Supporting Information

Strengthening ethylene-methacrylic acid ionomer with single-boron-based molecules as cross-linkers in dynamic networking

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1. General remarks of materials and analytical methods

Chemicals were purchased from Sigma-Aldrich, Adamas or TCI unless otherwise specified. Butylboronic acid, 1,2-propanediol, triethylphosphine oxide, butyl borate (**B1**), isopropyl borate (**B2**), ethyl borate (**B3**) and 4-(butylthio)-2,3,5,6-tetrafluoripyridine (**B4**) were used as received without further purification. Ethylene-(meth)acrylic acid ionomers (EMAAI) obtained from Dow company. Tetrahydrofuran (THF) was freshly distilled over sodium wire, ethanol was freshly distilled from anhydrous $CaCl_2$ before use. Pentane was used as received without further purification.

Differential scanning calorimetry (DSC) was measured by a TA Instrument Q2000 and heated for two cycles from -10 °C to 150 °C (heating rate = 10 °C/min) under a nitrogen atmosphere. Nuclear magnetic resonance (NMR) was recorded by a Bruker Advance III HD 600 at 25 °C. ^{1}H NMR signals were recorded relative to the signal of residual deuterochloroform ($CDCl_3$, 7.26 ppm) and reported in δ units, parts per million (ppm). ^{31}P NMR shifts were measured by using triethylphosphine oxide ($Et_3P=O$) as a reference.

The mechanical mixing process of boron compound (2.5 wt%) and Surlyn was conducted on an internal mixer (Rheomix 600) at 160 °C for 20 min. Rheology measurements were tested by a HAAKE MARS III rheometer using a 20 mm parallel plate in a convection oven. Oscillatory shear experiments were performed at strain γ = 1% and different frequencies (ω) from 0.1 Hz to 10 Hz at 70 °C. Tensile tests were studied with dumbbell-shaped samples (effective gauge length = 12 mm, width = 2.8 mm, thickness = 0.6 mm) via an Instron 5966 universal testing machine equipped with an oven.

Binding energies of boron compounds and sodium acetate were determined by density functional theory (DFT) calculations, which were performed in Gaussian (G16) suite of program with Becke's three-parameter hybrid method using the Lee–Yang–Parr correlation functional (B3LYP). The geometrical structures and the vibrational modes were calculated at 6-311+G(d, p) level. The binding energy (E_b) between two components was defined as follows:

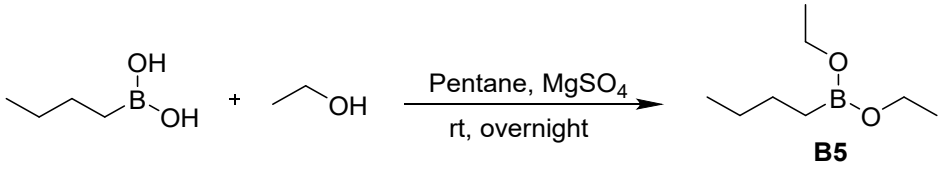
$$E_b = E_{total} - E_A - E_B$$ **Equation S1**

E_{total} presents the energy of the A–B complex, E_A and E_B present the energies of A component and B component, respectively. A and B components are the boron compound and sodium acetate.

2. Synthesis and characterization of boron compounds

2.1. Preparation of diethyl *n*-butylboronate (B5)

A round-bottom flask equipped with a stir bar was charged with butylboronic acid (1.0 eq), anhydrous ethanol (2.0 eq), anhydrous $MgSO_4$ (3.0 eq) and pentane. After stirring at room temperature overnight, the mixture was filtered. The collected organic layer was concentrated under vacuum to give the crude product. The crude product was then purified by distillation under vacuum at 50 °C to give the target compound as a colorless oil. ^{1}H NMR (400 MHz, $CDCl_3$) δ: 3.89-3.84 (m, 4 H,), 1.37-1.28 (m, 4 H), 1.19 (t, J = 8.0 Hz, 6 H), 0.89 (t, J = 8.0 Hz, 3 H), 0.75 (t, J = 8.0 Hz, 2 H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$) δ: 58.8, 26.4, 25.6, 17.3, 13.9 ppm. Similar to previous reports, carbon adjacent to boron is not detected.[1, 2] IR (KBr, cm^{-1}): 2962, 2925, 2860, 1365, 1302, 1148, 1044.

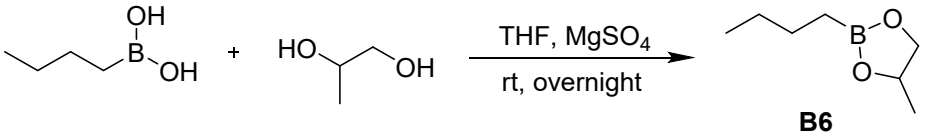


Scheme S1. Synthetic route of **B5**.

2.2. Preparation of 2-butyl-4-methyl-1,3,2-dioxaborolane (B6)

A round-bottom flask equipped with a stir bar was charged with butylboronic acid (1.0 eq), diol (1.0 eq), anhydrous $MgSO_4$ (3.0 eq) and anhydrous THF. After stirring at room temperature overnight, the mixture was filtered. The collected organic layer was concentrated under vacuum to give the crude product. The crude product was extracted

by anhydrous hexane and concentrated under vacuum to give the target compound as a colorless oil. ^{1}H NMR (400 MHz, $CDCl_3$) δ: 4.58-4.49 (m, 1 H,), 4.29-4.25 (m, 1 H), 3.73-3.69 (m, 1 H), 1.46-1.38 (m, 2 H), 1.36-1.30 (m, 5 H), 0.90 (t, J = 8.0 Hz, 3 H), 0.86-0.82 (m, 2 H) ppm; ^{13}C NMR (100 MHz, $CDCl_3$) δ: 72.9, 71.8, 26.1, 25.3, 21.7, 13.7 ppm. Similar to previous reports, carbon adjacent to boron is not detected.[1, 2] IR (KBr, cm^{-1}): 2956, 2925, 2855, 1381, 1301, 1148, 1036.



