Supplementary Information

Solution state and dissolution process of cellulose in ionic liquids-based solvents with different hydrogen-bonding basicity and microstructure

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Supporting information content

Number of pages: 8
Number of figures: 9
Number of tables: 2

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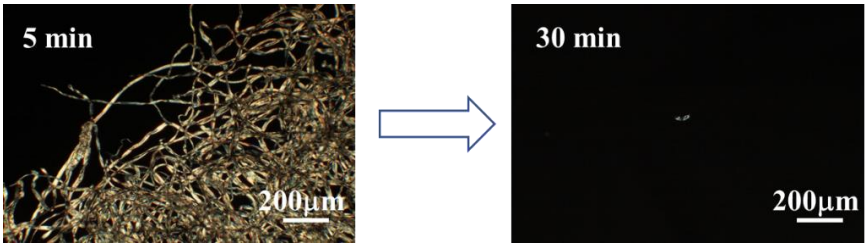

Fig. S1 Polarized photos of cellulose dissolution in BmimAc/DMAc (1:9, w/w).

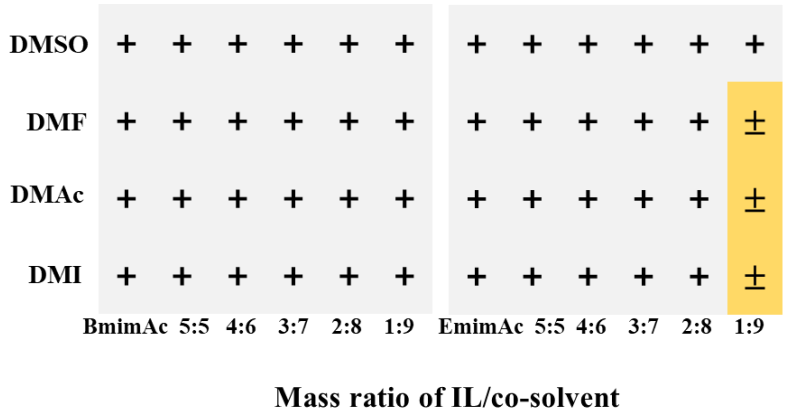

Fig. S2 Solubility of cellulose in different acetate-based ILs/co-solvent systems by directly dissolving cellulose in ILs/co-solvent systems. (+, soluble; ±, partially soluble)

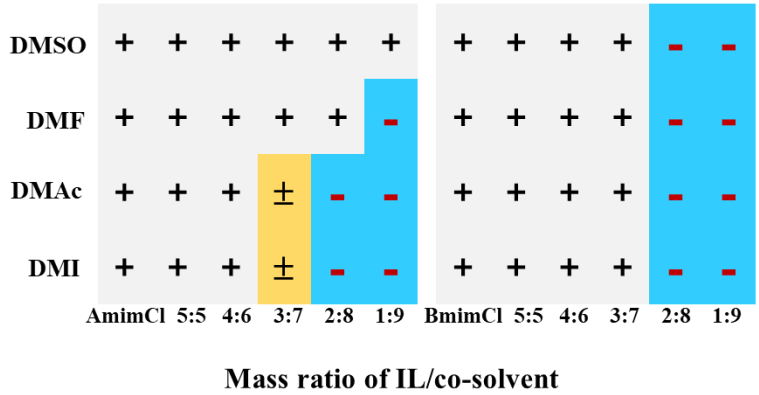

Fig. S3 Solubility of cellulose in different chloride-based ILs/co-solvent systems by adding co-solvents into cellulose/ILs solutions. (+, homogeneous solution; ±, partial precipitation; -, complete precipitation)

