Supporting Information

Synthesis, Characterization, and Properties of Ether-Functionalized 1,3-Dialkylimidazolium Ionic Liquids

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1. Synthesis procedure of the ether-functionalized 1,3 –dialkylimidazolium ILs

1.1. Synthesis of ether-substituted imidazoles

1.1.1. 1-methoxymethylimidazole (IM1o1)

100 mL dichloromethane was used to dissolve imidazole (13.2 g, 0.2 mol) in a 500 mL flask, and then chloromethyl methyl ether (17.7 g, 0.22 mmol) was added dropwise in icebath. After stirring for 4 hours, the solvent was removed by rotary evaporation at room temperature. The white crude product was washed with 10 mL ethanol / 100 mL diethyl ether for 3 times. The solid product was dissolved in ethanol and then reacted with triethylamine (20.2 g, 0.2 mol) in a 100 mL autoclave at 85 ^{o}C for 24 hours. The mixture in the autoclave was washed with 150 mL diethyl ether amd the solid salts were removed through filtration. After the diethyl ether was removed, 1-methoxymethylimidazole (IM1o1) was purified via reduced pressure distillation with a 15 cm Vigreux column and the product was collected at 105-107 ^{o}C when the pressure was about 5 Pa. Colorless liquid (yield: 55 %). ^{1}H NMR (400 MHz, $CDCl_3$): δ (ppm) 7.62-7.52 (s, 1H), 7.11-7.04 (d, 1H), 7.03-6.95 (d, 1H), 5.26-5.15 (s, 2H), 3.30-3.15 (s, 3H).

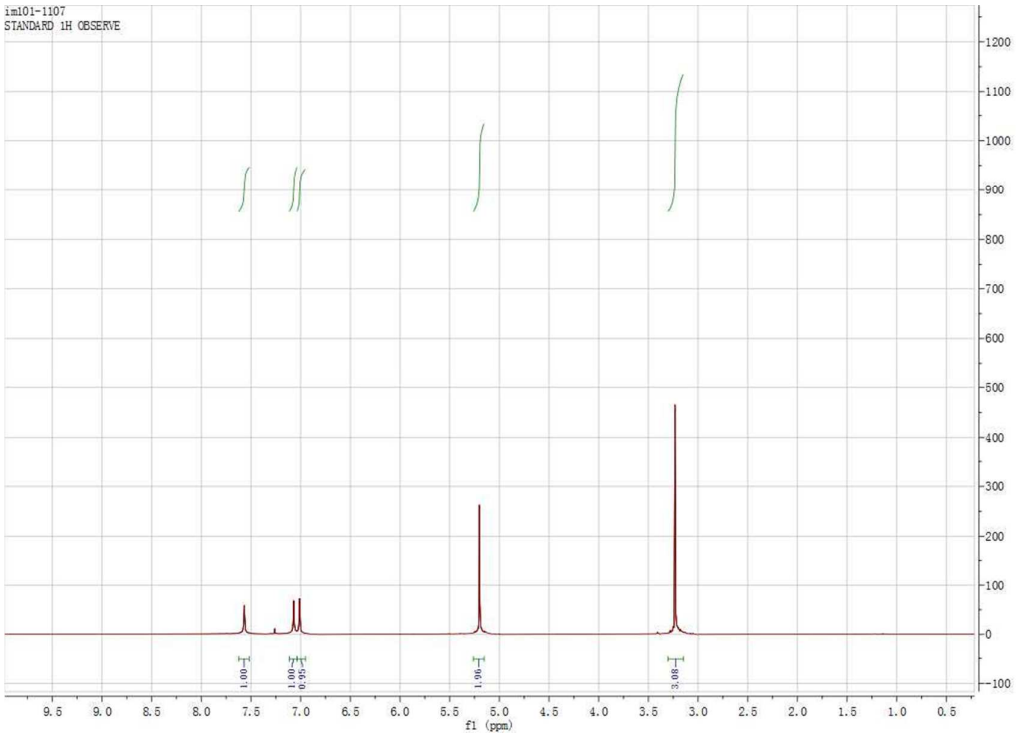

Fig. S1 ^{1}H NMR spectrum of 1-methoxymethylimidazole.

1.1.2. 1-ethoxymethylimidazole (IM1o2)

Imidazole (13.2 g, 0.2 mol) was dissolved in 100 mL dichloromethane in a 500 mL flask, and chloromethyl ethyl ether (20.8 g, 0.22 mol) was added dropwise to the solution at 0 ^{o}C. The following steps were similar with the synthesis of 1-methoxymethylimidazole. The product was collected at about 110-112 ^{o}C when the pressure is 5 Pa.Colorless liquid (yield: 60 %). ^{1}H NMR (400 MHz, $CDCl_3$): δ (ppm) 7.60-7.55 (s, 1H), 7.10-7.06 (d, 1H), 7.04-7.00 (d, 1H), 5.27-5.25 (s, 2H), 3.46-3.37(m, 2H), 1.18-1.11 (t, 3H).

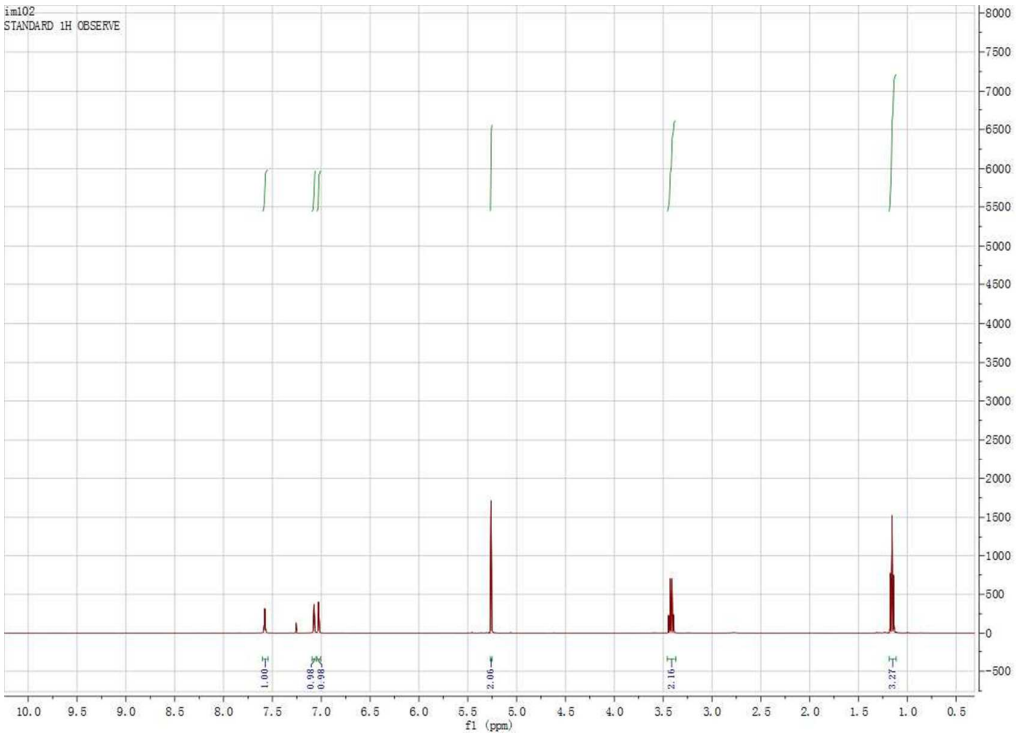

Fig. S2 ^{1}H NMR spectrum of 1-ethoxymethylimidazole.

1.1.3. 1-(2-methoxyethyl) imidazole (IM2o1)

The synthesis procedure of IM2o1 had been described in the Experimental part of the paper. Colorless liquid (yield: 80 %). ^{1}H NMR (400 MHz, $CDCl_3$): δ (ppm) 7.53-7.50 (s, 1H), 7.05-7.02 (d, 1H), 6.97-6.95 (d, 1H), 4.10-4.06 (t, 2H), 3.63-3.59 (t, 2H), 3.33-3.31 (s, 3H).

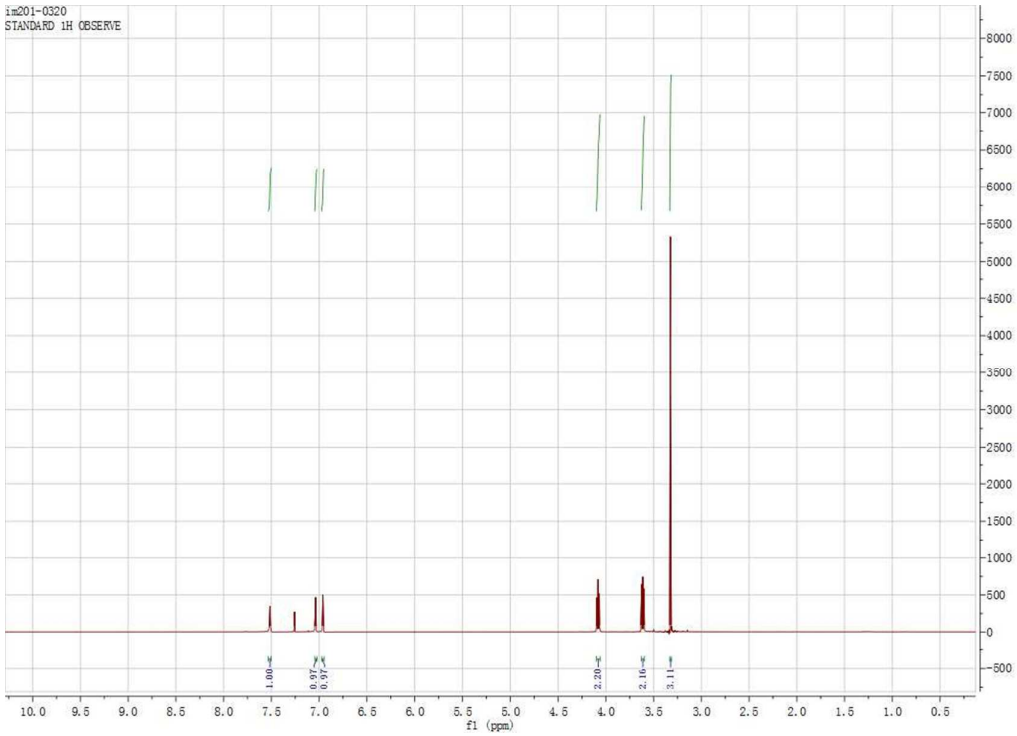

Fig. S3 ^{1}H NMR spectrum of 1-(2-methoxyethyl) imidazole.

1.1.4. 1-(2-methoxymethyl) imidazole (IM2o2)

Imidazole (10.2 g, 0.15 mol), powdered potassium hydroxide (10.08 g, 0.18 mol) and tetrabutylammonium bromide (TBAB, 3.22 g, 0.01 mol) were mixed together and stirred at 40 ^{o}C for 30 min. The following steps were similar with the synthesis of 1-(2-methoxyethyl) imidazole. The product was collected at 130-132 ^{o}C when the pressure is about 5 Pa. Colorless liquid (yield: 75 %). ^{1}H NMR (400 MHz, $CDCl_3$): δ (ppm) 7.51-7.48 (s, 1H), 7.03-6.99 (d, 1H), 6.98-6.92 (d, 1H), 4.09-4.03 (t, 2H), 3.67-3.60 (t, 2H), 3.47-3.40 (m, 2H), 1.19-1.12 (t, 3H).

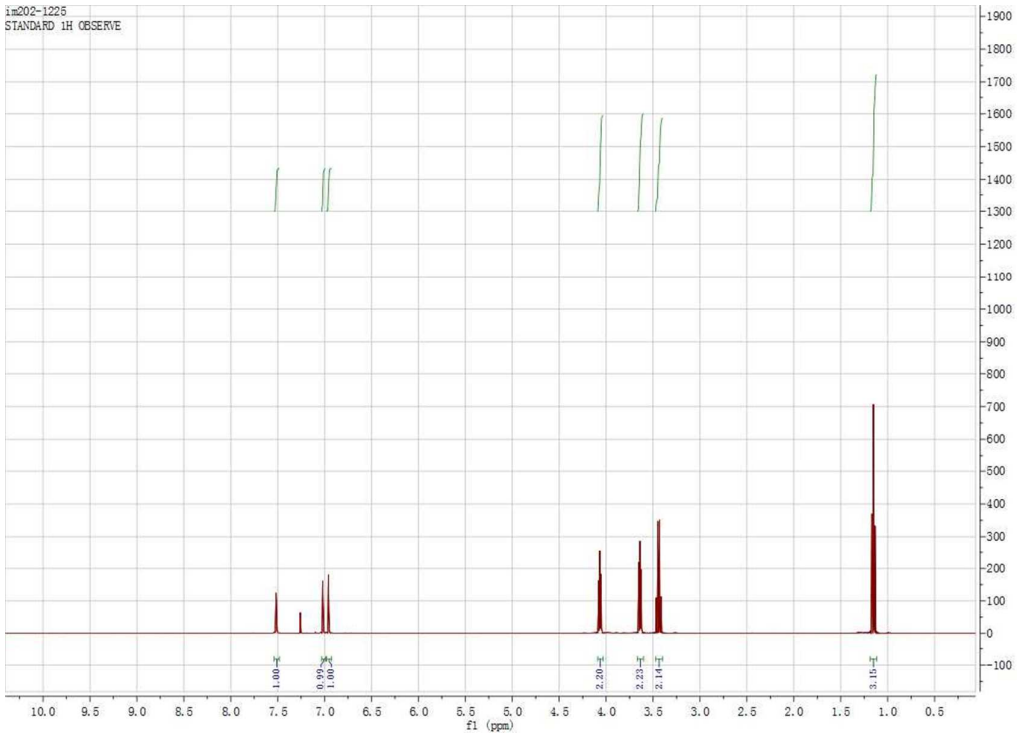

Fig. S4 ^{1}H NMR spectrum of 1-(2-methoxymethyl) imidazole.

