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Electrical conductivity behavior of various ionic liquids

J. Gómez-Santana, A. F. Estrada-Alexanders, I. Dávila-Ortega, P. Díaz-Leyva and R. Sánchez
Departamento de Física, Universidad Autónoma Metropolitana Unidad Iztapalapa,
Av. Ferrocarril San Rafael Atlixco 186, Col. Leyes de Reforma 1A Sección C.P. 09310, Ciudad de México, Mexico.

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The present work examines the experimental electrical conductivities as a function of temperature for a variety of ionic liquids near room temperature. Three analytic models are used to describe them, the simple Arrhenius equation, the Vogel-Tamman-Fulcher equation and a novel semi-empirical modified form based on precedents for electrolyte solutions. Patterns are determined that relate the model that best describes the experimental conductivity of a given ionic liquid and its specific chemical structure.

Keywords: Ionic liquids; non-Arrhenius behavior; non-Arrhenius conductivity; conductivity of liquid.

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1. Introduction

Ionic liquids (ILs) are defined as salts whose melting points are below 100°C [1,2] and are often liquids at room temperature [1–3]. They are of considerable scientific interest and have a wide range of potential applications, such as “green” solvents [4–7] and rheological modifiers [7]. This is especially true given the vast number of possible ILs and their amenability to structural and functional design [8]. Their mobile ions make them electrical conductors, and they are generally electrochemically stable, making them of interest as solvents for electrochemical reactions [1,7]. Since they often exhibit good thermal stability [1,7,9], their properties’ thermal dependence is of particular practical interest. However, partly due to their relatively recent discovery and also due to the complexity of their intermolecular interactions, the temperature dependence of many of their properties-including conductivity-is still poorly understood [2]. On the other hand, the simplest model typically used to describe the temperature dependence of the conductivity of electrolyte solutions in organic solvents is typically the simple Arrhenius model [10]:

$$\sigma\left(T\right) = \sigma_0 \exp\left(-\frac{E}{k_B T}\right) , \tag{1}$$

where σ_0 is a constant, E is the activation energy per molecule, k_B is Boltzmann’s constant (1.380649×10^{-23} JK^{-1}) and T is the absolute temperature [10]. Experimentally, many solutions do not follow this model [10], and although various modified models have been used, there is no consensus on the best model nor on the physical insights provided by the more common models [10–16]. Nevertheless, for some ionic liquids and for mixtures thereof and of conventional solvents, the simple Arrhenius model provides a good fit [17, 18]. In other cases, a Vogel-Tamman-Fulcher (VFT) behavior, itself a modified Arrhenius model [11–13], has been found [18, 19], corresponding to the form:

$$\sigma\left(T\right) = \sigma_{VFT} \exp\left(-\frac{E_{VFT}}{k_B\left(T - T_0\right)}\right) , \tag{2}$$

where T_0 is the temperature at which the conductivity tends to zero. This all suggests that a suitable model should readily simplify to Eq. (1) in some conditions.

On the other hand, it has been found that at least for some cases a remarkably good fit to the experimental data for ionic solutions can be obtained using a compensated form of the Arrhenius form [10, 20]:

$$\sigma\left(T\right) = \sigma_0'\left(\epsilon_r\left(T\right)\right) \exp\left(-\frac{E}{k_B T}\right) , \tag{3}$$

where ϵ_r is the dielectric constant, which is generally temperature-dependent [10, 21–23]. An extremely good fit to the experimental conductivities of various solutions was obtained by scaling the prefactor $\sigma_0\left(\epsilon_r\left(T\right)\right)$ using the experimentally measured values of the dielectric constant [10]. The assumption that it only depends on temperature via the dielectric constant’s temperature-dependence is thus borne out. There is also precedent of the dielectric constant being involved in the electrical conductivity of solid nancomposites as a multiplying factor in the absolute temperature in the Arrhenius form [23], though since that form was used for systems very different from ILs we are using the approach of Eq. (3) where the simple Arrhenius and the VFT forms are not applicable. However, aside from different ILs not necessarily exhibiting the same behaviour, this approach involves measuring the dielectric constant’s temperature variation for a given liquid, and different sources in the literature are not always in close agreement in the reported values as a function of temperature.

