Table S1 AMBER force field parameters for [emim][Gly]/H_2O mixture.[a]

nonbonded parameters			nonbonded parameters		
Atom	ε_{ii} (kJ·mol^{-1})	r_{ii} (Å)	Atom	ε_{ii} (kJ·mol^{-1})	r_{ii} (Å)
CR	0.3598	1.908	H1	0.0657	1.387
NA	0.7113	1.824	H	0.0657	0.600
CW	0.3598	1.908	CT	0.4577	1.908
H4	0.0628	1.409	C	0.3598	1.908
H5	0.0628	1.000	O2	0.8786	1.661
CT	0.4561	1.908	N3	0.7113	1.875
H1	0.0669	1.387	OW	0.6360	1.7683
HC	0.0669	1.487	HW	0.0000	0.0001
bonds	K_r (kJ·mol^{-1}·Å^2)	r_0 (Å)	bonds	K_r (kJ·mol^{-1}·Å^2)	r_0 (Å)
CT-CT	1297	1.53	CW-CW	1715	1.34
CT-H1	1423	1.08	CT-HC	1423	1.09
CW-H4	1611	1.07	CT-C	1326	1.55
CR-H5	1590	1.07	CT-N3	1536	1.46
CR-NA	1674	1.33	C-O2	2092	1.24
CW-NA	1506	1.38	N3-H	1883	1.00
CT-NA	1172	1.47	OW-HW	2314	0.96
angles	K_θ (kJ·mol^{-1}·rad^2)	θ(deg)	angles	K_θ (kJ·mol^{-1}·rad^2)	θ(deg)
CT-CT-H1	158.992	109.5	H4-CW-NA	125.980	122.1
CT-CT-HC	155.017	109.5	H5-CR-NA	125.980	125.7
CT-CT-NA	293.006	112.2	CW-CW-H4	125.980	130.7
H1-CT-H1	145.980	109.5	C-CT-N3	376.811	116.46
HC-CT-HC	142.005	109.5	C-CT-H1	242.839	108.05
H1-CT-NA	229.994	109.5	N3-CT-H1	251.207	108.84
CW-NA-CT	208.991	125.7	CT-C-O2	334.971	115.44
CR-NA-CT	208.991	126.3	O2-C-O2	355.891	129.11
CW-CW-NA	501.996	107.0	CT-N3-H	209.367	107.34
CR-NA-CW	501.996	108	H-N3-H	209.367	103.38
NA-CR-NA	501.996	109.9	HW-OW-HW	418.400	105.07
	Proper Torsions						
torsions	K_φ/2 (kJ·mol^{-1})	γ(deg)	n	torsions	K_φ/2 (kJ·mol^{-1})	γ(deg)	n
NA-CR-NA-CW	50.208	180	2	NA-CT-CT-HC	0.669	0	3
NA-CR-NA-CT	8.368	180	2	H1-CT-CT-HC	0.628	0	3
H5-CR-NA-CW	6.276	180	2	HC-CT-CT-HC	0.628	0	3
H5-CR-NA-CT	6.276	180	2	H1-CT-NA-CW	1.004	0	3
CW-CW-NA-CR	50.208	180	2	H1-CT-NA-CR	0.686	0	3
CW-CW-NA-CT	8.368	180	2	CT-CT-NA-CW	-0.745	0	1
H4-CW-NA-CR	8.368	180	2	CT-CT-NA-CR	-0.987	0	1
H4-CW-NA-CT	6.276	180	2	N3-CT-C-O2	0.962	0	2
NA-CW-CW-H4	6.276	180	2	H1-CT-C-O2	0.000	0	2
NA-CW-CW-NA	50.208	180	2	H1-CT-N3-H	0.921	0	3
H4-CW-CW-H4	6.276	180	2				

		Improper Torsions					
NA-NA-CR-H5	4.598	180	2	CR-CW-NA-CT	8.368	180	2
CW-NA-CW-H4	4.598	180	2	CT-O2-C-O2	43.932	180	2

[a]The labels of atoms refer to those used in Scheme 1.

Table S2 Values of surface tension, $\gamma(10^3, \mathrm{N{\cdot}m^{-1}})$, of ionic liquid [emim][Gly] of contained various amount of water.

w_{H2O}[a]	0.7693 %	0.9243 %	1.0799 %	1.2277 %	1.3773	0	*r*	*s*
γ	54.9	55.2	55.5	55.8	56.0	53.5	0.998	0.03