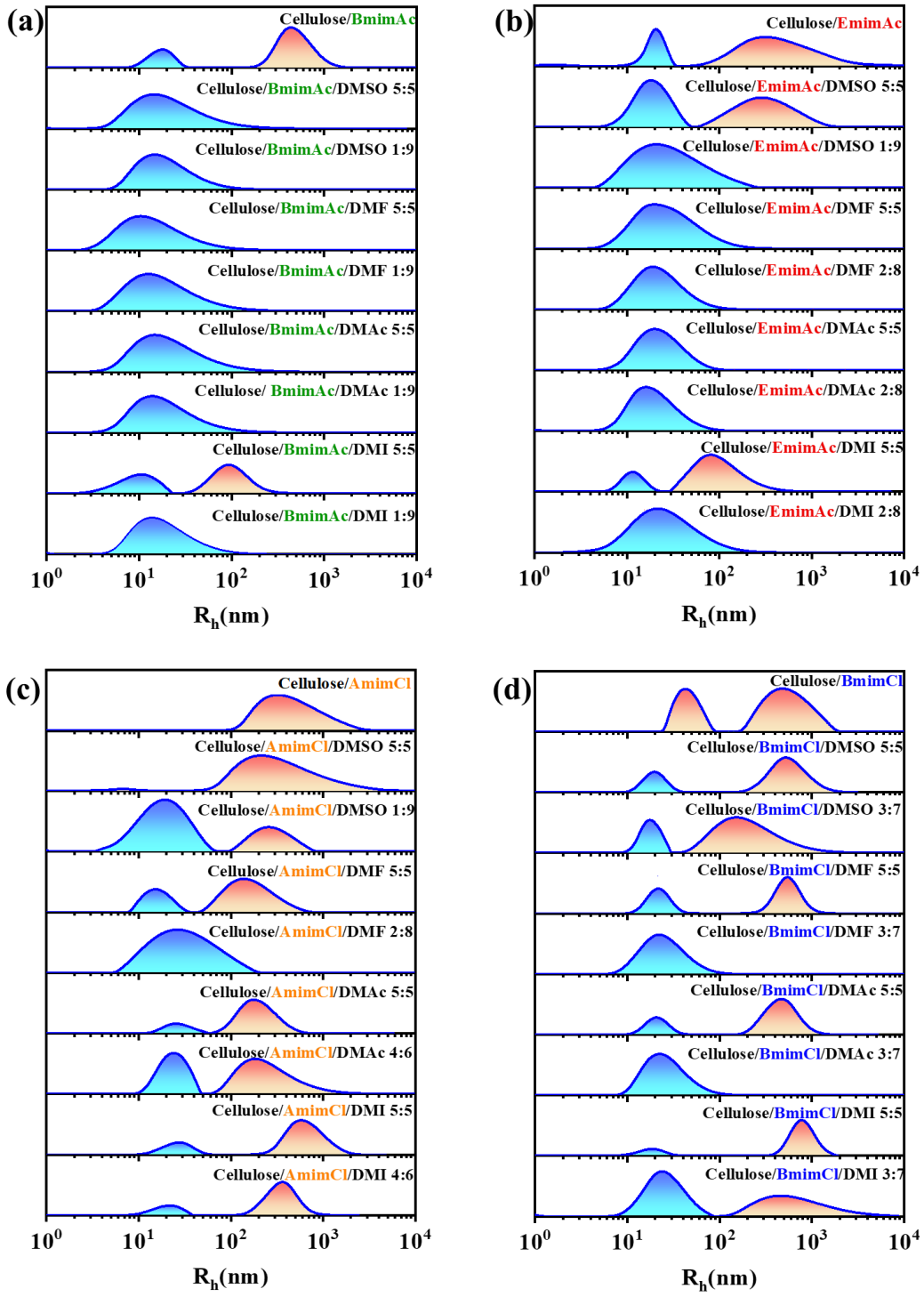

Fig. S4 The R_h distribution curves of different cellulose/ILs/co-solvent systems, (a) cellulose/BmimAc/co-solvents, (b) cellulose/EmimAc/co-solvents, (c) cellulose/AmimCl/co-solvents, and (d) cellulose/BmimCl/co-solvents.