1. 2. Synthesis of the 1, 3-dialkylimidazolium ILs with a 1oR group

1.2.1. 1-methoxymethyl-3-methylimidazolium bis(trifluoromethanesulfonyl)imide (IM1o1-1-TFSA)

IM1o1 (5.0 g, 0.045 mol) and iodomethane (7 g, 0.05 mol) were reacted in a 250 mL flask at 0 ^{o}C for 24 h with acetonitrile (8 mL) as the solvent. The product was washed with diethyl ether (100 mL *3). Then the same molar amount of LiTFSA and the iodate were mixed together in the deionized water, and stirred for 24 h at room temperature. The crude IL was extracted by dichloromethane and washed with deionized water for 3 times until no halide residual could be detected by $AgNO_3$ solution. The solvent was removed through rotary evaporation. The IL was dried under high vacuum for 48 h at 100 ^{o}C. Colorless liquid (yield: 82 %). ^{1}H NMR (400 MHz, acetone-d_3): δ (ppm) 9.26-9.19 (s, 1H), 7.89-7.85 (d, 1H), 7.83-7.78 (d, 2H), 5.76-5.66 (s, 2H), 4.17-4.09 (s, 3H), 3.49-3.42 (s, 3H).

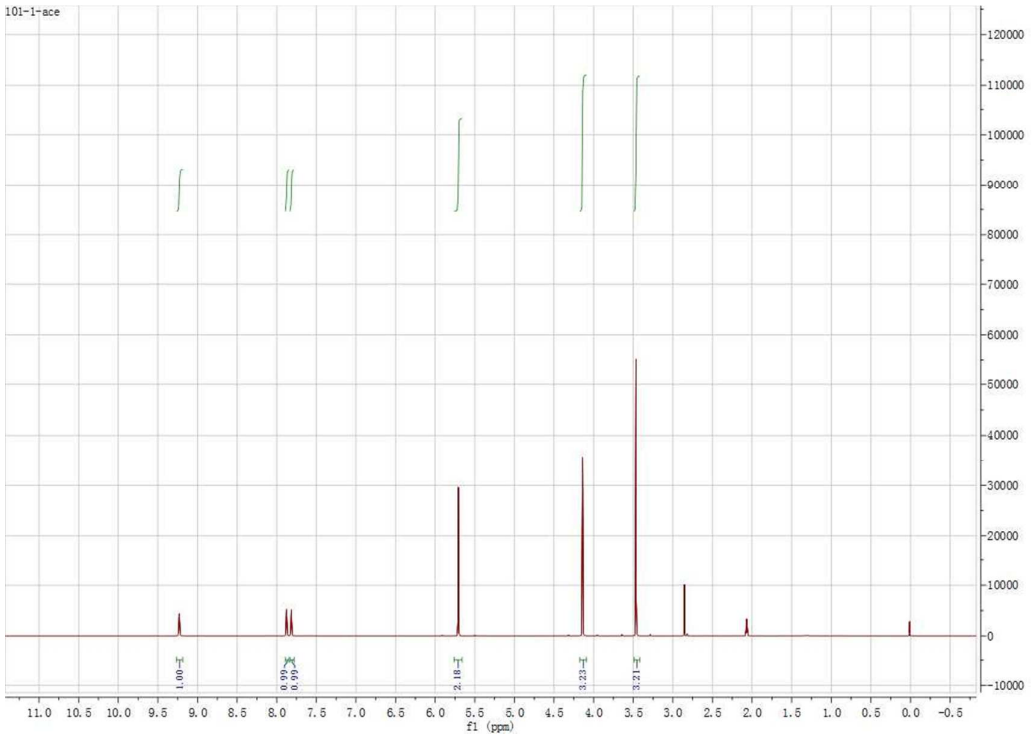

Fig. S5 ^{1}H NMR spectrum of IM1o1-1-TFSA.

1.2.2. 1-methoxymethyl-3-ethylimidazolium bis(trifluoromethanesulfonyl)imide (IM1o1-2-TFSA)

IM1o1 (5.0 g, 0.045 mol) and bromoethane (9.8 g, 0.09 mol) were reacted in a 250 mL flask at 30 ^{o}C for 48 h with acetonitrile (8 mL) as the solvent. The product was washed with diethyl ether (100 mL *3). Then the crude bromine was washed by active carbon with ethanol (100 mL) as the solvent. After the active carbon and ethanol were removed, the bromine was mixed with LiTFSA (same molar amount) and stirred for 24 h. Dichloromethane was used to extract the IL from the mixture and then the solvent was washed with deionized water for 3 times until no halide residual could be detected by $AgNO_3$ solution. The dichloromethane was removed by rotary evaporation. The IL was dried under high vacuum for 48 h at 100 ^{o}C. Colorless liquid (yield: 74 %). ^{1}H NMR (400 MHz, $CDCl_3$): δ (ppm) 8.98-8.93 (s, 1H), 7.47-7.42 (d, 1H), 7.42-7.39 (d, 1H), 5.52-5.45 (s, 2H), 4.34 4.25 (m, 2H), 3.46-3.37 (s, 3H), 1.62-1.53 (t, 3H).

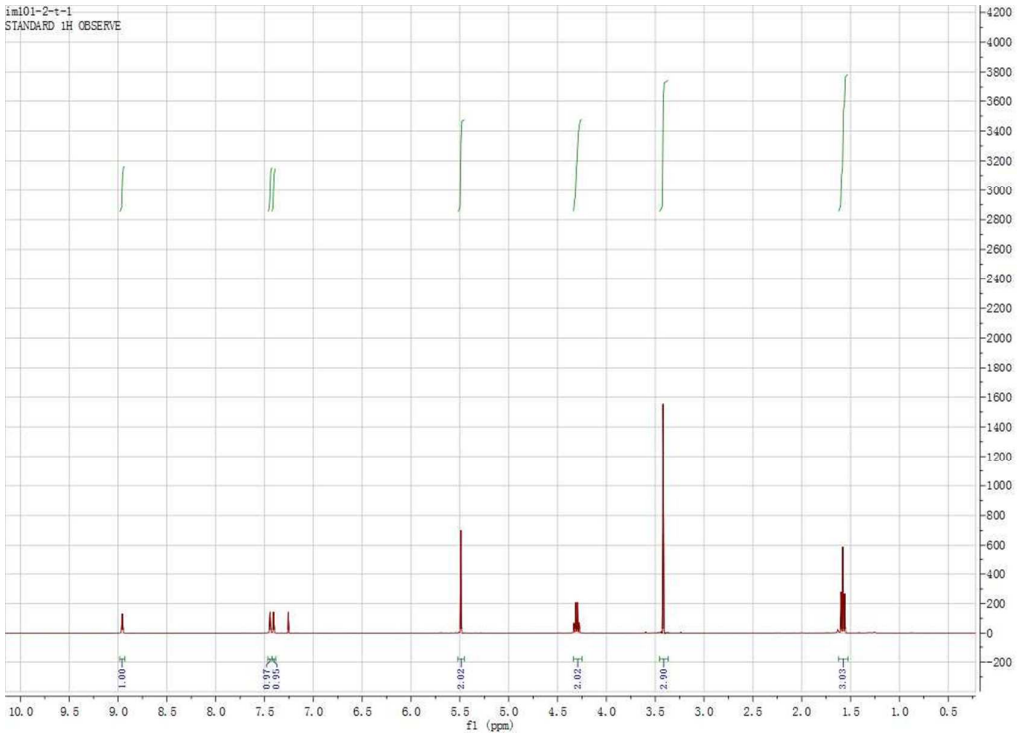

Fig. S6 ^{1}H NMR spectrum of IM1o1-2-TFSA.

1.2.3. 1-methoxymethyl-3-propylimidazolium bis(trifluoromethanesulfonyl)imide (IM1o1-3-TFSA)

IM1o1 (5 g, 0.045 mol) and 1-bromopropane (11.1 g, 0.09 mol) were reacted in a 250 mL flask at 60 ^{o}C for 48 h under argon atmosphere with acetonitrile (8 mL) as the solvent. The following steps were similar with IM1o1-2-TFSA. Colorless liquid (yield: 78 %). ^{1}H NMR (400 MHz, $CDCl_3$): δ (ppm) 9.00-8.93 (s, 1H), 7.48-7.43 (d, 1H), 7.42-7.34 (d, 1H), 5.55-5.39 (s, 2H), 4.24-4.14 (t, 2H), 3.47-3.33 (s, 3H), 2.00-1.84 (m, 2H), 1.05-0.90 (t, 3H).

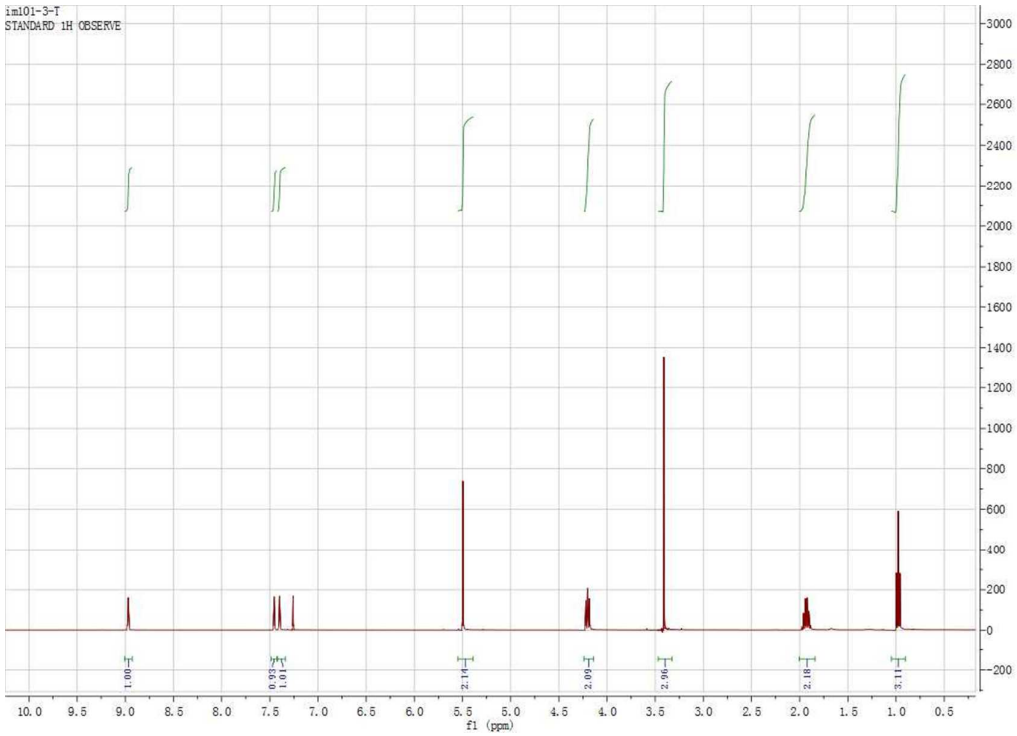

Fig. S7 ^{1}H NMR spectrum of IM1o1-3-TFSA.

1.2.4. 1-methoxymethyl-3-butylimidazolium bis(trifluoromethanesulfonyl)imide (IM1o1-4-TFSA)

IM1o1 (5 g, 0.045 mol) and 1-bromobutane (12.3 g, 0.09 mol) were reacted in a 250 mL flask at 60 ^{o}C for 48 h under argon atmosphere with acetonitrile (8 mL) as the solvent. The following steps were similar with IM1o1-2-TFSA. Colorless liquid (yield: 75 %). ^{1}H NMR (400 MHz, $CDCl_3$): δ (ppm) 8.98-8.91 (s, 1H), 7.47-7.42 (d, 1H), 7.42-7.37 (d, 1H), 5.51-5.45 (s, 2H), 4.26-4.18 (t, 2H), 3.43-3.35 (s, 3H), 1.92-1.82 (m, 2H), 1.42-1.31 (m, 2H), 0.99-0.92 (t, 3H).