[a] w_{H2O} is the water mass fraction

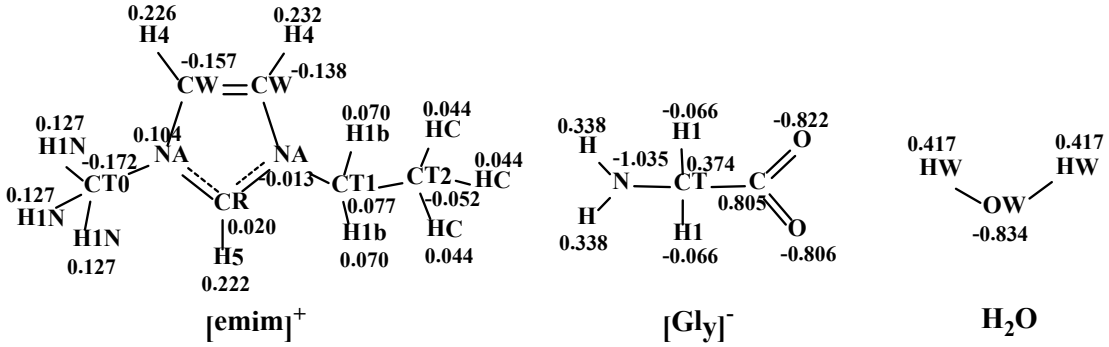


Fig. S1 RESP charge for $[emim]^+$, $[Gly]^-$ and H_2O.

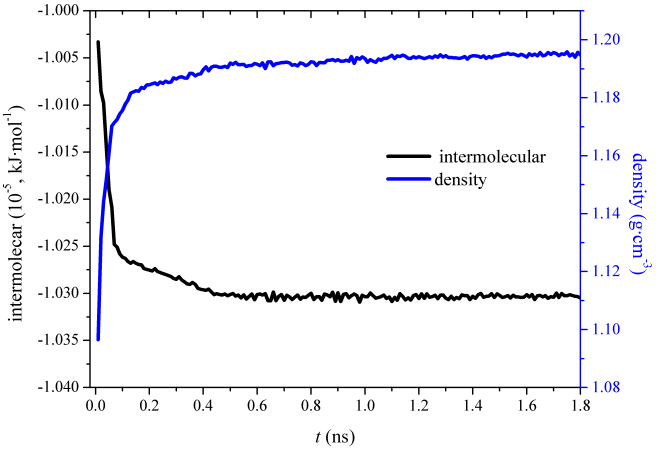
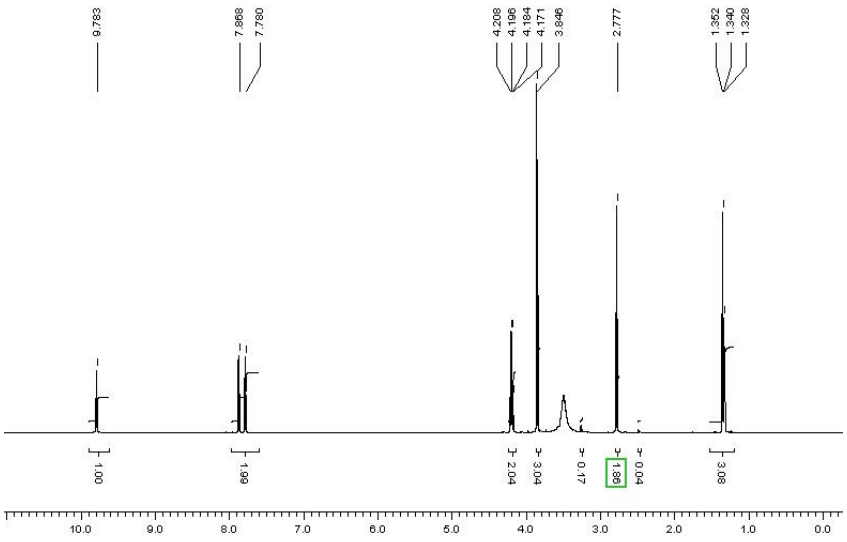


Fig. S2 The evolution of the pure [emim][Gly] system as a function of time.

Section A ^{1}H NMR spectra



The ^{1}H NMR spectrum δ_H (300 MHz，DMSO) of IL [emim][Gly]

chemical shift	hydrogen number	radical	chemical shift	hydrogen number	
1.328 ~ 1.352 (t)	3	$NCH_2\boldsymbol{CH_3}$	7.780 ~ 7.785 (d)	1	C(4)***H***
2.777(s)	2	$NH_2\boldsymbol{CH_2}$	7.863 ~ 7.868 (d)	1	C(5)***H***
3.846 (s)	3	$N\boldsymbol{CH_3}$	9.783 (s)	1	C(2)***H***
4.171 ~ 4.208 (m)	2	$N\boldsymbol{CH_2}$			

Section B DSC trace of IL [emim][Gly]. Calorimetric data were obtained with a differential scanning calorimeter DSC (Mettler-Toledo Co., Switzerland). The temperature range was −120 ~ 140 °C with heating rate of 10°C/min. Then samples were incubated at −120 °C for 5 min and were then heated to 140 °C.

