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Physical Properties of Selected Ionic Liquids for Use as Electrolytes and Other Industrial Applications

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The physical properties of selected room temperature ionic liquids (RTILs) were measured using differential scanning calorimetry, du Nouy tensiometry, and rheometry primarily to determine their suitability for use in electrochemical double layer capacitors. RTILs have other uses, however, such as liquid−liquid extraction, catalysis, and heat transfer, and these measurements may also be of use to researchers working in other fields. The ionic liquids used were 1-ethyl-3-methylimidazolium dicyanamide [emim][N(CN)$_2$], 1,2-dimethyl-3-propylimidazolium bis(trifluoromethylsulfonyl)imide [dmpim][Tf$_2$N], and 1-butyl-1-methylpyrrolidinium tris(pentafluoroethyl)trifluorophosphate [bmpyr][Pf$_3$PF$_3$]. Glass transition temperatures, crystallization and melting temperatures, surface tensions, and viscosities are reported. The three ionic liquids displayed a range of crystallization behaviors when cooled to 123.15 K and reheated to 403.15 K. All liquids were Newtonian over a range of shear rates from (10 to 1000) s^{-1}. Viscosities decreased with temperature, and this has been modeled using the Williams−Landel−Ferry equation.

Introduction

Ionic liquids are essentially molten salts composed entirely of a mixture of anions and cations with a melting temperature below the boiling temperature of water. Room temperature ionic liquids (RTILs) are a relatively new class of compound usually defined as having a melting point at or below room temperature, although the exact maximum limit of melting point in the definition of both ionic liquids and RTILs is sometimes extended upward. RTILs have industrial uses including liquid−liquid extraction, catalysis, heat transfer fluids, and the main interest of the authors of this article, as electrolytes in batteries and capacitors. The range over which a RTIL is liquid is important for almost every industrial application, and ionic liquids have a tendency to supercool by many degrees below the melting temperature. It is therefore necessary to make sure a RTIL is a stable liquid as opposed to a supercooled liquid liable to crystallize. Differential scanning calorimetry (DSC) was used to determine the melting temperature of each ionic liquid. Viscosity is important for many applications where an ionic liquid is in motion, and it is important for electrolytes where ions move toward a charged electrode. The glass transition temperature can be used to predict the dependence of viscosity on temperature. Surface tension may affect the manner in which these electrolytes are adsorbed into the structure of porous electrodes during capacitor manufacture.

Three RTILs were chosen specifically for use as electrolytes in electrochemical double layer capacitors. First, 1-ethyl-3-methylimidazolium dicyanamide [emim][N(CN)$_2$], shown in Figure 1, was selected; unlike many RTILs used as electrolytes, it does not contain fluorine and therefore cannot produce either fluorine or hydrogen fluoride by reaction, degradation, or combustion. This may present a problem in a domestic or commercial situation particularly if large quantities are being used. In addition to safety considerations, hydrogen fluoride and fluorine may damage the materials of construction of a battery or capacitor.

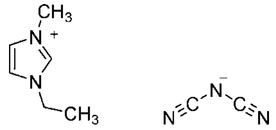

Figure 1. 1-Ethyl-3-methylimidazolium dicyanamide.

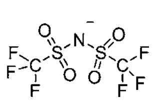

Figure 2. 1,2-Dimethyl-3-propylimidazolium bis(trifluoromethylsulfonyl)imide.

1-Ethyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide was of interest owing to a reported low viscosity, high diffusivity, and relatively high ionic conductivity. This cation−anion combination was found to have the optimum properties for electrolyte use in an extensive series of studies by Tokuda et al.,[1−3] varying anion, cation, and alkyl chain length, and has been patented in the U.S. by Covalent Associates, Inc. However, 1,2,-dimethyl-3-propylimidazolium bis(trifluoromethylsulfonyl)imide [dmpim][Tf$_2$N], shown in Figure 2, was chosen in preference for this study owing to a reported[4] slight increase in anodic voltage potential before breakdown occurs. This is an important advantage for electrolyte use in capacitors as the energy stored is proportional to the square of the voltage applied.