Due to their relative novelty, and to the non-trivial nature of their intermolecular interactions and molecular behaviour, the behaviour of ILs’ conductivity, and its relationship to other properties, such as their dielectric constants, is poorly understood and it is unclear which of these models, if any, is best applicable. On the other hand, the basis of the compensated Arrhenius form and its implied relationship between the dielectric constant and the conductivity are not

TABLE I. The labels used in this work for the various ionic liquids used, and their full names, are shown.

Label	Full Compound Name
IL1	1-Hexyl-3-methylimidazolium bis(trifluormethylsulfonyl)imide
IL2	1-Butyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide
IL3	1-Butyl-3-methylimidazolium hexafluorophosphate
IL4	1-Butyl-3-methylimidazolium tetrafluoroborate
IL5	Triisobutylmethylphosphonium Tosylate
IL6	1-Butyl-1-methylpiperidinium bis(trifluoromethylsulfonyl)imide
IL7	1-Methyl-1-propylpiperidinium bis(trifluoromethylsulfonyl)imide
IL8	Tributyltetradecylphosphonium dodecylbenzenesulfonate
IL9	Methyl-trioctylammonium bis(trifluoromethylsulfonyl)imide
IL10	Tributylmethylammonium bis(trifluoromethylsulfonyl)imide

well understood and thus any evidence of its applicability to ILs may shed light on its origins. Furthermore, while the simple Arrhenius and the VFT forms can be at least qualitatively understood as originating from a resistance to ion mobility governed by the inverse viscosity [24], the compensated Arrhenius form is not readily compatible with such a basis, making it a particularly intriguing yet relatively little explored model. In the present work, experimental electric conductivities are compared to the simple Arrhenius model, a form based of the compensated Arrhenius model and to the VFT form, seeking to shed light on these issues on the basis of the nature of the molecular structures involved.

2. Methods

The room temperature ionic liquids used were all supplied by Ionic Liquid Technologies, and they were used as received; the ILs used are listed in Table I, while the molar masses and purities are shown in Table II.

Conductivities were measured using a HI 2550 conductivity meter (Hanna Instruments), which allows for simultaneous, real-time conductivity and temperature measurements. Samples were placed in a Pyrex cylinder and the conductivity meter's probe was submerged into the liquid. The samples were then heated well above 100°C under a nitrogen atmosphere at 1.0 atm in a glove box using a heating plate, and they were then allowed to cool spontaneously until reaching room temperature. To minimize temperature gradients, only data from the cooling process were used, as it is much slower than the heating process and thus the temperature has more time to homogenize. Furthermore, the greater delay involved between pouring the sample and taking data allows for relaxation of any effects induced on the sample by its flow. As additional precautions against moisture, uncovered drierite, previously dehydrated by heat, was kept in the glove box during each experiment, and any trace humidity on the Pyrex cylinder was removed by heat prior to use. For each IL, measurements were repeated at least three times; the data was averaged in ranges for which the results are highly reproducible (standard deviation $\lesssim$ 3% of the mean, except for IL8, which yielded a standard deviation of $\sim 5\%$ of the mean at the higher temperatures used for fitting, but exhibited a highly consistent trend; for most of the data shown the standard deviation is well under 2% of the mean). For the fitted functions, the non-linear least-squares Marquardt-Levenberg algorithm was used, as implemented by Gnuplot. The obtained uncertainties in the resulting values of fitted parameters are asymptotic standard errors obtained from the variance-covariance matrix after the final iteration.

TABLE II. The molar masses and purities, according to the manufacturer, are shown for the various ionic liquids used.