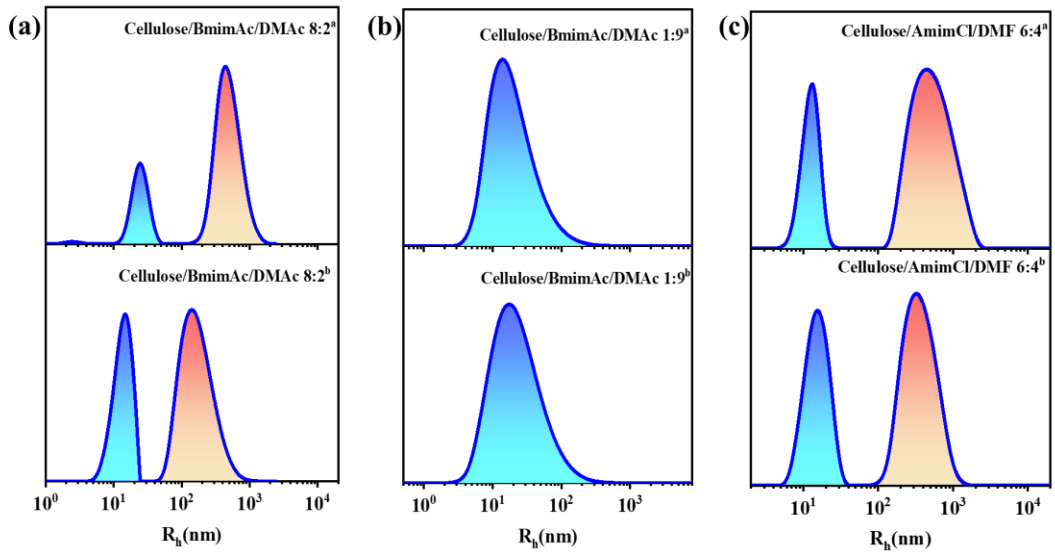

Fig. S5 Effect of different preparation methods on the solution state of cellulose/ILs/co-solvent systems. (a) Cellulose/BmimAc/DMAc (BmimAc:DMAc, 8:2), (b) cellulose/BmimAc/DMAc (BmimAc:DMAc, 1:9), and (c) cellulose/AmimCl/DMF (AmimCl:DMF, 6:4).

Note: [a] The dissolution method is Method 1 in which the mixture of ILs and co-solvents is used to dissolve cellulose.

[b] The dissolution method is Method 2 in which the ILs dissolve cellulose firstly followed the addition of the co-solvents.