1-butyl-1-methylpyrrolidinium tris(pentafluoroethyl)trifluorophosphate [bmpyr][Pf$_3$PF$_3$], shown in Figure 3, was a new ionic liquid and was selected owing to a reported[5] extremely

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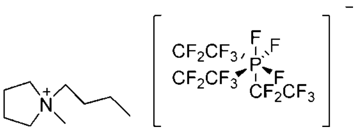

Figure 3. 1-Butyl-1-methylpyrrolidinium tris(pentafluoroethyl)trifluorophosphate.

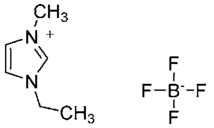

Figure 4. 1-Ethyl-3-methylimidazolium tetrafluoroborate.

large electrochemical window of 6.8 V total, 4.2 V anodic. Note that Figure 3 shows only one of several possible stereoisomers.

Additionally, a fourth ionic liquid, 1-ethyl-3-methylimidazolium tetrafluoroborate [emim][BF_4], shown in Figure 4, was used as a reference fluid to allow comparison with literature values of viscosity and check that the measurement methods reported here were producing credible results. [emim][BF4] has been of interest as an electrolyte in the past but reacts to produce hydrogen fluoride in aqueous environments.[5]

Experimental Section

Chemicals. All ionic liquids used in this study were obtained at the highest purity commercially available at the time; [emim][$N(CN)_2$], Sigma-Aldrich U.K., > 98.5 % purity, water mass fraction $w_{H_2O} \leq 10^{-3}$; [dmpim][Tf_2N] Strem Chemicals U.K., > 99.0 % purity; [bmpyr][Pf_3PF_3], Merck KGaA, Germany, > 99 % purity, $w_{H_2O} \leq 10^{-4}$, halide mass fraction $\leq 10^{-4}$; [emim][BF_4], Sigma-Aldrich U.K., > 99.0 % purity, $w_{H_2O} \leq 2 \cdot 10^{-4}$, $w_{Br^-} \leq 10^{-5}$, $w_{Cl^-} \leq 10^{-5}$, $w_{NO_3^-} \leq 10^{-5}$, $w_{SO_4^{2-}} \leq 10^{-5}$. No independent measurement of water content was made; the manufacturers' data were taken as correct.

Differential Scanning Calorimetry. Samples of RTILs were placed in intermediate sample bottles and sealed under argon inside a drybox. Between (10 and 18) mg of RTIL was removed from the intermediate sample container and immediately placed inside an aluminum DSC crucible. The crucible was weighed; a lid with two small perforations to allow water vapor to evaporate was crimped in place, and thermal analysis commenced as quickly as possible. Thermal analysis was carried out between (123 and 403.15) K at a scan rate of 10 $K \cdot min^{-1}$ using a nitrogen flushed Mettler Toledo DSC 30 calibrated with a standard sample of indium. RTIL samples were dried inside the DSC chamber in a similar manner to Crosthwaite et al.[6] and Fredlake et al.[7] by holding at 403.15 K until the temperature of any phase transitions observed remained constant. For most samples this occurred after three hours at 403.15 K. All measurements were repeated in triplicate. Although the DSC measured temperature to an uncertainty of ± 0.01 K, reproducibility of phase transitions for RTILs can be poor; therefore, temperatures are only reported to the nearest 1 K.

Rheometry. Viscosity measurements were made using a TA Instruments AR1000-N Rheolyst rheometer with 0.02 m diameter stainless steel parallel plates set at a gap of 200 μm. Shear stress was measured for shear rates between (10 and 1000) s^{-1} at a temperature of 298.15 K. Shear stress was then measured at a constant shear rate of 100 s^{-1} for temperatures from (288.15 to 368.15) K at a ramp rate of 6 $K \cdot min^{-1}$. The estimated uncertainty of viscosity measurements for the range of shear rates used was ± 0.0005 $Pa \cdot s$. All measurements were carried out in triplicate. Samples were sealed under argon inside a drybox, opened, and rapidly transferred to the plates of the rheometer. Measurements were commenced as quickly as possible to minimize water absorption. The size of gap between the plates only allowed a small specific surface area for moisture absorption from the atmosphere. Reproducibility between the first and the third measurements was good, indicating only slow water absorption into the sample.

