Electronic Supplementary Material (ESI) for RSC Advances.

Supporting Information

Mechanical and self-recovery properties of supramolecular ionic liquid elastomers based on host-guest interaction and correlation with ionic liquid content

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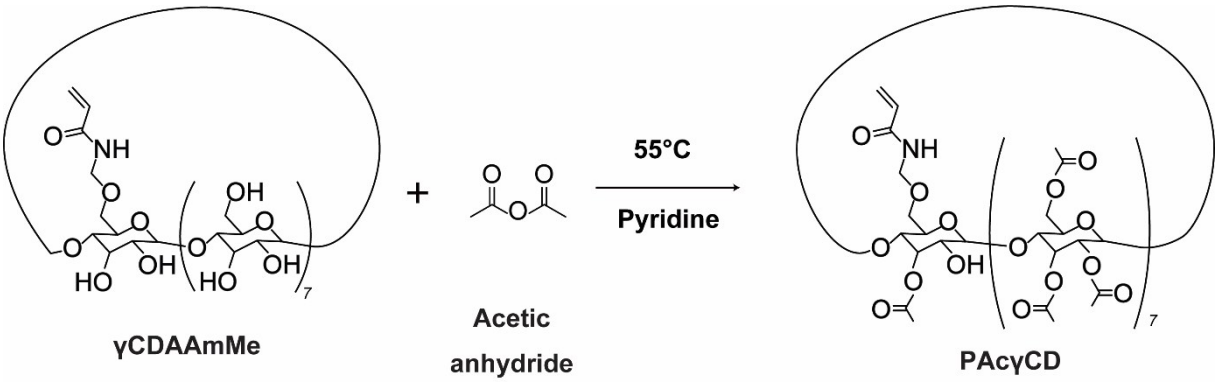
References

1. Preparation of peracetylated 6–acrylamido methylether–γCD (PAcγCD)[1]



Scheme S1. Preparation of PacγCD.

γCDAAmMe[2] (20 g, 15 mmol) and acetic anhydride (0.17 kg, 1.7 mol) were dissolved in pyridine (0.30 L) and stirred at 55 ºC for overnight. The solution was cooled to room temperature and put in ice bath and then methanol (50 mL) was added to quench the reaction. The solution was evaporated until 0.20 L and then precipitated in cooled water (1.5 L). Precipitate was filtered and dissolved again in acetone (0.20 L) and then precipitated again in cooled water (1.5 L). After second filtration, the precipitate was washed twice with water (0.50 L) and then dried under reduced pressure at 40 ºC for 48 hours. Yield: 81%.

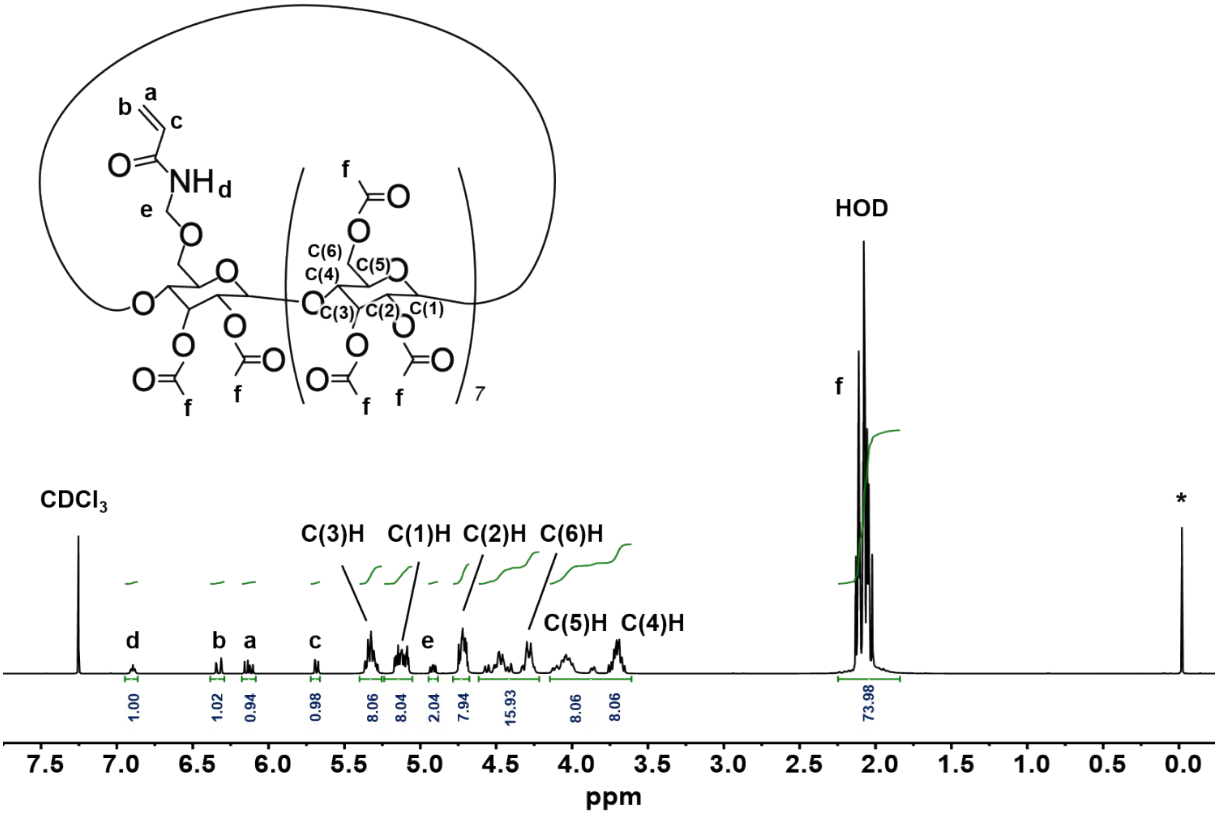


Figure S1. 500 MHz ^{1}H NMR spectrum of PAcγCD in chloroform-*d*.

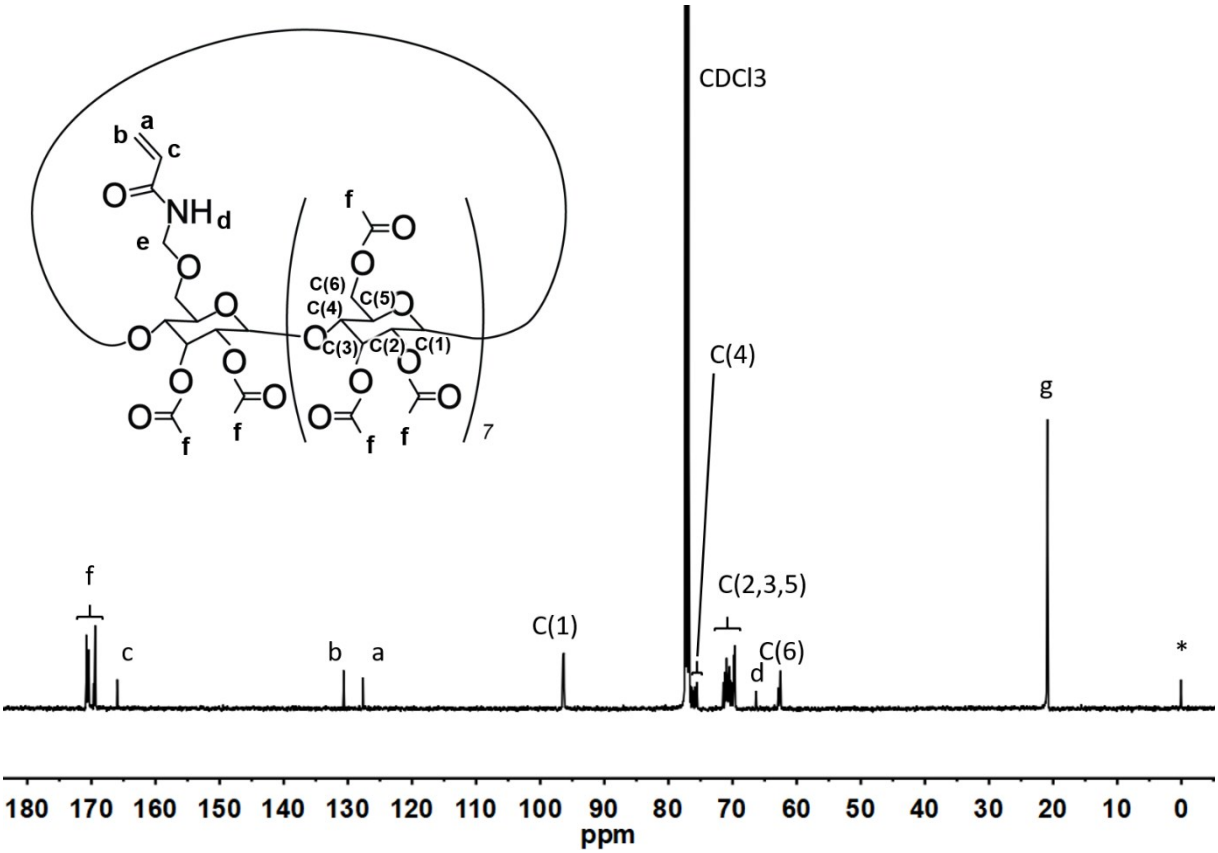


Figure S2. 125 MHz ^{13}C NMR spectrum of PAcγCD in chloroform-*d*.

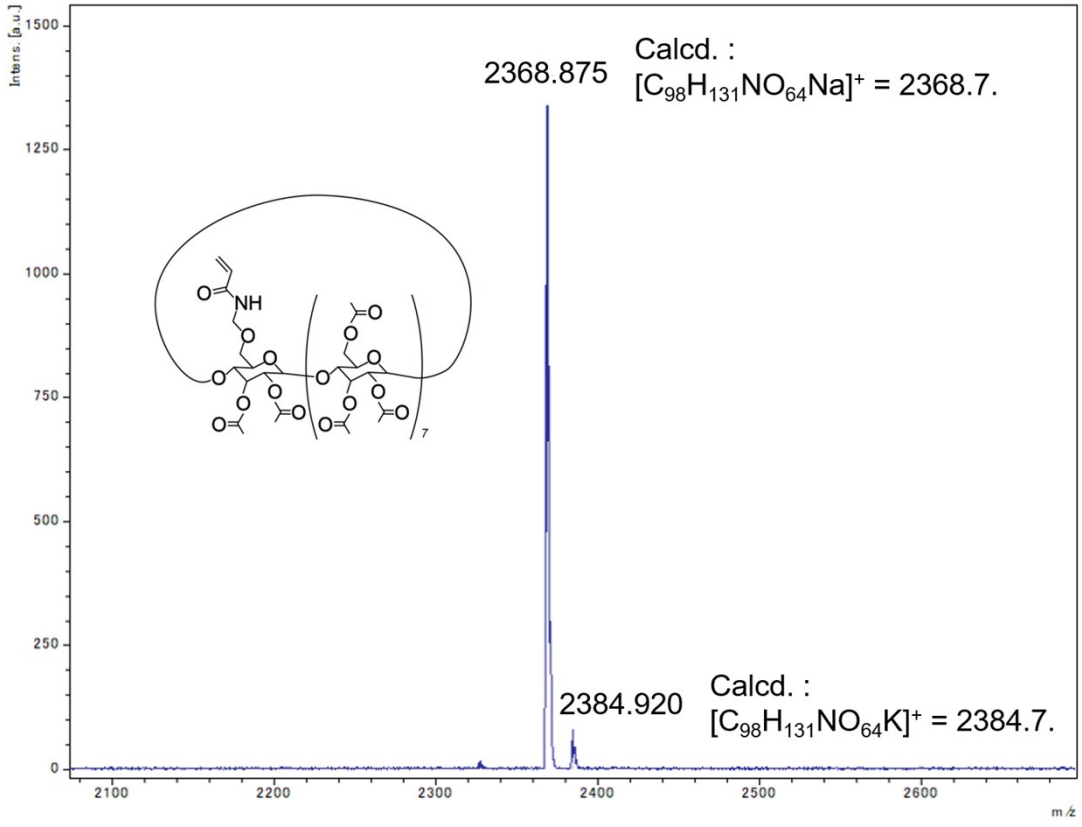
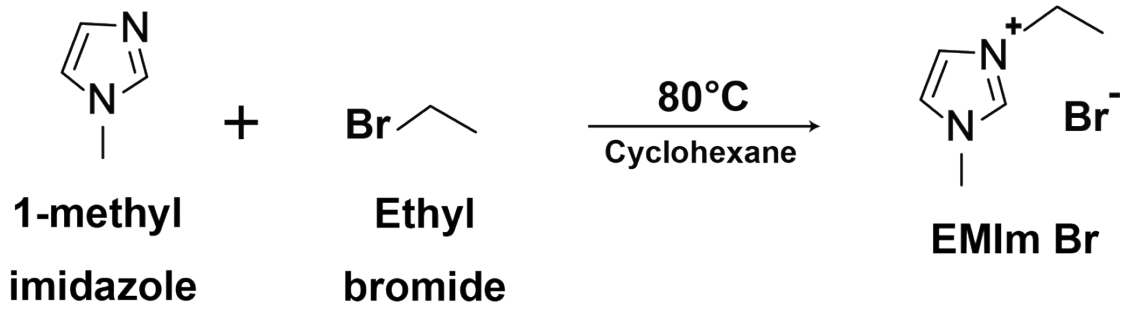


Figure S3. MALDI TOF mass spectrum of PAcγCD.

2. Preparation of ionic liquid (EMIm TFSI)[3]

Preparation of 1-ethyl-3-methylimidazolium bromide (EMIm Br)



Scheme S2. Preparation of EMIm Br.

1-Methyl imidazole (37 g, 0.50 mol) and ethyl bromide (0.15 kg, 1.4 mol) were dissolved in cyclohexane (0.20 L) and the solution was stirred at 80 °C for 24 h. After reaction, the mixture separated into 2 layers and then poured into beaker glass to room temperature. Lower layer turned into solid phase and then poured away the liquid phase. Solid phase was recrystallized using ethyl acetate : 2-propanol (1:1 by volume). The crystalized sample was melted at 83 °C for 24 h under vacuum to give EMIm Br. Yield: 91%.

^{1}H NMR (500 MHz, DMSO-d_6) of EMIm Br: δ = 9.37 (s, 1H,-NC*H*N-), 7.86 (t, *J* = 1.8 Hz, 1H,-NC*H*CH-), 7.76 (t, *J* = 1.8 Hz, 1H,-NCHC*H*-), 4.19 (q, *J* = 7.3 Hz, 2H,-NCH_2-), 3.84 (s, 3H,-NCH_3), 1.36 (t, *J* = 7.3 Hz, 3H, -CH_2CH_3).

^{13}C NMR (125 MHz, DMSO-d_6) of EMIm Br: δ = 136.8, 124.0, 122.5, 44.6, 36.3, 15.7.