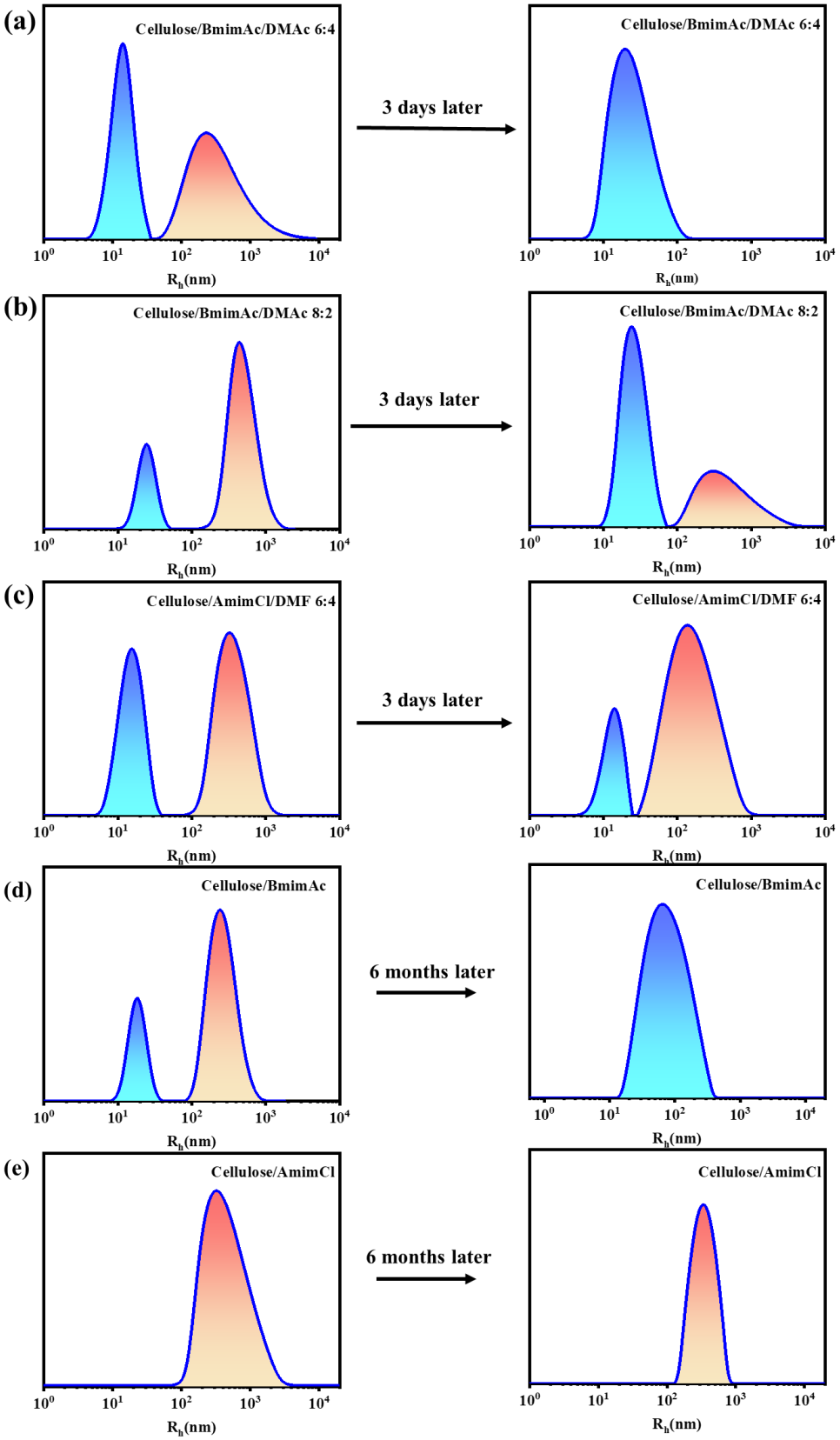

Fig. S6 R_h distribution curves of cellulose/ILs and cellulose/ILs/co-solvent solutions before and after placement. (a) Cellulose/BmimAc/DMAc (BmimAc/DMAc, 6:4), (b) cellulose/BmimAc/DMAc (BmimAc/DMAc, 8:2), (c) cellulose/AmimCl/DMF

(AmimCl/DMF, 6:4), (d) cellulose/BmimAc, (e) cellulose/AmimCl.