Table 1. Melting, T_m, Freezing, T_{fus}, Crystallization, T_{cr}, and Glass Transition, T_g, Temperatures

	T_m	T_m lit.	T_{fus}	T_{cr}	T_g	T_g lit.
RTIL	K	K	K	K	K	K
[emim][$N(CN)_2$]	255	252[11]	232	214, 229, 245	177	169[11]
[dmpim][Tf_2N]	281	284.44,[15] 288[14]	N/A	231	164	191,[7,14] 192[13]
[bmpyr][Pf_3PF_3]	277		246	233	157	

Surface Tension Measurements. The surface tension measurements were performed using the du Nouy method using a type OS tensiometer torsion balance (White Electrical Instrument Co. Ltd., U.K.) fitted with a platinum ring with a total contact length of 0.08 m. Both the ring and the glass sample dish were cleaned thoroughly with propylene carbonate, acetone, 5 $mol \cdot L^{-1}$ nitric acid, and deionized water and dried overnight in a drying cabinet at 333 K before use. The tensiometer was calibrated using deionized water at 293 K. For the four ionic liquids, all measurements of surface tension were taken inside an argon-filled drybox at a temperature of (293 ± 0.5) K. The uncertainty of measurement for surface tension was estimated by the tensiometer manufacturer to be less than ± $4.8 \cdot 10^{-4}$ $N \cdot m$.

Results and Discussion

Differential Scanning Calorimetry. Ionic liquids are reported[2,6−9] to follow several different patterns of behavior during heating and cooling. Some RTILs cool to form a glass at the glass transition and on heating melt to form a liquid. Other RTILs, when cooled, crystallize then undergo a glass transition at a lower temperature. When heated, some of these RTILs melt to form a liquid; others "cold crystallize" to form a crystal which on further heating melts to form a liquid. Furthermore, certain types of RTILs, particularly those containing the dicyanamide anion, exhibit multiple crystalline phases as discussed by Macfarlane et al.[10,11] Another feature of many RTILs is the ability to supercool below the melting temperature by as much as 50 K or even 200 K, see Crosthwaite et al.,[6] Fredlake et al.,[7] Galiński et al.,[12] McEwen et al.,[13] and Ngo et al.[14] Therefore, it is important to ensure that a novel RTIL is a thermodynamically stable liquid over the range of temperatures of use and not a supercooled liquid liable to crystallize. Table 1 summarizes the DSC results for the three RTILs tested.

Glass transition temperatures were measured as the inflection point of the change in the DSC trace. Melting temperatures, freezing temperatures, and cold crystallization onset temperatures were calculated using Graphware TA72 thermal analysis software from Mettler-Toledo AG.

Figure 5 shows the DSC heating trace for [emim][$N(CN)_2$] with a glass transition, three cold crystallization exothermic peaks, and a melting trough. The melting temperature for [emim][$N(CN)_2$] is slightly higher than the value reported by MacFarlane et al.[11] The melting temperature is difficult to determine for [emim][$N(CN)_2$] as the third cold crystallization peak overlaps with the onset of melting, shown in Figure 5. MacFarlane et al.[11] also reported multiple crystalline states. The glass transition temperature was 3 K higher than the value reported by MacFarlane.[11]

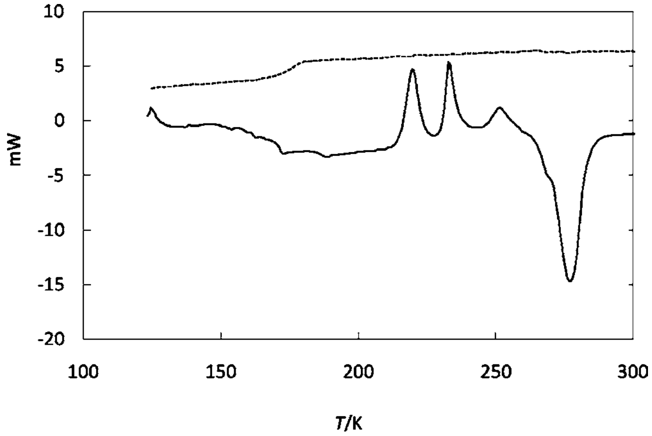

Figure 5. DSC trace for 1-ethyl-3-methylimidazolium dicyanamide. Solid line, heating; dashed line, cooling.