Scheme S2. Synthetic route of **B6**.

2.3. NMR spectra of B5 and B6

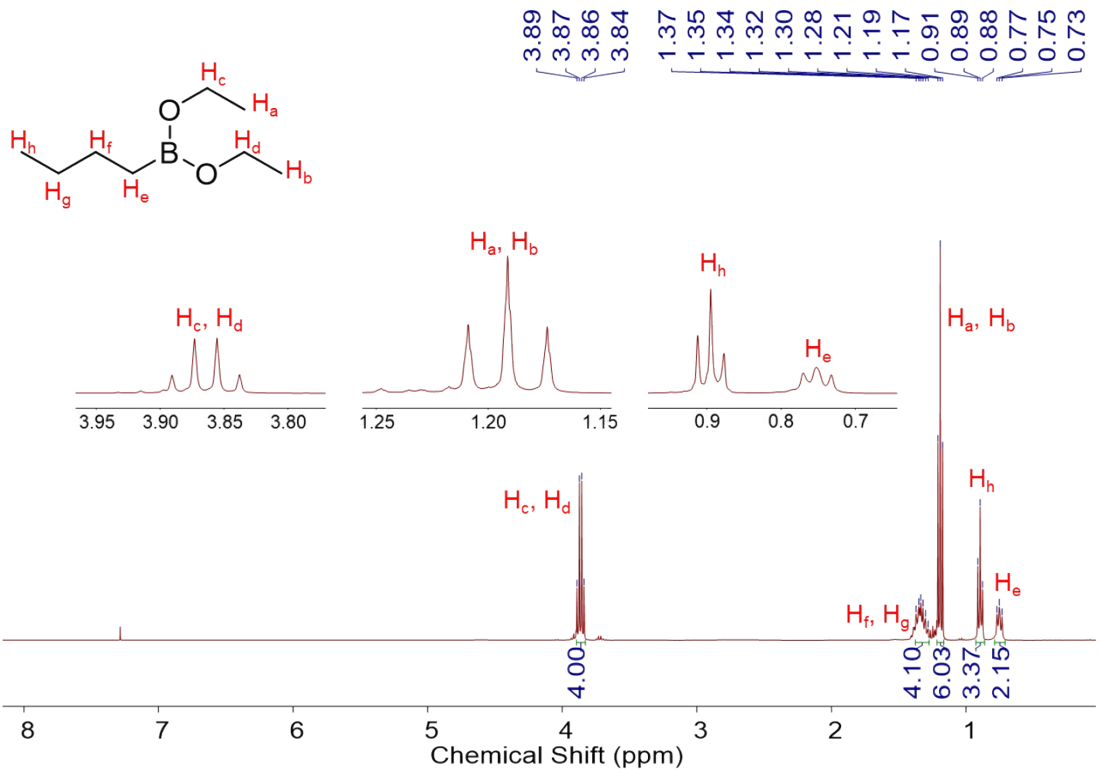


Figure S1. ^{1}H NMR spectrum of **B5** (400 MHz, $CDCl_3$).

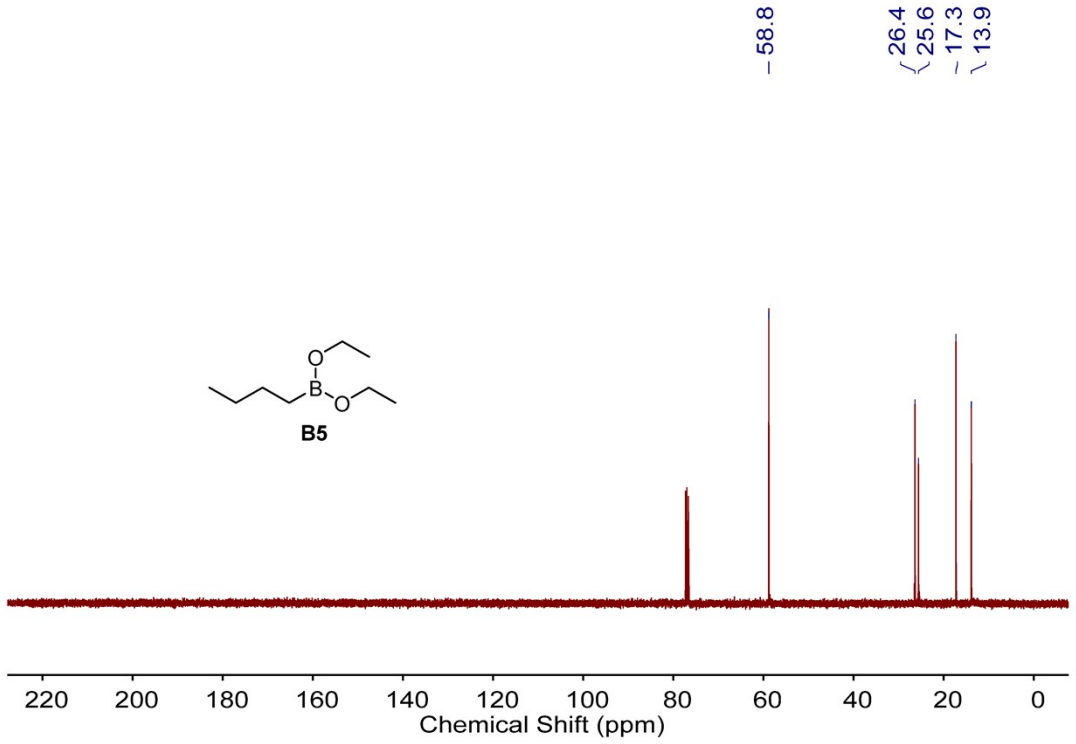


Figure S2. ^{13}C NMR spectrum of **B5** (100 MHz, $CDCl_3$).