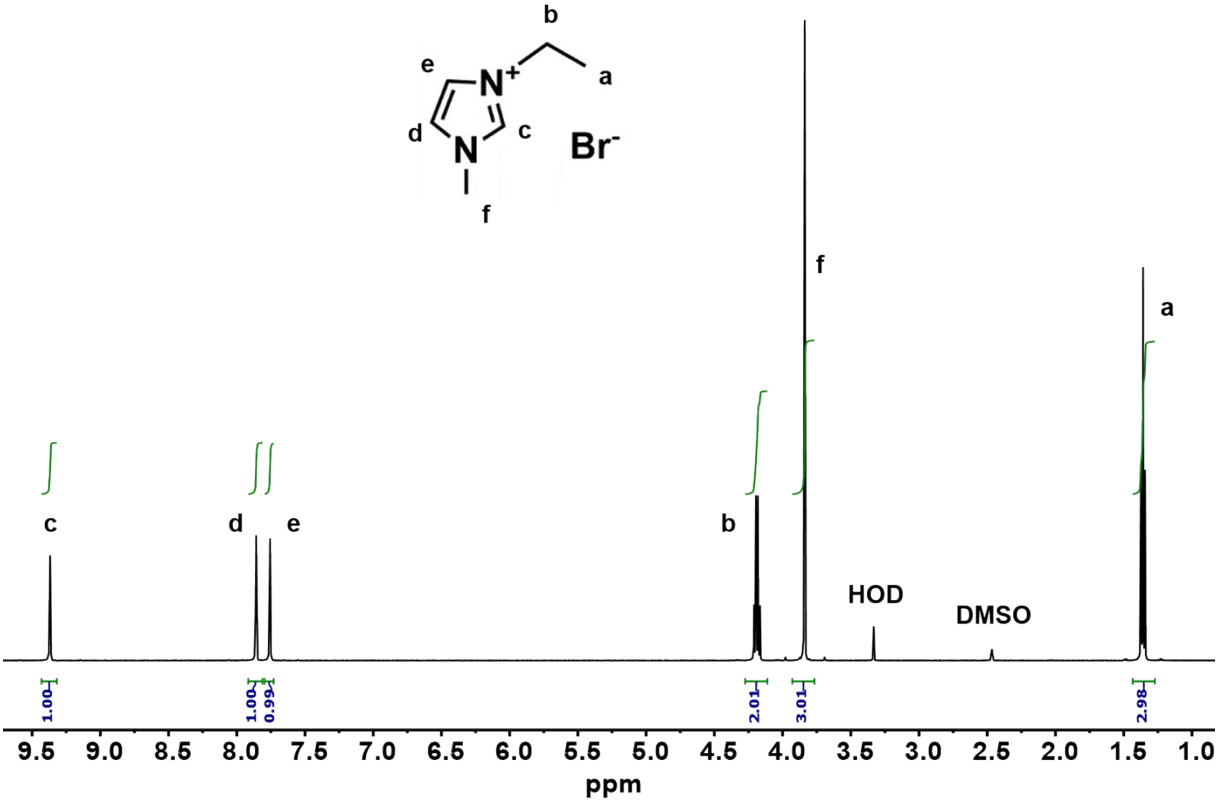


Figure S4. 500 MHz ^{1}H NMR spectrum of EMIm Br in DMSO–d_6.

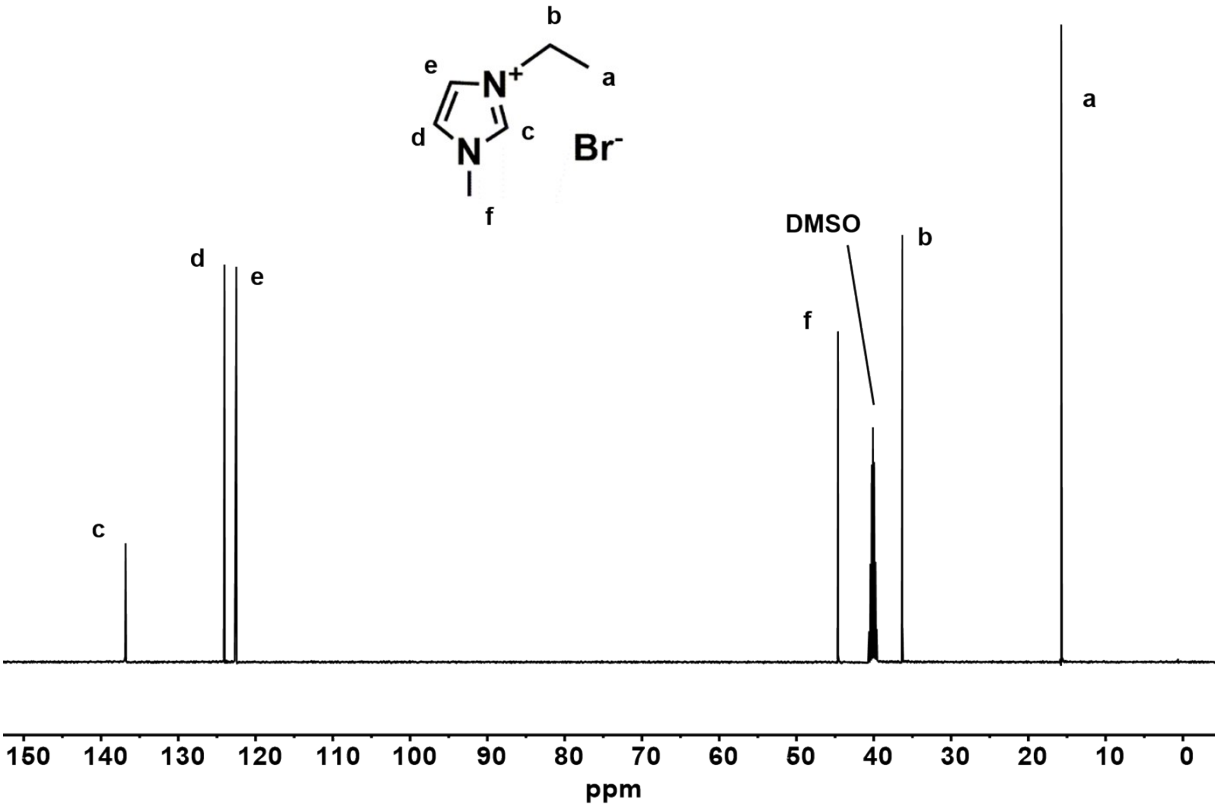
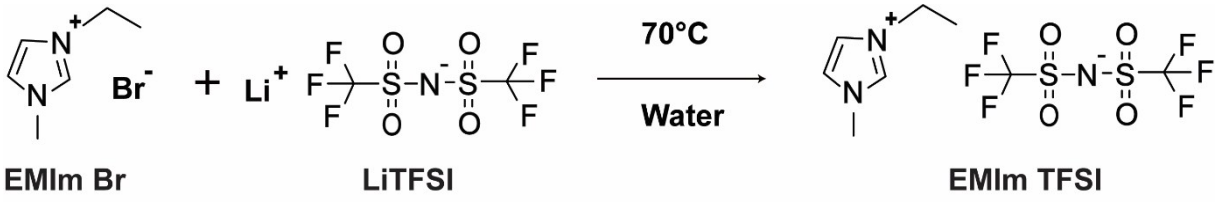


Figure S5. 125 MHz ^{13}C NMR spectrum of EMIm Br in DMSO–d_6.

Preparation of 1-ethyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide (EMIm TFSI)



Scheme S3. Preparation of EMIm TFSI.

EMIm Br (5.0 g, 26 mmol) was dissolved in water (10 mL), and then bis(trifluoromethylsulfonyl)imide lithium salt (LiTFSI, 8.3 g, 29 mmol) was also dissolved in water (10 mL). These solutions were mixed together and stirred at 70 °C for 24 h. The mixture separated into 2 layers and poured into separating funnel. The lower layer was washed three times with water. After washing, it was dried under reduced pressure at 120 °C for 72 h to obtain EMIm TFSI ionic liquid. Yield: 84%.

^{1}H NMR (500 MHz, DMSO-d_6) of EMIm TFSI: δ = 9.06 (s, 1H,-NC*H*N-), 7.71 (t, *J* = 1.8 Hz, 1H,-NC*H*CH-), 7.63 (t, *J* = 1.8 Hz, 1H,-NCHC*H*-), 4.20 (q, *J* = 7.3 Hz, 2H,-NCH_2-), 3.83 (s, 3H,-NCH_3), 1.42 (t, *J* = 7.3 Hz, 3H,-CH_2CH_3).

^{13}C NMR (125 MHz, DMSO-d_6) of EMIm TFSI: δ = 136.76, 124.05, 122.43, 121.31, 118.75, 44.67, 36.12, 15.42.

^{19}F NMR (470 MHz, DMSO-d_6) of EMIm TFSI: δ = -78.80.

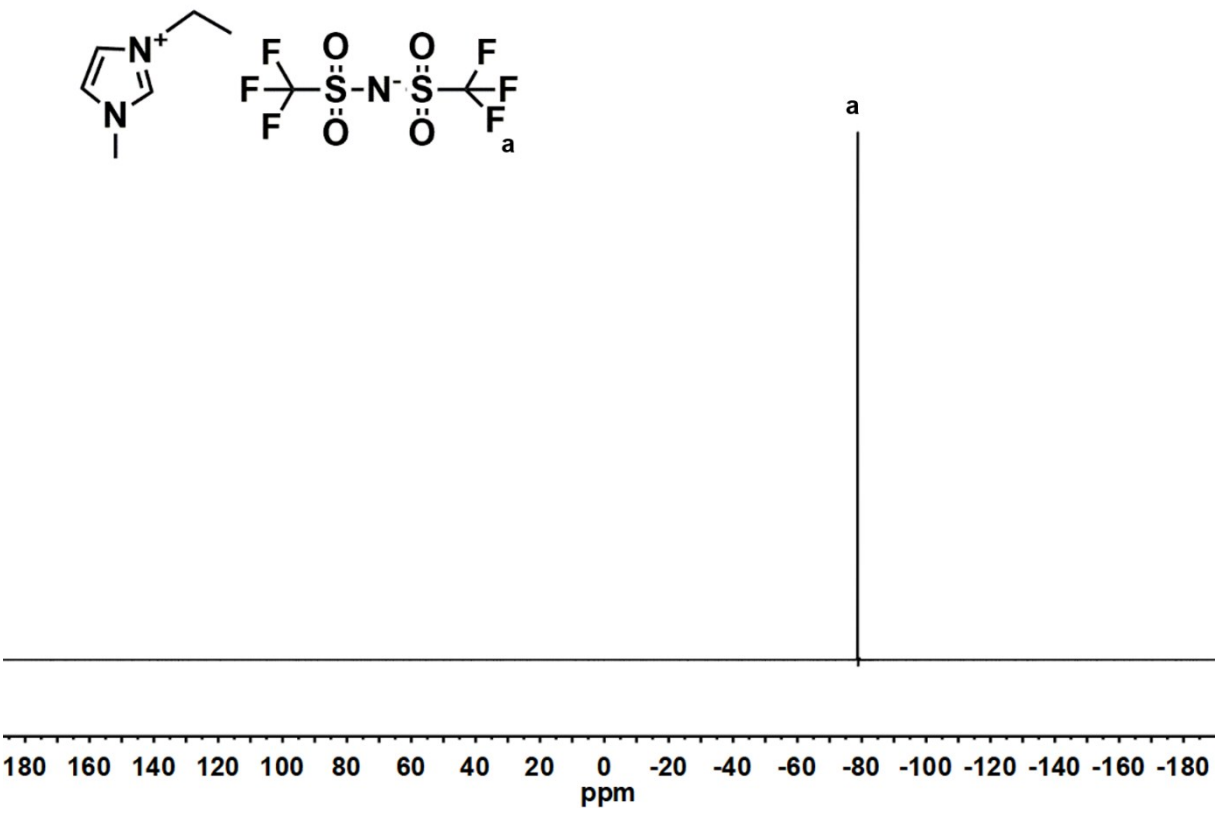


Figure S6. 470 MHz ^{19}F NMR spectrum of EMIm TFSI in DMSO–d_6.

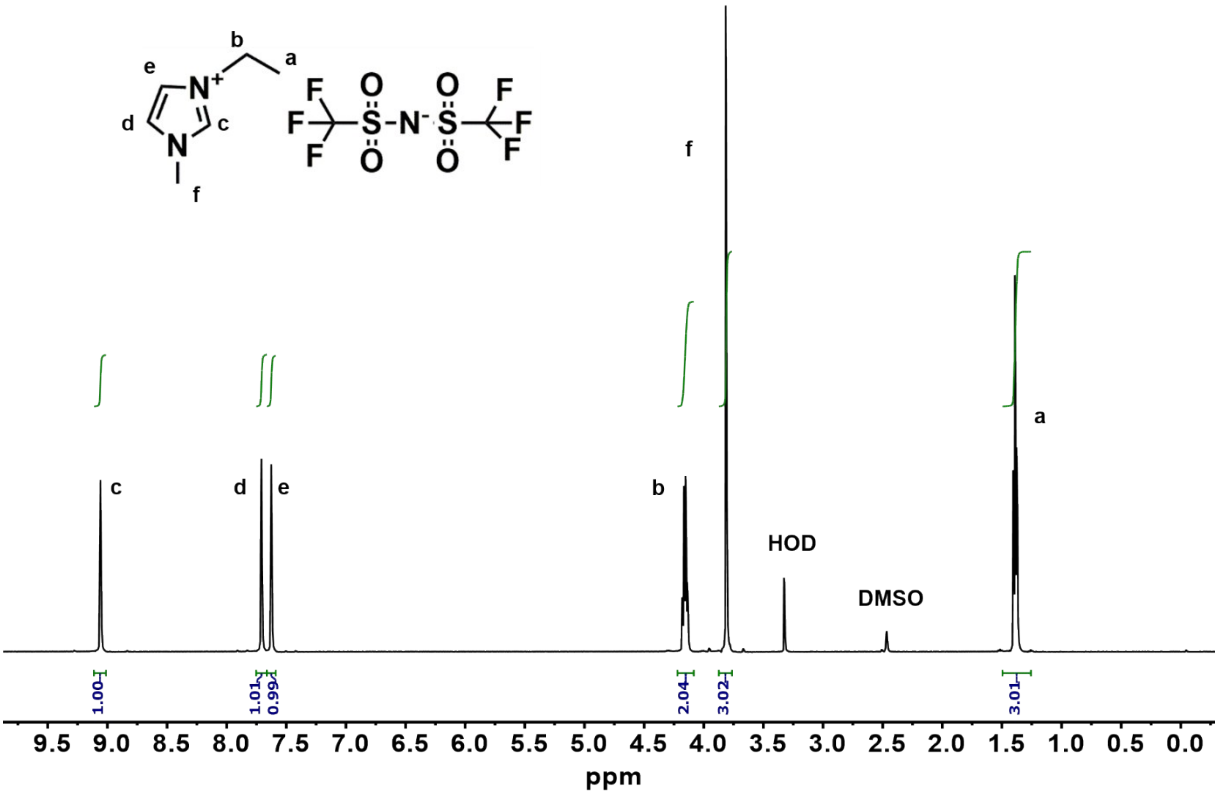


Figure S7. 500 MHz ^{1}H NMR spectrum of EMIm TFSI in DMSO–d_6.