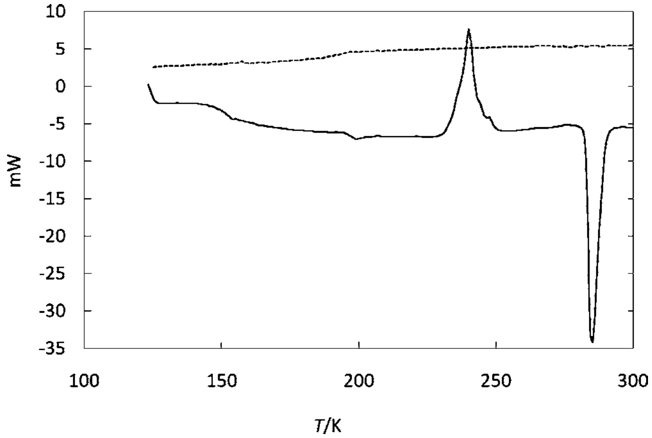

Figure 6. DSC trace for 1,2-dimethyl-3-propylimidazolium bis(trifluoromethylsulfonyl)imide. Solid lines, heating; dashed line, cooling.

[dmpim][Tf_2N] appeared to undergo a slow glass transition over a comparatively large temperature range, as shown in Figure 6. The glass transition is lower than the reported literature values from Fredlake et al.,[7] Ngo et al.,[14] and McEwen et al.[13] The melting temperature is 3 K lower than the value given by van Valkenberg et al.[15] and 7 K lower than the value reported by Ngo et al.[14] [dmpim][Tf_2N] crystallized during some but not all DSC measurements; interestingly, Fredlake et al.[7] found [dmpim][Tf_2N] did not crystallize but cooled to form a glass which reformed as a liquid on heating. A slight increase in water content has been reported by Fredlake et al.,[7] to produce crystalline structures in RTILs which in drier samples are either not present or produce much smaller exothermic peaks. This was confirmed by a few DSC runs with samples that had been left exposed to the atmosphere and samples that were dried in the DSC for an insufficient time at 403.15 K. However, this would not explain the appearance, disappearance, and reappearance of crystals that occurred for repeat DSC heating and cooling cycles with the same sample continuously flushed with dry nitrogen. Possibly very slight differences in the temperature history of the RTIL, for example, a very small difference in the heating/cooling rate or the time the sample was held to equilibrate at 123.15 K before the heating scan commenced, may determine whether crystallization occurs. Another possibility is that the process is stochastic in origin.

[bmpyr][Pf_3PF_3], as shown in Figure 7, had a freezing temperature of 246 K, glass transition temperature of 157 K, cold crystallization temperature of 233 K, and melting temperature of 277 K. The freezing temperature below the melting temperature shows an ability to supercool often displayed by ionic liquids;, see Fredlake et al.,[7] Galiński et al.,[12] McEwen et al.,[13] and Ngo et al.[14] This is a newly available ionic liquid, and no literature values could be found for comparison.

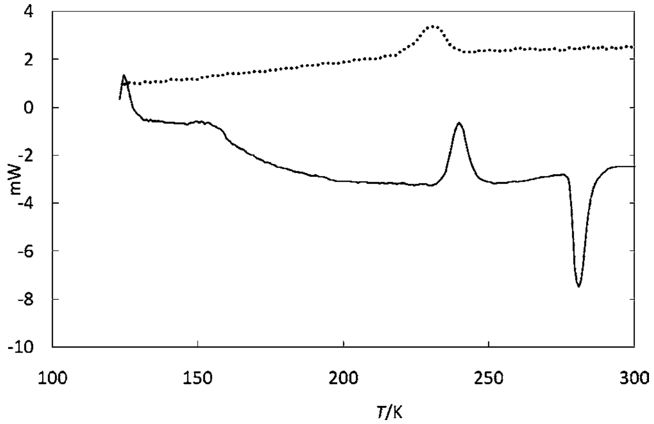

Figure 7. DSC trace for heating 1-butyl-1-methylpyrrolidinium tris(pentafluoroethyl) trifluorophosphate. Solid line, heating; dashed line, cooling.