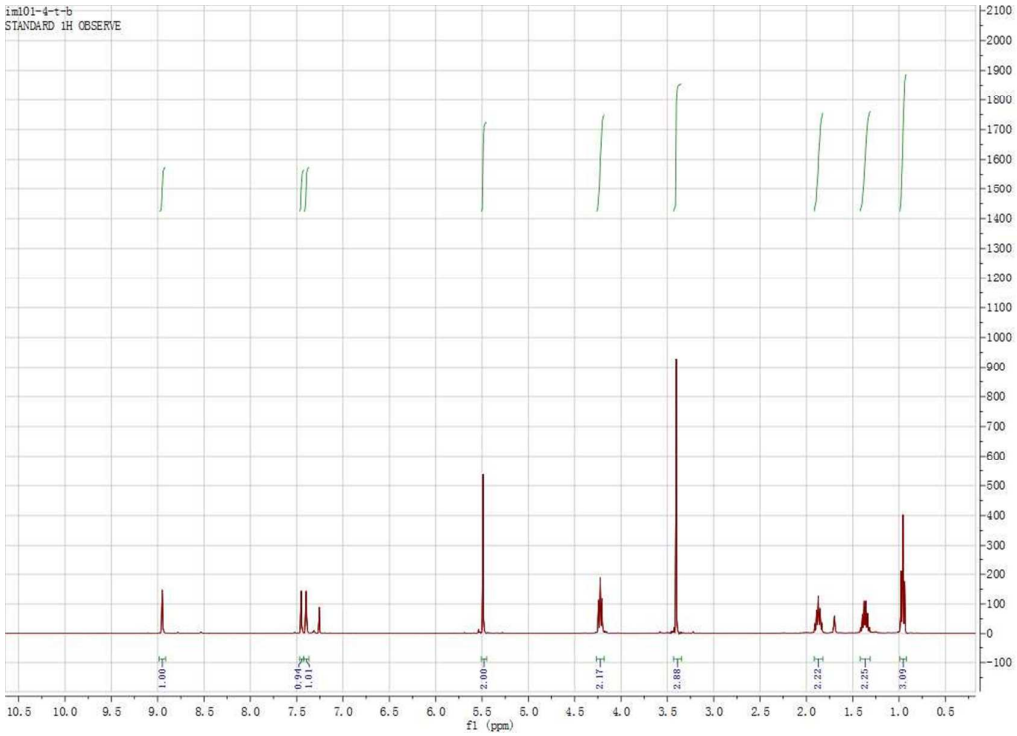

Fig. S8 ^{1}H NMR spectrum of IM1o1-4-TFSA.

1.2.5. 1-methoxymethyl-3-(2-ethoxymethyl)imidazolium bis(trifluoromethanesulfonyl)imide (IM1o1-2o1-TFSA)

IM1o1 (5 g, 0.045 mol) and 2-methoxyethyl bromide (7.5 g, 0.054 mol) were reacted in a 250 mL flask at 60 ^{o}C for 48 h under argon atmosphere with acetonitrile (8 mL) as the solvent. The following steps were similar with IM1o1-2-TFSA. Colorless liquid (yield: 62 %). ^{1}H NMR (400 MHz, $CDCl_3$): δ (ppm) 8.91-8.85 (s, 1H), 7.53-7.48 (d, 1H), 7.46-7.42 (d, 1H), 5.53-5.46 (s, 2H), 4.43-4.38 (t, 2H), 3.76-3.72 (t, 2H), 3.44-3.41 (s, 3H), 3.38-3.39 (s, 3H).

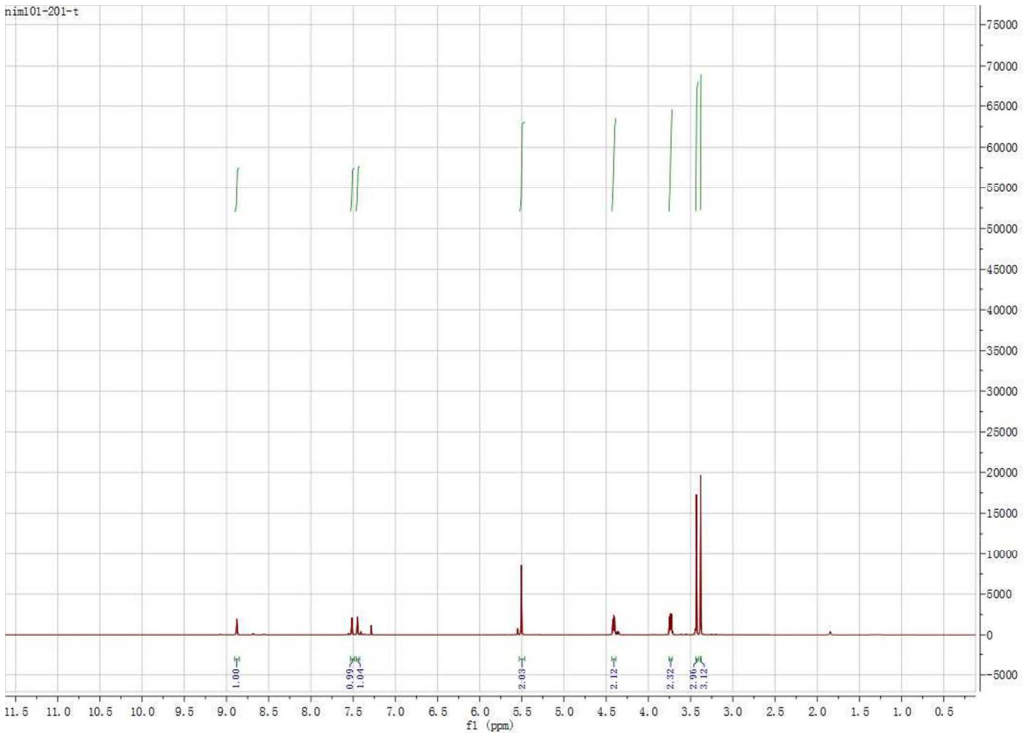

Fig. S9 ^{1}H NMR spectrum of IM1o1-2o1-TFSA.

1.2.6. 1-methoxymethyl-3-(2-ethoxyethyl)imidazolium bis(trifluoromethanesulfonyl)imide (IM1o1-2o2-TFSA)

IM1o1 (5 g, 0.045 mol) and 2-ethoxyethyl bromide (8.3 g, 0.054 mol) were reacted in a 250 mL flask at 60 ^{o}C for 48 h under argon atmosphere with acetonitrile (8 mL) as the solvent. The following steps were similar with IM1o1-2-TFSA. Colorless liquid (yield: 60 %). ^{1}H NMR (400 MHz, $CDCl_3$): δ (ppm) 8.92-8.83 (s, 1H), 7.54-7.51 (d, 1H), 7.48-7.43 (d, 1H), 5.52-5.48 (s, 2H), 4.43-4.38 (t, 2H), 3.79-3.75 (t, 2H), 3.55-3.49 (m, 2H), 3.44-3.42 (s, 3H), 1.19-1.15 (t, 3H).

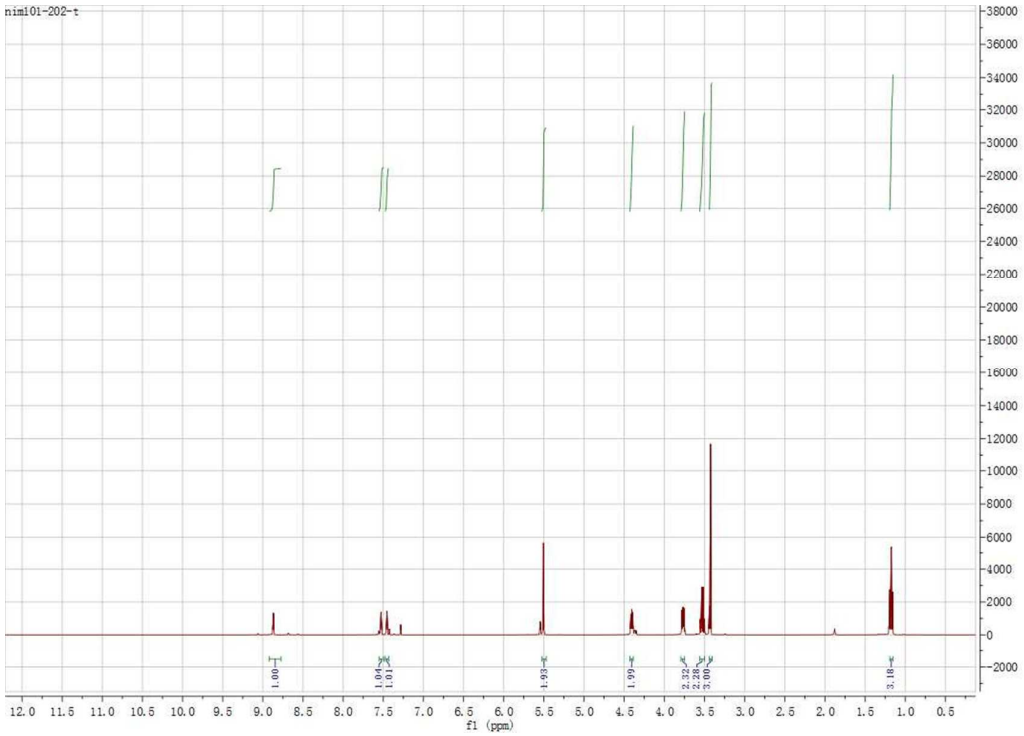

Fig. S10 ^{1}H NMR spectrum of IM1o1-2o2-TFSA.

1.2.7. 1-ethoxymethyl-3-methylimidazolium bis(trifluoromethanesulfonyl)imide (IM1o2-1-TFSA)

IM1o2 (5.0 g, 0.04 mol) and iodomethane (6.2 g, 0.044 mol) were reacted in a 250 mL flask at 0 ^{o}C for 24 h with acetonitrile (8 mL) as the solvent. The following steps were similar with IM1o1-1-TFSA. Colorless liquid (yield: 80%). ^{1}H NMR (400 MHz, $CDCl_3$): δ (ppm) 8.93-8.86 (s, 1H), 7.48-7.42 (d, 1H), 7.42-7.36 (d, 1H), 5.62-5.74 (s, 2H), 4.05-3.92 (s, 3H), 3.66-3.53 (m, 2H), 1.28-1.16 (t, 3H).

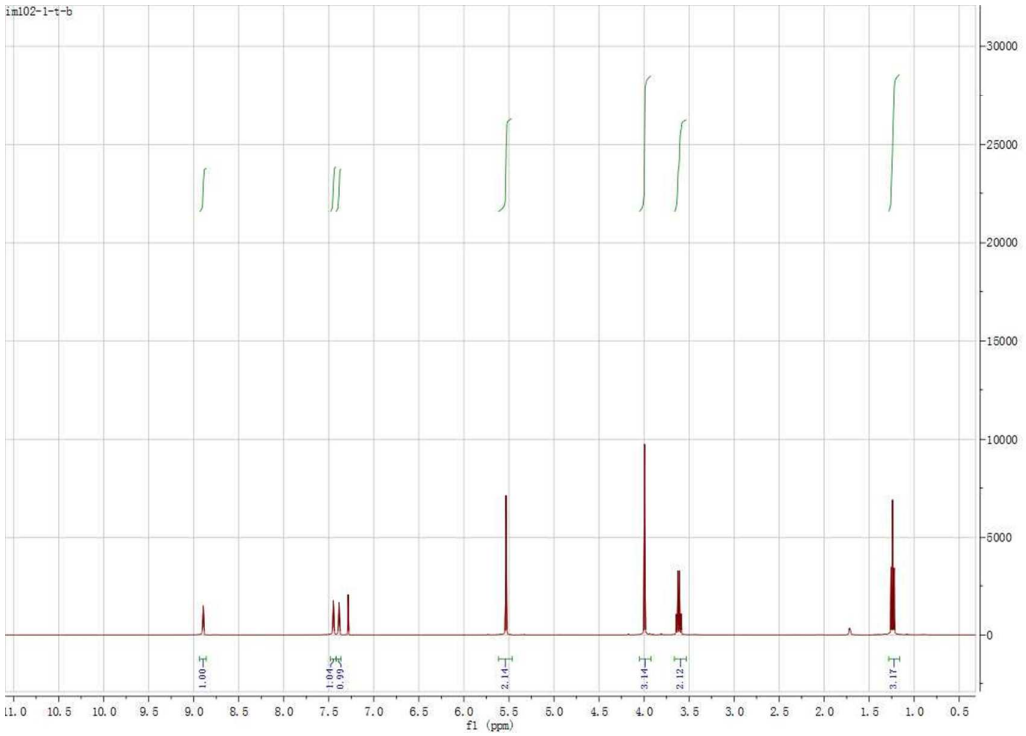

Fig. S11 ^{1}H NMR spectrum of IM1o2-1-TFSA.