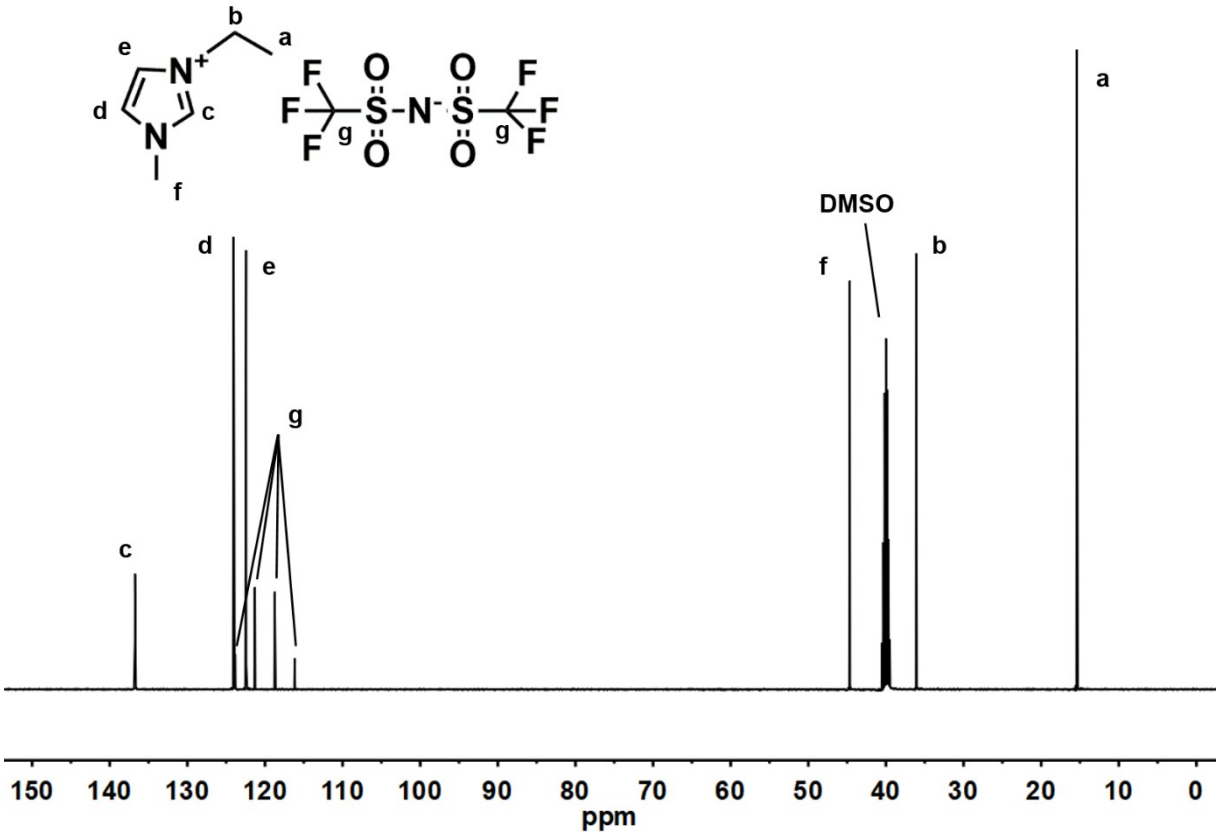
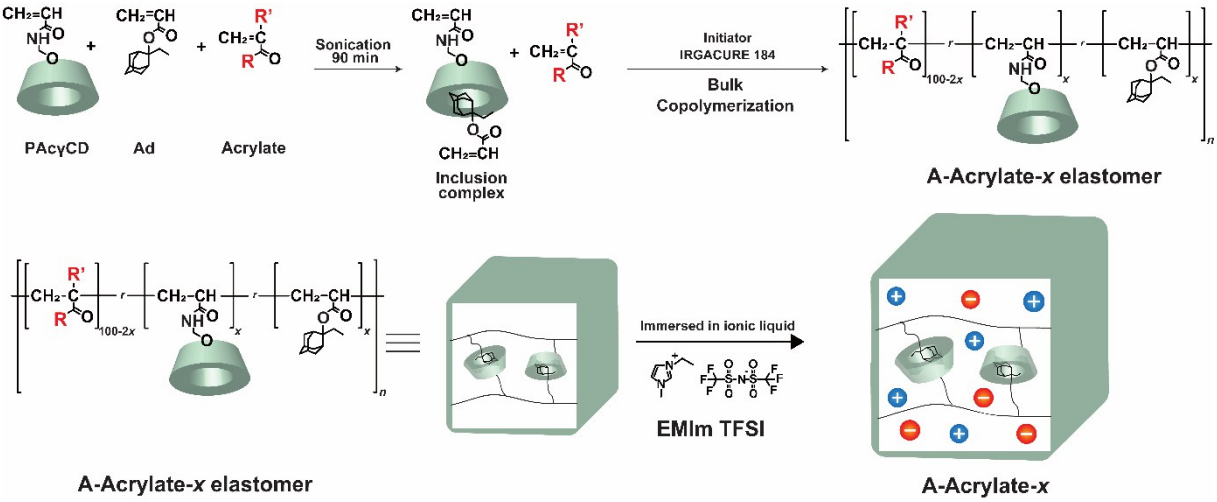


Figure S8. 125 MHz ^{13}C NMR spectrum of EMIm TFSI in DMSO–d_6.

3. Schematic preparation of supramolecular ionic liquid elastomers (A-Acrylate-*x*)



Scheme S4. Preparation of the **A-Acrylate-*x*** elastomer by bulk polymerization then the **A-Acrylate-*x* elastomer** was immersed in the EMIm TFSI to become **A-Acrylate-*x***, and *x* indicate the mol% of cross-linker from host guest inclusion complex between PAcγCD and Ad units. Group of side chain monomers (Acrylate): methyl acrylate (1), ethyl acrylate (2), butyl acrylate (3), and methyl methacrylate (4).

4. Preparation of elastomers

Table S1. Preparation of **A-1-*x* elastomer**.

x	PAcγCD / g	Ad / g	1 / g	IRGACURE 184 / g
0.5	0.69	0.07	5.00	0.02
1	1.38	0.14	5.00	0.02
2	2.78	0.28	5.00	0.02

Table S2. Preparation of **A-2-*x* elastomer**.

x	PAcγCD / g	Ad / g	2 / g	IRGACURE 184 / g
0.5	0.59	0.06	5.00	0.02
1	1.20	0.12	5.00	0.02
2	2.44	0.24	5.00	0.02

Table S3. Preparation of **A-3-*x* elastomer**.

x	PAcγCD / g	Ad /g	3 / g	IRGACURE 184 / g
0.5	0.46	0.05	5.00	0.02
1	0.93	0.09	5.00	0.02
2	1.91	0.19	5.00	0.02

Table S4. Preparation of **A-4-*x* elastomer**.

x	PAcγCD / g	Ad /g	4 / g	IRGACURE 184 / g
0.5	0.59	0.06	5.00	0.02
1	1.20	0.12	5.00	0.02
2	2.44	0.24	5.00	0.02

Table S5. Preparation of **B-1-*y* elastomer**.

y	BDA / g	1 / g	IRGACURE 184 / g
0.5	0.06	5.00	0.02
1	0.12	5.00	0.02
2	0.23	5.00	0.02

Table S6. Preparation of **B-2-*y* elastomer**.

y	BDA / g	2 / g	IRGACURE 184 / g
0.5	0.05	5.00	0.02
1	0.10	5.00	0.02
2	0.20	5.00	0.02

Table S7. Preparation of **B-3-*y* elastomer**.

y	BDA / g	3 / g	IRGACURE 184 / g
0.5	0.04	5.00	0.02
1	0.08	5.00	0.02
2	0.16	5.00	0.02

Table S8. Preparation of **B-4-*y* elastomer**.

y	BDA / g	4 / g	IRGACURE 184 / g
0.5	0.05	5.00	0.02
1	0.10	5.00	0.02
2	0.20	5.00	0.02

5. Chemical structures and abbreviation of ionic liquid elastomers

Table S9. Chemical structures of supramolecular ionic liquid elastomers (**A-Acrylate-*x***)

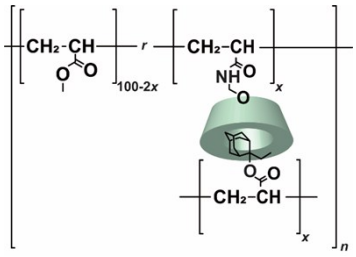
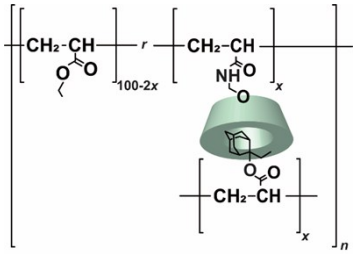
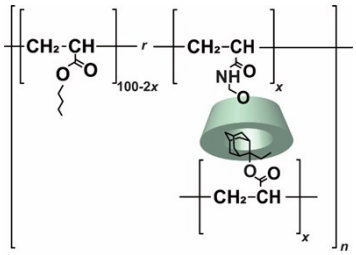
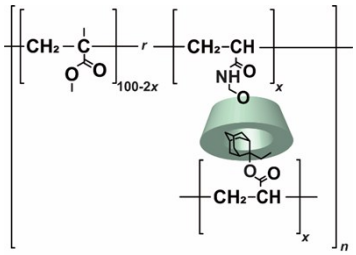
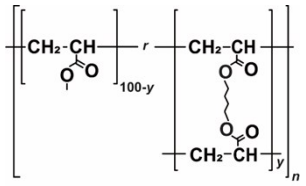
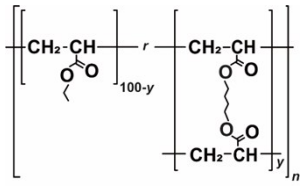
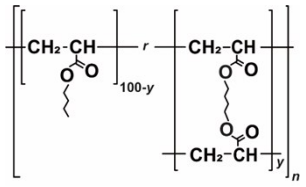
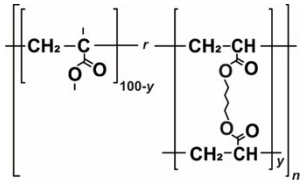
Chemical structures	%mol cross-linker (*x*)	Abbreviation
 **A-1-*x***	0.5	A-1-0.5
	1	A-1-1
	2	A-1-2
 **A-2-*x***	0.5	A-2-0.5
	1	A-2-1
	2	A-2-2
 **A-3-*x***	0.5	A-3-0.5
	1	A-3-1
	2	A-3-2
 **A4-*x***	0.5	A-4-0.5
	1	A-4-1
	2	A-4-2

Table S10. Chemical structures of chemically cross-linked ionic liquid elastomers (**B-Acrylate-*y***)

Chemical structures	%mol cross-linker (*y*)	Abbreviation
 **B-1-*y***	**0.5**	**B-1-0.5**
	1	**B-1-1**
	2	**B-1-2**
 **B-2-*y***	**0.5**	**B-2-0.5**
	1	**B-2-1**
	2	**B-2-2**
 **B-3-*y***	**0.5**	**B-3-0.5**
	1	**B-3-1**
	2	**B-3-2**
 **B4-*y***	**0.5**	**B-4-0.5**
	1	**B-4-1**
	2	**B-4-2**

6. Characterization of elastomers and ionic liquid elastomers

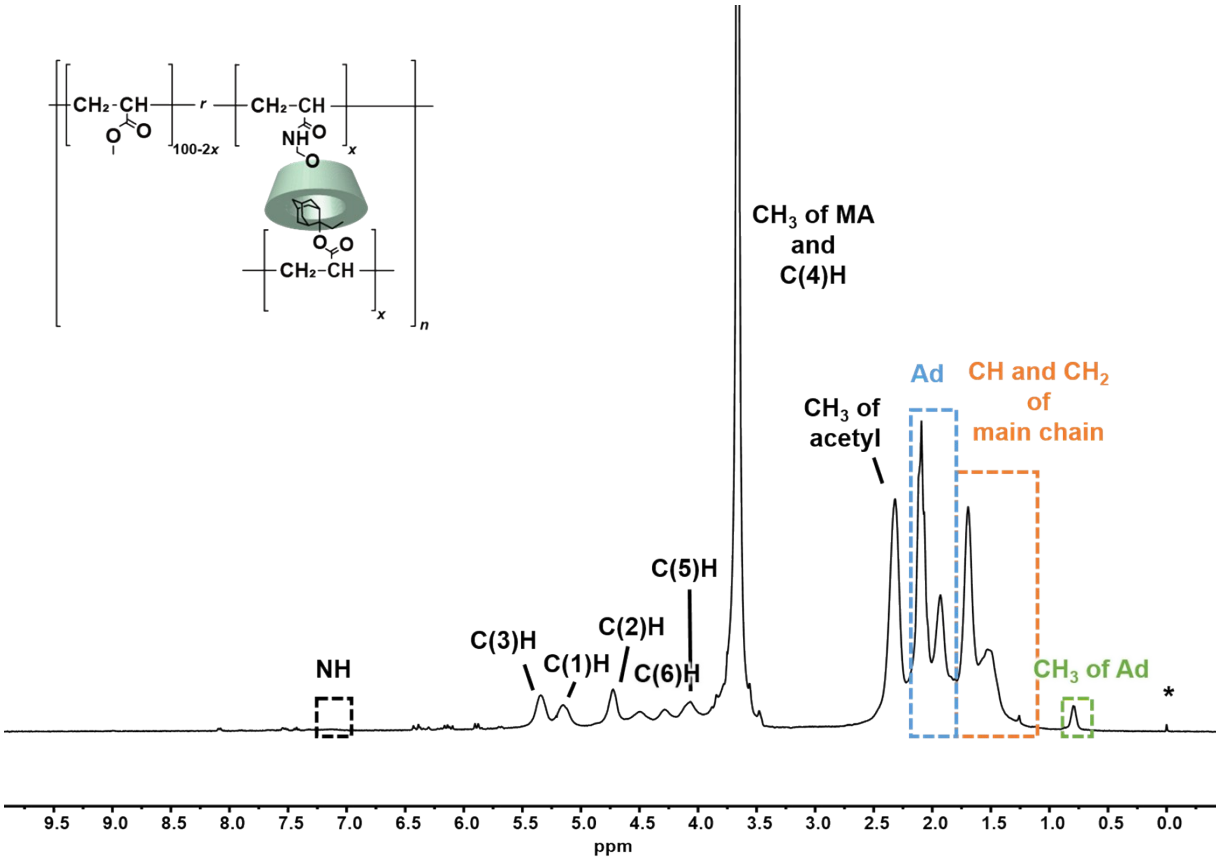


Figure S9. Solid-state ^{1}H FGMAS NMR spectrum of **A-1-1 elastomer** (TMS for standard, 400 MHz, 25 ^{0}C, rotation frequency = 7 kHz).