Label	Compound	Molar Mass (g/mol)	Purity
IL1	$[C_6\text{mim}]\,[\text{NTf}_2]$	447.42	99.5%
IL2	$[C_4\text{mim}]\,[\text{NTf}_2]$	419.36	99%
IL3	$[C_4\text{mim}]\,[\text{PF}_6]$	284.18	99%
IL4	$[C_4\text{mim}]\,[\text{BF}_4]$	226.02	99%
IL5	$[C_{13}H_{30}P^+]\,[\text{TosO}]$	394.23	>95%
IL6	$[C_{10}H_{22}N^+]\,[\text{NTf}_2]$	436.43	99%
IL7	$[C_9H_{20}N^+]\,[\text{NTf}_2]$	422.41	99%
IL8	$[C_{26}H_{56}P^+]\,[\text{DBS}]$	725.18	>95%
IL9	$[C_{25}H_{54}N^+]\,[\text{NTf}_2]$	648.85	99%
IL10	$[C_{13}H_{30}N^+]\,[\text{NTf}_2]$	480.53	99%

3. Results and discussion

Figure 1 shows that a simple Arrhenius fit is sufficient to describe the conductivities of some ILs and Table III shows the corresponding parameter values. IL1's conductivity has been reported to be well-described empirically by a polynomial [9], but this approach involved four adjustable parameters, whereas our approach has the advantage of only using two adjustable parameters. For this IL, our results are very

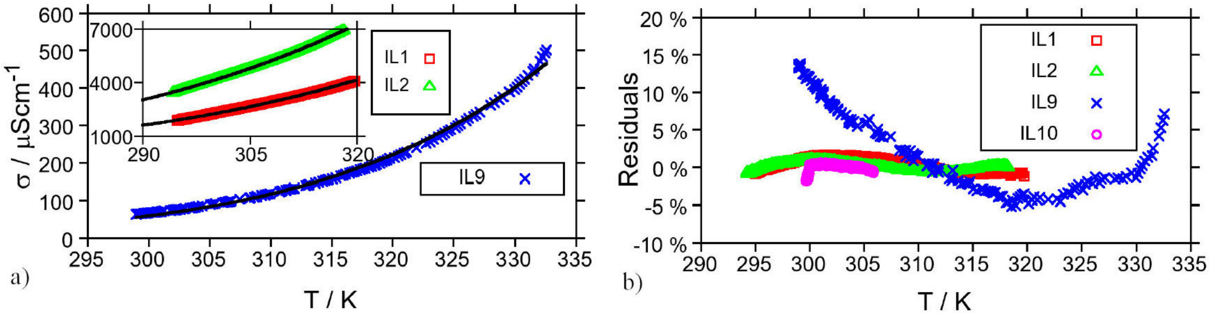

FIGURE 1. Various examples are shown of ILs whose conductivities are well-fitted by an Arrhenius relationship ($\sigma = \sigma_0 \exp(-E/k_B T)$); the inset's axes' units are the same as for the main figure a). The residuals (the experimental value minus the fitted function, as a percentage of the experimental value) are also shown for all ILs well-fitted by an Arrhenius relationship b).

TABLE III. The ionic liquids fitted by the simple Arrhenius equation ($\sigma = \sigma_0 \exp(-E/k_B T)$), and the fitted parameters, are shown.

Label	Compound	σ_0/μS cm^{-1}	E/J
IL1	[C_6mim] [NTf_2]	$4.13 \times 10^7 \pm 1.0\%$	$4.07 \times 10^{-20} \pm 0.1\%$
IL2	[C_4mim] [NTf_2]	$3.48 \times 10^7 \pm 0.8\%$	$3.74 \times 10^{-20} \pm 0.09\%$
IL9	[$C_{25}H_{54}N^+$] [NTf_2]	$7.36 \times 10^{10} \pm 13\%$	$8.67 \times 10^{-20} \pm 0.7\%$
IL10	[$C_{13}H_{30}N^+$] [NTf_2]	$1.22 \times 10^{10} \pm 6.6\%$	$7.27 \times 10^{-20} \pm 0.4\%$

