

## Electronic Supplementary Information for

### Ultra-stretchable Ion Gels Based on Physically Cross-linked Polymer Networks

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**Table S1.** Feed ratio of P(EA-co-AA).

Sample	AIBN (mg)	EA (g)	AA (g)	Dioxane (mL)
P(EA-co-AA)-1	10.0	5.25	0.540	12
P(EA-co-AA)-2	10.0	5.00	0.720	12
P(EA-co-AA)-3	10.0	4.50	1.08	12
P(EA-co-AA)-4	10.0	4.00	1.44	12
P(EA-co-AA)-5	10.0	3.00	2.16	12
P(EA-co-AA)-6	10.0	2.00	2.88	12

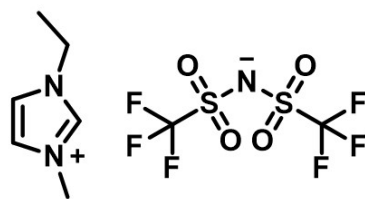
**Table S2.** Sample composition and tensile properties of different P(EA-co-AA-co-AANa) ion gels.

Sample <sup>a)</sup>	Polymer <sup>b)</sup>	NaOH (mg)	EA (mol%)	AA (mol%)	AANa (mol%)	Stress (kPa)	Strain (%)
78-22-0	P(EA-co-AA)-3	0.00	78	22	0	-	-
78-19-3	P(EA-co-AA)-3	5.00	78	19	3	-	-
78-16-6	P(EA-co-AA)-3	10.0	78	16	6	60	> 6500
78-10-12	P(EA-co-AA)-3	20.0	78	10	12	980	2600
78-5-17	P(EA-co-AA)-3	30.0	78	5	17	1100	790
78-0-22	P(EA-co-AA)-3	40.0	78	0	22	1270	470
88-0-12	P(EA-co-AA)-1	20.0	88	0	12	180	2700
85-3-12	P(EA-co-AA)-2	20.0	85	3	12	740	2500
70-18-12	P(EA-co-AA)-4	20.0	70	18	12	2600	910
52-36-12	P(EA-co-AA)-5	20.0	52	36	12	-	-
38-50-12	P(EA-co-AA)-6	20.0	38	50	12	-	-

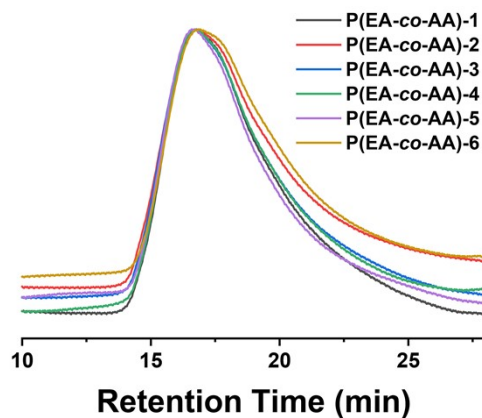
<sup>a)</sup> Named based on the mole percentages of the three monomers, <sup>b)</sup> 400 mg of polymer in each sample.

**Table S3.** Parameters for the VTF fitting of the conductivity data

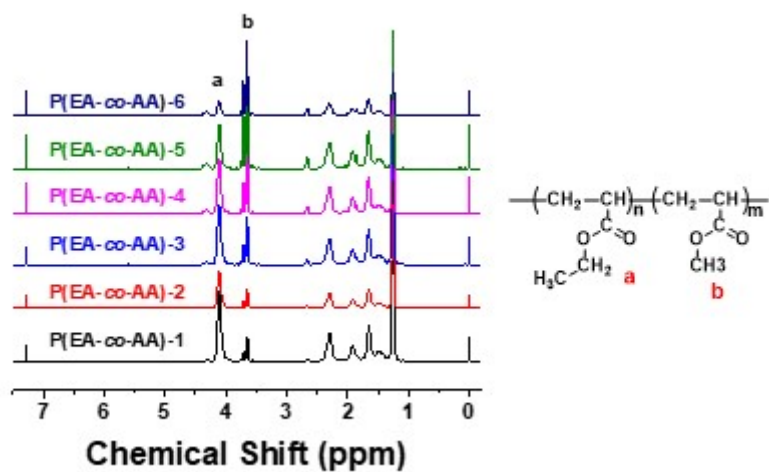
$\sigma_0$ (S cm <sup>-1</sup> )	$E_a$ (kJ mol <sup>-1</sup> )	$T_0$ (K)
0.086	2.04	157



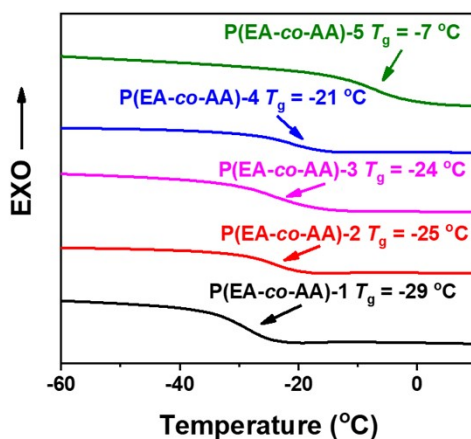
**Scheme S1.** Chemical structure of [EMIM][TFSI].