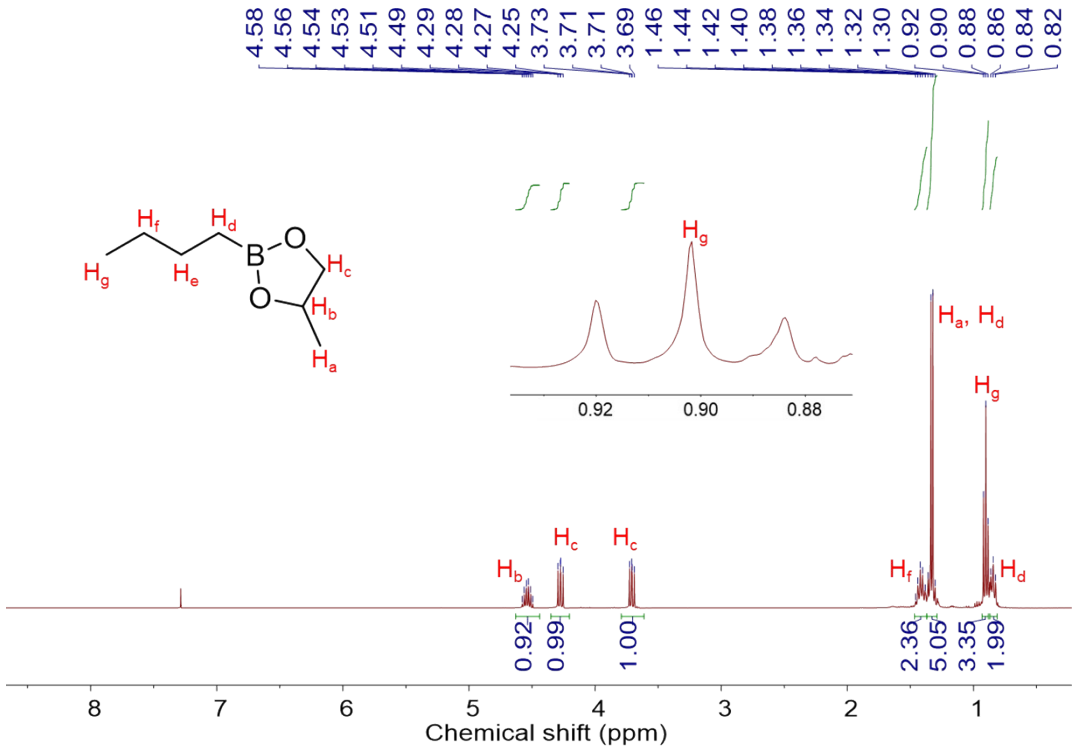


Figure S3. ^{1}H NMR spectrum of **B6** (400 MHz, $CDCl_3$).

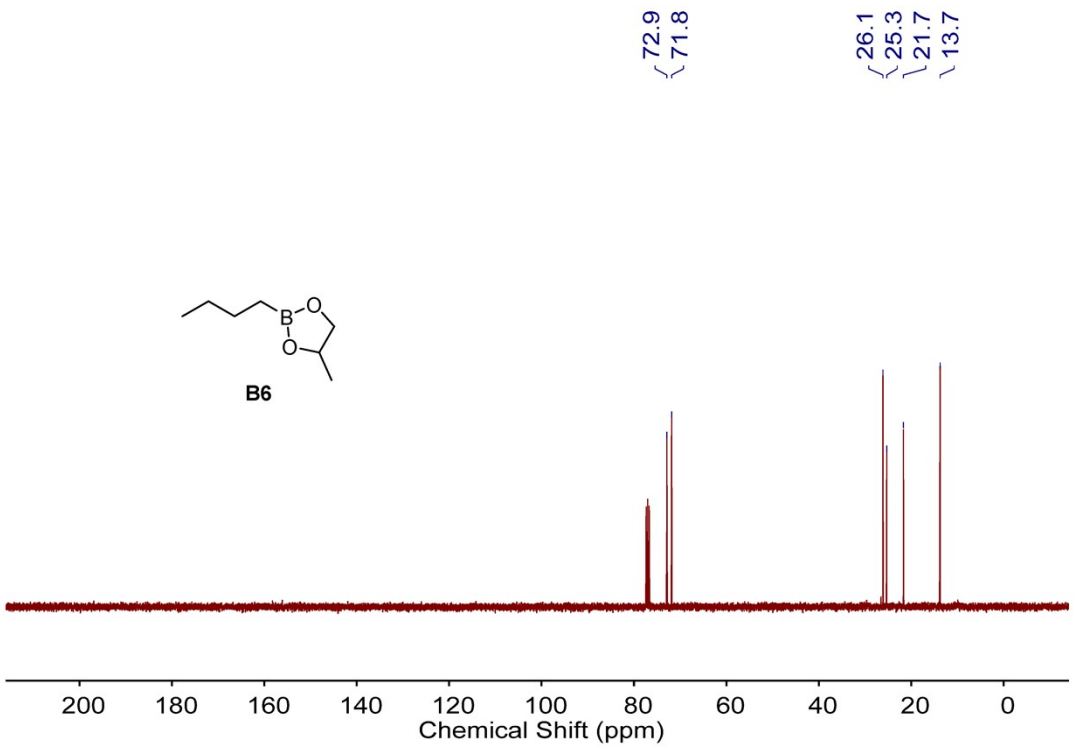


Figure S4. ^{13}C NMR spectrum of **B6** (100 MHz, $CDCl_3$).

3. Measurement of Lewis acidity

The Lewis acidity of six boron compounds was determined by the Gutmann-Beckett method.[3] ^{31}P NMR shifts upon complexation with borane compounds were recorded using $Et_3P{=}O$ as areference . The ^{31}P NMR shifts are converted to acceptor number (AN).

