.72	[51]
$[C_4py]$ NTf_2	824	299	298–338	3.00	[51]
$[C_3mim]$ NTf_2	839	289	312–473	2.82	[52, 66]
Average				2.98	

[a] The first reference is for the surface tension and other for melting point data, respectively

Table 2 AAD(%) results for surface tension predicted by Eq. (4) for samples not to be used in correlation

Ionic liquid	ΔT/K	AAD(%)	Refs.
[C_5mim] NTf_2	312–512	6.43 (15.71)	[52, 66]
[C_{12}mim] NTf_2	333–532	13.23 (31.94)	[52, 66]
[C_7mim] NTf_2	293–353	2.23 (4.33)	[53, 66]
[P6$_3$14] NTf_2	298–349	5.52 (9.33)	[39, 68]

Values in parentheses represent maximum deviations

Corresponding states theory

The corresponding states theory (CST) for simple spherical fluids was first proposed by van der Waals [16] in an empirical form and later demonstrated by Pitzer [17]. Following its initial empirical formulation by van der Walls, the corresponding states principle has evolved in order to deal with several non-spherical and polar molecules. Its success is grounded on the accuracy obtained in broad temperature and pressure conditions and its strong theoretical basis [17–23], while mathematically simple and using a minimum amount of experimental information. One of its advantages is that the same formalism can be used for different properties whereas the only changes are the definition of the reduced property and the equation used for the reference system, or the reference system itself. Since these models do not use any experimental data of the estimating property of the target fluid, they are also predictive. The only experimental information required from the target fluid are the critical properties and the Pitzer acentric factor. The application of corresponding states models extends from equilibrium properties such as vapor pressure [24–28], liquid density [26, 28–30], or surface tension [31–36].

Correlation

Empirical relationships for the surface tension of ILs are shown to follow from the principle of corresponding states and are defined as [14, 15]:

$$\gamma^* = \left(\frac{\gamma}{\gamma_\mathrm{m}}\right)\left(\frac{T_\mathrm{b}}{T}\right) \tag{1}$$

$$T^* = \left(\frac{T_\mathrm{b}-T}{T_\mathrm{b}-T_\mathrm{m}}\right)\left(\frac{T_\mathrm{b}}{T}\right) \tag{2}$$

where γ and γ_m are the surface tension at temperature T and the melting temperature, T_m, respectively, and T_b is the boiling temperature. Dimensionless quantities were introduced by choosing the corresponding quantities at the melting temperature.

One distinct advantage of this set (γ_m, T_m) is that these are standard parameters that are readily available, as compared with critical parameters. Figure 1 shows the correlations suggested in this work. In Fig. 1, the solid line is the linear fit to the experimental data, which was obtained by linear regression as:

$$\gamma^* = 0.819\ T^* + 0.500 \tag{3}$$

The linear correlation coefficient for Eq. (3) is 0.984. From Eq. (3), by using definitions (1) and (2) for the surface tension, γ, we can write:

$$\gamma = 0.819\left(\frac{T_\mathrm{b}-T}{T_\mathrm{b}-T_\mathrm{m}}\right)\gamma_\mathrm{m} + 0.500\left(\frac{T}{T_\mathrm{b}}\right)\gamma_\mathrm{m} \tag{4}$$

Results and discussion

In Table 1, the absolute average percent deviation, AAD (%), was calculated in the range from the melting point to the boiling point.

The AAD(%), defined as,

$$\mathrm{AAD}(\%) = \frac{1}{N}\sum_{N}^{1}\left|\frac{\gamma_{\exp t.}-\gamma_{\mathrm{calc.}}}{\gamma_{\exp t.}}\right| \times 100 \tag{5}$$

where N represents the number of data points.

Reduced surface tension, as well as with the reduced temperature, can be expressed as a unique straight-line plot with a linear correlation coefficient of 0.984. In a database of 385 data points for 30 ILs, the mean percent deviation observed was only 2.98% (Table 1). We would like to

Table 3 Comparisons of the AAD(%) of surface tension correlated by this work and QSPR method [13]

Ionic liquid	ΔT/K	AAD(%), this work	AAD(%), QSPR method
[C_4mim]PF_6	288–352	2.35	3.50
[C_2mim] $EtSO_4$	278–328	0.86	2.64
[C_6mim] NTf_2	293–343	1.95	6.59
[C_4mim] NTf_2	293–343	0.98	5.33
[C_2mim] NTf_2	283–351	4.00	5.33
[C_4mim]BF_4	288–343	6.60	6.00
[C_8mim] PF_6	293–313	1.50	11.93
[C_8mim] BF_4	293–343	0.80	13.96
[C_4mim] $MeSO_4$	283–313	2.60	6.41
[C_4mim]$TFSO_3$	293–343	4.30	10.46
[C_4mim]I	298	0.64	0.13
[C_7mim] NTf_2	293–353	2.23	7.67
Average		2.40	6.66