similar to those of Kandil and Marsh [25]. We fitted their published data for the same IL to the simple Arrhenius form, obtaining a good fit, and the two fitted functions are very similar at temperatures below ∼303 K, while at higher temperatures the fit to their reported conductivities tends to be somewhat higher, for a maximum discrepancy $\approx 20\%$ at the top of our measured temperature range (data not shown). For their data, we obtained $E = 5.03 \times 10^{-20}$ J $\pm$ 2.4% and $\sigma_0 = 4.37 \times 10^8$ μS cm^{-1} $\pm$ 27.5%. Note that the two data sets correspond to different temperature ranges and Kandil and Marsh [25] used a sample synthesized in-house whereas we used a commercial sample, among other methodological differences. For IL2, a simulational study [26] has found that above ∼300 K its structural relaxation times for both cations and anions obey the simple Arrhenius form, consistent with our findings for the conductivity.

For ILs whose conductivities are poorly described by the simple Arrhenius equation, we assume that they are either described by the VFT equation or, inspired by the approach of Petrowsky and Frech [10], that their non-Arrhenius behavior is dominated by the dielectric constant's temperature dependence and that the conductivity is proportional to the product of a simple Arrhenius and the dielectric constant. According to the experimental data in the literature, for a variety of ILs, the dielectric constant itself varies roughly linearly with temperature [28] or is nearly temperature-independent [6] (which would recover simple Arrhenius behavior) at comparable temperature ranges. On this basis, we begin by assuming that the dielectric constant has a linear temperature-dependence and substituting accordingly yields:

$$\sigma(T) = (mT + b) \exp\left(-\frac{E}{k_B T}\right) , \qquad (4)$$

where m and b depend on the substance and $m = 0$ recovers the simple Arrhenius equation.

However, for the conductivities for which Eq. (4) was used, the fitted b values were consistently small and with uncertainties greater than the parameter itself; *i.e.*, zero was always within the fitted value's margin of error. This suggests that b can be set to zero. Since this eliminates one adjustable parameter, this allows us to consider an additional T^2 term without increasing the overall number of adjustable parameters; the following form is thus obtained:

$$\sigma(T) = \left(c_1 T + c_2 T^2\right) \exp\left(-\frac{E}{k_B T}\right) , \qquad (5)$$

which has only one more adjustable parameter than the simple Arrhenius equation and has the same number of adjustable parameters as the VFT equation. A power law and a purely $c_2 T^2$ form were also tried for $\sigma_0'(T)$ (*i.e.* for the prefactor modifying the Arrhenius form), but both consistently yielded poorer fits than Eq. (5) and than the VFT equation (data not shown).

Figure 2 shows the data and the corresponding fits to Eq. (5) for various ILs, for which it provides a better fit than either the simple Arrhenius or the VFT equations. Table IV shows the fitted parameters. Despite Eq. (5) simplicity and the rough approximation involved in describing the dielectric constant's temperature-dependence, very reasonable fits are obtained.

It must be emphasized that Eq. (5) is a novel expression and distinct from both the VFT and the simple Arrhenius equations; setting $b = 0$ ensures that it remains distinct from them for any values of c_1 and c_2. Regarding the physical interpretation of these parameters, E can be interpreted as the energetic cost of ions becoming available for conduction,