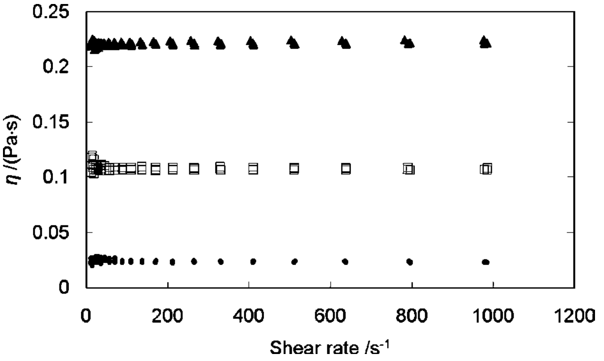

Figure 8. Viscosity versus shear rate: ▲, [bmpyr][Pf_3PF_3]; □, [dmpim]-[Tf_2N]; ●, [emim][$N(CN)_2$].

Viscometry. Viscosity measurements at 298 K for shear rates between (10 and 1000) s^{-1} are shown in Figure 8 and Table 3. Each of the three RTILs exhibited Newtonian behavior with no dependence of viscosity on shear rate. The pyrrolidinium salt [bmpyr][Pf_3PF_3] was the most viscous followed by [dmpim][Tf_2N] followed by [emim][$N(CN)_2$]. For comparison, [emim][BF4] at 298 K had a viscosity of 0.037 Pa·s in good agreement with the values given by McEwen et al.,[13] van Valkenburg et al.,[15] Nishida et al.,[16] and Noda et al.[17]

The viscosity measured for [dmpim][Tf_2N] is in agreement with the value reported by van Valkenburg et al.[15] but higher than the value measured at (299 ± 1) K by McEwen et al.[13] Discrepancies in viscosity values for ionic liquids can usually be explained by the presence of impurities, particularly water, which is readily absorbed by most RTILs. Even very small quantities of water can reduce the viscosity quite dramatically; Van Valkenburg et al.,[15] Widegren et al.,[18] Seddon et al.,[19] and McEwen et al.[13] however, reported a water mass fraction of $< 20 \cdot 10^{-5}$ for their sample of [dmpim][Tf_2N] and performed viscosity measurements with a cone and plate viscometer under a N_2 atmosphere. This would tend to increase rather than decrease the viscosity and suggests differences in impurities other than water between samples.

The measured viscosity for [emim][$N(CN)_2$] is in good agreement with the value reported at 298 K by both MacFarlane et al.[11] and Leong et al.[20]

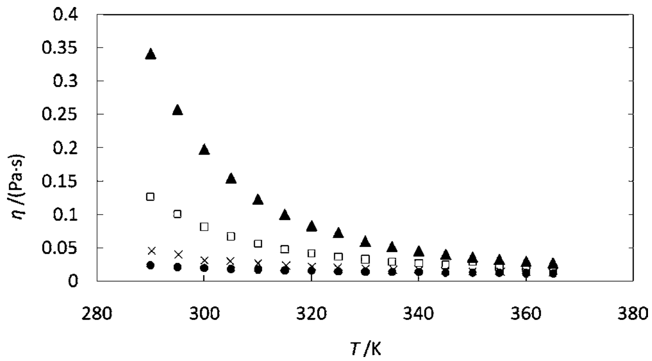

Figure 9. Viscosity versus temperature: ▲, [bmpyr][Pf$_3$PF$_3$]; □, [dmpim]-[Tf$_2$N]; x, [emim][BF$_4$]; ●, [emim][N(CN)$_2$].

Table 2. Viscosity, *η*, for Each Ionic Liquid at 298 K

	η	*η* lit.
	Pa·s	Pa·s
[emim][N(CN)$_2$]	0.021	0.021[11,20]
[dmpim][Tf$_2$N]	0.09	0.06,[13] 0.09005[15]
[bmpyr][Pf$_3$PF$_3$]	0.221	
[emim][BF$_4$]	0.037	0.032,[17] 0.03607,[15] 0.037,[16] 0.043[13]

Table 3. Viscosity, *η*, for Each Ionic Liquid at (290 to 365) K

	[emim][N(CN)$_2$]	[dmpim][Tf$_2$N]	[bmpyr][Pf$_3$PF$_3$]	[emim][BF$_4$]
T	*η*	*η*	*η*	*η*
K	Pa·s	Pa·s	Pa·s	Pa·s
290	0.024	0.127	0.355	0.046
295	0.022	0.101	0.272	0.041
300	0.020	0.082	0.209	0.032
305	0.018	0.067	0.163	0.031
310	0.017	0.056	0.129	0.027
315	0.016	0.048	0.105	0.024
320	0.015	0.042	0.087	0.022
325	0.015	0.036	0.072	0.021
330	0.014	0.032	0.062	0.019
335	0.014	0.029	0.053	0.018
340	0.013	0.027	0.047	0.018
345	0.013	0.024	0.041	0.016
350	0.012	0.029	0.037	0.015
355	0.012	0.021	0.034	0.015
360	0.012	0.020	0.030	0.015
365	0.012	0.019	0.028	0.013

