# Supporting Information

## Evaluating the Contributions to Conductivity in Room Temperature Ionic Liquids

Emily Simonis and G. J. Blanchard*
Michigan State University
Department of Chemistry
578 S. Shaw Lane, East Lansing, MI 48824 USA

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* Author to whom correspondence should be addressed. Email: blanchard@chemistry.msu.edu, Tel: +1 517 303 1105.

**1.**

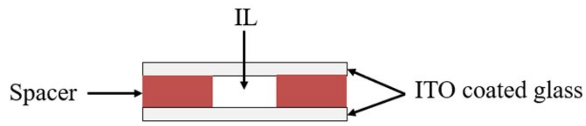


**Figure S1.** Diagram of the sample cell design used for TCSPC measurements.

**2.**

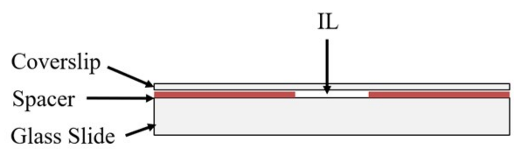


**Figure S2.** Diagram of sample cell design for FRAP measurements.

## 3. $D_T$ values determined from $D_R$ data for CV as a function of temperature.

**Table S1**. Average values of $D_R$ for CV and $D_T$ values for the IL ions. Uncertainties in $D_T$ values are ± 5%.

| Temp (K) | CV $D_R$ (1/ps) | BMIM $D_T$ (Å$^2$/ps) | TFSI $D_T$ (Å$^2$/ps) | Λ (S cm$^2$/mol) |
|---|---|---|---|---|
| 291.27 | 3.12E-05 | 6.70E-04 | 6.28E-04 | 4.99E-01 |
| 291.89 | 3.17E-05 | 6.81E-04 | 6.38E-04 | 5.06E-01 |
| 292.77 | 3.42E-05 | 7.34E-04 | 6.88E-04 | 5.44E-01 |
| 297.56 | 4.48E-05 | 9.62E-04 | 9.02E-04 | 7.01E-01 |
| 312.75 | 9.97E-05 | 2.14E-03 | 2.01E-03 | 1.49 |

## 4. Reaction Dominant Model

Three replicates were taken at spot sizes (diameters) of 10 µm, 20 µm, and 30 µm, and the corresponding $k_d$ values were determined by fitting the recovery curves to the reaction dominant model. The reaction dominant model is not an appropriate model for ionic liquids since there exists some $k_d$ dependence on spot size.

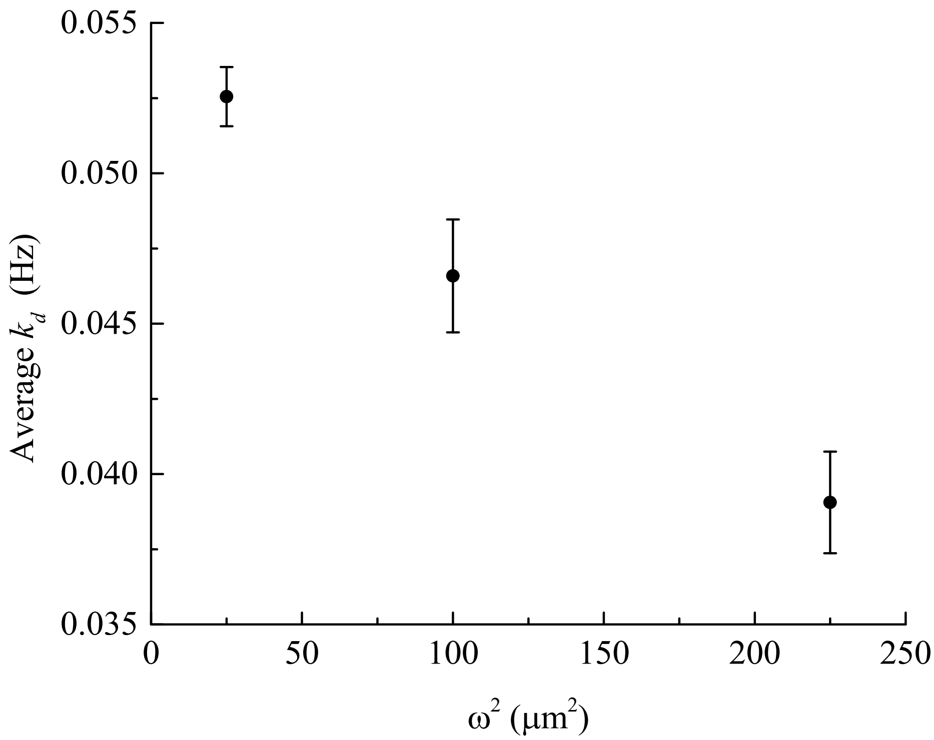


**Figure S3.** Average $k_d$ vs. photobleach spot size, fitted using the reaction dominant model for three RB in BMIM TFSI samples. Error bars represent the standard error of the mean.

