## **Supporting Information**

## Enhanced Ionic Mobility in NanoPorous Silica by Controlled Surface Interactions

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**Figure S1.** 1D <sup>13</sup>C and <sup>1</sup>H NMR spectra of diethyl-methyl-ammonium methanesulfonate (DEMAOMs) and 1-methyl-3-hexyl imidazolium bis(trifluoro methanesulfonate) (C<sub>6</sub>C<sub>1</sub>ImTFSI).

**Table S1.** Silica gel composition for both functionalized and untreated silica with the ionic liquidsDEMAOMs and  $C_6C_1$ ImTFSI.

Pore filling factor (%)	Volume fraction of silica <sup>a</sup> $(\Phi SiO_2)$	Ionic liquid (mL)
25	0.682	0.211
50	0.518	0.422
75	0.417	0.633
100	0.350	0.844
125	0.301	1.055
150	0.264	1.266
200	0.212	1.688

<sup>*a*</sup>The amount of nanoporous silica used to prepare each silica gel is 1 gram

Volume fraction silica ( $\Phi$ SiO2) (further details on these calculations can be found in reference [20])

$$\Phi \text{SiO}_2 = \frac{V_{SiO_2}}{V_{SiO_2} + V_{IL}}$$

 $\rho_{SiO2} = 2.2$  g/cm<sup>3</sup>,  $\rho_{C6CIImTFSI} = 1.372$  g/cm<sup>3</sup> and  $\rho_{DEMAOMs+H2O} = 1.096$  g/cm<sup>3</sup>

Silica particles contain 65% of free space, and 35% of dense silica matrix.  $V_{\text{max}}$ 

 $V_{SiO_2 = 35}$ ,  $V_{IL =>}$   $V_{Free space} = x$  65, where x = % of pore filling Complete filling of ionic liquids in silica accounts to the 100% pore filling.

$$i.e. \Phi SiO_2 = \frac{35}{35 + \frac{x}{100}65}$$
  
e.g. x = 200% pore filling  
$$\frac{35}{35 + \frac{200}{100}65} = 0.21$$



Figure S2. Thermogravimetric analysis showing the mass loss phenomena in gels containing the ionic liquid (at 100% pore filling) diethyl-methyl-ammonium methanesulfonate (DEMAOMs) and 1-methyl-3-hexylimidazolium bis(trifluoromethanesulfonate) ( $C_6C_1$ ImTFSI) for untreated (red) and functionalized (black) silica.



Figure S3. 2D <sup>13</sup>C {<sup>1</sup>H} HETCOR NMR spectra of functionalized nanoporous silica gels with (a)
C<sub>6</sub>C<sub>1</sub>ImTFSI and (b) DEMAOMs ionic liquids collected at a magnetic field of 14.1 T at a MAS rate of 10 kHz. The <sup>1</sup>H MAS (and <sup>1</sup>H projection) and <sup>13</sup>C {<sup>1</sup>H} CP-MAS (and <sup>13</sup>C projection) NMR spectra are shown on left and top of the 2D spectrum, respectively.



**Figure S4.** Raman shift of the S–O stretching mode in the OMs anion as a function of pore filling factor for gels based on untreated (red) and functionalized (black) silica.



Figure S5.  $T_g$  values recorded for the ionic liquid C<sub>6</sub>C<sub>1</sub>ImTFSI in untreated (red diamonds) and functionalized (black circles) gels.