

# **Tough and Stretchable Inorganic/Organic Double Network Ion Gel Containing Gemini-Type Ionic Liquid as a Multiple Hydrogen Bond Cross-Linker**

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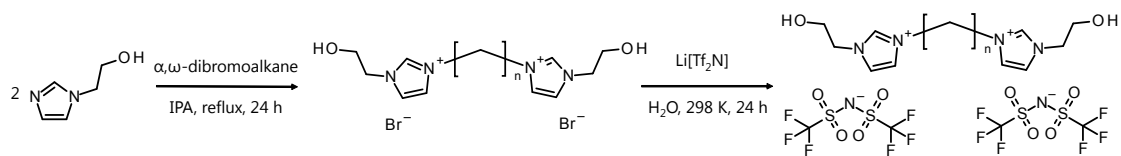
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## **SUPPLEMENTARY INFORMATION**

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## (1) Reaction scheme of synthesis of gemini-type ionic liquids

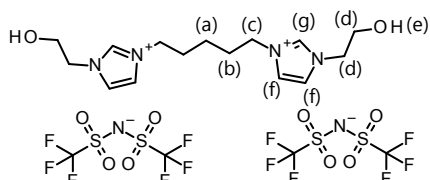


Scheme S1 Reaction scheme of synthesis of gemini-type ionic liquids

## (2) $^1\text{H}$ NMR results of gemini-type ionic liquids

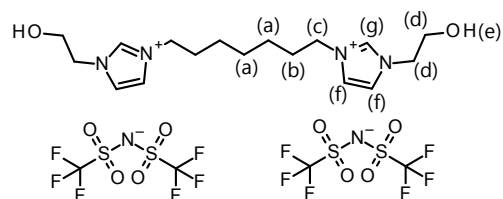
The chemical structures and  $^1\text{H}$  NMR measurement results of the synthesized gemini-type ionic liquids are shown in Fig. S1. The gemini-type ionic liquids were successfully synthesized because we observe that the  $^1\text{H}$  NMR measurement results agreed well with the previous report.<sup>1,2</sup>

### (a) $[\text{C}_5(\text{C}_2\text{OHim})_2][\text{Tf}_2\text{N}]_2$



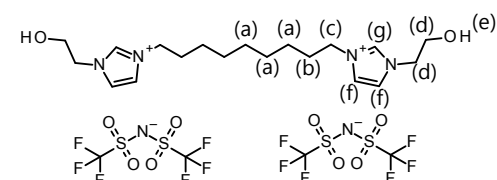
$^1\text{H}$  NMR (400 MHz,  $\text{DMSO-}d_6$ ):  $\delta$  (ppm) 1.23 (m, 2H, (a)), 1.83 (m, 4H, (b)), 3.76 (m, 4H, (c)), 4.19 (m, 8H, (d)), 5.18 (t, 2H, (e)), 7.76 (m, 4H, (f)), 9.13 (s, 2H, (g))

### (b) $[\text{C}_7(\text{C}_2\text{OHim})_2][\text{Tf}_2\text{N}]_2$



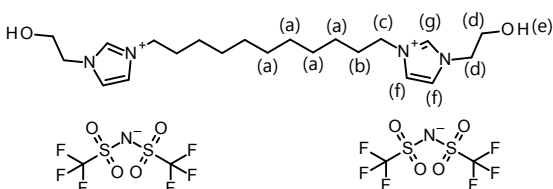
$^1\text{H}$  NMR (400 MHz,  $\text{DMSO-}d_6$ ):  $\delta$  (ppm) 1.28 (m, 6H, (a)), 1.78 (m, 4H, (b)), 3.73 (m, 4H, (c)), 4.19 (m, 8H, (d)), 5.18 (t, 2H, (e)), 7.76 (m, 4H, (f)), 9.13 (s, 2H, (g))