^{1}H NMR (400 MHz, Chloroform-*d*): δ = 7.05 (-N*H*-), 5.42-5.28 (C(3)*H* of CD), 5.19-5.11 (C(1)*H* of CD), 4.81-4.66 (C(2)*H* of CD), 4.50-4.29 (C(6)*H* of CD), 4.13-4.01 (C(5)*H* of CD), 3.85-3.50 (-CH_3 of MA and C(4)*H* of CD), 2.45-2.20 (-CH_3 of acetyl), 2.15-1.88 (-C*H*-, -CH_2-, and -CH_2CH$_3$ of Ad), 1.78-1.17 (-CH_2C*H*- of side chain), 0.83-0.73 (-CH_3 of Ad).

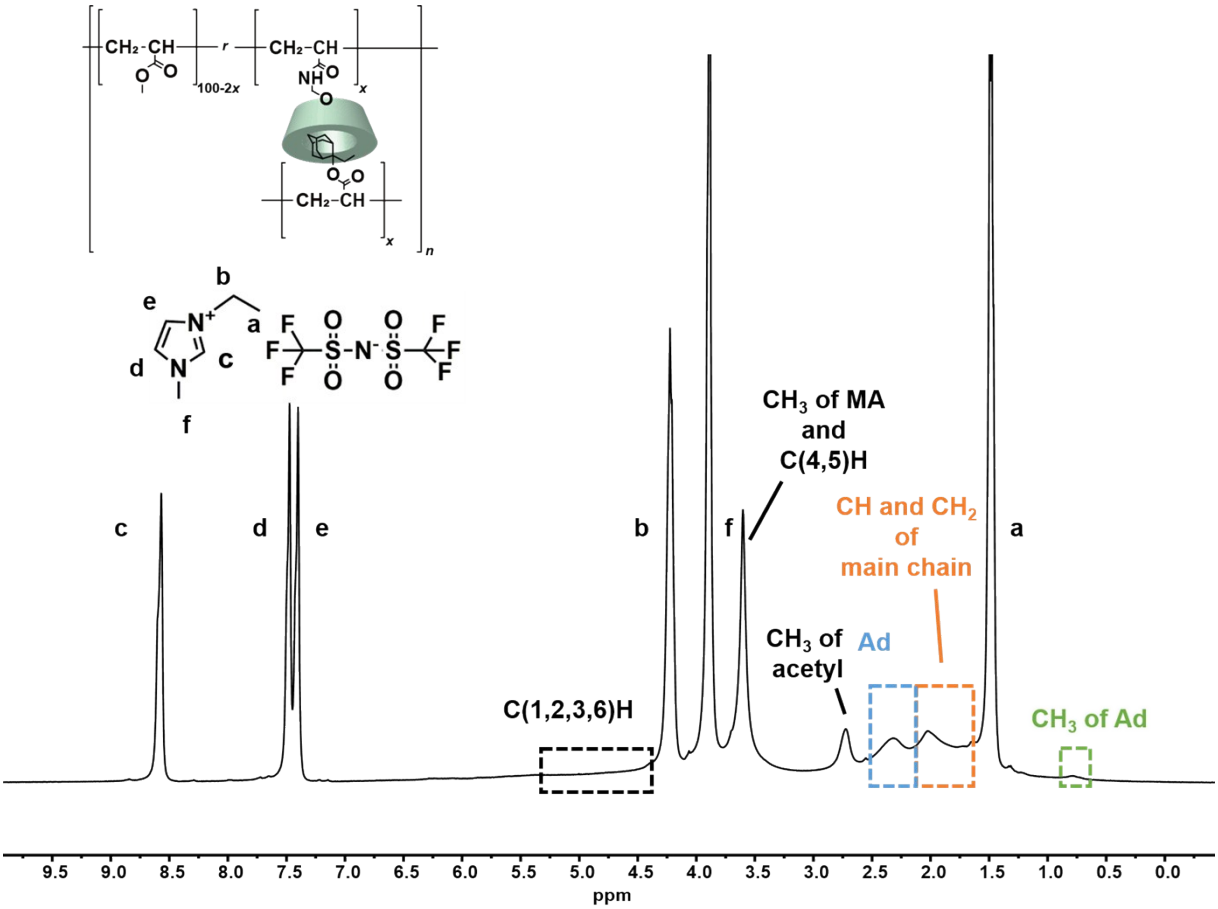


Figure S10. Solid-state ^{1}H FGMAS NMR spectrum of **A-1-1** (TMS for standard, 400 MHz, 25 ^{0}C, rotation frequency = 7 kHz).

^{1}H NMR (400 MHz, Chloroform-*d*): δ = δ 8.58 (-NC*H*N^{+}- of EMIm TFSI), 7.49 (-NC*H*CH- of EMIm TFSI), 7.40 (-N^{+}C*H*CH- of EMIm TFSI), 5.42-4.29 (C(1,2,3,6)*H* of CD), 4.22 (-N^{+}CH_2- of EMIm TFSI), 3.90 (-NCH_3 of EMIm TFSI), 3.70-3.43 (-CH_3 of MA and C(4)*H* of CD), 2.79-2.55 (-CH_3 of acetyl), 2.51-2.13 (-C*H*-, -CH_2-, and -CH_2CH$_3$ of Ad), 2.10-1.59 2.02 (-CH_2C*H*- of side chain), 1.48 (-CH$_2$CH_3 of EMIm TFSI), 0.88-0.68 (-CH_3 of Ad).

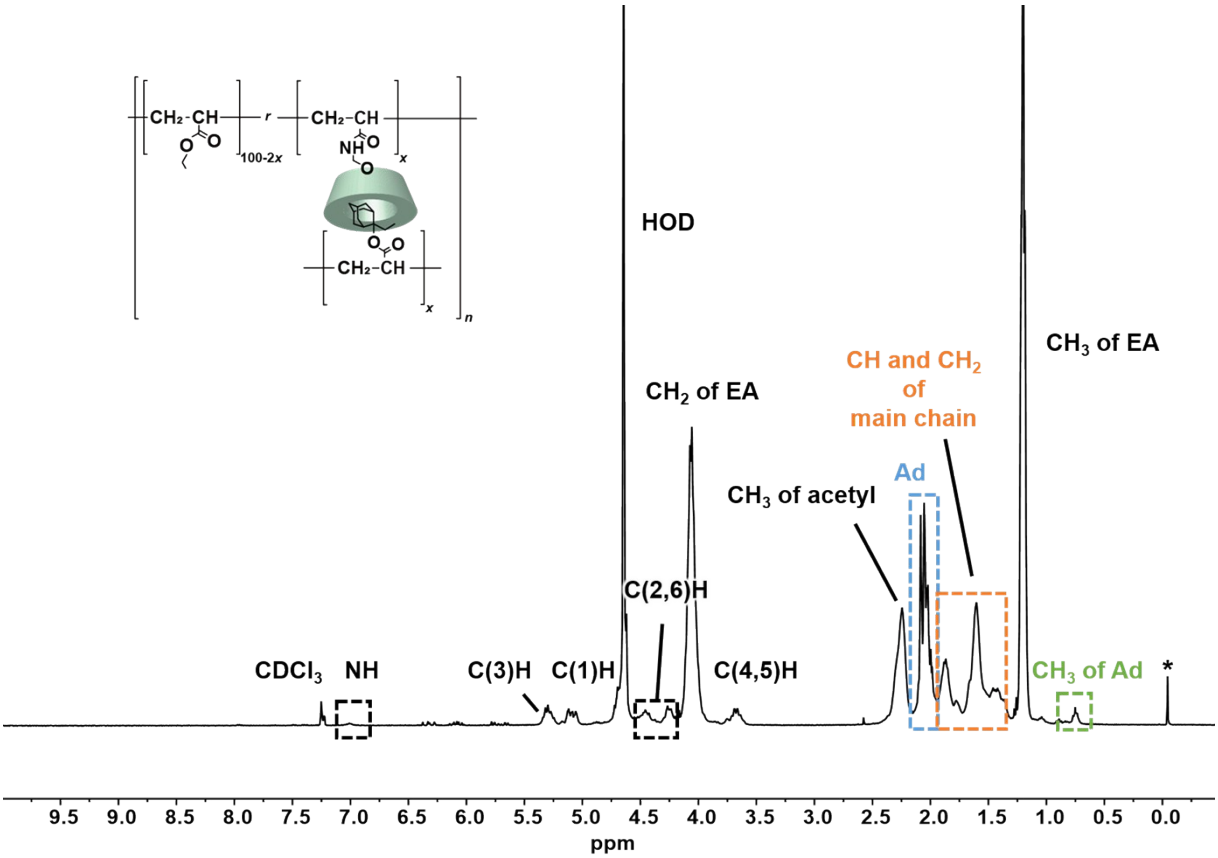


Figure S11. Solid-state ^{1}H FGMAS NMR spectrum of **A-2-1 elastomer** (TMS for standard, 400 MHz, 25 ^{0}C, rotation frequency = 7 kHz).

^{1}H NMR (400 MHz, Chloroform-*d*): δ = 7.05 (-N*H*-), 5.38-5.29 (C(3)*H* of CD), 5.17-5.10 (C(1)*H* of CD), 4.52-4.27 (C(2,6)*H* of CD), 4.16-4.04 (-CH_2CH$_3$ of EA), 3.80-3.68 (C(4,5)*H* of CD), 2.35-2.29 (-CH_3 of acetyl), 2.15-2.03 (-C*H*-, -CH_2-, and -CH_2CH$_3$ of Ad), 1.97-1.41 (-CH_2C*H*- of side chain), 1.30-1.23 (-CH_3 of EA), 0.82-0.78 (-CH_3 of Ad).

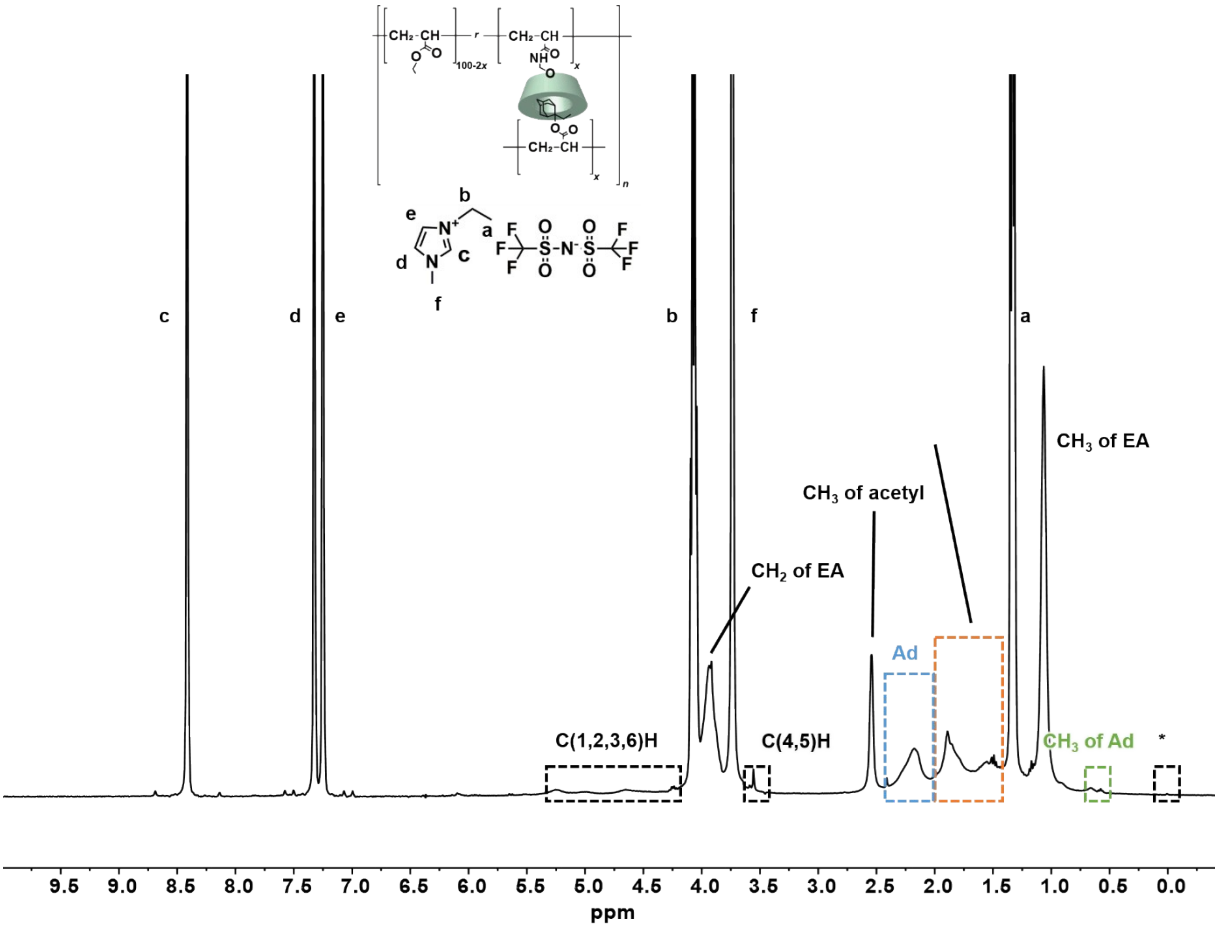


Figure S12. Solid-state ^{1}H FGMAS NMR spectrum of **A-2-1** (TMS for standard, 400 MHz, 25 ^{0}C, rotation frequency = 7 kHz).

^{1}H NMR (400 MHz, Chloroform-*d*): δ = 8.47 (-NC*H*N^{+}- of EMIm TFSI), 7.38 (-NC*H*CH- of EMIm TFSI), 7.31 (-N^{+}C*H*CH- of EMIm TFSI), 5.31-4.70 (C(1,2,3,6)*H* of CD), 4.14 (-N^{+}C*H*$_2$- of EMIm TFSI), 3.99-3.95 (-C*H*$_2$CH$_3$ of EA), 3.80 (-NC*H*$_3$ of EMIm TFSI), 2.59 (-C*H*$_3$ of acetyl), 2.24 (-C*H*-, -C*H*$_2$-, and -C*H*$_2$CH$_3$ of Ad), 1.95-1.61 (-C*H*$_2$C*H*- of side chain), 1.39 (-CH$_2$C*H*$_3$ of EMIm TFSI), 1.12 (-C*H*$_3$ of EA), 0.68 (-C*H*$_3$ of Ad).