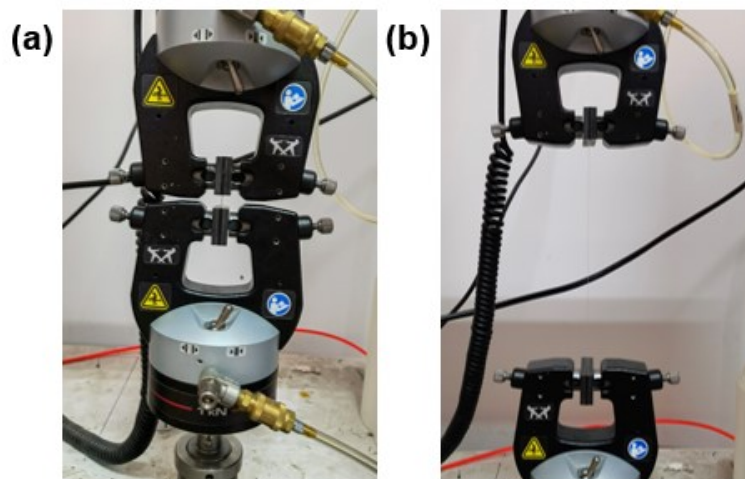
**Figure S1.** GPC curves of P(EA-co-AA).



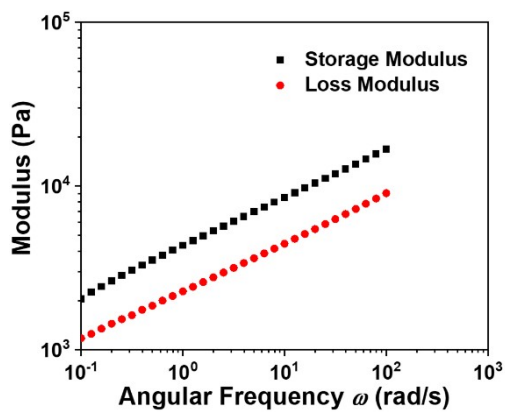
**Figure S2.**  $^1\text{H}$  NMR spectra of P(EA-co-AA) samples with  $\text{CDCl}_3$  as the solvent.



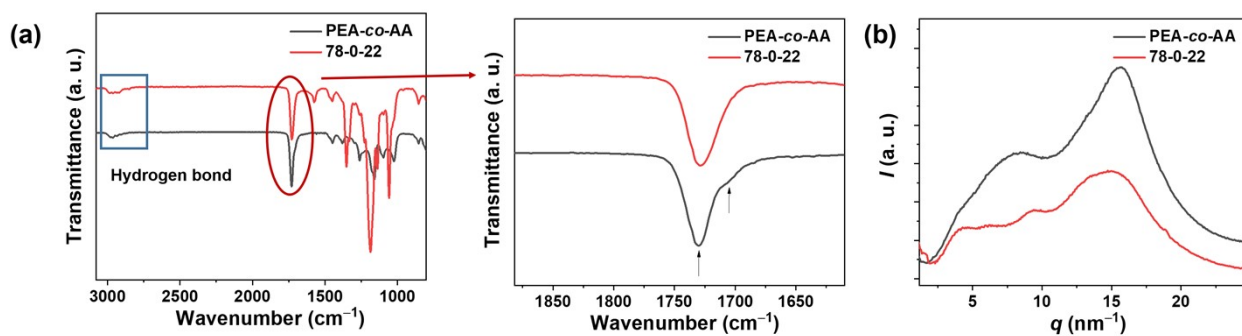
**Figure S3.** DSC thermograms of P(EA-co-AA) with different EA/AA ratios.



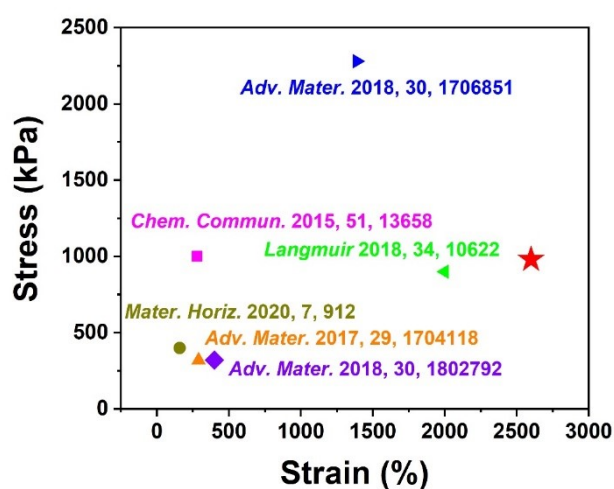
**Figure S4.** Photographs of the P(EA-co-AA-co-AANa) ion gel before (a) and after (b) stretching.