General procedure: a boron compound (1 eq) and $Et_3P{=}O$ (0.6 eq) were dissolved in $CDCl_3$ (0.5 mL). Then, the mixture was transfered into an NMR tube and analyzed by ^{31}P NMR spectroscopy.[4]

Table S1. ^{31}P NMR shifts and acceptor number of boron compounds.[a]

Boron compound	^{31}P NMR shift (ppm)	[a]AN
B1	48.49	16.63
B2	48.54	16.74
B3	48.70	17.09
B4	49.06	17.89
B5	50.18	20.38
B6	50.56	21.22

[a]AN = (δ – 41.0) [100/(86.14 – 41.0)]. δ: ^{31}P NMR chemical shift of Et_3P=O after complexation with boron compounds.

4. DFT calculations

4.1 DFT calculations of one boron center with two sodium acetate

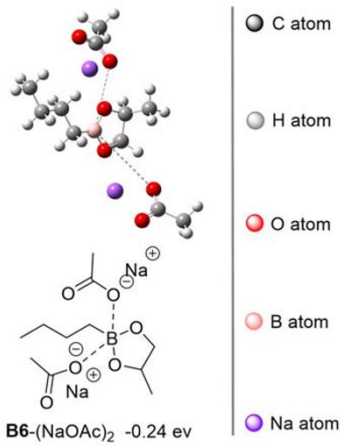


Figure S5. DFT calculations of binding energies between **B6** and two sodium acetate.

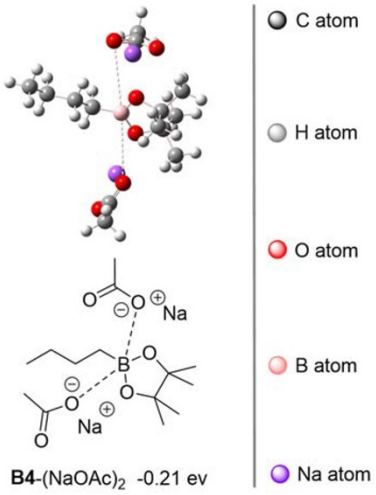


Figure S6. DFT calculations of binding energies between **B4** and two sodium acetate.

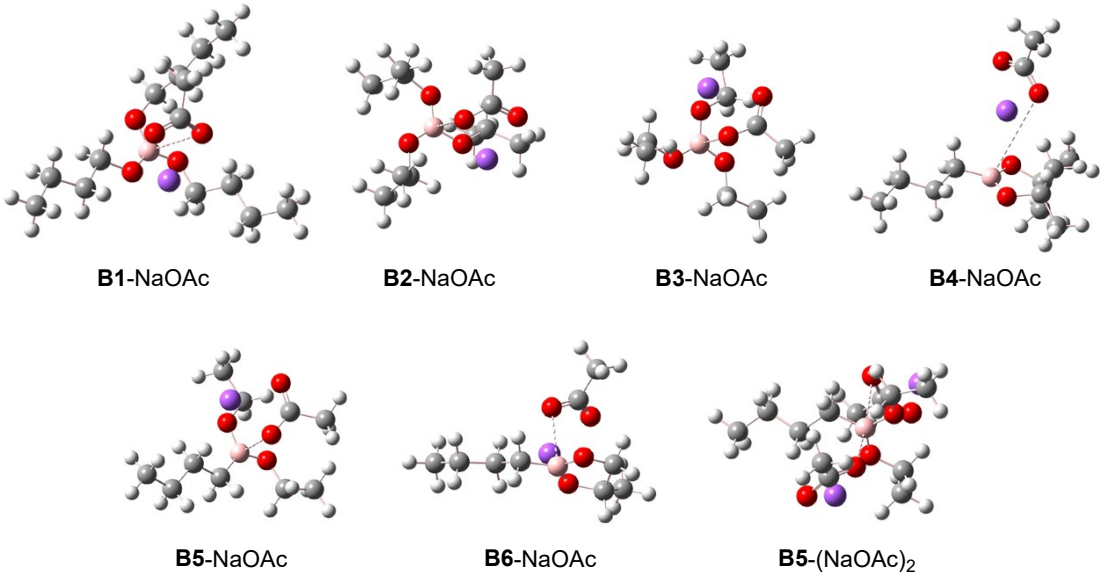


Figure S7. Optimized geometries for sodium acetate (NaOAc) coordinated with different boron compounds calculated by using B3LYP/6-31+G(d, p).

4.2 Cartesian coordinates of structures in manuscript

XYZ-Coordinates for the **B1**-NaOAc structure

B	0.16025300	-0.10105700	-1.54059100
O	0.02552200	-1.43689300	-1.74767200
O	-0.83440100	0.20244900	1.74835600
O	1.36475100	0.38059700	-1.04788300

C	-1.27572600	-2.01563700	-1.95500400
H	-1.87571200	-1.36154500	-2.59477400
H	-1.10853100	-2.95792600	-2.48292100
C	-1.96815900	-2.24813100	-0.61816300
H	-2.01158800	-1.29952100	-0.07298400
H	-1.34831400	-2.92186800	-0.01604800
C	2.45185000	-0.56336000	-0.84376900
H	2.62492200	-1.10438500	-1.77730000
H	2.15413900	-1.27865900	-0.07382100
C	3.69560600	0.19870600	-0.42144200
H	3.92752100	0.96169300	-1.17331600
H	3.50163900	0.72355100	0.52437400
O	-0.86438200	0.75202900	-1.82297900
C	-0.87445200	2.14752500	-1.53170300
H	0.10904800	2.47168900	-1.16384600
C	-1.97364900	2.46281600	-0.52323500
H	-1.87126400	1.78767600	0.33196300
H	-2.94152900	2.23648600	-0.98347300
C	-0.06849800	-0.67388600	2.25556400
C	-0.69601200	-1.90797600	2.88741300
H	-0.13887300	-2.80009900	2.59534100
H	-0.62265400	-1.81842700	3.97524800
H	-1.74430800	-2.00335300	2.60775500
O	1.19349700	-0.57516200	2.27266600
H	-1.04202800	2.68069500	-2.47306000
C	-1.94392000	3.92190500	-0.05833900
H	-2.02091800	4.59238400	-0.92329100
H	-0.97072200	4.13975200	0.40435400
C	-3.05885800	4.24033700	0.94187200