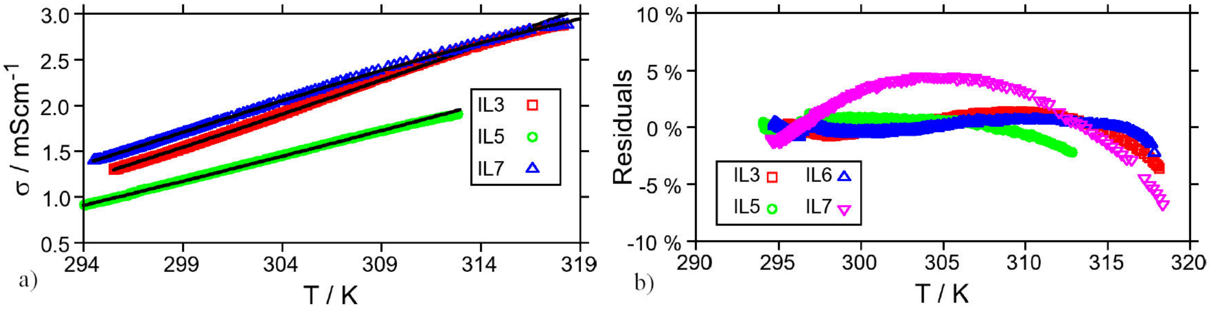

FIGURE 2. a) Examples are shown of ILs whose conductivities are well-fitted by Eq. (5) ($\sigma(T) = \left(c_1 T + c_2 T^2\right) \exp(-E/k_B T)$). The residuals (the experimental value minus the fitted function, as a percentage of the experimental value) are also shown for all ILs well-fitted by this equation b).

TABLE IV. The ionic liquids fitted by Eq. (5) (modified Arrhenius equation: $\sigma(T) = \left(c_1 T + c_2 T^2\right) \exp(-E/k_B T)$), and the fitted parameters, are shown.

Label	c_1/μS cm^{-1} K^{-1}	c_2/μS cm^{-1} K^{-2}	E/J
IL3	$-60.8 \pm 8.6\%$	$0.221 \pm 8.7\%$	$2.68 \times 10^{-26} \pm 1.5 \times 10^{6}\%$
IL5	$-45.7 \pm 2.8\%$	$0.167 \pm 2.8\%$	$1.06 \times 10^{-27} \pm 1.2 \times 10^{7}\%$
IL6	$2.96 \times 10^{5} \pm 2.0\%$	$-3.92 \times 10^{3} \pm 2.2\%$	$3.45 \times 10^{-20} \pm 0.23\%$
IL7	$4.79 \times 10^{4} \pm 167\%$	$-889 \pm 205\%$	$4.36 \times 10^{-20} \pm 11\%$

much as in the original simple Arrhenius equation. It follows that for small values, essentially all ions must be considered to be available for conduction. As for the other constants, Eq. (5) goes to zero at a temperature of $-c_1/c_2$, and for an ionic liquid this in turn can be interpreted as a temperature at which it freezes, becomes viscoelastic or the conductivity otherwise drops dramatically. For all the ILs shown in Table IV, this temperature is positive and below room temperature and thus physically plausible.

For the remaining compounds, the VFT equation was found to be best. Figure 3 shows the fits while Table V shows the fitted parameters; Table VI summarizes which model best describes each compound's conductivity. IL4 has been reported to be well-described by the VFT form before by Leys *et al.* [28] for a different temperature range, although with different values for the fitted parameters; the VFT equation, evaluated at 293.15 K using our parameters an those of Leys *et al.* yields values that differ by 8.5%.

Our approach of using an approximate description of the dielectric constant's temperature dependence is advantageous since it is non-trivial to measure experimentally [5,30] and its temperature-dependence is not well understood nor widely reported in the literature for ILs. Even for ILs for which data is available, the dielectric constant is usually reported only for a few temperatures. Our approach would allow, for such cases, the use of interpolations or extrapolations, not necessarily linear ones, to fit the conductivities. In addition, if analytic or numerical models are developed that predict how the dielectric constant scales with temperature, these could be used instead of the model used here. Thus, it provides a promising avenue to explore connections between electrical conductivity and the dielectric properties that the VFT form does not provide.