1.2.8. 1-ethoxymethyl-3-ethylimidazolium bis(trifluoromethanesulfonyl)imide (IM1o2-2-TFSA)

IM1o2 (5 g, 0.04 mol) and bromoethane (8.7 g, 0.08 mol) were reacted in a 250 mL flask at 30 ^{o}C for 48 h with acetonitrile (8 mL) as the solvent. The following steps were similar with IM1o1-2-TFSA. Colorless liquid (yield: 78 %). ^{1}H NMR (400 MHz, $CDCl_3$): δ (ppm) 8.90-8.85 (s, 1H), 7.48-7.46 (d, 2H), 7.46-7.45 (d, 2H), 5.55-5.47 (s, 2H), 4.35-4.23 (m, 2H), 3.64-3.55 (m, 2H), 1.60-1.54 (t, 3H), 1.25-1.19 (t, 3H).

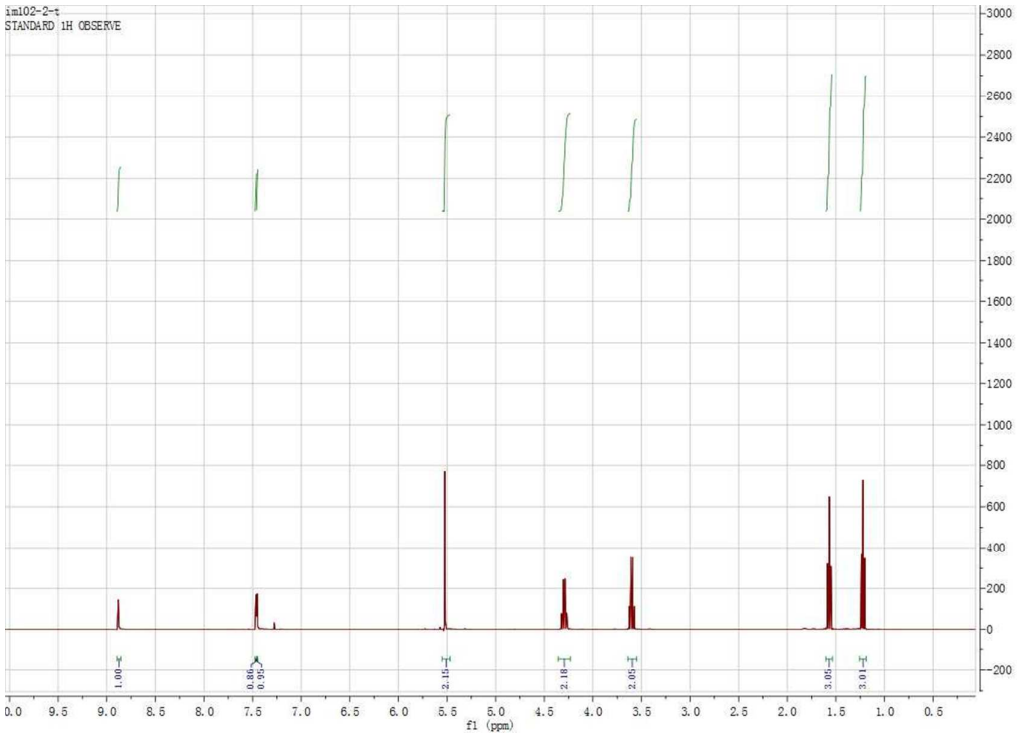

Fig. S12 ^{1}H NMR spectrum of IM1o2-2-TFSA.

1.2.9. 1-ethoxymethyl-3-propyllimidazolium bis(trifluoromethanesulfonyl)imide (IM1o2-3-TFSA)

IM1o2 (5 g, 0.04 mol) and 1-bromopropane (9.8 g, 0.08 mol) were reacted in a 250 mL flask at 60 ^{o}C for 48 h with acetonitrile (8 mL) as the solvent. The following steps were similar with IM1o1-2-TFSA. Colorless liquid (yield: 74 %). ^{1}H NMR (400 MHz, $CDCl_3$): δ (ppm) 9.00-8.93 (s, 1H), 7.48-7.42 (d, 1H), 7.40-7.33 (d, 1H), 5.56-5.49 (s, 2H), 4.24-4.13 (t, 2H), 3.66-3.54 (m, 2H), 1.99-1.86 (m, 2H), 1.25-1.17 (t, 3H), 1.02-0.95 (t, 3H).

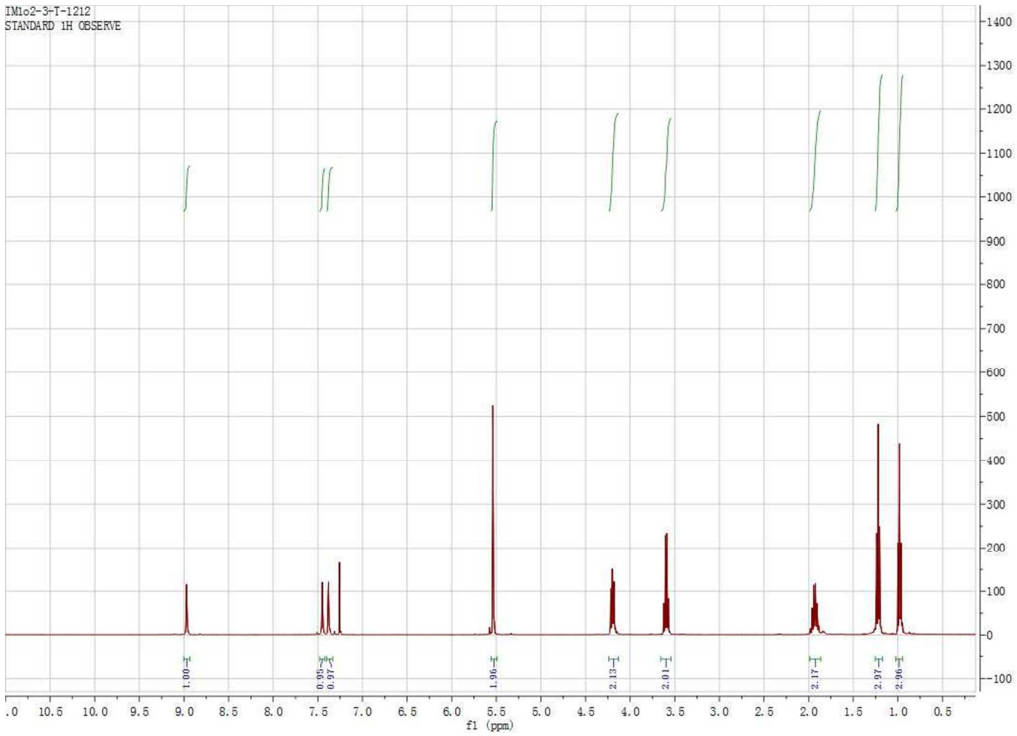

Fig. S13 ^{1}H NMR spectrum of IM1o2-3-TFSA.

1.2.10. 1-ethoxymethyl3-butylimidazolium bis(trifluoromethanesulfonyl)imide (IM1o2-4-TFSA)

IM1o2 (5 g, 0.04 mol) and 1-bromobutane (11.0 g, 0.08 mol) were reacted in a 250 mL flask at 60 ^{o}C for 48 h with acetonitrile (8 mL) as the solvent. The following steps were similar with IM1o1-2-TFSA. Colorless liquid (yield: 70 %). ^{1}H NMR (400 MHz, $CDCl_3$): δ (ppm) 8.97-8.93 (s, 1H), 7.48-7.41 (d, 1H), 7.41-7.34 (d, 1H), 5.55-5.49 (s, 2H), 4.25-4.17 (t, 2H), 3.64-3.55 (m, 2H), 1.91-1.81 (m, 2H), 1.41-1.31 (m, 2H), 1.25-1.17 (t, 3H), 0.99-0.92 (t, 3H).

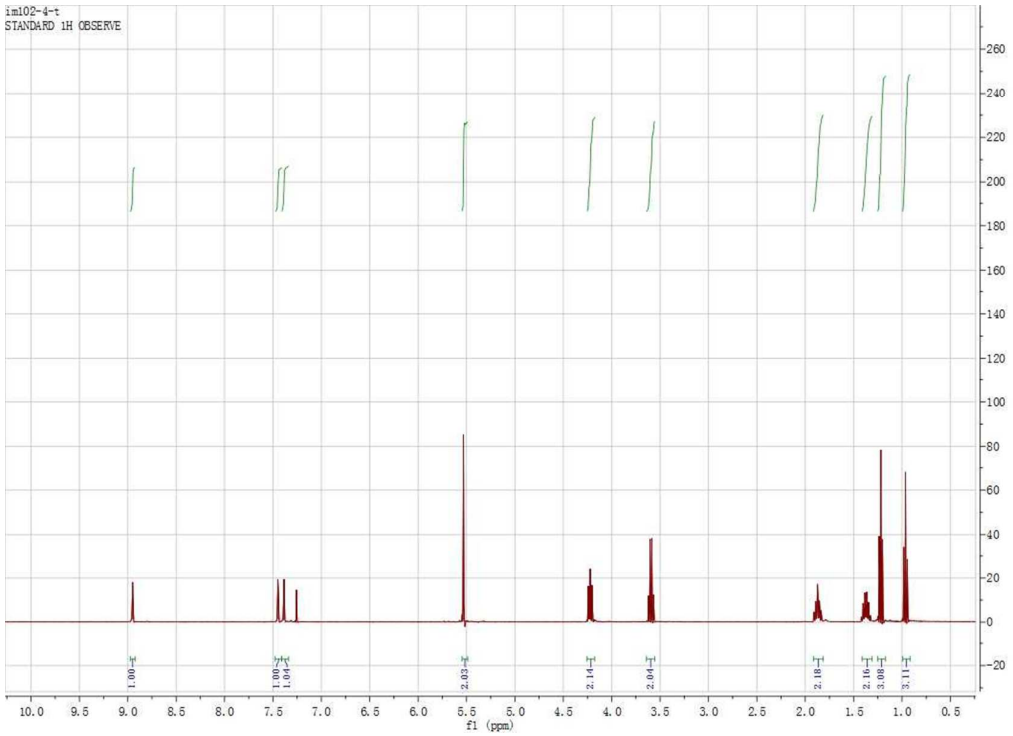

Fig. S14 ^{1}H NMR spectrum of IM1o2-4-TFSA.

1.2.11. 1-ethoxymethyl-3-(2-ethoxymethyl)imidazolium bis(trifluoromethanesulfonyl)imide (IM1o2-2o1-TFSA)

IM1o2 (5 g, 0.04 mol) and 2-methoxyethyl bromide (6.7 g, 0.048 mol) were reacted in a 250 mL flask at 60 ^{o}C for 48 h under argon atmosphere with acetonitrile (8 mL) as the solvent. The following steps were similar with IM1o1-2-TFSA. Colorless liquid (yield: 65 %).^{1}H NMR (400 MHz, $CDCl_3$): δ (ppm) 8.94-8.77 (s, 1H), 7.53-7.47 (d, 1H), 7.47-7.41 (d, 1H), 4.45-4.36 (t, 2H), 3.78-3.71 (t, 2H), 3.66-3.58 (m, 2H), 3.39-3.32 (s, 3H), 1.26-1.19 (t, 3H).