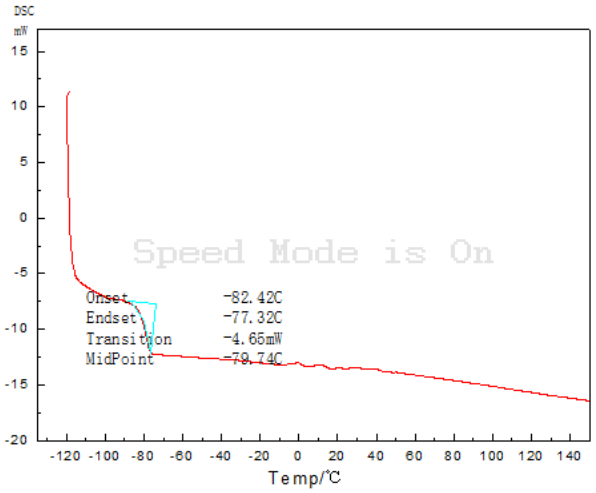


Fig. S3 ^{1}H NMR spectra (section A) and thermal analysis (section B)

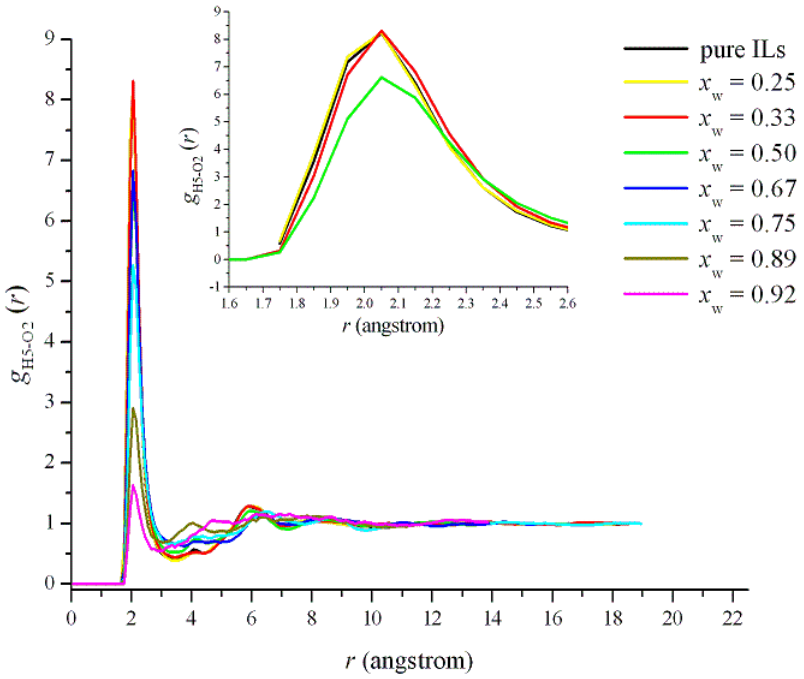


Fig. S4 Atomic partial radial distribution between H5 atom of $[emim]^+$ and O2 atom of $[Gly]^-$. The focus curves correspond to the different water mole fraction.

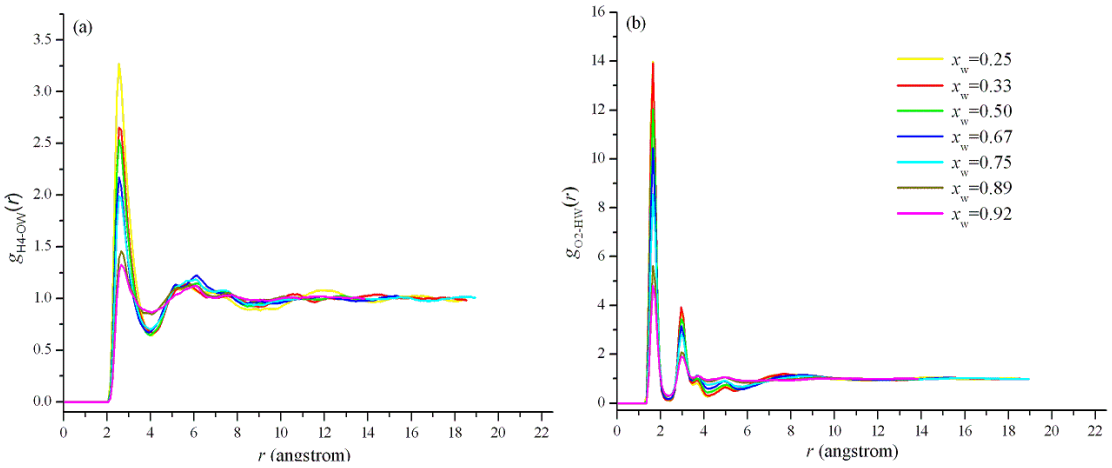


Fig. S5 Atomic partial radial distribution between H4 atom of $[emim]^+$ and OW atom of H_2O (a) and O2 atom of $[Gly]^-$ and OW atom of H_2O (b). The focus curves correspond to the different water mole fraction.