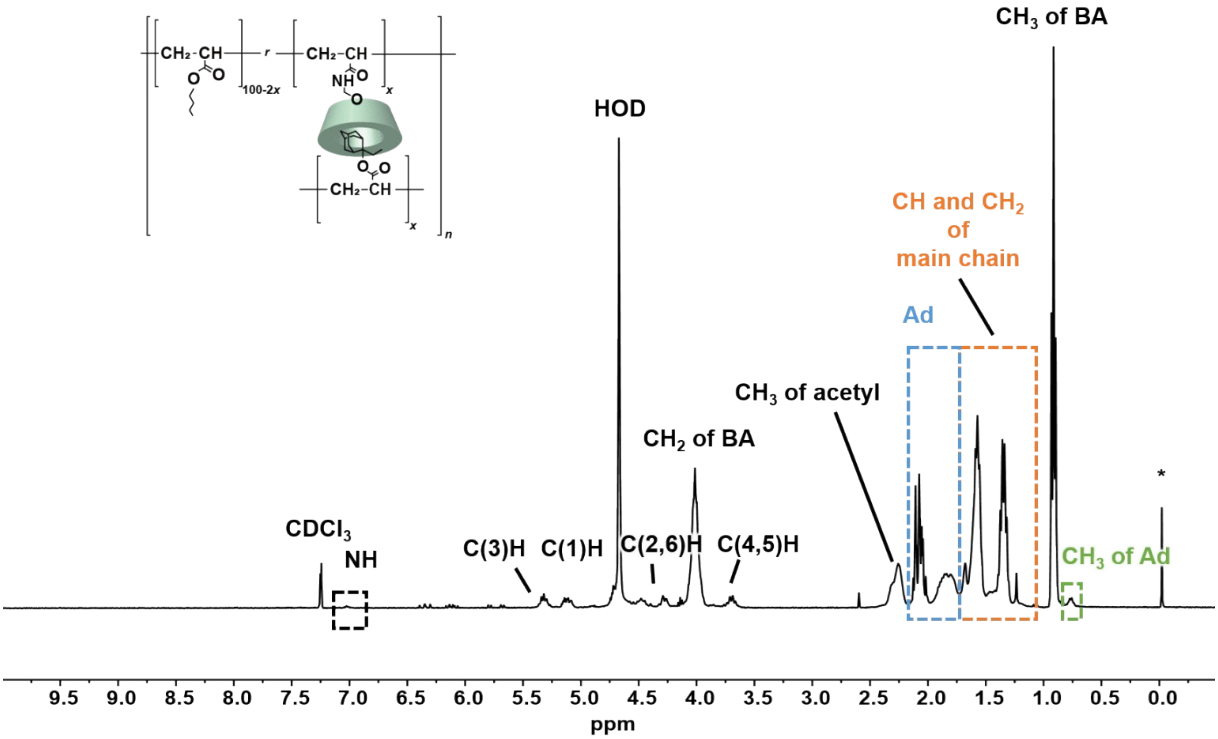


Figure S13. Solid-state ^{1}H FGMAS NMR spectrum of **A-3-1 elastomer** (TMS for standard, 400 MHz, 25 ^{0}C, rotation frequency = 7 kHz).

^{1}H NMR (400 MHz, Chloroform-*d*): δ = 7.03 (-N*H*-), 5.38-5.32 (C(3)*H* of CD), 5.18-5.10 (C(1)*H* of CD), 4.50-4.16 (C(2,6)*H* of CD), 4.09-3.98 (-CH_2CH_2- of BA), 3.76-3.69 (C(4,5)*H* of CD), 2.28-2.26 (-CH_3 of acetyl), 2.15-1.70 (-C*H*-, -CH_2-, and -CH_2CH$_3$ of Ad), 1.66-1.26 (-CH_2C*H*- of side chain), 0.95-0.92 (-CH_3 of BA), 0.79 (-CH_3 of Ad).

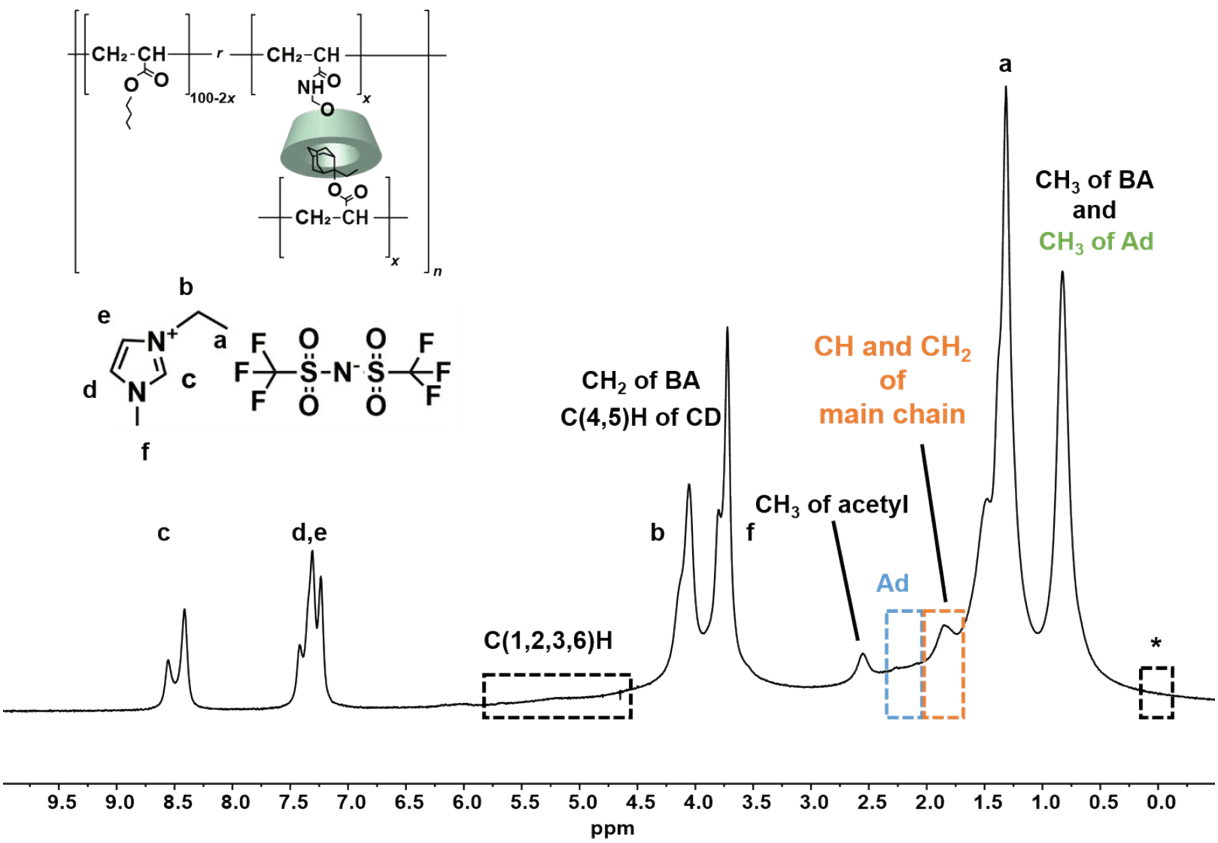


Figure S14. Solid-state ^{1}H FGMAS NMR spectrum of **A-3-1** (TMS for standard, 400 MHz, 25 ^{0}C, rotation frequency = 7 kHz).

^{1}H NMR (400 MHz, Chloroform-*d*): δ = 8.63-8.50 (-NC*H*N^{+}- of EMIm TFSI), 7.50-7.31 (-NC*H*C*H*N^{+}- of EMIm TFSI), 5.80-4.71 (C(1,2,3,6)*H* of CD), 4.14 (-N^{+}CH_2- of EMIm TFSI), 3.88-3.80 (-CH_2CH_2- of BA, C(4,5)*H* of CD, and -NCH_2- of EMIm TFSI), 2.64 (-CH_3 of acetyl), 2.25 (-C*H*-, -CH_2-, and -CH_2CH$_3$ of Ad), 1.92 (-CH_2C*H*- of side chain), 1.39 (-CH$_2$CH_3 of EMIm TFSI), 0.91-0.70 (-CH_3 of BA and -CH_3 of Ad).

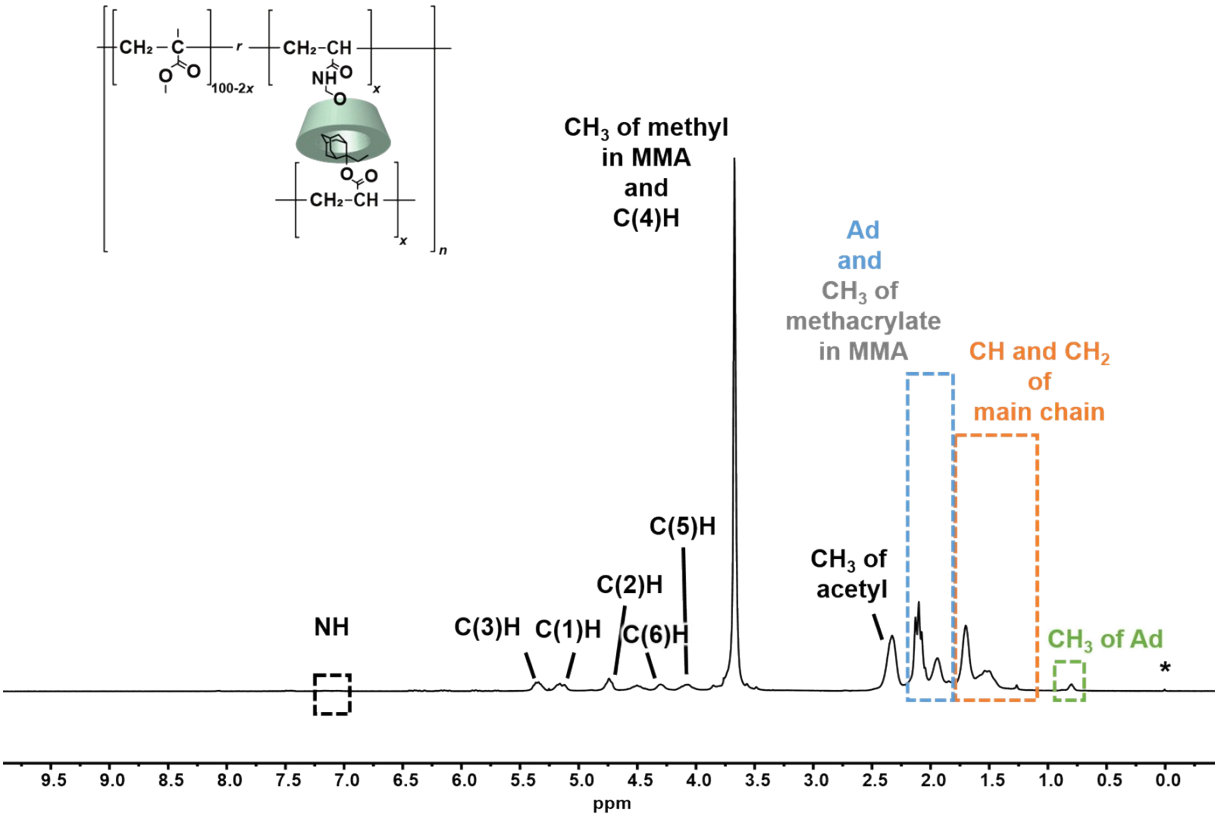


Figure S15. Solid-state ^{1}H FGMAS NMR spectrum of **A-4-1 elastomer** (TMS for standard, 400 MHz, 25 ^{0}C, rotation frequency = 7 kHz).

^{1}H NMR (400 MHz, Chloroform-*d*): δ = 7.05 (-N*H*-), 5.42-5.29 (C(3)*H* of CD), 5.24-5.10 (C(1)*H* of CD), 4.80-4.71 (C(2)*H* of CD), 4.56-4.23 (C(6)*H* of CD), 4.15-4.04 (C(5)*H* of CD), 3.80-3.57 (-C*H*$_3$ of methyl in MMA and C(4)*H* of CD), 2.43-2.18 (-C*H*$_3$ of acetyl), 2.18 – 1.83 (-C*H*-, -C*H*$_2$-, -C*H*$_2$CH$_3$ of Ad, and -C*H*$_3$ of methacrylate in MMA), 1.80-1.19 (-C*H*$_2$C*H*- of side chain), 0.85-0.74 (-C*H*$_3$ of Ad).

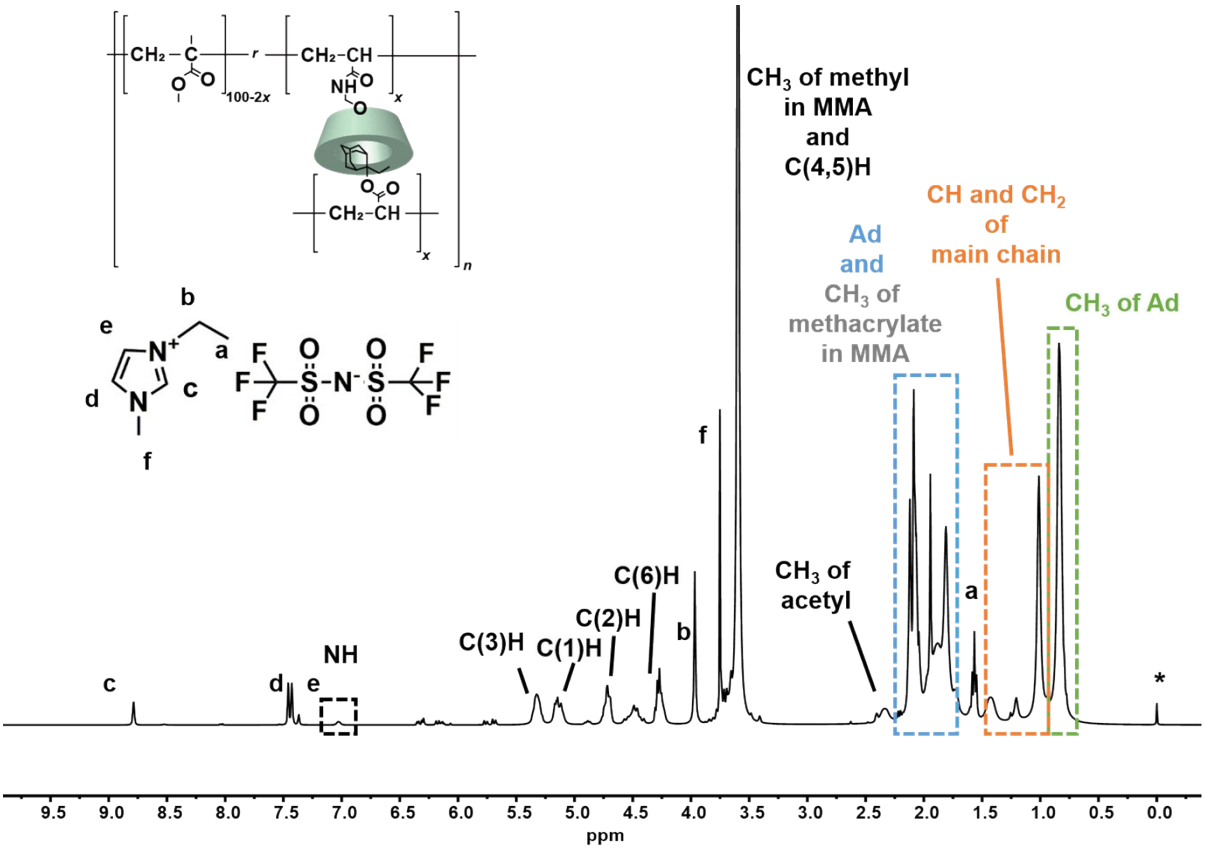


Figure S16. Solid-state ^{1}H FGMAS NMR spectrum of **A-4-1** (TMS for standard, 400 MHz, 25 ^{0}C, rotation frequency = 7 kHz).