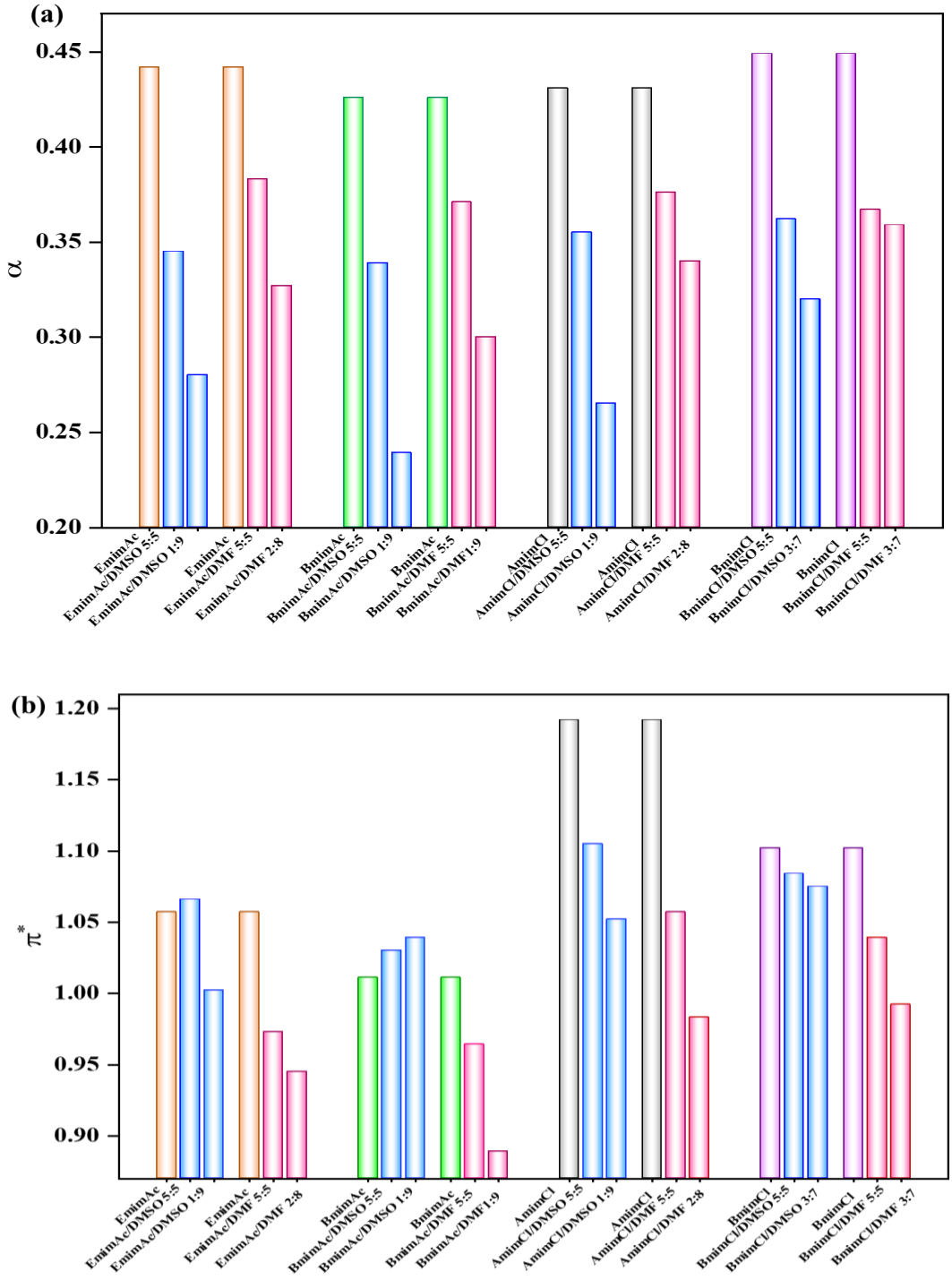

Fig. S7 Solvent parameters of different ILs/co-solvent systems. (a) α, (b) π*.

Table S1 Viscosity, cation self-diffusion coefficient (D_+), anion self-diffusion coefficient (D_-), conductivity Λ_{NMR}, molar conductivity Λ_{imp} and ion dissociation degree ($\Lambda_{imp}/\Lambda_{NMR}$) of BmimAc/DMAc system.

Solvents	Viscosity (cP)	D_+ ($\times10^{-7}$ cm^2/S)	D_- ($\times10^{-7}$ cm^2/S)	Λ_{NMR} ($S\cdot cm^2/mol$)	Conductivity (mS/cm)	Λ_{imp} ($S\cdot cm^2/mol$)	$\Lambda_{imp}/\Lambda_{NMR}$
BmimAc	477.2	0.31	0.31	0.23	0.62	0.12	0.51
BmimAc/DMAc 8:2	38.44	1.81	1.87	1.38	3.12	0.59	0.42
BmimAc/DMAc 7:3	18.44	3.50	3.68	2.70	4.21	0.79	0.29
BmimAc/DMAc 6:4	10.23	6.18	6.59	4.79	4.86	0.91	0.19
BmimAc/DMAc 5:5	6.16	10.0	10.6	7.75	5.35	1.01	0.13
BmimAc/DMAc 4:6	3.92	15.4	16.3	11.9	5.46	1.03	0.09
BmimAc/DMAc 3:7	2.52	23.0	23.4	17.4	4.90	0.92	0.05
BmimAc/DMAc 2:8	1.72	33.0	33.4	24.9	4.07	0.77	0.04
BmimAc/DMAc 1:9	1.23	47.7	48.8	36.2	3.25	0.61	0.02