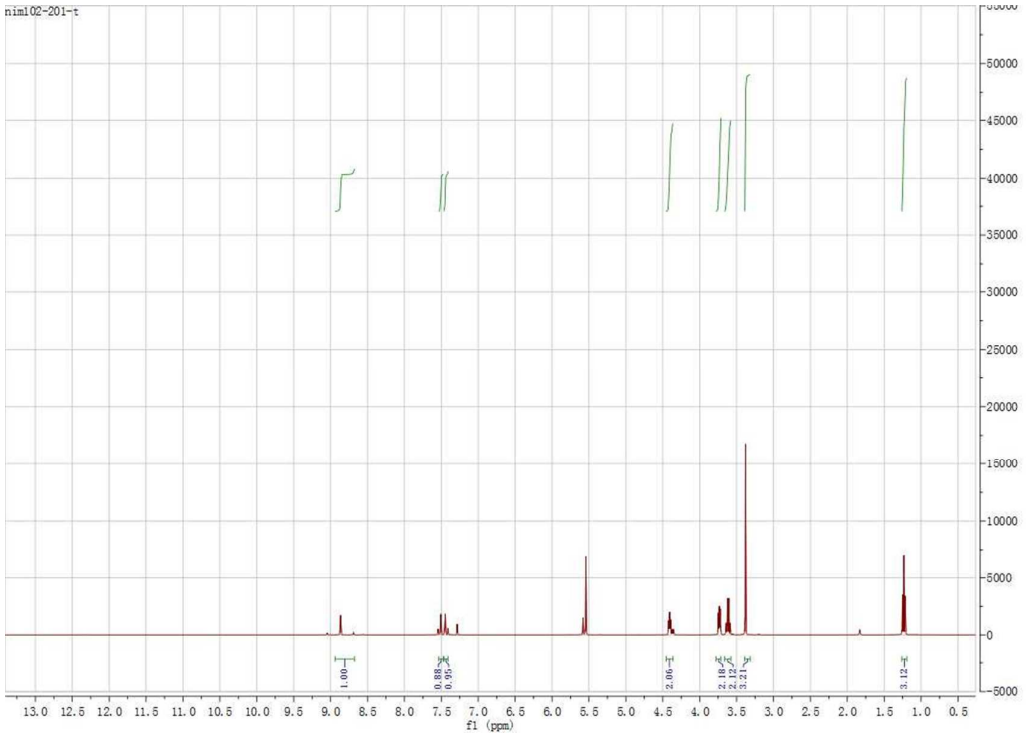

Fig. S15 ^{1}H NMR spectrum of IM1o2-2o1-TFSA.

1.2.12. (IM(1O1)2(2o2)-TFSA) 1-ethoxymethyl-3-(2-ethoxyethyl)imidazolium bis(trifluoromethanesulfonyl)imide (IM1o2-2o2-TFSA)

IM1o2 (5 g, 0.04 mol) and 2-ethoxyethyl bromide (7.3 g, 0.048 mol) were reacted in a 250 mL flask at 60 ^{o}C for 48 h under argon atmosphere with acetonitrile (8 mL) as the solvent. The following steps were similar with IM1o1-2-TFSA. Colorless liquid (yield: 63 %). ^{1}H NMR (400 MHz, $CDCl_3$): δ (ppm) 9.08-8.95 (s, 1H), 7.57-7.51 (d, 1H), 7.47-7.40 (d, 1H), 5.65-5.49 (s, 2H), 4.50-4.33 (t, 2H), 3.83-3.72 (t, 2H), 3.68-3.57(m, 2H), 3.57-3.46 (m, 2H), 1.28-1.21 (t, 3H), 1.21-1.15 (t, 3H).

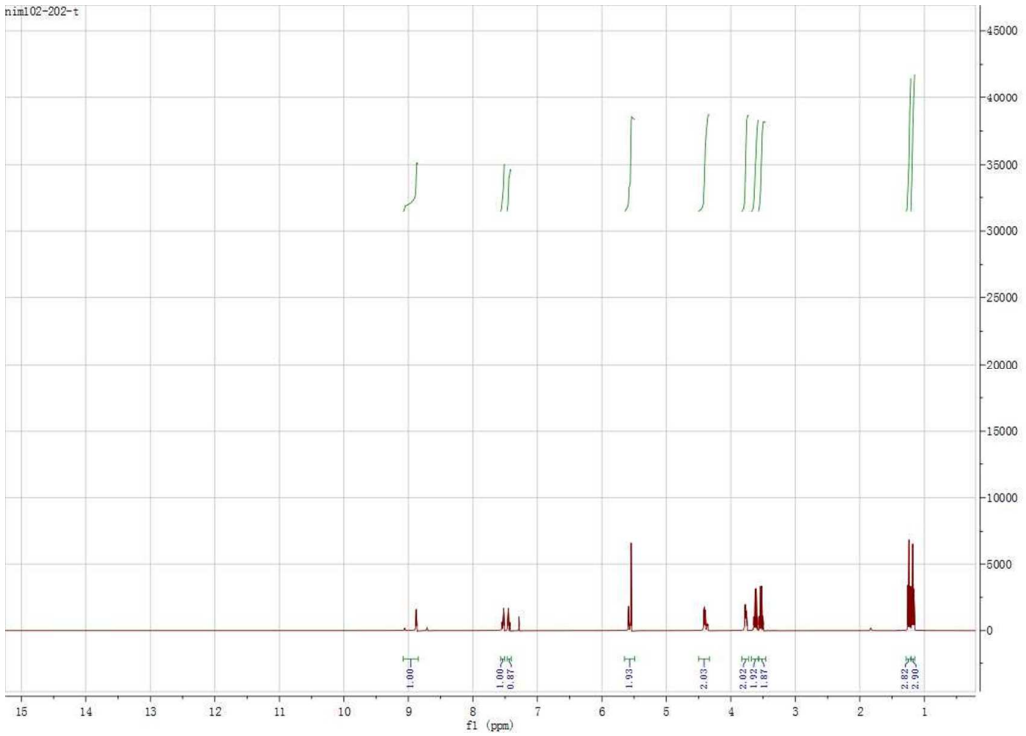

Fig. S16 ^{1}H NMR spectrum of IM1o2-2o2-TFSA.

1.3 Synthesis of the 1, 3-dialkylimidazolium ILs with a 2oR group

1.3.1 1-methoxyethyl-3-methylimidazolium bis(trifluoromethanesulfonyl)imide (IM2o1-1-TFSA)

IM2o1 (5.0 g, 0.04 mol) and iodomethane (6.2 g, 0.044 mol) were reacted in a 250 mL flask at 0 ^{o}C for 24 h with acetonitrile (8 mL) as the solvent. The following steps were similar with IM1o1-1-TFSA. Colorless liquid (yield: 82%). ^{1}H NMR (400 MHz, $CDCl_3$): δ (ppm) 8.70-8.62 (s, 1H), 7.44-7.37 (d, 1H), 7.31-7.28 (d, 1H), 4.36-4.31 (t, 2H), 3.96-3.72 (s, 3H), 3.72-3.67 (t, 2H), 3.38-3.33 (s, 3H).

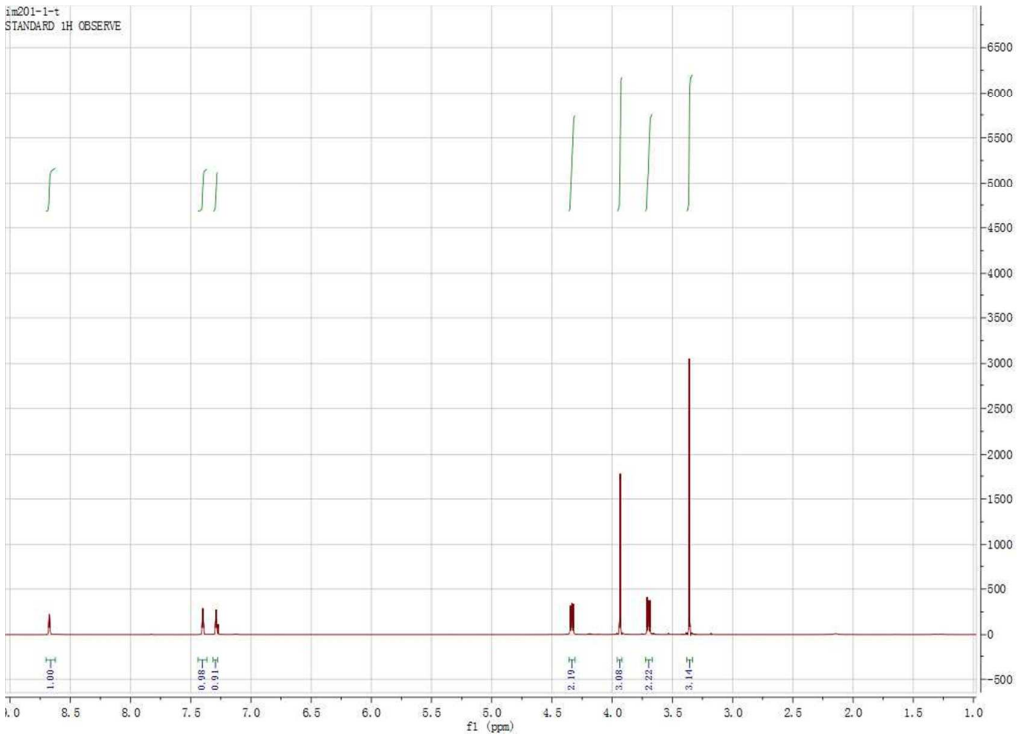

Fig. S17 ^{1}H NMR spectrum of IM2o1-1-TFSA.

1.3.2 1-methoxyethyl-3-ethylimidazolium bis(trifluoromethanesulfonyl)imide (IM2o1-2-TFSA)

IM2o1 (5 g, 0.04 mol) and bromoethane (8.7 g, 0.08 mol) were reacted in a 250 mL flask at 30 ^{o}C for 48 h with acetonitrile (8 mL) as the solvent. The following steps were similar with IM1o1-2-TFSA. Colorless liquid (yield: 70 %). ^{1}H NMR (400 MHz, $CDCl_3$): δ (ppm) 8.76-8.70 (s, 1H), 7.46-7.39(d, 1H), 7.36-7.30 (d, 1H), 4.40-4.31 (t, 2H), 4.31-4.22 (m, 2H), 3.74-3.68 (t, 2H), 3.40-3.33 (s, 3H), 1.61-1.51 (s, 3H).

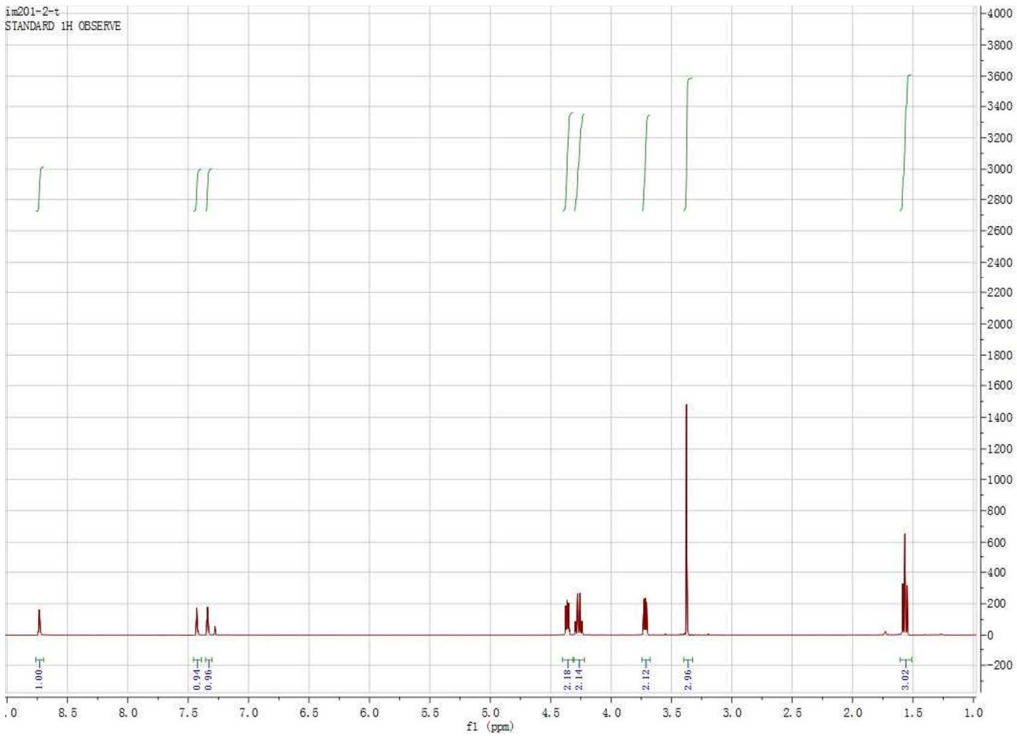

Fig. S18 ^{1}H NMR spectrum of IM2o1-2-TFSA.

1.3.3 1-methoxyethyl-3-propyllimidazolium bis(trifluoromethanesulfonyl)imide (IM2o1-3-TFSA)

IM2o1 (5 g, 0.04 mol) and 1-bromopropane (9.8 g, 0.08 mol) were reacted in a 250 mL flask at 60 ^{o}C for 48 h with acetonitrile (8 mL) as the solvent. The following steps were similar with IM1o1-2-TFSA. Colorless liquid (yield: 74 %). ^{1}H NMR (400 MHz, $CDCl_3$): δ (ppm) 8.79-8.74 (s, 1H), 7.47-7.41 (d, 1H), 7.34-7.30 (d, 1H), 4.42-4.35 (t, 2H), 4.21-4.13 (t, 2H), 3.75-3.69 (t, 2H), 3.41-3.35 (s, 3H), 1.99-1.88 (m, 2H), 1.02-0.96 (t, 3H).