Regarding which model is best for a given IL and its structure, the simple Arrhenius equation adequately describes the conductivities in the ranges examined for compounds lacking aromatic groups and having alkyl chains with up to

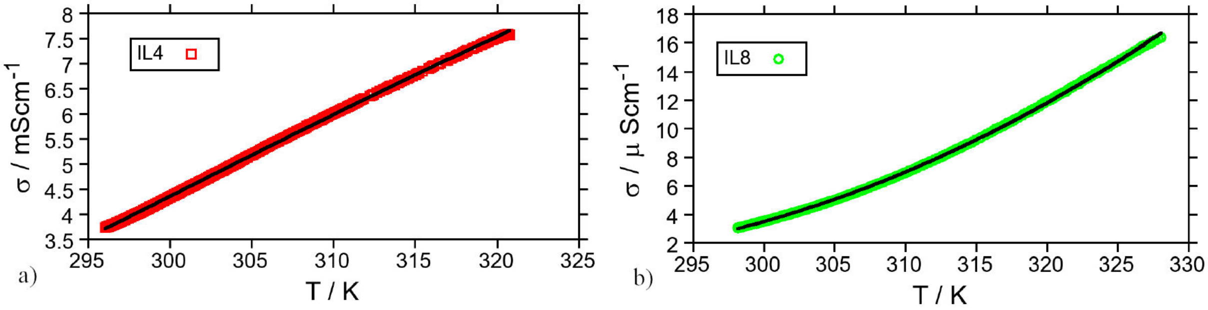

FIGURE 3. The conductivities of the two ILs best fitted by the VFT form are shown, along with the fits to the VFT form.

TABLE V. The ionic liquids fitted by the VFT equation, and the fitted parameters, are shown.

Label	σ_{VFT}/S cm^{-1}	E_{VFT}/J	T_0/K
IL4	$3.32 \times 10^4 \pm 0.99\%$	$1.52 \times 10-21 \pm 1.1\%$	$246 \pm 0.13\%$
IL8	$627 \pm 3.5\%$	$4.68 \times 10^{-21} \pm 1.6\%$	$235 \pm 2.7\%$

TABLE VI. The ionic liquids used, and the model best fitting the data, on the basis of resulting in the smallest reduced χ^2 value, are shown.

Label	Compound	Best Model
IL1	[C_6mim] [NTf_2]	Simple Arrhenius
IL2	[C_4mim] [NTf_2]	Simple Arrhenius
IL3	[C_4mim] [PF_6]	Modified Arrhenius
IL4	[C_4mim] [BF_4]	VFT
IL5	[$C_{13}H_{30}P^+$] [TosO]	Modified Arrhenius
IL6	[$C_{10}H_{22}N^+$] [NTf_2]	Modified Arrhenius
IL7	[$C_9H_{20}N^+$] [NTf_2]	Modified Arrhenius
IL8	[$C_{26}H_{56}P^+$] [DBS]	VFT
IL9	[$C_{25}H_{54}N^+$] [NTf_2]	Simple Arrhenius
IL10	[$C_{13}H_{30}N^+$] [NTf_2]	Simple Arrhenius

8 carbon atoms or having ammonia (IL9, IL10) cations, or an amide anion and an imidazolium cation (IL1, IL2), whereas the VFT equation is more suitable for compounds containing long alkyl chains (IL8) or a BF_4 anion (IL4), and the modified Arrhenius equation tends to be best for compounds with a PF_6 anion (IL3), or an amide anion and a non-imidazolium cation (IL6, IL7), as well as for the sole aromatic compound studied (IL5) (see Table VI).

4. Conclusions

A modified Arrhenius equation, based on a simple approximation of the dielectric constant's temperature dependence, has been shown to describe a variety of ionic liquids that are not well-fitted by the simple Arrhenius equation and can provide a better fit than the VFT form, which has the same number of adjustable parameters. It has been established that the modified Arrhenius form is advantageous for an aromatic IL and for an IL with a hexafluorophosphate anion. For ILs with bis(trifluoromethylsulfonyl)imide anions and nitrogen-based (ammonia-based or piperidinium-based) cations, a pattern emerges: lower molecular weight, piperidinium-based cations require the modified Arrhenius form, whereas higher molecular weight, ammonia-based cations lead to conductivites that can be modeled with the simple Arrhenius form. Whether the difference is due to the cation structure or due to the alkyl chain lengths would require further research to elucidate, but either possibility would help establish more general patterns, and could shed light on the relationship between the dielectric constant and the conductivity.

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