H	-3.01356000	5.28041000	1.27620300
H	-4.04299200	4.07583000	0.49283200
H	-2.98655800	3.59648200	1.82322500
C	-3.37584100	-2.82988000	-0.77238000
H	-3.32847700	-3.77455000	-1.32873000
H	-3.98613100	-2.14778600	-1.37695600
C	-4.05923400	-3.06442700	0.57832500
H	-5.06682000	-3.47088500	0.45355900
H	-3.48684200	-3.76903500	1.18961400
H	-4.13926400	-2.12980300	1.14150000
C	4.89827900	-0.72892000	-0.21802700
H	4.63860600	-1.49365400	0.52202800
H	5.10626000	-1.25962400	-1.15456200
C	6.15246300	0.02225000	0.23631400
H	5.98125500	0.52995400	1.19072500
H	6.99731700	-0.65837800	0.36956600
H	6.44638900	0.78014800	-0.49685900
Na	0.99597500	1.32235800	1.08474200

XYZ-Coordinates for the **B2**-NaOAc structure

B	0.80021300	0.39223300	-0.28998600
O	1.56906300	-0.28515600	0.60801500
O	-0.04126200	1.41507900	0.14320300
O	0.85173200	0.12189800	-1.61946700
C	-0.63103800	2.32183400	-0.83793900
H	-0.89768800	1.73110900	-1.71666500
C	1.57304600	-0.09423200	2.02791300
C	1.55147200	-0.99471300	-2.22044400
H	1.27679400	-0.92110300	-3.27695600

C	1.23540900	-1.43017800	2.68118800
H	2.01779400	-2.15897500	2.45265600
H	1.17393000	-1.32260300	3.76836800
H	0.28973400	-1.81649800	2.29522600
C	-1.89079300	2.91741700	-0.22576900
H	-1.65590000	3.43964700	0.70897100
H	-2.32771400	3.64866700	-0.90998000
H	-2.64695700	2.14545200	-0.05348700
C	0.40408400	3.37265500	-1.21605100
H	0.69915500	3.95139300	-0.33586200
H	1.29085000	2.89720700	-1.63982600
H	-0.00545900	4.05911400	-1.96200400
C	3.06229800	-0.82108700	-2.09381000
H	3.37031800	-0.89331400	-1.04963600
H	3.57542000	-1.59948800	-2.66557400
H	3.36969100	0.15166900	-2.48628800
C	1.04445800	-2.32484000	-1.67190200
H	-0.04540900	-2.36387400	-1.70661900
H	1.44800300	-3.14639700	-2.27090000
H	1.34726000	-2.46094000	-0.63360000
H	0.81834200	0.65914800	2.29560000
C	2.93778400	0.44851500	2.43565300
H	3.13436600	1.40452500	1.94469400
H	2.98773600	0.59419000	3.51827400
H	3.71950800	-0.25761000	2.14366000
O	-1.55622500	-1.60215400	0.69874200
C	-2.64824100	-1.35067000	0.10426800
C	-3.16033100	-2.36391600	-0.91004800
H	-2.83235700	-3.37055900	-0.64890500

H	-4.24680300	-2.31675900	-0.98803400
H	-2.73965800	-2.11182600	-1.88907400
O	-3.31470800	-0.28781800	0.27870500
Na	-1.64179100	0.46519600	1.56763100

XYZ-Coordinates for the **B3**-NaOAc structure

B	0.13342000	0.84985800	0.83385700
O	1.35066600	0.31484000	1.11628700
O	-0.04950300	-0.37332400	-1.88021800
O	-1.00581300	0.14224900	1.20355000
C	2.54328800	0.91095500	0.57750300
H	2.46421100	0.91772600	-0.51363800
H	2.61619900	1.94922500	0.91613600
C	3.73632100	0.09747200	1.04094400
H	3.65409200	-0.93613400	0.69685100
H	4.66126300	0.52221900	0.64063300
H	3.80060100	0.09206300	2.13197600
C	-0.83622500	-1.05591800	2.00958100
H	-0.20237600	-0.81245900	2.86502800
H	-0.33141700	-1.81454200	1.40760700
C	-2.20253500	-1.53145400	2.46480700
H	-2.09411700	-2.40139200	3.11757800
H	-2.72781900	-0.74644800	3.01425100
H	-2.82415900	-1.84325700	1.61830800
O	0.03605000	2.07819600	0.25556500
C	-1.17605600	2.67577000	-0.19811000
H	-2.03906900	2.06507400	0.09720000
C	-1.11614700	2.87303000	-1.70647800
H	-0.92851400	1.92169500	-2.20940300

H	-2.05081700	3.30979200	-2.07280500
H	-0.29740600	3.54897500	-1.96297200
C	0.17508400	-1.59664600	-1.62584700
C	1.56159900	-2.14715300	-1.92178100
H	1.51170600	-3.20955200	-2.16185300
H	2.03814400	-1.58763600	-2.72704200
H	2.16726200	-2.03255600	-1.01718600
O	-0.67584100	-2.36956000	-1.09515300
H	-1.28156600	3.63518400	0.31757900
Na	-2.01452400	-0.59298300	-0.81558800