^{1}H NMR (400 MHz, Chloroform-*d*): δ = δ 8.78 (-NC*H*N^{+}- of EMIm TFSI), 7.48 (-NC*H*CH- of EMIm TFSI), 7.42 (-N^{+}C*H*CH- of EMIm TFSI), 7.32 (-N*H*-), 6.39-27 (C(3)*H* of CD), 6.22-6.06 (C(1)*H* of CD), 4.77-4.64 (C(2)*H* of CD), 4.69-4.18 (C(6)*H* of CD), 3.97 (-N^{+}C*H*$_2$- of EMIm TFSI), 3.76 (-NC*H*$_3$ of EMIm TFSI), 3.69-3.37 (-C*H*$_3$ of methyl in MMA and C(4,5)*H* of CD), 2.42-2.26 (-C*H*$_3$ of acetyl), 2.19-1.88 (-C*H*-, -C*H*$_2$-, -C*H*$_2$CH$_3$ of Ad, and -C*H*$_3$ of methacrylate in MMA), 1.68 (-CH$_2$C*H*$_3$ of EMIm TFSI), 1.50-0.94 (-C*H*$_2$C*H*- of side chain), 0.92-0.74 (-C*H*$_3$ of Ad).

7. Calculation for controlled EMIm TFSI content

EMIm TFSI content in IE type in Fig. 4c and 5c was controlled by first calculating to mass of elastomer (W_E) and then EMIm TFSI that required for immersion (W_{IL}) was calculated to obtain controlled EMIm TFSI content in the sample.

$$IL = \frac{W_{IE} - W_E}{W_{IE}} \times 100\% \quad \text{where} \quad W_{IE} = W_{IL} + W_E$$

$$IL = \frac{(W_{IL} + W_E) - W_E}{(W_{IL} + W_E)} \times 100\%$$

IL is EMIm TFSI content in weight percent. W_{IE} is weight of elastomers after immersion in ionic liquid. W_E is weight of elastomers. W_{IL} is the weight of EMIm TFSI required for immersion. By solving the equation, W_{IL} required for the controlled EMIm TFSI content can be obtained.

8. Stress-strain curves of ionic liquid elastomers

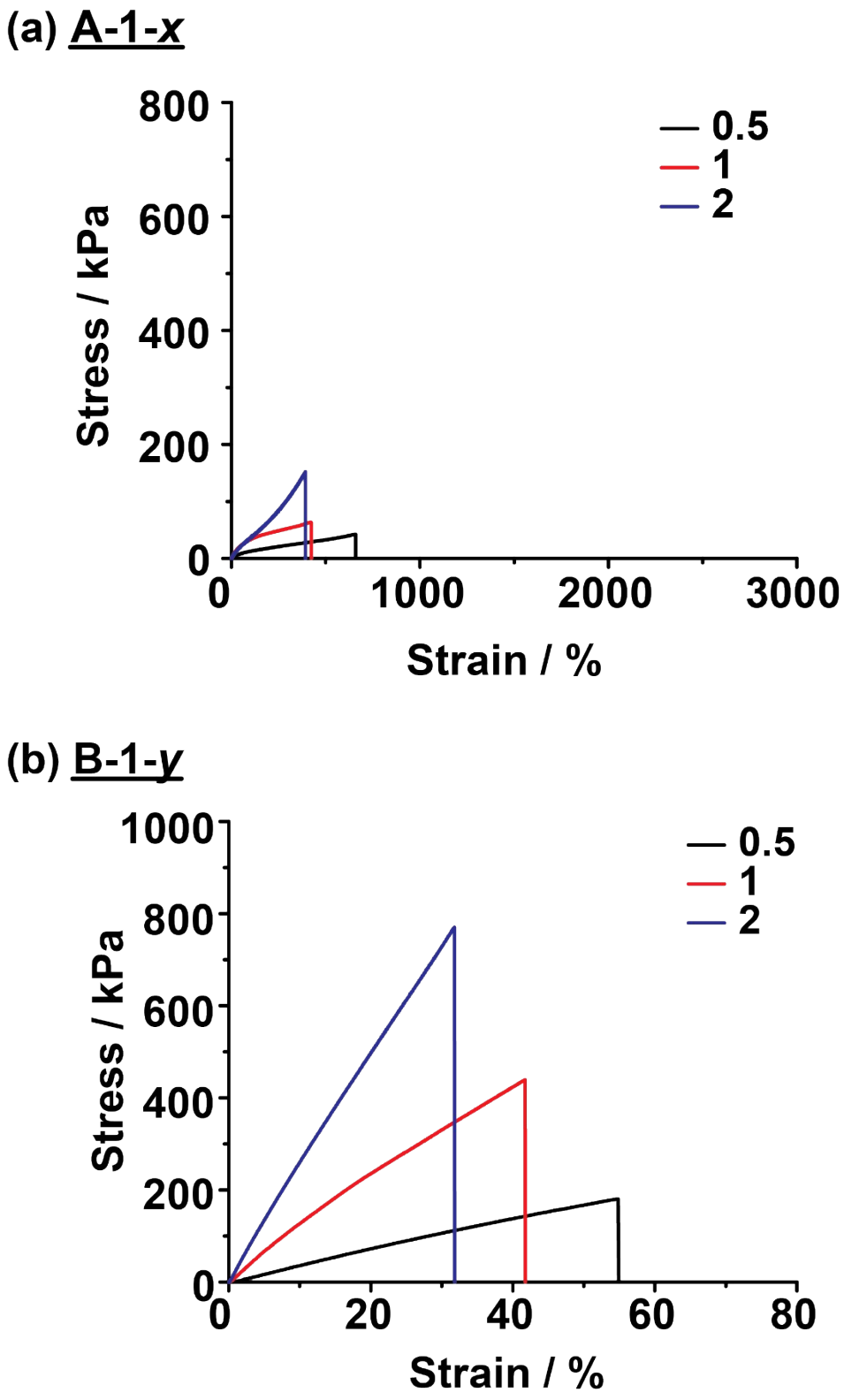


Figure S17. Stress-strain curves of (**a**) supramolecular ionic liquid elastomers (**A-1-*x***) and (**b**) chemically cross-linked ionic liquid elastomers (**B-1-*y***). 1 indicates the methyl acrylate side chain. *x* and *y* indicate the mol% of cross-linker from host-guest inclusion complex (PAcγCD and Ad) and BDA units, respectively.

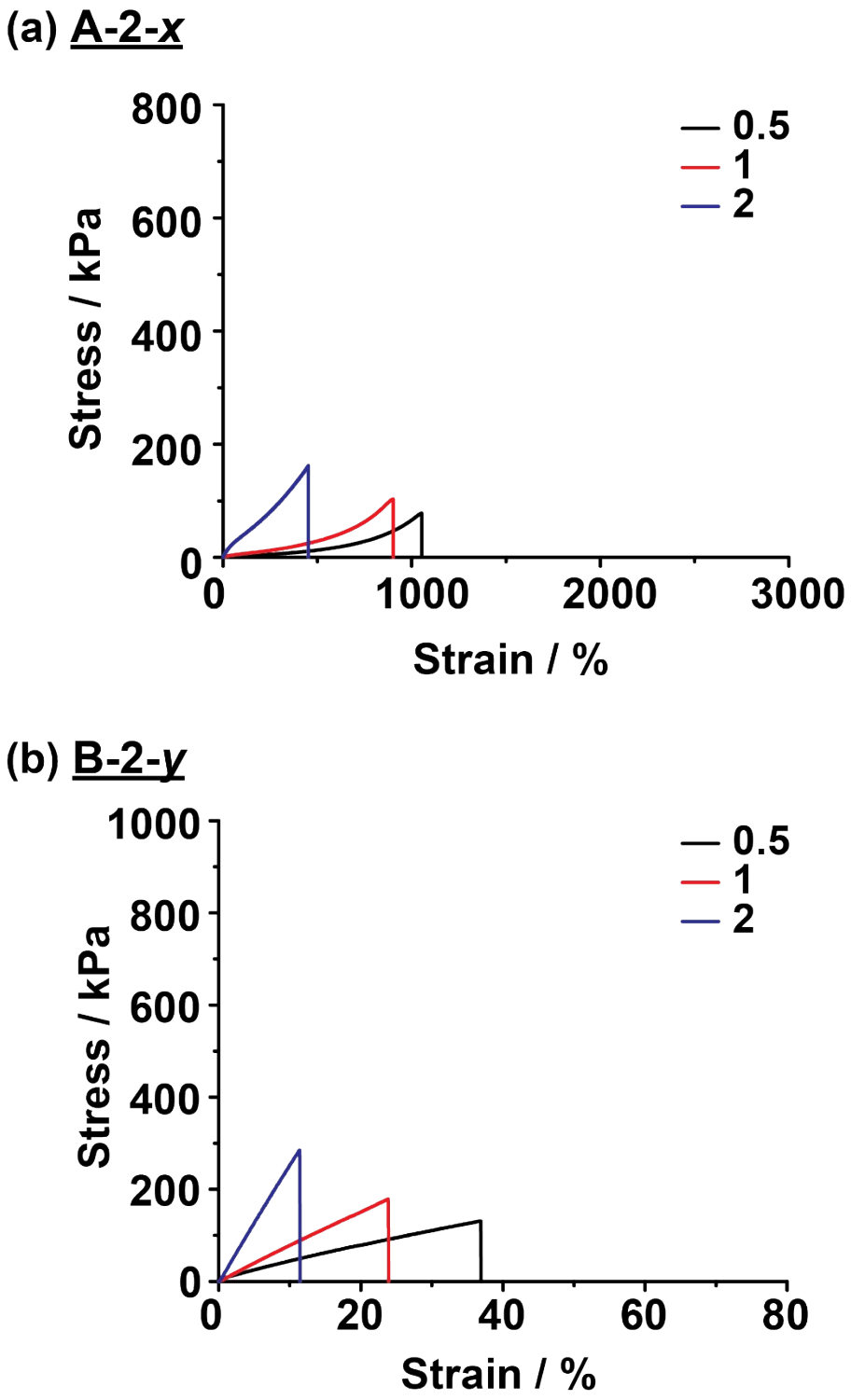


Figure S18. Stress-strain curves of (**a**) supramolecular ionic liquid elastomers (**A-2-*x***) and (**b**) chemically cross-linked ionic liquid elastomers (**B-2-*y***). 2 indicates the ethyl acrylate side chain. *x* and *y* indicate the mol% of cross-linker from host-guest inclusion complex (PAcγCD and Ad) and BDA units, respectively.

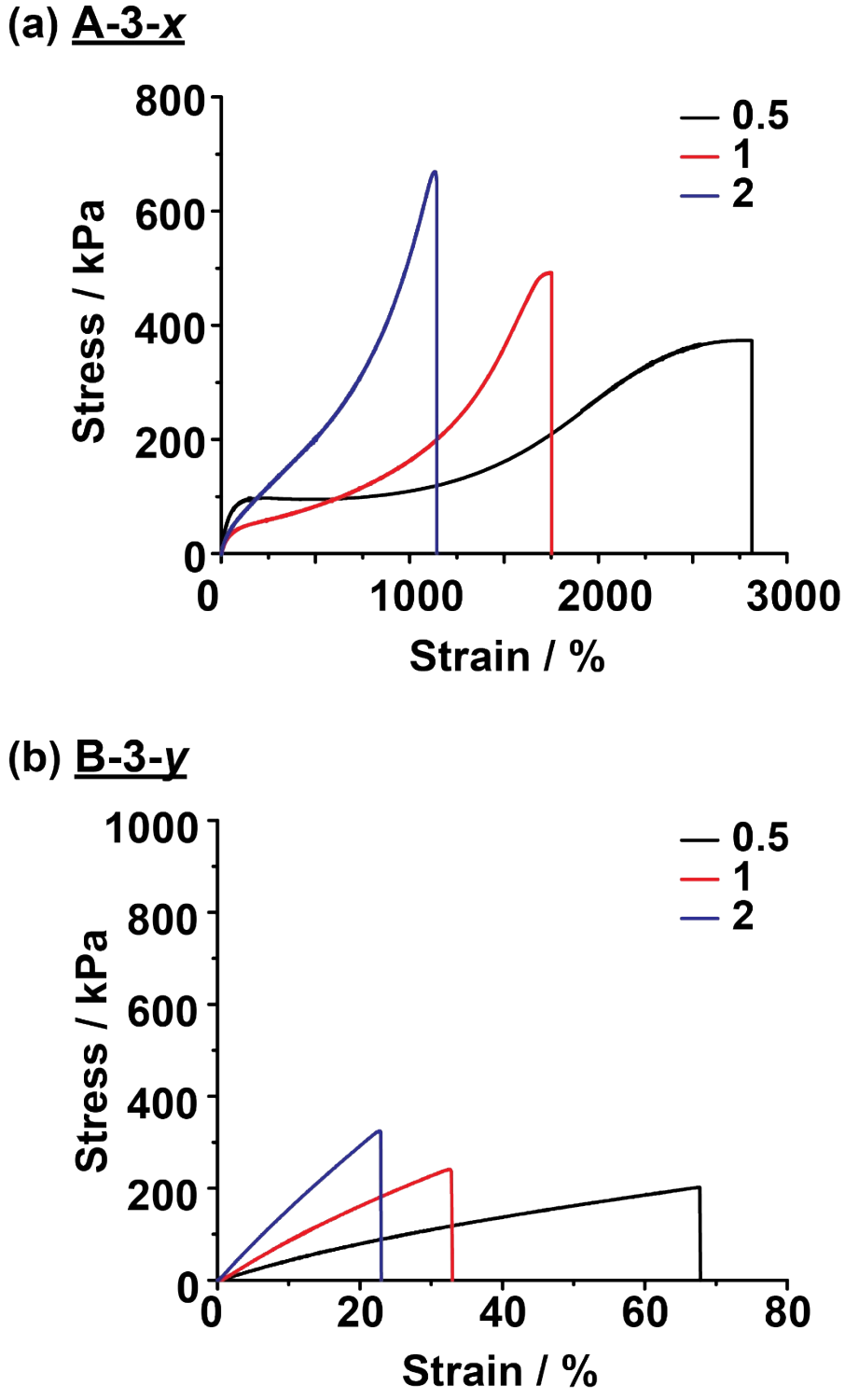


Figure S19. Stress-strain curves of (**a**) supramolecular ionic liquid elastomers (**A-3-*x***) and (**b**) chemically cross-linked ionic liquid elastomers (**B-3-*y***). 3 indicates the butyl acrylate side chain. *x* and *y* indicate the mol% of cross-linker from host-guest inclusion complex (PAcγCD and Ad) and BDA units, respectively.