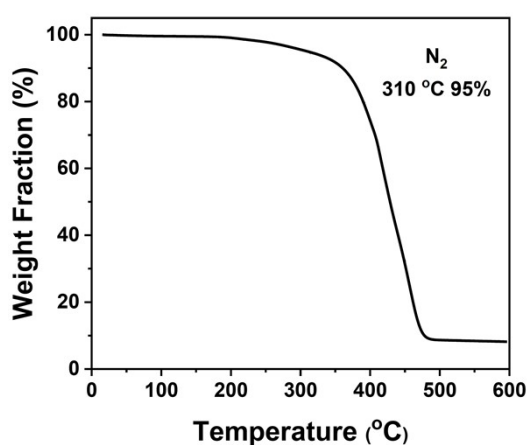
**Figure S5.** Storage and loss moduli of the ion gel from sample 78-16-6.



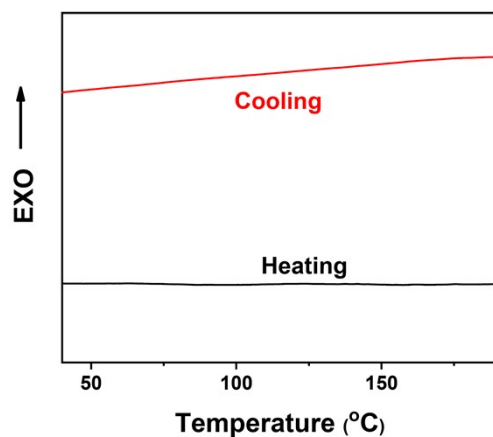
**Figure S6.** FTIR (a) and WAXS (b) results of PEA-co-AA and 78-0-22.



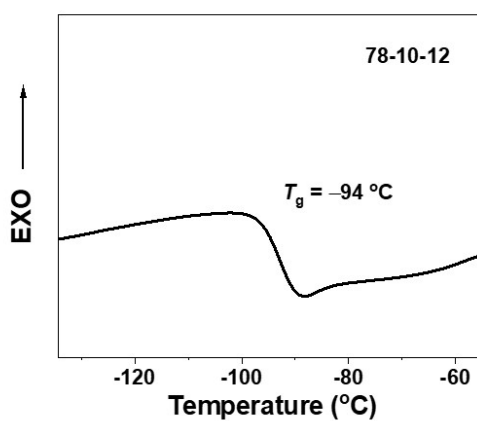
**Figure S7.** Breaking elongation and failure tensile stress of high-performance ion gels reported in recent years.<sup>1-6</sup> The red star indicates ion gel from sample 78-10-12 in this work.



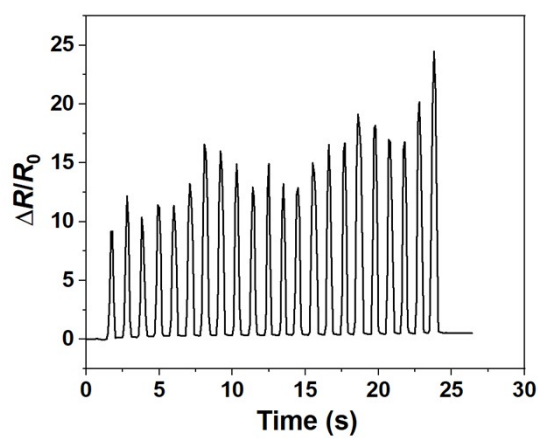
**Figure S8.** Thermogravimetric curve of the P(EA-co-AA-co-AANa) ion gel (78-10-12) at a heating rate of  $10\text{ }^{\circ}\text{C min}^{-1}$  under nitrogen.



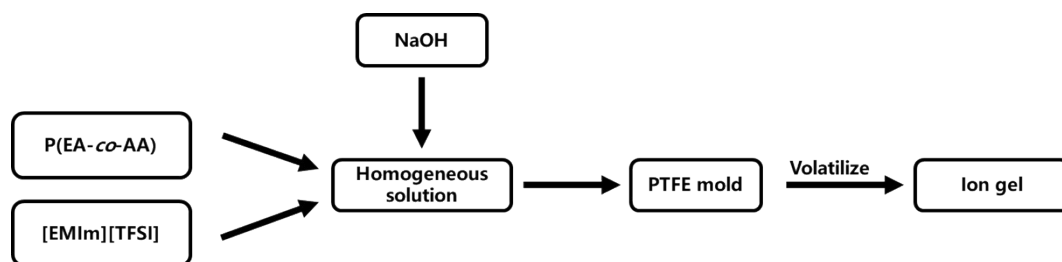
**Figure S9.** DSC thermogram of the P(EA-co-AA-co-AANa) ion gel at a heating and cooling rate of  $20\text{ }^{\circ}\text{C min}^{-1}$  under a nitrogen atmosphere.



**Figure S10.** DSC thermogram of the ion gel from sample 78-10-12 at a heating and cooling rate of  $20\text{ }^{\circ}\text{C min}^{-1}$  under a nitrogen atmosphere.



**Figure S11.** Sensing performance of the strain sensor in large strains.



**Figure S12.** Schematic illustration of the preparation of the P(EA-co-AA-co-AANa) ion gel.

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