## 5. Sample Thickness

In order to evaluate the thickness of this sample cell for FRAP measurement, the cell was made with multiple spacers. The samples were all made using the rhodamine B chromophore and

BMIM TFSI. A second and third spacer was added onto the first spacer and the wells lined up. A sample cell with one, two, or three spacers created a cell with a well that was 0.12 mm, 0.24 mm or 0.36 mm deep respectively. These samples were measured under the same parameters as earlier and results are displayed in Table S2. There is a significant difference between the molar conductivity of a sample with one spacer and a sample with two. But there is no significant difference between a sample with two spacers and a sample with three. These data suggest a boundary between two dimensional dynamics in the diffusion and three-dimensional dynamics.

**Table S2.** Average molar conductivity calculated using the Pd or Frd models for samples with one, two, or three spacers.

| | Pd model (Scm$^2$ / mol) | Frd model (Scm$^2$ / mol) |
|---|---|---|
| One spacer | 0.23 ± 0.012 | 0.39 ± 0.024 |
| Two spacers | 0.28 ± 0.018 | 0.45 ± 0.032 |
| Three spacers | 0.28 ± 0.023 | 0.42 ± 0.029 |

## 6. Capped samples

A solution of 0.1 M dichlorodimethylsilane (Sigma-Aldrich, ≥99.5%) in toluene (J.T. Baker, ACS Reagent) was made and poured into a glass dish with a clean glass slide and coverslip. This was agitated periodically for a total of 15 minutes before removing the glass slide and coverslip. The glass slide and coverslip were rinsed with ethanol and dried under nitrogen before assembling the sample cell with a spacer as before and with the BMIM TFSI and rhodamine B solution. FRAP measurements were carried out as before and results are displayed in Table S2. The average molar conductivity of the samples that were capped, by completing the silyation reaction of the glass, was not significantly different from the average molar conductivity of the samples prepared with untreated glass. This holds true for both the Pd and Frd models calculations. This signifies that the charge of the glass does not effect the ionic liquid charge density gradient on a large enough scale to change the conductivity.

**Table S2.** Average molar conductivity calculated using the Pd or Frd models for the capped samples and untreated samples

| | Pd model ($Scm^2$ / mol) | Frd model ($Scm^2$ / mol) |
|---|---|---|
| Capped | 0.22 ± 0.01 | 0.34 ± 0.018 |
| Untreated | 0.22 ± 0.009 | 0.34 0.022 |

## 7. Water Contamination

Solutions of 1% and 5% water in BMIM TFSI with the rhodamine B chromophore were prepared. Sample cells were assembled as before and with three replicate cells of each water concentration. The values for molar conductivity in the unaltered samples, which are displayed as 0% water in Figure S5, compared to 1% and 5% water content are within the error margins. This indicates that any water contamination from glassware or the air when assembling the sample cells does not significantly alter the molar conductivity.

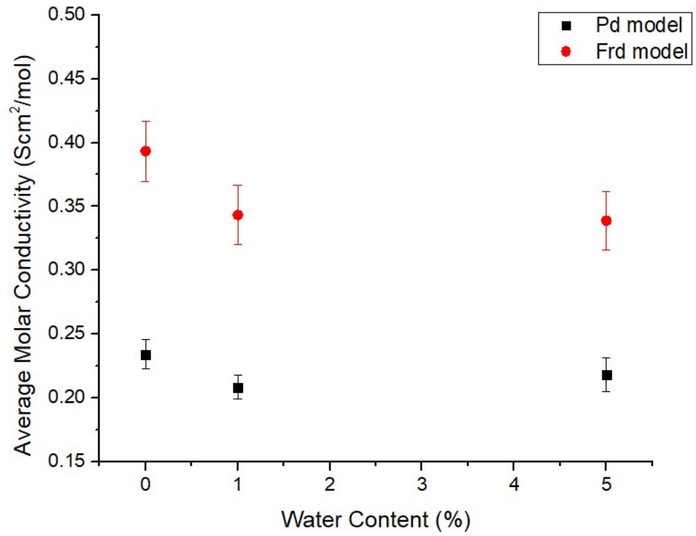


**Figure S5.** Average molar conductivity calculated from FRAP derived diffusion values for BMIM TFSI under different water content amounts: unaltered (0%), 1%, and 5%.