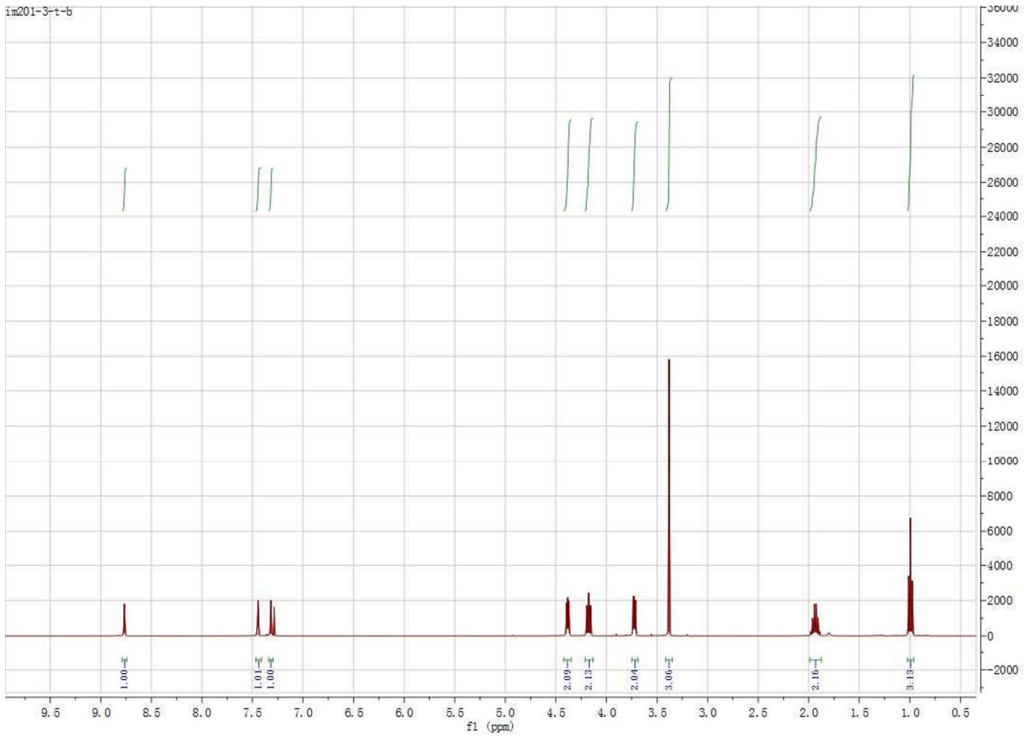

Fig. S19 ^{1}H NMR spectrum of IM2o1-3-TFSA.

1.3.4 1-methoxyethyl3-butylimidazolium bis(trifluoromethanesulfonyl)imide (IM1o2-4-TFSA)

IM1o2 (5 g, 0.04 mol) and 1-bromobutane (11.0 g, 0.08 mol) were reacted in a 250 mL flask at 60 ^{o}C for 48 h with acetonitrile (8 mL) as the solvent. The following steps were similar with IM1o1-2-TFSA. Colorless liquid (yield: 78 %). ^{1}H NMR (400 MHz, $CDCl_3$): δ (ppm) 8.78-8.68 (s, 1H), 7.48-7.41 (d, 1H), 7.37-7.30 (d, 1H), 4.40-4.34 (t, 2H), 4.23-4.15 (t, 2H), 3.74-3.69 (t, 2H), 3.40-3.34 (s, 3H), 1.93-1.81(m, 2H), 1.43-1.32 (m, 2H), 1.00-0.93 (t, 3H).

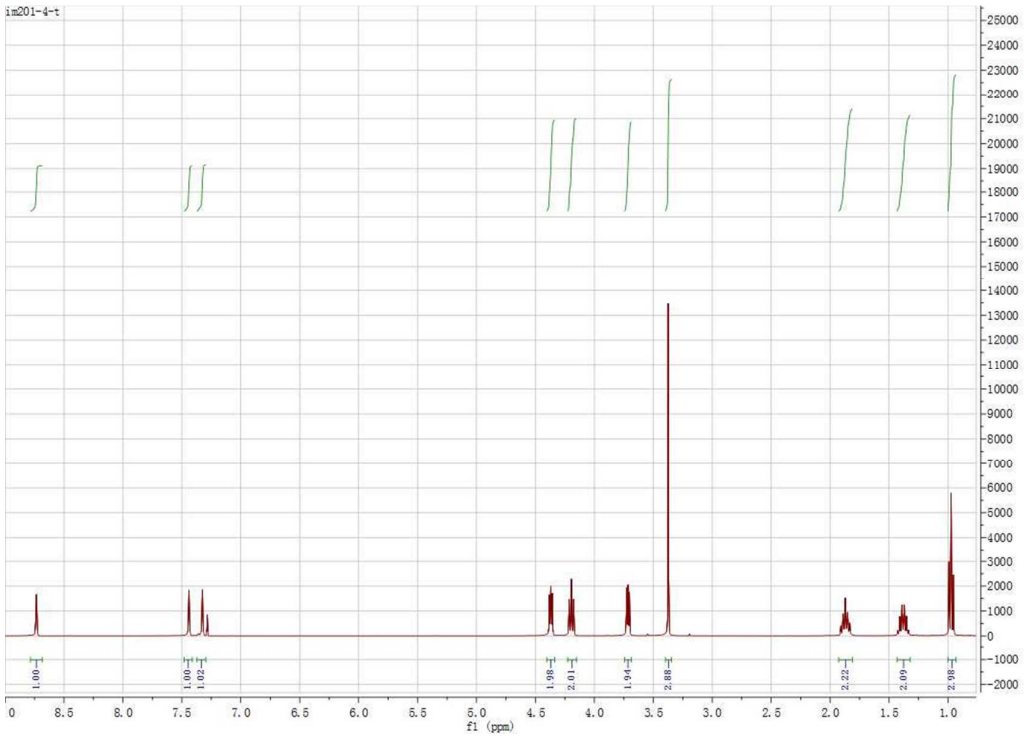

Fig. S20 ^{1}H NMR spectrum of IM2o1-4-TFSA.

1.3.5 1-methoxyethyl-3-(2-ethoxymethyl)imidazolium bis(trifluoromethanesulfonyl)imide (IM2o1-2o1-TFSA)

IM2o1 (5 g, 0.04 mol) and 2-methoxyethyl bromide (6.7 g, 0.048 mol) were reacted in a 250 mL flask at 60 ^{o}C for 48 h under argon atmosphere with acetonitrile (8 mL) as the solvent. The following steps were similar with IM1o1-2-TFSA. Colorless liquid (yield: 72 %). ^{1}H NMR (400 MHz, $CDCl_3$): δ (ppm) 8.85-8.70 (s, 1H), 7.48-7.35 (d, 2H), 4.44-4.33 (t, 4H), 3.82-3.68 (t, 4H), 3.44-3.33 (s, 6H).

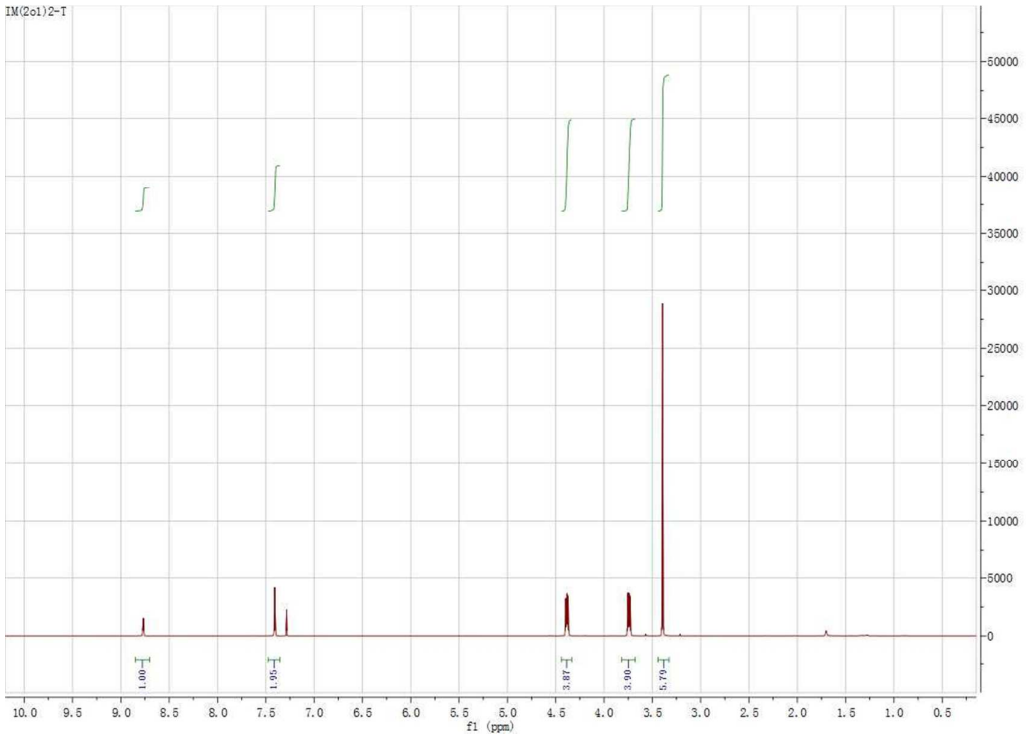

Fig. S21 ^{1}H NMR spectrum of IM2o1-2o1-TFSA.

1.3.6 1-ethoxyethyl-3-methylimidazolium bis(trifluoromethanesulfonyl)imide (IM2o2-1-TFSA)

IM2o2 (5.0 g, 0.035 mol) and iodomethane (5.5 g, 0.039 mol) were reacted in a 250 mL flask at 0 ^{o}C for 24 h with acetonitrile (8 mL) as the solvent. The following steps were similar with IM1o1-1-TFSA. Colorless liquid (yield: 82%). ^{1}H NMR (400 MHz, $CDCl_3$): δ (ppm) 8.63-8.54 (s, 1H), 7.37-7.28 (d, 1H), 7.24-7.20 (d, 1H), 4.28-4.22 (t, 2H), 3.88-3.84 (s, 3H), 3.68-3.62 (t, 2H), 3.47-3.39 (m, 2H), 1.12-1.06 (t, 3H).

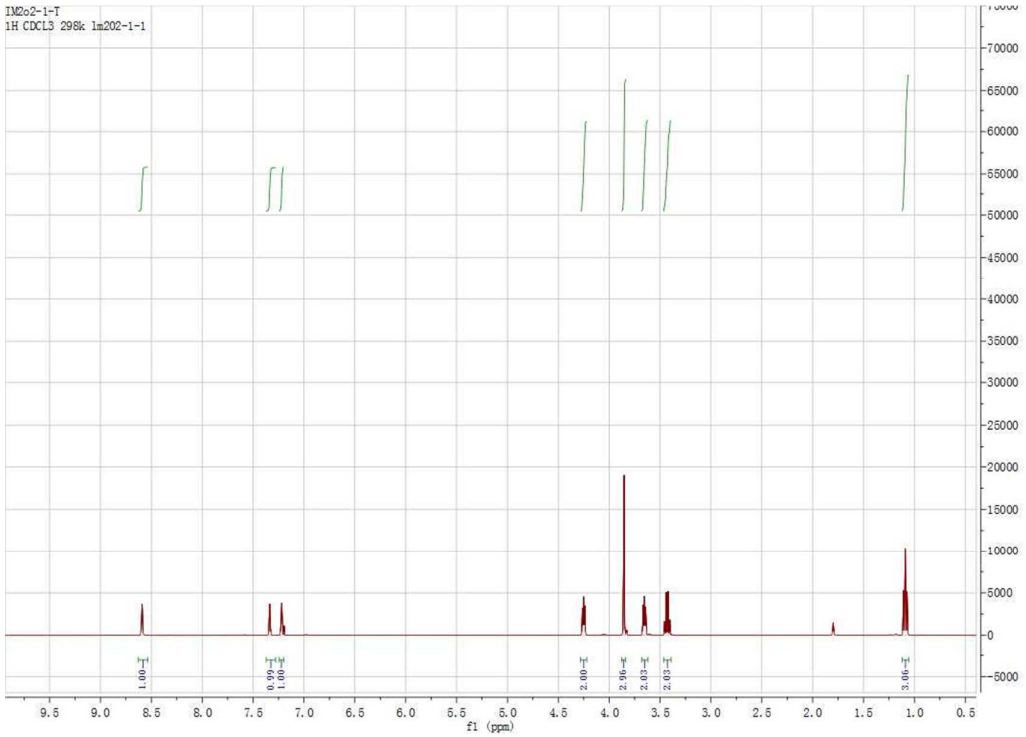

Fig. S22 ^{1}H NMR spectrum of IM2o2-1-TFSA.