XYZ-Coordinates for the **B4**-NaOAc structure

B	1.03174300	0.35091900	-0.30684700
C	1.85941200	-0.94423400	-0.59945600
H	1.26860200	-1.80572100	-0.24934600
H	1.96046500	-1.08511300	-1.68375100
C	3.24184100	-0.98842100	0.07307400
H	3.12289300	-0.83108100	1.15046900
H	3.84839300	-0.14787500	-0.28513200
C	3.99614300	-2.29815400	-0.17469200
H	4.10814600	-2.45180900	-1.25532200
H	3.38934900	-3.13593800	0.19038200
C	5.37283400	-2.33012200	0.49454000
H	5.28498100	-2.20943200	1.57890700
H	5.88959800	-3.27496000	0.30499400
H	6.00984900	-1.52095200	0.12301700
O	1.27979100	1.22231400	0.70779400
O	-0.08244600	0.71815700	-1.05635800
C	0.13164300	2.12371100	0.83065500

C	-0.48422800	2.07078500	-0.61246300
C	0.64217200	3.49420200	1.25446300
H	-0.17496000	4.22059800	1.25974000
H	1.04871900	3.43219800	2.26562200
H	1.43038500	3.85418000	0.59369100
C	-0.78758100	1.52857700	1.89940300
H	-0.20895200	1.39116300	2.81510700
H	-1.61945900	2.20259100	2.11694800
H	-1.20298800	0.56524100	1.59695500
C	0.16895200	3.04542200	-1.59292800
H	-0.16316800	2.80934800	-2.60604600
H	-0.11401700	4.07606700	-1.36950400
H	1.25761000	2.96773900	-1.56315400
C	-2.00189000	2.18622600	-0.65994300
H	-2.34550300	2.16474400	-1.69868100
H	-2.48994600	1.38362900	-0.10649600
H	-2.31297300	3.14412500	-0.23495800
O	-2.73637700	-2.78776900	-0.77802400
C	-3.06654800	-2.16068900	0.27102000
C	-3.95649200	-2.86956100	1.28221600
H	-3.76992400	-3.94389900	1.27016300
H	-3.80853500	-2.46021100	2.28195800
H	-5.00067900	-2.70612400	0.99715200
O	-2.71356700	-0.96589700	0.51500100
Na	-1.56757700	-1.00308900	-1.44192000

XYZ-Coordinates for the **B5**-NaOAc structure

B	-0.02088100	0.62360000	0.83945100
O	1.17021600	1.27436500	0.95058300

O	0.45516800	-0.17493200	-1.76703300
O	-0.06354600	-0.67630500	1.32618600
C	1.41240700	2.53180100	0.30632900
H	1.26831200	2.40173700	-0.77088000
H	0.69199900	3.27703200	0.66320700
C	2.83176900	2.96792000	0.61817600
H	3.54683100	2.22434000	0.25860200
H	3.05073800	3.92244800	0.13138800
H	2.97191900	3.08622000	1.69551900
C	1.11733900	-1.23822200	1.95545900
H	1.42910500	-0.57444500	2.76533200
H	1.92258800	-1.28406800	1.21931600
C	0.77635000	-2.61916300	2.48190800
H	1.63450900	-3.03816200	3.01389900
H	-0.07207700	-2.57664600	3.16950100
H	0.53524000	-3.30849100	1.66620400
C	-2.52888500	0.35238400	0.01824500
H	-2.28491200	-0.22344600	-0.88685200
C	-3.85495400	1.07453900	-0.24422100
H	-3.71112800	1.80126600	-1.05212300
H	-4.12484500	1.65496300	0.64612300
C	1.51620300	-0.87236600	-1.77138800
C	2.79869000	-0.21130200	-2.25455900
H	3.52478300	-0.95681600	-2.57838900
H	2.58674100	0.49783400	-3.05592400
H	3.22681300	0.34642700	-1.41533200
O	1.57296000	-2.06500800	-1.35329000
H	-2.66948400	-0.36186400	0.83981800
C	-1.35791900	1.29247200	0.33374600

H	-1.14194500	1.90651300	-0.54592400
H	-1.67241200	1.99878200	1.11856000
C	-4.99635100	0.11985200	-0.60384500
H	-5.17393100	-0.60259600	0.19984600
H	-5.93194500	0.65790900	-0.77870700
H	-4.76499600	-0.44462000	-1.51344200
Na	-0.55488900	-1.88861600	-0.66160700

XYZ-Coordinates for the **B6**-NaOAc structure

B	0.14219200	-1.25193300	-0.69795200
C	1.63358600	-0.97409300	-1.06868000
H	2.09694300	-1.95174700	-1.27188400
H	1.65958300	-0.43066300	-2.01820200
C	2.46682900	-0.22917500	-0.01523900
H	2.36889600	-0.73131500	0.95890300
H	2.06767100	0.78747400	0.08411000
C	3.95611900	-0.13010500	-0.36132800
H	4.06001800	0.35348200	-1.33942300
H	4.36820700	-1.14035600	-0.47275200
C	4.76100900	0.64464600	0.68532600
H	4.69397200	0.16569600	1.66826500
H	5.81952500	0.70308500	0.41770400
H	4.38778700	1.66892100	0.78757000
O	-0.33681700	-1.31657400	0.61616600
O	-0.82523600	-1.58150200	-1.60801100
C	-2.07335800	-1.70122600	-0.90264300
H	-2.64786200	-2.53061300	-1.31749600
H	-2.63743200	-0.77008000	-1.02007700
C	-1.66708000	-1.92832500	0.56000100

H	-1.51774500	-2.99728100	0.74972000
C	-2.60499300	-1.33414500	1.59092800
H	-2.73121400	-0.25973800	1.43252700
H	-3.58494600	-1.81461100	1.51424600
H	-2.23097500	-1.51426400	2.60297400
O	-0.21467200	1.67495600	-0.52656500
C	-1.39698600	2.02911700	-0.24012800
C	-2.23965000	2.63764900	-1.35162800
H	-3.24386600	2.87596400	-1.00276700
H	-1.75020100	3.54329000	-1.71864200
H	-2.28735600	1.93826600	-2.19063500
O	-1.91036500	1.88744600	0.91042200
Na	-0.02891700	0.81486300	1.55544400