### (c) $[\text{C}_9(\text{C}_2\text{OHim})_2][\text{Tf}_2\text{N}]_2$



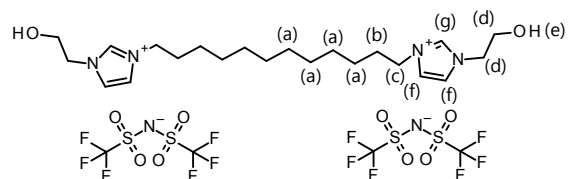
$^1\text{H}$  NMR (400 MHz,  $\text{DMSO-}d_6$ ):  $\delta$  (ppm) 1.26 (m, 14H, (a)), 1.78 (m, 4H, (b)), 3.73 (m, 4H, (c)), 4.19 (m, 8H, (d)), 5.18 (t, 2H, (e)), 7.76 (m, 4H, (f)), 9.13 (s, 2H, (g))

### (d) $[\text{C}_{11}(\text{C}_2\text{OHim})_2][\text{Tf}_2\text{N}]_2$



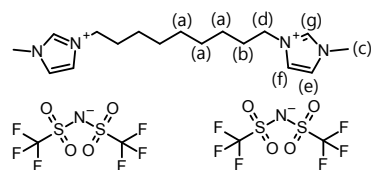
$^1\text{H}$  NMR (400 MHz,  $\text{DMSO-}d_6$ ):  $\delta$  (ppm) 1.27 (m, 10H, (a)), 1.78 (m, 4H, (b)), 3.73 (m, 4H, (c)), 4.18 (m, 8H, (d)), 5.17 (t, 2H, (e)), 7.76 (m, 4H, (f)), 9.13 (s, 2H, (g))

(e)  $[\text{C}_{12}(\text{C}_2\text{OHim})_2][\text{Tf}_2\text{N}]_2$



$^1\text{H}$  NMR (400 MHz,  $\text{DMSO}-d_6$ ):  $\delta$  (ppm) 1.25 (m, 16H, (a)), 1.78 (m, 4H, (b)), 3.73 (m, 4H, (c)), 4.18 (m, 8H, (d)), 5.18 (t, 2H, (e)), 7.76 (m, 4H, (f)), 9.14 (s, 2H, (g))

(f)  $[\text{C}_9(\text{mim})_2][\text{Tf}_2\text{N}]_2$



$^1\text{H}$  NMR (400 MHz,  $\text{DMSO}-d_6$ ):  $\delta$  (ppm) 1.25 (m, 10H, (a)), 1.77 (m, 4H, (b)), 3.84 (s, 6H, (c)), 4.14 (t, 4H, (d)), 7.69 (t, 2H, (e)), 7.75 (t, 2H, (f)), 9.08 (s, 2H, (g))

Fig. S1  $^1\text{H}$  NMR results of the synthesized gemini-type ionic liquids

(3) TEM image of inorganic/organic  $\mu$ -DN ion gel containing  $[\text{C}_9(\text{C}_2\text{OHim})_2][\text{Tf}_2\text{N}]_2$

The TEM image of the inorganic/organic  $\mu$ -DN ion gel containing  $[\text{C}_9(\text{C}_2\text{OHim})_2][\text{Tf}_2\text{N}]_2$  is shown in Fig. S2. The TEM image clearly demonstrated that the silica particle network clusters were formed in the inorganic/organic  $\mu$ -DN ion gel.

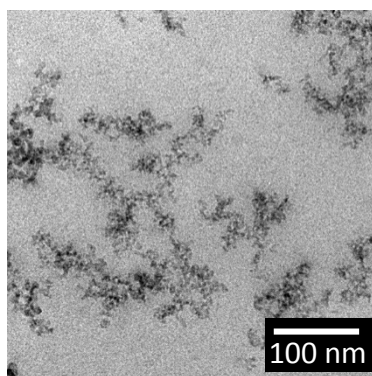


Fig. S2 TEM image of inorganic/organic  $\mu$ -DN ion gel containing  $[\text{C}_9(\text{C}_2\text{OHim})_2][\text{Tf}_2\text{N}]_2$

## References

- 1 T. Payagala, J. Huang, Z. S. Breitbach, P. S. Sharma and D. W. Armstrong, *Chem. Mater.*, 2007, **19**, 5848–5850.
- 2 Jared L. Anderson, Rongfang Ding, A. Arkady Ellern and D. W. Armstrong, *J. Am. Chem. Soc.*, 2004, **127**, 593–604.