1.3.7 1-ethoxyethyl-3-ethylimidazolium bis(trifluoromethanesulfonyl)imide (IM2o2-2-TFSA)

IM2o2 (5 g, 0.035 mol) and bromoethane (7.6 g, 0.07 mol) were reacted in a 250 mL flask at 30 ^{o}C for 48 h with acetonitrile (8 mL) as the solvent. The following steps were similar with IM1o1-2-TFSA. Colorless liquid (yield: 76 %). ^{1}H NMR (400 MHz, $CDCl_3$): δ (ppm) 8.81-8.75 (s, 1H), 7.41-7.36 (d, 1H), 7.24-7.22 (d, 1H), 4.37-4.29 (t, 2H), 4.28-4.18 (m, 2H), 3.74-3.68 (t, 2H), 3.51-3.43 (m, 2H), 1.56-1.49 (t, 3H), 1.16-1.10 (t, 3H).

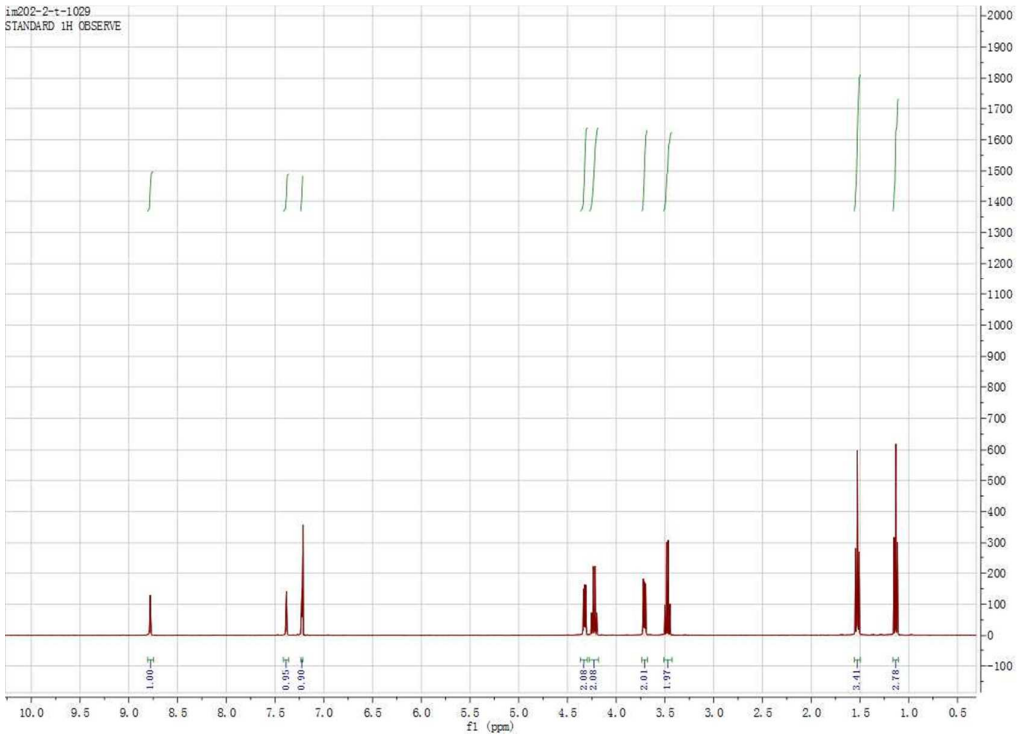

Fig. S23 ^{1}H NMR spectrum of IM2o2-2-TFSA.

1.3.8 1-ethoxyethyl-3-propyllimidazolium bis(trifluoromethanesulfonyl)imide (IM2o2-3-TFSA)

IM2o2 (5 g, 0.035 mol) and 1-bromopropane (8.6 g, 0.07 mol) were reacted in a 250 mL flask at 60 ^{o}C for 48 h with acetonitrile (8 mL) as the solvent. The following steps were similar with IM1o1-2-TFSA. Colorless liquid (yield: 78 %). ^{1}H NMR (400 MHz, CDCl3): δ (ppm) 8.81-8.75 (s, 1H), 7.45-7.41 (d, 1H), 7.30-7.26 (d, 1H), 4.40-4.33 (t, 2H), 4.19-4.11 (t, 2H) 3.77-3.70 (t, 2H), 3.54-3.47 (m, 2H), 1.98-1.86 (m, 2H), 1.19-1.13 (t, 3H), 1.00-0.93 (t, 3H).

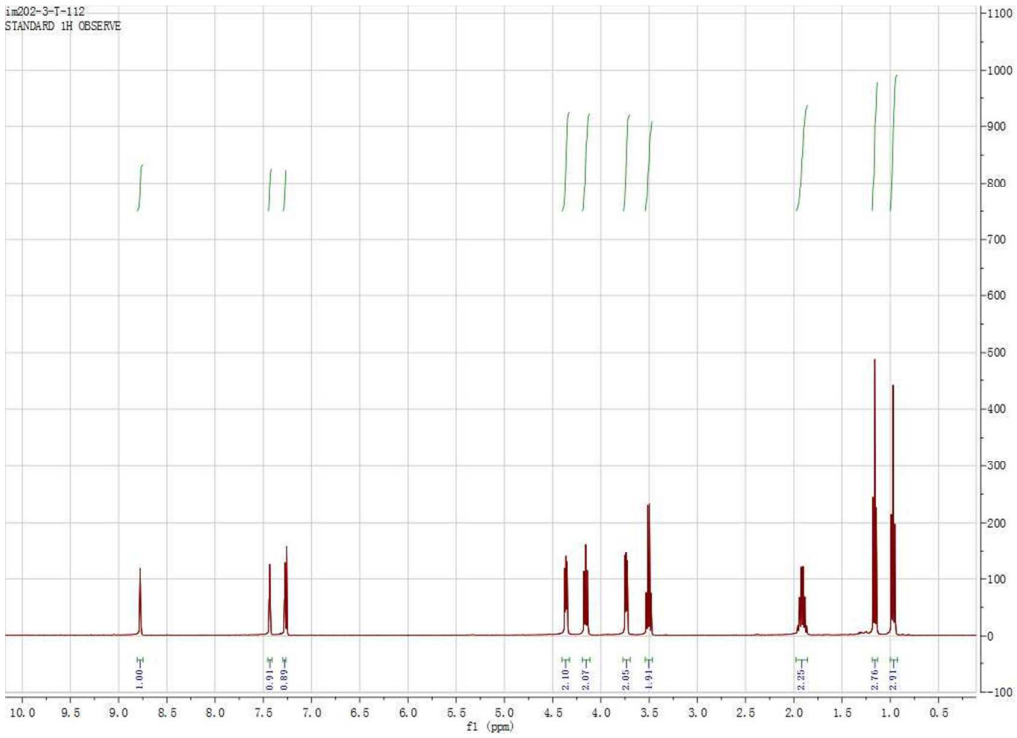

Fig. S24 ^{1}H NMR spectrum of IM2o2-3-TFSA.

1.3.9 1-ethoxyethyl3-butylimidazolium bis(trifluoromethanesulfonyl)imide (IM2o2-4-TFSA)

IM2o2 (5 g, 0.035 mol) and 1-bromobutane (9.6 g, 0.07 mol) were reacted in a 250 mL flask at 60 ^{o}C for 48 h with acetonitrile (8 mL) as the solvent. The following steps were similar with IM1o1-2-TFSA. Colorless liquid (yield: 78 %). ^{1}H NMR (400 MHz, $CDCl_3$): δ (ppm) 8.75-8.70 (s, 1H), 7.46-7.40 (d, 1H), 7.33-7.26 (d, 1H), 4.37-4.31 (t, 2H), 4.21-4.13 (t, 2H), 3.75-3.68 (t, 2H), 3.53-3.44 (m, 2H), 1.91-1.79 (m, 2H), 1.41-1.28 (m, 2H), 1.18-1.10 (t, 3H), 0.99-0.90 (t, 3H).

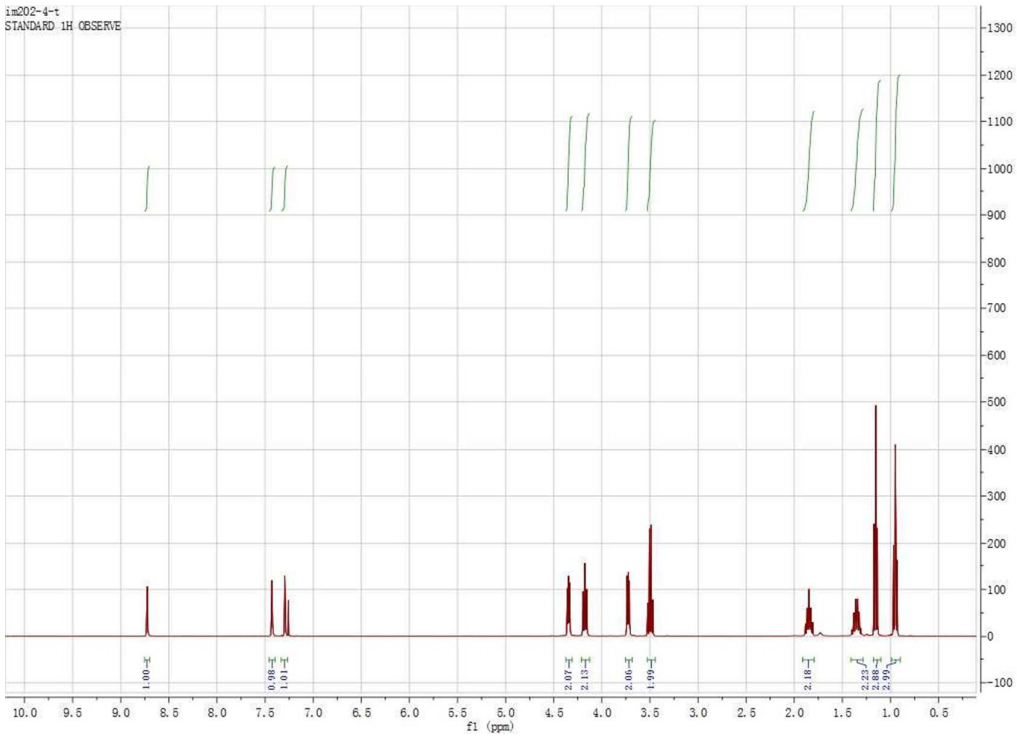

Fig. S25 ^{1}H NMR spectrum of IM2o2-4-TFSA.

1.2.11 1-ethoxyethyl-3-(2-ethoxymethyl)imidazolium bis(trifluoromethanesulfonyl)imide (IM2o2-2o1-TFSA)

IM2o2 (5 g, 0.035 mol) and 2-methoxyethyl bromide (5.8 g, 0.042 mol) were reacted in a 250 mL flask at 60 ^{o}C for 48 h under argon atmosphere with acetonitrile (8 mL) as the solvent. The following steps were similar with IM1o1-2-TFSA. Colorless liquid (yield: 61 %).^{1}H NMR (400 MHz, $CDCl_3$): δ (ppm) 8.71-8.66 (s, 1H), 7.35-7.32 (d, 2H), 7.32-7.30 (d, 2H), 4.33-4.24 (t*2, 4H), 3.74-3.59 (t*2, 4H), 3.50-3.40 (m, 2H), 3.34-3.26 (s, 3H), 1.16-1.07 (t, 3H).

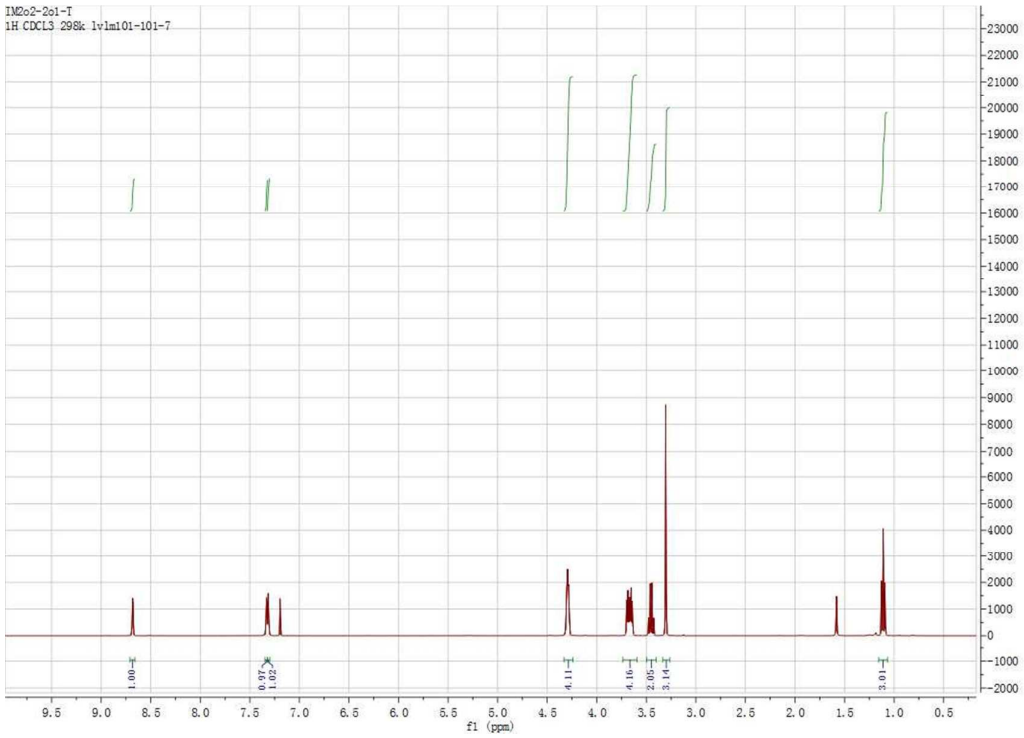

Fig. S26 ^{1}H NMR spectrum of IM2o2-2o1-TFSA.