XYZ-Coordinates for the **B4**-$(NaOAc)_2$ structure

B	0.19117200	0.19474700	-0.17553400
C	0.40195500	1.66279600	-0.66901500
H	1.42870800	1.72346700	-1.06158500
H	-0.24524600	1.87056700	-1.53070300
C	0.21105400	2.75870500	0.39979800
H	0.88036400	2.56742000	1.24857500
H	-0.80551800	2.70797500	0.80790800
C	0.47518000	4.17242200	-0.13003100
H	-0.19899700	4.36931400	-0.97241500
H	1.49263900	4.21730300	-0.53511500
C	0.29338800	5.25846800	0.93362600
H	0.98135700	5.11093500	1.77241100
H	0.48416000	6.25317500	0.52219900
H	-0.72479300	5.25495400	1.33526500

O	0.71050800	-0.30658200	1.00091900
O	-0.50800200	-0.76852900	-0.87288800
C	0.14077500	-1.65667400	1.22627000
C	-0.25309600	-2.08415700	-0.23458600
C	-1.05659900	-1.47506400	2.16042200
H	-1.47167500	-2.44433600	2.44697800
H	-0.72184800	-0.97212000	3.07081800
H	-1.85672100	-0.88487300	1.70848900
C	1.21537100	-2.52512200	1.87155100
H	1.42439000	-2.16078100	2.88205800
H	0.86100200	-3.55417900	1.97179200
H	2.14117500	-2.53394500	1.29430900
C	-1.52448900	-2.92004500	-0.32850800
H	-1.71791800	-3.18001500	-1.37380700
H	-1.39762400	-3.86052000	0.21397000
H	-2.39231000	-2.39857800	0.07832100
C	0.89051800	-2.73067000	-1.01816900
H	0.60900600	-2.78951800	-2.07209500
H	1.82401800	-2.16915300	-0.93706800
H	1.07435900	-3.74766100	-0.66363600
C	5.33098700	-0.33918600	-1.88481600
H	5.13942400	-1.29832100	-2.36723400
H	6.36530900	-0.34443300	-1.52656500
H	5.22610100	0.48059200	-2.59604100
C	4.40927900	-0.13988600	-0.68957700
O	4.10110000	1.04393800	-0.35962600
O	4.00796700	-1.16777000	-0.06169300
Na	2.93440400	0.21747300	1.36219800
C	-6.07913200	0.37893500	0.44241800

H	-5.89128500	0.52083200	1.50709700
H	-6.69074400	-0.52098900	0.32188400
H	-6.63906700	1.22031000	0.03303500
C	-4.77471400	0.18186700	-0.31747400
O	-4.74245000	0.49470800	-1.54326200
O	-3.78278300	-0.31131100	0.30470700
Na	-2.61692300	-0.17575700	-1.63655000

XYZ-Coordinates for the **B5**-$(NaOAc)_2$ structure

B	0.11761900	-0.43721700	0.18288700
O	0.52540000	-1.71850700	0.53799700
O	0.46365000	-0.00147900	-1.08105000
C	-0.21514800	-2.43824600	1.55018400
H	-0.06836900	-1.95845400	2.52452700
H	-1.27110200	-2.38665600	1.28064800
C	0.26701500	-3.87644800	1.56943600
H	1.32285900	-3.95066600	1.85627400
H	-0.30898300	-4.45857900	2.29317500
H	0.14414300	-4.32871200	0.58329400
C	1.08345300	-0.87124800	-2.05636000
H	0.32717000	-1.57540100	-2.42211800
H	1.88146500	-1.44423900	-1.58775400
C	1.67104500	-0.01363000	-3.15909000
H	2.10471500	-0.64815000	-3.93662100
H	0.91223900	0.62246900	-3.62887100
H	2.46015200	0.61161500	-2.74109500
C	-1.01208700	1.91572500	0.65238200
H	-0.30108400	2.31471600	-0.08332300
C	-1.24610100	2.98380800	1.72609000

H	-0.30089600	3.17386600	2.24706600
H	-1.93893300	2.58712500	2.47768500
H	-1.97064300	1.74756600	0.14239100
C	-0.47910100	0.58869100	1.21237400
H	0.36850400	0.80805200	1.87783600
H	-1.24796300	0.11602300	1.83152900
C	-1.79891900	4.29104500	1.15308700
H	-2.75772400	4.12543800	0.65132500
H	-1.95679000	5.03866600	1.93509000
H	-1.10864000	4.71991000	0.41896500
C	-4.39404800	-1.48077800	0.37459400
H	-4.14840200	-1.22875300	1.41130500
H	-4.31272900	-2.56396400	0.26845500
H	-5.41034700	-1.14903100	0.16598300
C	-3.39436700	-0.78583900	-0.54093300
O	-3.75178600	0.26731500	-1.13478400
O	-2.23161100	-1.29282300	-0.63234600
Na	-1.67710800	0.56150700	-1.89051000
C	3.27015900	2.33358800	-0.23278200
H	2.32441900	2.49853700	-0.75920600
H	4.08628000	2.50884600	-0.93440900
H	3.32131900	3.02791200	0.60566900
C	3.27775400	0.89193900	0.25699200
O	2.81473000	0.65516800	1.41072100
O	3.69285800	-0.00362800	-0.53808300
Na	2.85686800	-1.51624000	0.89708100