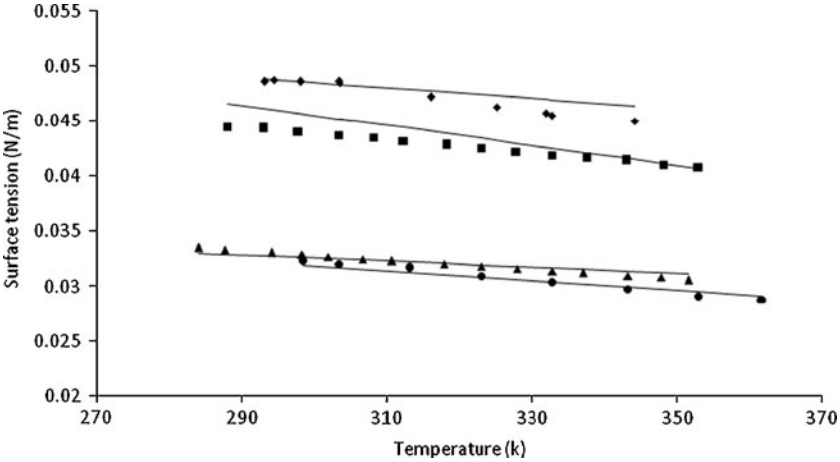

Fig. 2 Comparison of experimental and correlated surface tension data for [C_8mim]BF_4 (*black circle*), [C_4mim]NTf_2 (*black triangle*), [C_4mim]Pf_6 (*black square*), [C_4mim]dca (*black diamond*). *Dots* experimental data, *Solid line* predicted by Eq. (4)

illustrate the procedure by specific examples, [C_5mim]NTf_2, [C_7mim]NTf_2, [C_{12}mim]NTf_2, and [$P6_314$]NTf_2, that are not to be used in correlation. Table 2 shows the agreement with data reported from measurements. In Table 3, a comparison between the predicted surface tension values using Eq. (4) and the QSPR correlation [13] for the some ionic liquids is reported. It is apparent from Table 3 that our correlation results are in good overall agreement with the experimental data over the entire temperature range.

In the dimensionless representation γ^*, there is a unique function for ionic liquids (Fig. 1). With dimensions, γ gives a curve for [C_8mim]BF_4, [C_4mim]NTf_2, [C_4mim]Pf_6, [C_4mim]dca, resulting in good correlation with the experimental data (Fig. 2). The comparison of experimental values of surface tension at melting temperature γ_m and calculated ones using Eq. (4) is shown in Fig. 3. A good agreement is obtained, indicating that expression provides a satisfactory description for the surface tension at melting temperature. In this work, we employ kT_m as the characteristic energy, and no molecular parameters are involved. The proposed correlation has a simple form for easy calculation, requiring only the melting and boiling point parameters, and can predict the surface tension up to the boiling temperature accurately.

The central idea of this work is the existence of a universal relation between dimensionless parameters formed using the physical quantities of interest and the applicability of CST to the estimation of surface tension. Thus, it is clear from the above results that whether a “universal” relation exists between the reduced surface tension and temperature depends on the particular parameter set used.

Most existing correlations for the prediction of the thermodynamic properties of ordinary fluids [16–23, 37] use the critical point data. The critical properties of ionic liquids cannot be determined since many of these compounds start to decompose as the temperature approaches the normal boiling point [38]. Moreover, the T_b as a scaling parameter has not been measured, but the correlating methods for T_b are available and estimates with high accuracy as compared with T_c. One distinct advantage of this set (γ_m, T_m) is that these are standard parameters and are readily available. Thus, it would be interesting to see if

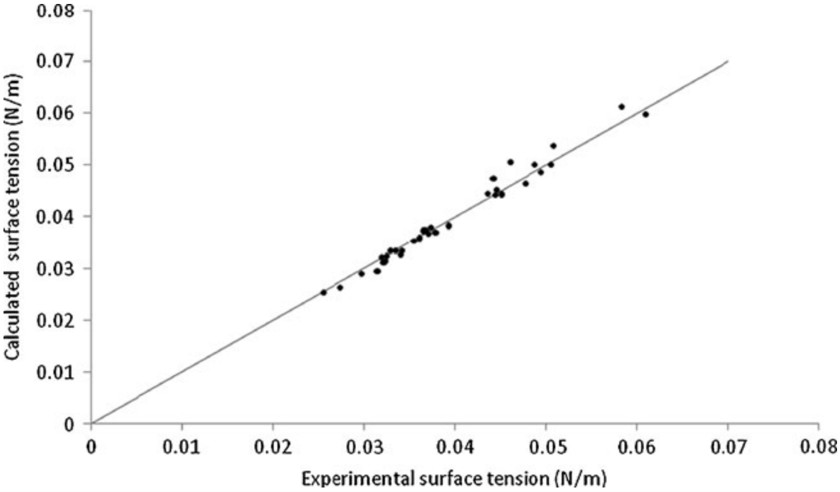

Fig. 3 Comparison of experimental and calculated surface tension at melting temperature

they do well. Moreover, the effectiveness of T_m, and T_b as scaling parameters has been observed for liquid alkali metals [14]. Thus, we may classify those sets that suggest a universal relation, as desirable from a CST viewpoint. This does not mean that the other parameter sets have to be rejected altogether but only that they are not available within the context of CST. Also, the correlation shows how the successful empirical regularities can be obtained from a simple equation. As far as our results show, the present correlation could be applicable to other ionic liquids, but lack of experimental data hinders us from testing it.

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