1.2.12 1-ethoxyethyl-3-(2-ethoxyethyl)imidazolium bis(trifluoromethanesulfonyl)imide (IM2o2-2o2-TFSA)

IM2o2 (5 g, 0.035 mol) and 2-ethoxyethyl bromide (6.4 g, 0.042 mol) were reacted in a 250 mL flask at 60 ^{o}C for 48 h under argon atmosphere with acetonitrile (8 mL) as the solvent. The following steps were similar with IM1o1-2-TFSA. Colorless liquid (yield: 63 %). ^{1}H NMR (400 MHz, $CDCl_3$): δ (ppm) 8.77-8.68 (s, 1H), 7.46-7.37 (d, 2H), 4.41-4.29 (t, 4H), 3.79-3.69 (t, 4H), 3.56-3.45 (m, 4H), 1.21-1.12 (t, 6H).

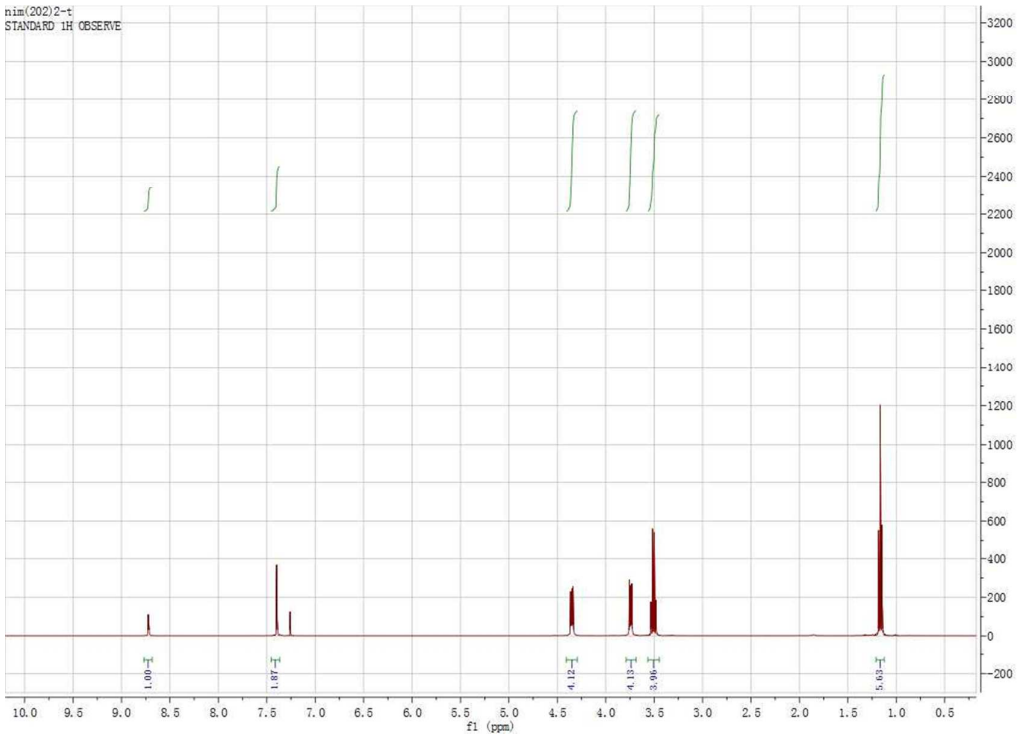

Fig. S27 ^{1}H NMR spectrum of IM2o2-2o2-TFSA.

Table S1

Adjustable parameters of VTF equation for viscosity.

ILs	η_0 (cP)	B (K)	T_0 (K)	R^2
IM1o1-1-TFSA	0.105 (±9%)	832.9(±4%)	162.9(±2%)	0.9999
IM1o1-2-TFSA	0.084(±7%)	869.1(±7%)	158.3(±1%)	0.99999
IM1o1-3-TFSA	0.107(±6%)	782.3(±6%)	172.4(±1%)	0.99999
IM1o1-4-TFSA	0.086(±12%)	853.7(±12%)	166.7(±2%)	0.99997
IM1o1-2o1-TFSA	0.150(±11%)	666.6(±11%)	185.2(±2%)	0.99996
IM1o1-2o2-TFSA	0.104(±13%)	762.3(±5%)	175.5(±2%)	0.99996
IM1o2-1-TFSA	0.147(±6%)	703.9(±3%)	176.1(±1%)	0.99999
IM1o2-2-TFSA	0.163(±8%)	660.1(±3%)	176.7(±1%)	0.99998
IM1o2-3-TFSA	0.087(±3%)	823.7(±1%)	168.8(±0%)	1
IM1o2-4-TFSA	0.166(±10%)	661.4(±4%)	184.4(±1%)	0.99997
IM1o2-2o1-TFSA	0.196(±11%)	578.2(±5%)	194.5(±1%)	0.99995
IM1o2-2o2-TFSA	0.098(±12%)	763.5(±4%)	175(±1%)	0.99997
IM2o1-1-TFSA	0.175(±9%)	660.5(±9%)	177.5(±2%)	0.99997
IM2o1-2-TFSA	0.167(±15%)	663.2(±15%)	174.7(±3%)	0.99993
IM2o1-3-TFSA	0.174(±13%)	654.0(±13%)	181.1(±2%)	0.99995
IM2o1-4-TFSA	0.172(±16%)	632.9(±16%)	186.9(±2%)	0.99991
IM2o1-2o1-TFSA	0.186(±15%)	622.4(±16%)	185.6(±2%)	0.99991
IM2o2-1-TFSA	0.070(±18%)	923.9(±28%)	149.5(±6%)	0.99986
IM2o2-2-TFSA	0.167(±28%)	654.1(±17%)	173.1(±3%)	0.99991
IM2o2-3-TFSA	0.066(±16%)	899.6(±21%)	159.7(±4%)	0.99992
IM2o2-4-TFSA	0.087(±21%)	833.2(±8%)	166.8(±1%)	0.99999
IM2o2-2o1-TFSA	0.177(±8%)	620.0(±7%)	186.5(±1%)	0.99998
IM2o2-2o2-TFSA	0.134(±7%)	724.9(±7%)	171.5(±1%)	0.99999
EMI-TFSA	0.182(±9%)	718.6(±9%)	160.8(±1%)	0.99991
PMI-TFSA	0.160(±8%)	654.5(±7%)	177.6(±1%)	0.99992
BMI-TFSA	0.195(±6%)	670.0(±2%)	176.0(±1%)	0.99999

The percentage standard errors for η_0, B and T_0 have been included, and R^2 is the VTF fitting parameter.

Table S2

Adjustable parameters of VTF equation for conductivity.

ILs	σ_0 (cP)	B (K)	T_0 (K)	R^2
IM1o1-1-TFSA	238.9(±5%)	393.3(±3%)	199.2(±1%)	0.99997
IM1o1-2-TFSA	235.2(±4%)	399.8(±3%)	194.1(±1%)	0.99998
IM1o1-3-TFSA	240.9(±2%)	431.8(±2%)	197(±0%)	0.99999
IM1o1-4-TFSA	270.8(±5%)	494.5(±3%)	190.8(±1%)	0.99998
IM1o1-2o1-TFSA	235.0(±2%)	429.4(±1%)	199.8(±0%)	1
IM1o1-2o2-TFSA	319.0(±5%)	553.0(±3%)	181.7(±1%)	0.99998
IM1o2-1-TFSA	211.5(±2%)	384.7(±1%)	200.8(±0%)	1
IM1o2-2-TFSA	198.1(±4%)	381.7(±3%)	197.7(±1%)	0.99998
IM1o2-3-TFSA	259.1(±4%)	478.3(±2%)	191.0(±1%)	0.99999
IM1o2-4-TFSA	268.5(±3%)	522.4(±2%)	187.7(±1%)	0.99999
IM1o2-2o1-TFSA	234.6(±7%)	450.1(±4%)	196.7(±1%)	0.99995
IM1o2-2o2-TFSA	240.2(±9%)	482.2(±5%)	192.7(±2%)	0.99994
IM2o1-1-TFSA	215.9(±2%)	394.7(±1%)	195.5(±0%)	1
IM2o1-2-TFSA	228.8(±5%)	419.4(±3%)	187.8(±1%)	0.99997
IM2o1-3-TFSA	204.9(±2%)	405.3(±1%)	199.0(±0%)	0.99999
IM2o1-4-TFSA	169.8(±3%)	380.5(±2%)	205.4(±1%)	0.99999
IM2o1-2o1-TFSA	210.4(±2%)	427.1(±1%)	196.7(±0%)	1
IM2o2-1-TFSA	219.6(±7%)	425.2(±5%)	190.0(±2%)	0.99995
IM2o2-2-TFSA	201.4(±2%)	409.0(±1%)	189.5(±0%)	1
IM2o2-3-TFSA	220.5(±5%)	453.2(±3%)	192.2(±1%)	0.99998
IM2o2-4-TFSA	236.3(±3%)	504.1(±2%)	187.6(±1%)	0.99999
IM2o2-2o1-TFSA	212.1(±2%)	446.5(±1%)	194.2(±1%)	0.99999
IM2o2-2o2-TFSA	214.4(±8%)	488.1(±5%)	186.9(±2%)	0.99995
EMI-TFSA	191.7(±6%)	331.2(±5%)	191.3(±2%)	0.99993
PMI-TFSA	247.7(±8%)	404.1(±5%)	190.9(±2%)	0.99994
BMI-TFSA	287.1(±8%)	495.4(±5%)	182.4(±2%)	0.99995

The percentage standard errors for σ_0, B and T_0 have been included, and R^2 is the VTF fitting parameter.

Table S3

Cathodic and anodic limiting potentials and electrochemical windows values at 25 ^{o}C.

ILs	Cathodic limiting potential	Anodic limiting potential	Electrochemical window
	E vs (Ag)/V	E vs (Ag)/V	/V
IM1o1-1-TFSA	-2.0	2.2	4.2
IM1o1-2-TFSA	-1.9	2.1	4.0
IM1o1-3-TFSA	-1.7	2.3	4.0
IM1o1-4-TFSA	-1.8	2.3	4.1
IM1o1-2o1-TFSA	-1.8	2.2	4.0
IM1o1-2o2-TFSA	-1.8	2.2	4.0
IM1o2-1-TFSA	-1.8	2.3	4.1
IM1o2-2-TFSA	-1.7	2.4	4.1
IM1o2-3-TFSA	-1.8	2.3	4.1
IM1o2-4-TFSA	-1.8	2.4	4.2
IM1o2-2o1-TFSA	-1.8	2.2	4.0
IM1o2-2o2-TFSA	-1.9	2.3	4.2
IM2o1-1-TFSA	-1.9	1.6	3.5
IM2o1-2-TFSA	-2.0	2.2	4.2
IM2o1-3-TFSA	-2.2	2.0	4.2
IM2o1-4-TFSA	-2.0	2.1	4.1
IM2o1-2o1-TFSA	-2.0	2.1	4.1
IM2o2-1-TFSA	-1.9	1.6	3.5
IM2o2-2-TFSA	-2.0	2.0	4.0
IM2o2-3-TFSA	-2.3	1.8	4.1
IM2o2-4-TFSA	-2.4	1.8	4.2
IM2o2-2o1-TFSA	-1.9	2.1	4.0
IM2o2-2o2-TFSA	-1.9	2.1	4.0
EMI-TFSA	-2.0	2.2	4.2
PMI-TFSA	-2.1	1.9	4.0
BMI-TFSA	-2.1	2.2	4.3

Working electrode: glassy carbon; counter electrode: platinum wire; reference electrode: silver wire; scan rate: 10 mV s^{-1}.