Table 4. Surface Tension Measurements, *τ*, for Each Ionic Liquid at (293 ± 0.5) K

	τ	*τ* lit.
	N·m^{-1}	N·m^{-1}
[emim][N(CN)$_2$]	0.064	0.0436,[23] 0.0426[a,24]
[dmpim][Tf$_2$N]	0.041	
[emim][Tf$_2$N]		0.0352,[a,24] 0.04162[25]
[bmpyr][Pf$_3$PF$_3$]	0.038	
[emim][BF$_4$]	0.053	0.0544,[b,22] 0.0467[a,24]

[a] Room temperature, (295 to 298) K. [b] 298.15 K.

There are currently no published viscosity measurements for [bmpyr][Pf$_3$PF$_3$]. Pyrrolidinium salts are, however, known to be considerably more viscous than the imidazolium equivalents,[3] and the value of 0.221 Pa·s is within the expected range of viscosities for this type of RTIL.

The effect of temperature on viscosity is shown in Figure 9 and summarized in Table 2. As the liquids were Newtonian, it was legitimate to use a constant shear rate, rather than constant shear stress, for the temperature sweep. For each RTIL, viscosity decreased rapidly with increasing temperature, although the order of viscosities from least viscous to most viscous remained unchanged. Attempts were made to fit viscosity versus temperature data, which were fitted to both the Arrhenius and the Williams−Landel−Ferry[21] (WLF) equations for each RTIL.

$$\eta = \eta_g \exp[-C_1(T - T_g)/(C_2 + (T - T_g))] \tag{1}$$

where η_g is the viscosity at the glass transition, C_1 and C_2 are the WLF constants, T is temperature, and T_g is the glass transition temperature. Both the Arrhenius and the WLF equations, however, failed to provide a satisfactory fit over this range of temperatures. Traditionally, the WLF equation only applies to substances at temperatures less than 100 K above the glass transition point, and the range of temperatures used was considerably higher than this. A simple Arrhenius equation only provided an acceptable fit over a restricted range of temperatures.

Surface Tension. Surface tension measurements are shown in Table 4. The surface tension measured for [emim][BF4] is in reasonable agreement with the value reported by Zhou et al.[22] but higher than the value reported by Martino et al.[24] Martino et al.,[24] however, recorded measurements at a slightly higher temperature, from (295 to 298) K.

The surface tension for [emim][N(CN)$_2$] was higher than the values reported by Froba et al.[23] and Martino et al.[24] Surface tension data for RTILs are relatively scarce, and no literature values could be found for [dmpim][Tf$_2$N] or [bmpyr][Pf$_3$PF$_3$]. Literature values[24,25] for [emim][Tf$_2$N] are included in Table 4 for comparison and are not dissimilar to the measured value for the related [dmpim][Tf$_2$N].

Conclusions

The heating and cooling phase change behavior for each RTIL for the temperature range from (123.15 to 403.15) K was measured. All three RTILs tested had melting points below 285 K, allowing for their use as electrolytes in electrochemical double layer capacitors and other ambient temperature applications.

Viscosity was measured over a range of shear rates at 298 K and over a range of temperatures from (288 to 368) K at a shear rate of 100 s^{-1}. All RTILs were found to be Newtonian with viscosities that decreased with temperature. [emim][N(CN)$_2$] was the least viscous followed by [emim][BF4], followed by [dmpim][Tf$_2$N], and followed by [bmpyr][Pf$_3$PF$_3$]. The high viscosity of the pyrrolidinium ionic liquid may adversely affect the performance of an electrochemical capacitor. For [bmpyr][Pf$_3$PF$_3$] these are previously unreported data.

Surface tensions are reported for (293 ± 0.5) K and appear to be previously unreported for two of the four RTILs tested. It is anticipated that surface tension may affect the performance of electrochemical capacitors when porous electrodes are used.

Acknowledgment

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