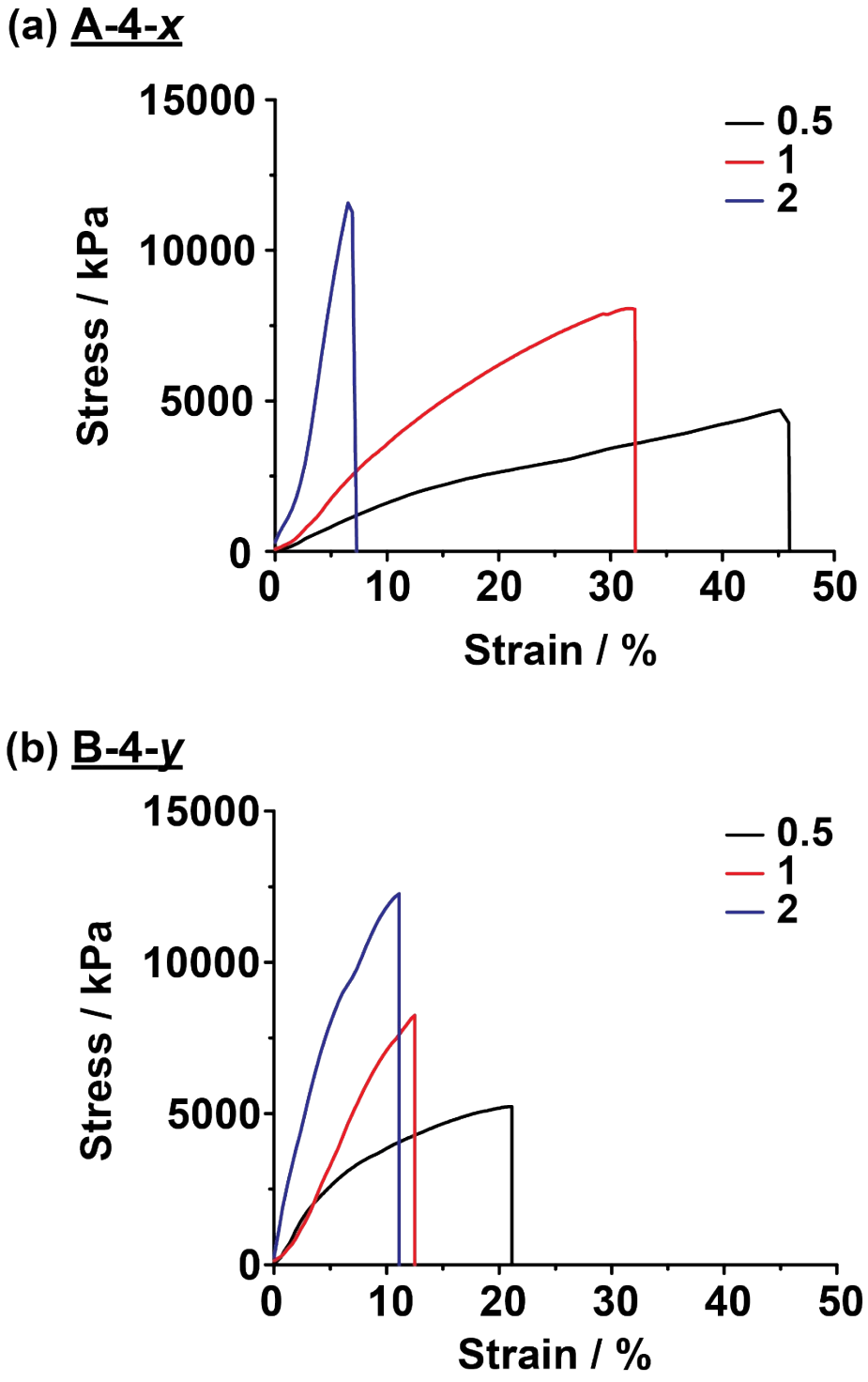


Figure S20. Stress-strain curves of (**a**) supramolecular ionic liquid elastomers (**A-4-*x***) and (**b**) chemically cross-linked ionic liquid elastomers (**B-4-*y***). 4 indicates the methyl methacrylate side chain. *x* and *y* indicate the mol% of cross-linker from host-guest inclusion complex (PAcγCD and Ad) and BDA units, respectively.

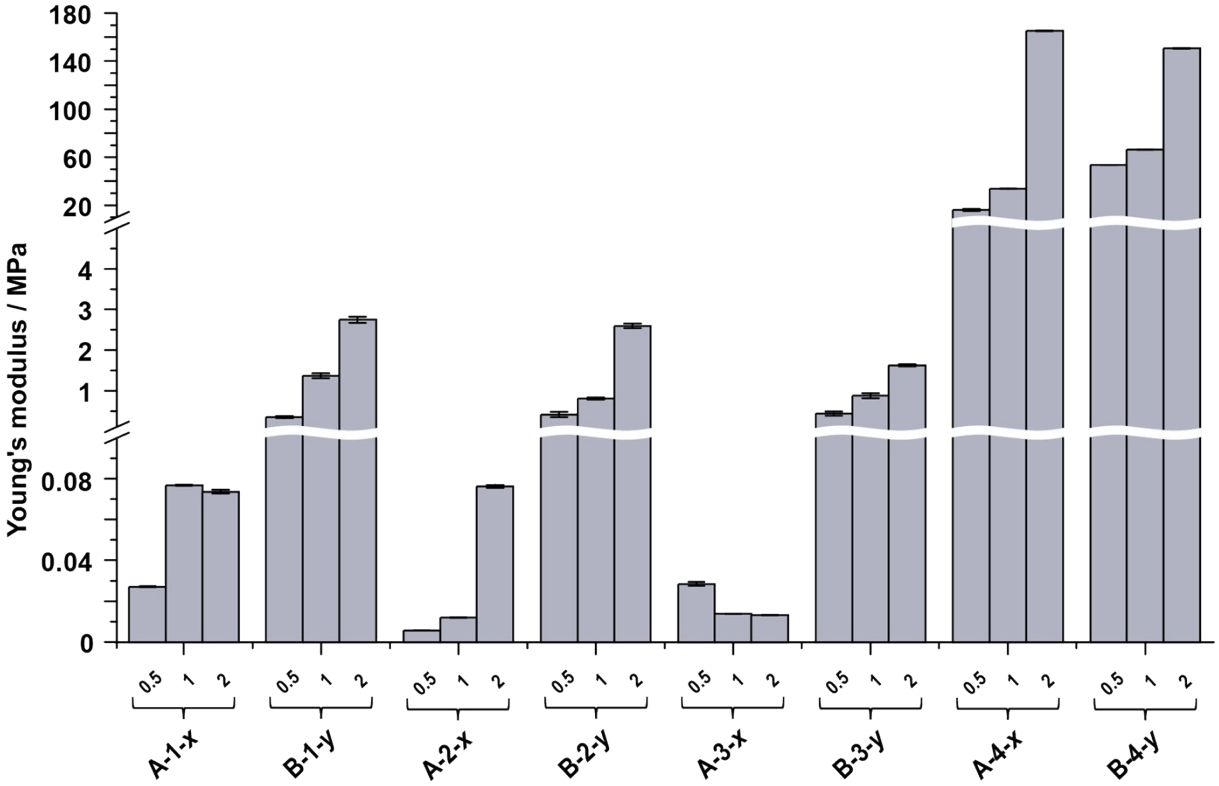


Figure S21. Young's modulus of supramolecular ionic liquid elastomer (**A-Acrylate-*x***) and chemically cross-linked ionic liquid elastomer (**B-Acrylate-*y***). Acrylate: methyl acrylate (1), ethyl acrylate (2), butyl acrylate (3), and methyl methacrylate (4) *x* and *y* indicate the mol% of cross-linker from host-guest inclusion complex (PAcγCD and Ad) and BDA units, respectively.

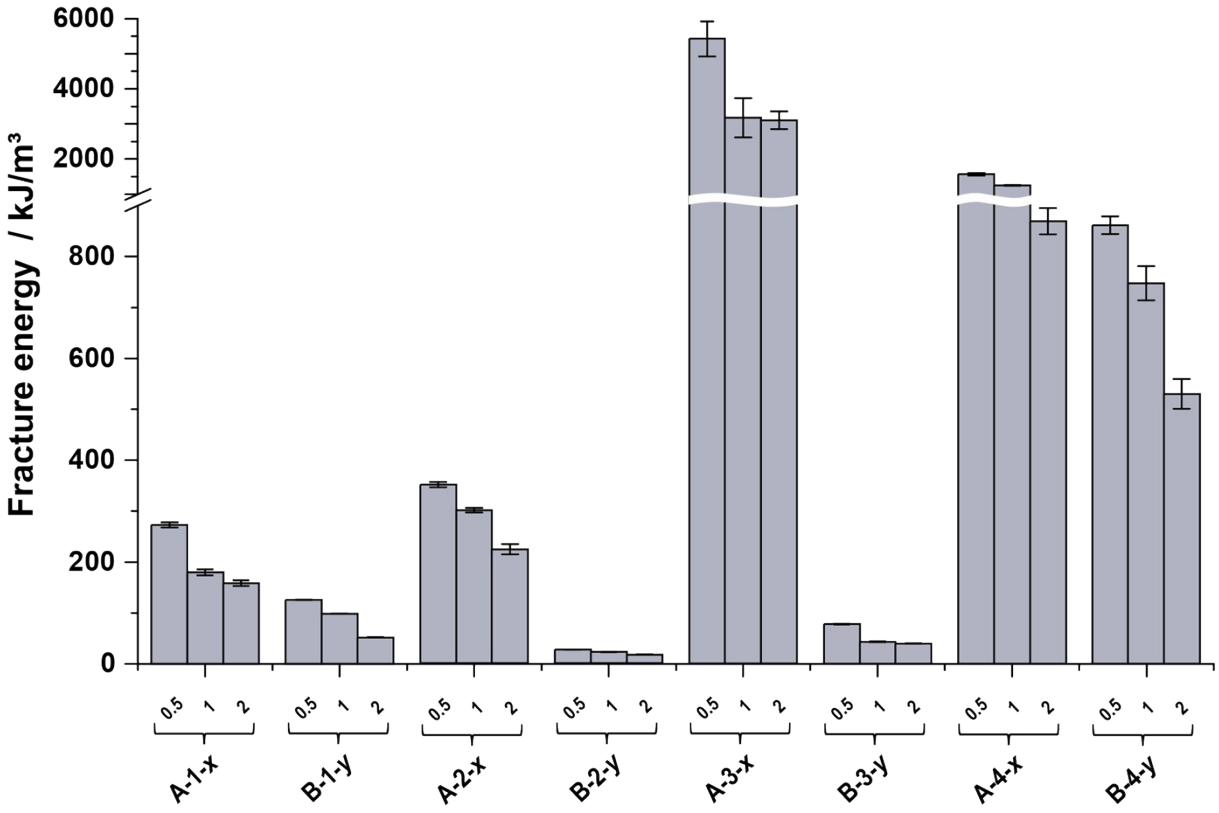


Figure S22. Fracture energy of supramolecular ionic liquid elastomer (**A-Acrylate-*x***) and chemically cross-linked ionic liquid elastomer (**B-Acrylate-*y***). Acrylate: methyl acrylate (1), ethyl acrylate (2), butyl acrylate (3), and methyl methacrylate (4) *x* and *y* indicate the mol% of cross-linker from host-guest inclusion complex (PAcγCD and Ad) and BDA units, respectively.

9. Stress relaxation time constants of ionic liquid elastomers

Data acquired from universal tensile test machine then fitted with Maxwell method curve fitting. The stress-relaxation curves of supramolecular polymeric ionic liquid elastomers (**A-Acrylate-1**) were fitted with two-order exponential equation (τ' and τ) whereas chemically cross-linked ionic liquid elastomers (**B-Acrylate-1**) were fitted with one-order exponential equation (τ) whereas

Fitting equation:

A-Acrylate-1

$$\sigma_t = \sigma_0 + G_1 e^{-\left(\frac{t}{\tau}\right)} + G_2 e^{-\left(\frac{t}{\tau'}\right)}$$

B-Acrylate-1

$$\sigma_t = \sigma_0 + G_1 e^{-\left(\frac{t}{\tau}\right)}$$

σ_t is the stress at a certain time *t*; σ_0 is the peak stress; G_i is constant; τ is slow relaxation time constant for **A-Acrylate-1** and **B-Acrylate-1**, respectively; τ' is rapid relaxation time constant for **A-Acrylate-1**.

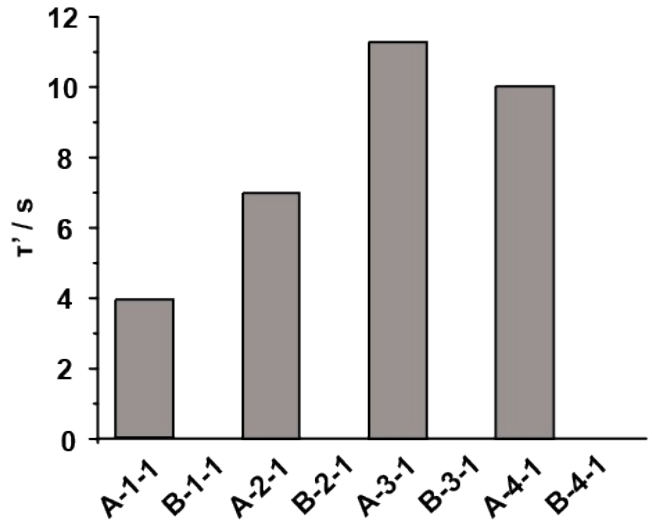


Figure S23. Rapid relaxation time constant of supramolecular ionic liquid elastomer (**A-Acrylate-1**) and chemically cross-linked ionic liquid elastomer (**B-Acrylate-1**). Acrylate: methyl acrylate (1), ethyl acrylate (2), butyl acrylate (3), and methyl methacrylate (4) *x* and *y* indicate the mol% of cross-linker from host-guest inclusion complex (PAcγCD and Ad) and BDA units, respectively.

10. Calculation of ionic liquid elastomers conductivity

Data acquired from LCR metre then calculated as using equation to obtain conductivity in AC current (σ_{ac}) as follows:

$$\sigma_{ac} = \omega\varepsilon_0\varepsilon' \tan(\delta) \qquad \text{where} \qquad \tan(\delta) = \frac{\varepsilon''}{\varepsilon'}$$

σ_{ac} is conductivity of AC current. ω is angular frequency. ε_0 is vacuum permittivity with value 8.85×10^{-12} F m^{-1}. ε' is dielectric constant and ε" is dielectric loss factor. Then, the result of σ_{ac} was plotted with frequency (f) in log graph to obtain graph that accordance to *Jonscher's Universal Power Law*[4] shows the sample calculation for conductivity of EMIm TFSI).[5] According to *Jonscher's Universal Power Law*, equation of plotted graph is $\sigma_{ac} = \sigma_{dc} + A\omega^n$ where in the high frequency conductivity there will be "plateau region" which means conductivity that independent to frequency (σ_{dc}).