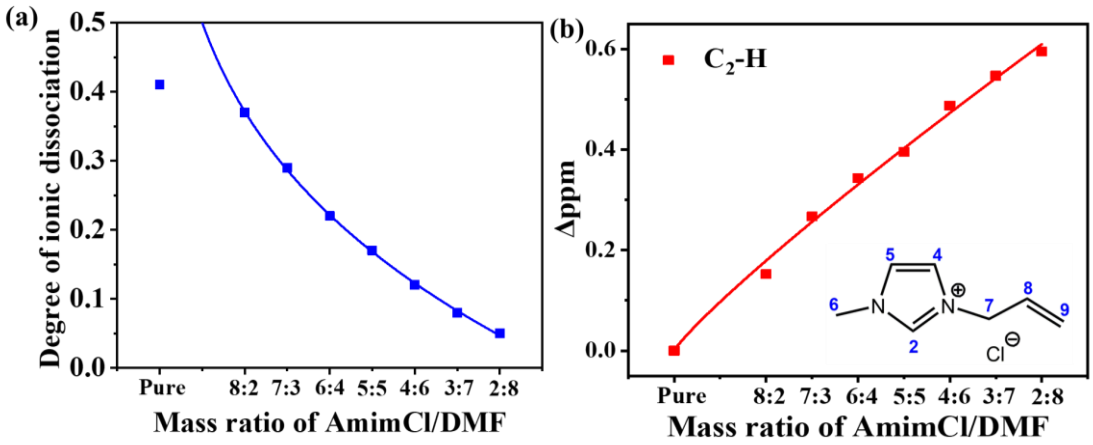

Fig. S8 Microstructure of the AmimCl/DMF system. (a) Degree of ion dissociation of the AmimCl/DMF system. (b) Chemical shift change of C_2-H in AmimCl.

Table S2 Viscosity, cation self-diffusion coefficient (D_+), molar conductivity Λ_{NMR}, molar conductivity Λ_{imp} and ion dissociation degree ($\Lambda_{imp}/\Lambda_{NMR}$) of AmimCl/DMF system.

Solvents	Viscosity (cP)	D_+ ($\times10^{-7}$ cm^2/S)	Λ_{NMR} ($S\cdot cm^2/mol$)	Conductivity (mS/cm)	Λ_{imp} ($S\cdot cm^2/mol$)	$\Lambda_{imp}/\Lambda_{NMR}$
AmimCl	2086	0.09	0.07	0.20	0.03	0.41
AmimCl/DMF 8:2	128.7	1.13	0.84	2.28	0.32	0.37
AmimCl/DMF 7:3	50.07	2.78	2.09	4.35	0.60	0.29
AmimCl/DMF 6:4	21.77	5.76	4.32	7.01	0.96	0.22
AmimCl/DMF 5:5	9.23	10.4	7.77	9.42	1.29	0.17
AmimCl/DMF 4:6	4.79	17.2	12.9	11.2	1.53	0.12
AmimCl/DMF 3:7	2.81	26.5	19.8	11.9	1.63	0.08
AmimCl/DMF 2:8	1.69	39.6	29.7	11.2	1.53	0.05

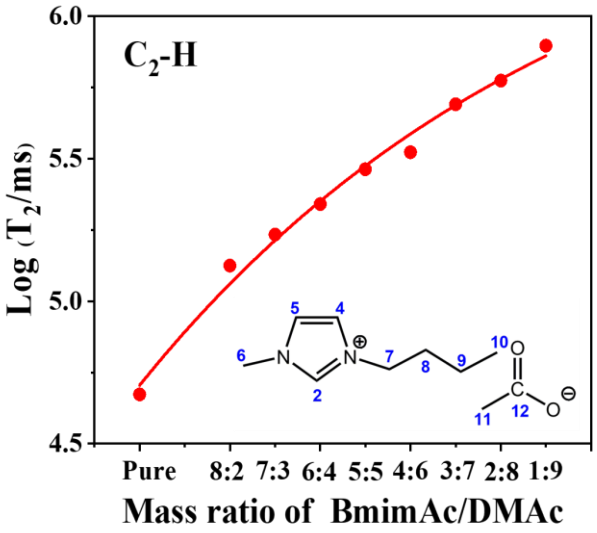

Fig. S9 Average T_2 values for C_2-H in BmimAc of the BmimAc/DMAc system.

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