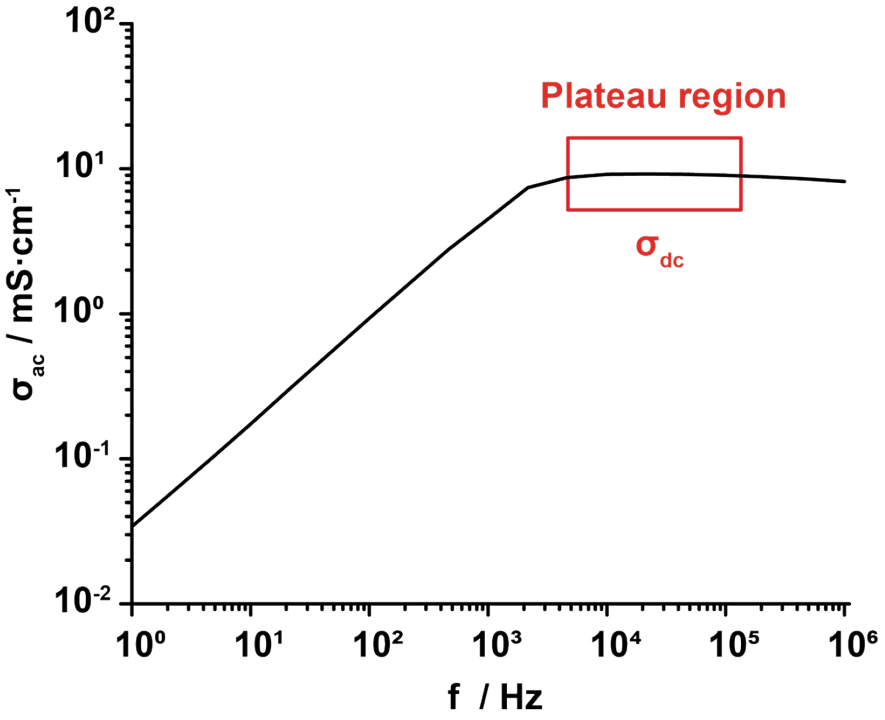


Figure S24. Plotted graph for conductivity calculation of native ionic liquid [1-ethyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide (EMIm TFSI)].

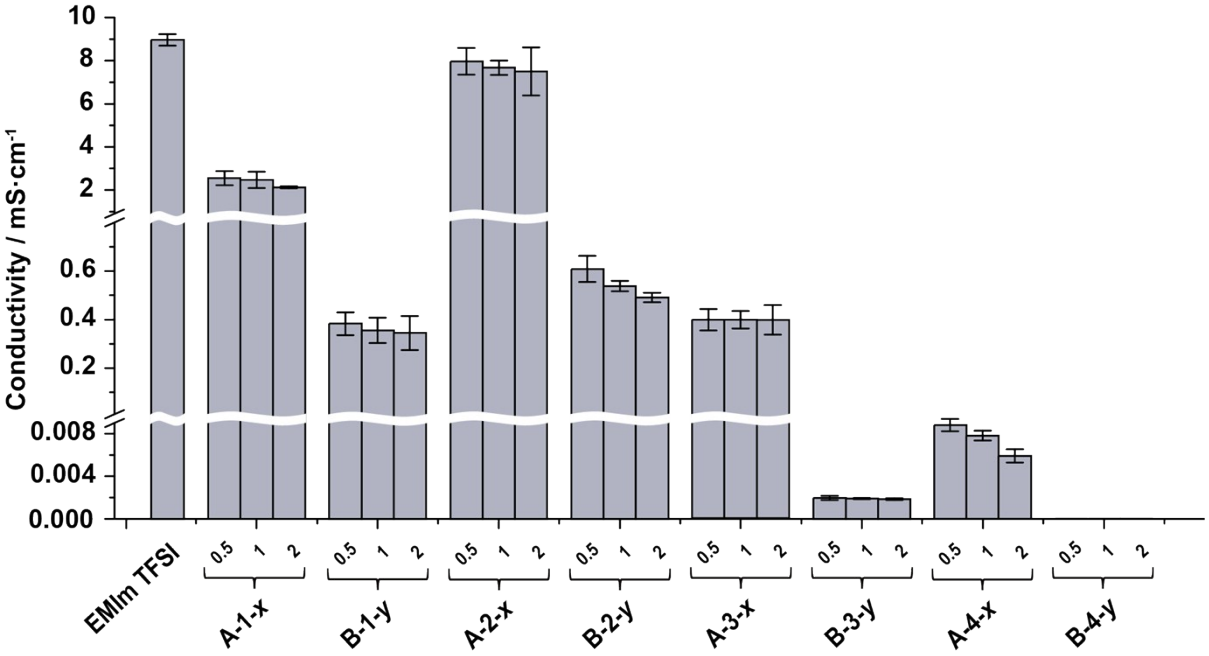


Figure S25. Conductivity of 1-ethyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide (EMIm TFSI), supramolecular ionic liquid elastomer (**A-Acrylate-*x***), and chemically cross-linked ionic liquid elastomer (**B-Acrylate-*y***). Acrylate: methyl acrylate (1), ethyl acrylate (2), butyl acrylate (3), and methyl methacrylate (4) *x* and *y* indicate the mol% of cross-linker from host-guest inclusion complex (PAcγCD and Ad) and BDA units, respectively.

11. Cyclic test for self-recovery calculation

Recovery ratio of supramolecular polymeric IE were calculated using hysteresis curve. First, supramolecular polymeric IE samples were strecthed until half of the plastic deformable region then let it back to initial state. From this first cycle we obtained hysteresis curve and we calculate the are under the curve. Then, supramolecular polymeric IE samples were held either at RT or 80 ^{0}C for 12 hours (different sample for both temperature). After 12 hours, we performed similar procedure as same as first cycle to obtain second cycle's hysteresis curve and we also calculate the are under the curve. Last, we divide the result of second cycle hysteresis area with first cycle hysteresis area and times it with 100% to obtain recovery ratio.

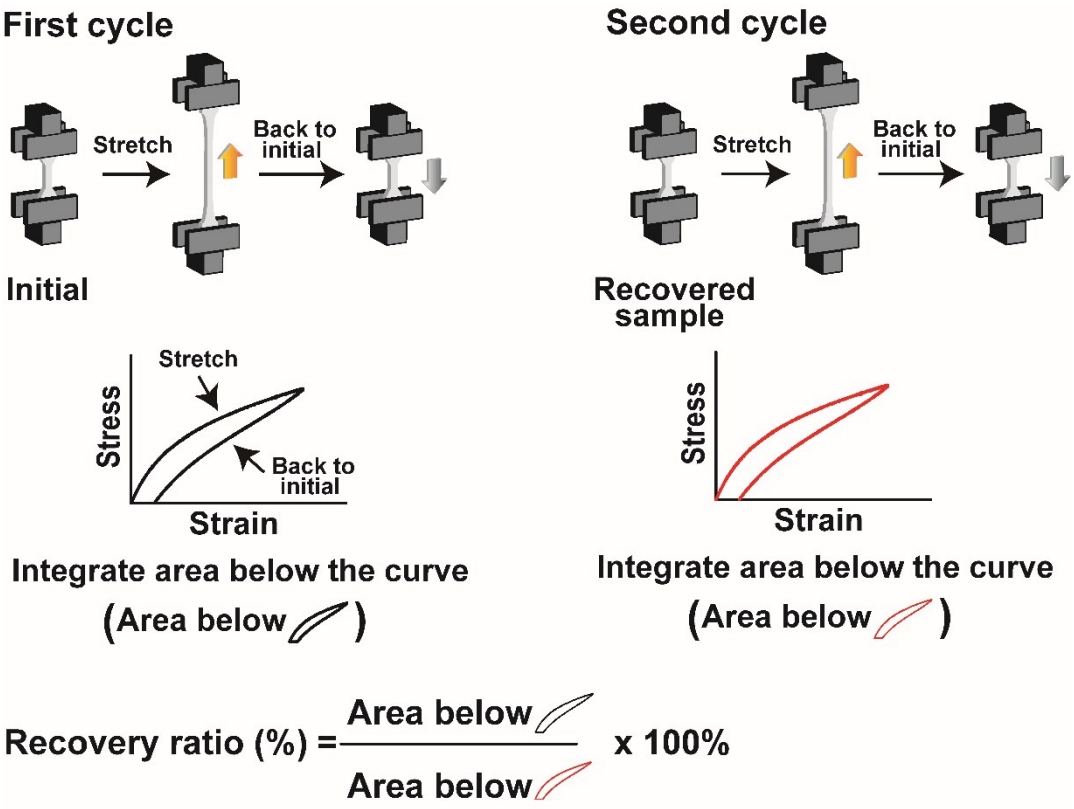


Figure S26. Recovery ratio calculation procedure

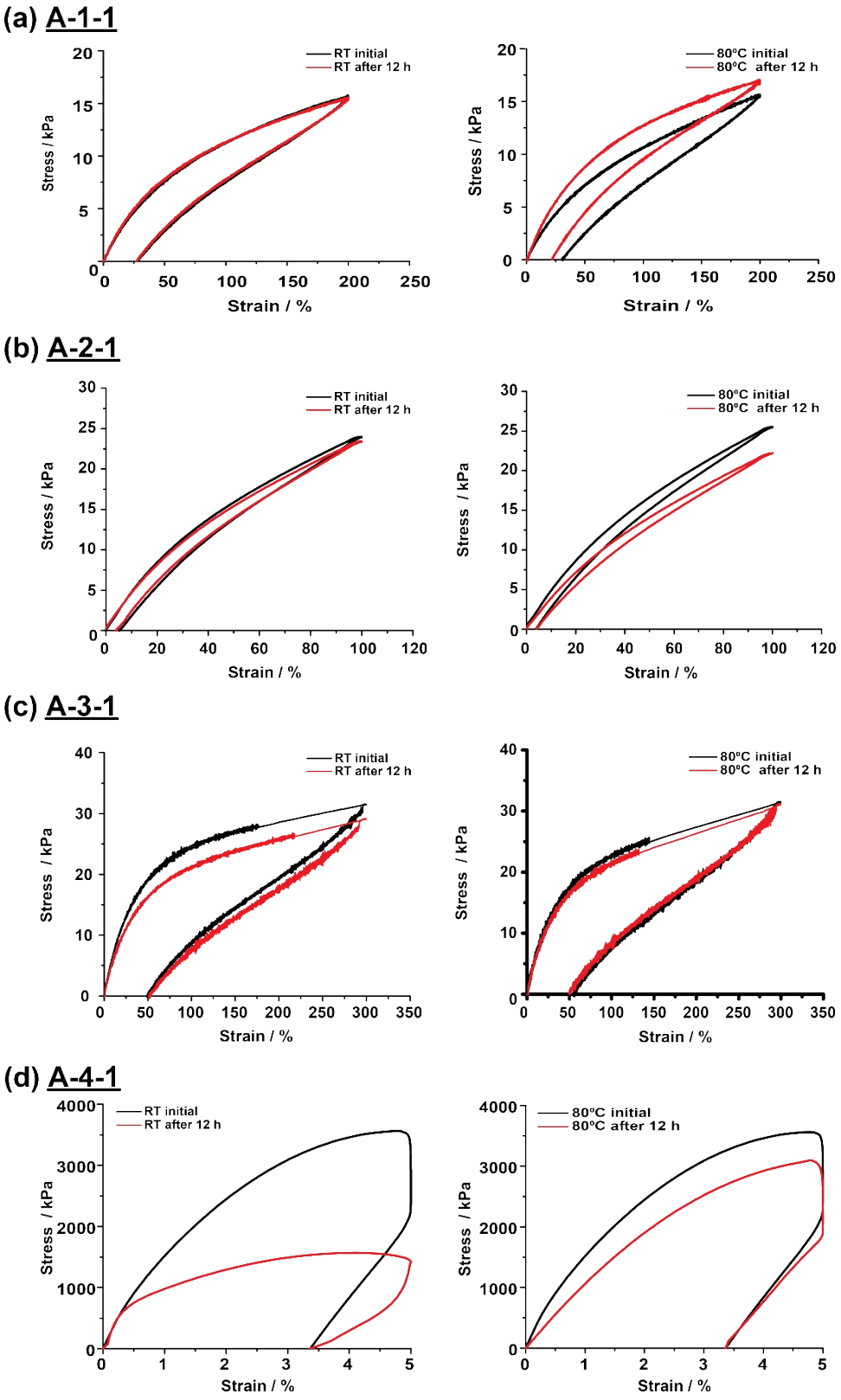


Figure S27. Cyclic test for first cycle / initial state (black line) and after 12 h rest (red line) at room temperature (right figure) and 80 ^{0}C (left figure): (**a**) **A-1-1**, (**b**) **A-2-1**, (**c**) **A-3-1**, and (**d**) **A-4-1**.

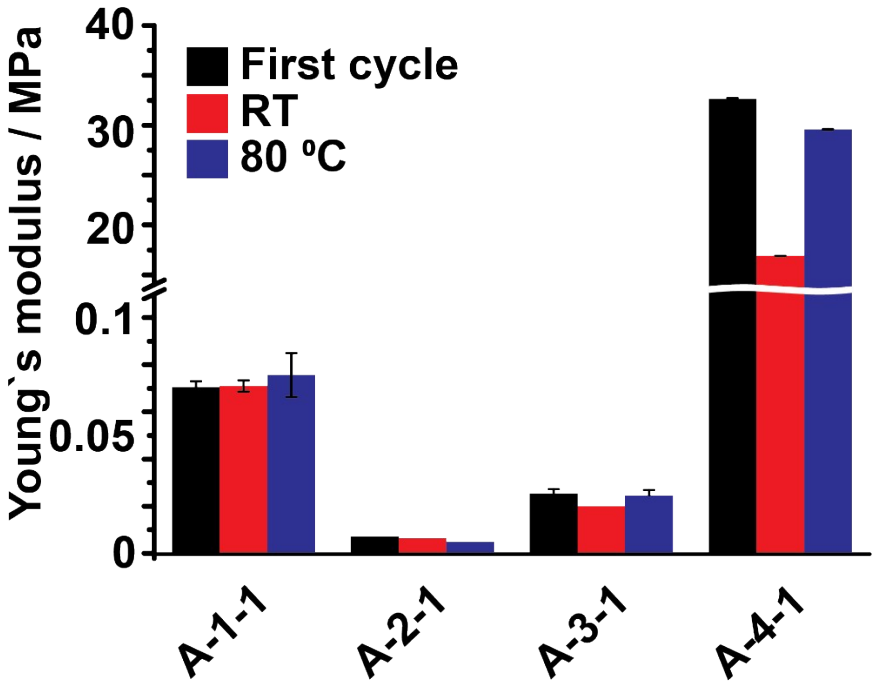


Figure S28. Young's modulus for self-recovery for **A-1-1**, **A-2-1**, **A-3-1**, and **A-4-1**. First cycle/initial state (black bar), after 12 h rest in at room temperature (red bar) and at 80 ^{0}C (blue bar).

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5. A. K. Jonscher, *Nature*, 1977, **267**, 673.