XYZ-Coordinates for the **B6**-$(NaOAc)_2$ structure

B	-0.13135600	0.88716000	-0.24338700

C	0.14885500	2.42813700	-0.27043100
H	-0.12443400	2.82587100	-1.25627200
H	-0.54726200	2.91136300	0.43094100
C	1.58709200	2.84942100	0.08013100
H	2.28699500	2.39558500	-0.63554800
H	1.84888900	2.46011300	1.07306300
C	1.80648000	4.36706700	0.07508700
H	1.10866800	4.82935200	0.78334400
H	1.54401500	4.76179400	-0.91350300
C	3.24028300	4.77221300	0.42623000
H	3.95829500	4.35125600	-0.28446000
H	3.36002600	5.85836700	0.41146600
H	3.51844300	4.42360900	1.42588100
O	0.73325400	-0.04426000	0.27538000
O	-1.28421100	0.30840500	-0.73352400
C	-1.12843300	-1.15173400	-0.66011300
H	-0.93016500	-1.50750200	-1.67292200
H	-2.06392100	-1.57314700	-0.29161200
C	0.07035600	-1.36994900	0.27862100
H	0.79860200	-2.07310800	-0.12534600
C	-0.30237900	-1.74285400	1.70141300
H	-1.01160700	-1.03018700	2.13169400
H	-0.76971200	-2.73093000	1.70643900
H	0.58778200	-1.79118000	2.33192900
C	-6.57871300	-1.44499500	0.63479500
H	-6.20781400	-2.15701000	1.37347300
H	-7.02027800	-2.01885100	-0.18604000
H	-7.35282700	-0.81003100	1.06529200
C	-5.43608300	-0.60745900	0.07921000

O	-5.66388500	0.60017700	-0.22432100
O	-4.30584000	-1.16629300	-0.07776400
Na	-3.54740800	0.79822200	-0.88783000
C	5.12414300	-3.55432300	-0.64395600
H	4.50726000	-4.23648800	-1.23044800
H	5.42107100	-4.07660900	0.27107300
H	6.02762900	-3.28354900	-1.19031700
C	4.33277700	-2.31320200	-0.25641400
O	4.95095700	-1.21353100	-0.14965400
O	3.08890100	-2.44062500	-0.03284000
Na	3.03626700	-0.22635200	0.42175300

5. Photos of creep tests

5.1. Photos of ionomer and I-B5

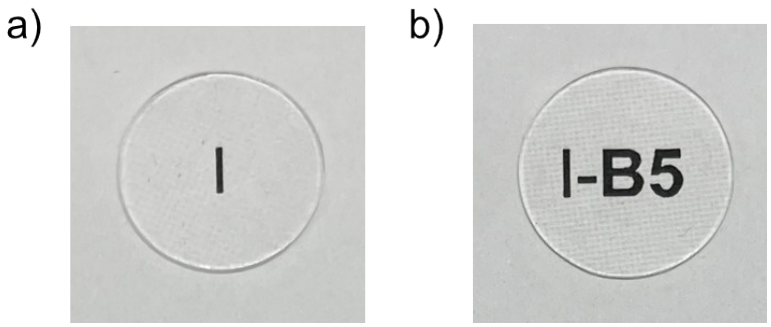


Figure S8. Photos of a) ionomer and b) **I-B5** at room temperature.

5.2. Photos of creep tests of ionomer, I-B3, I-B5, I-B6

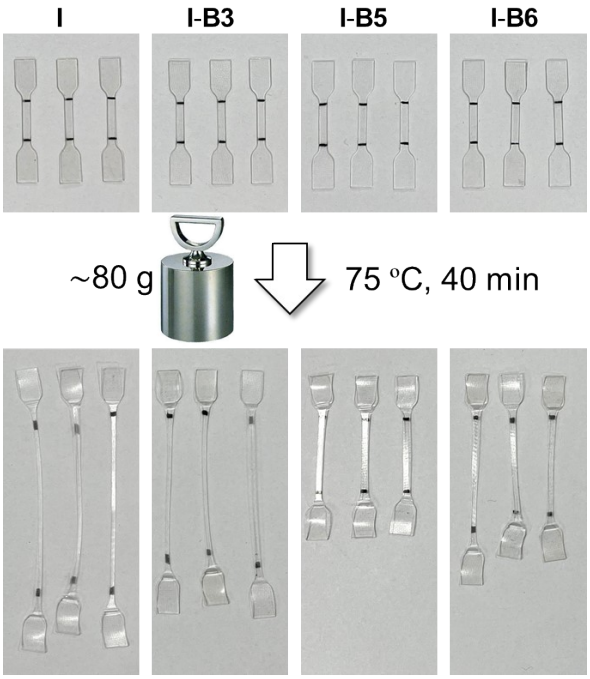


Figure S9. Photos of creep tests of ionomer, **I-B3**, **I-B5**, **I-B6** at 75 °C.

6. DSC results of ionomer and I-B materials.

6.1. Summarized results for ionomer and I-B materials.

Table S2. DSC results of ionomer and **I-B** materials.

	Ionomer	**I-B1**	**I-B2**	**I-B3**	**I-B4**	**I-B5**	**I-B6**
T_{m1} (°C)	93.7	93.1	93.2	93.9	88.8	90.6	89.7
ΔH_{m1} (J/g)	40.56	40.26	39.84	40.52	41.05	37.05	41.07
[b]X_{m1} (%)	13.8	13.7	13.6	13.8	14.2	12.6	14.0
T_{m2} (°C)	50.6	50.3	50.2	50.3	46.1	52.3	40.5
ΔH_{m2} (J/g)	8.23	9.23	9.98	9.53	6.08	6.15	6.97
[a]X_{m2} (%)	2.8	3.2	3.4	3.3	2.1	2.1	2.4

T_{m1} and T_{m2} represent melting points of primary and secondary crystallinities, respectively. ΔH_{m1} and ΔH_{m2} represent the heat of fusion of primary and secondary crystallinities, respectively. X_{m1} and X_{m2} represent the degrees of primary and secondary crystallinities, which are calculated from ΔH_{m1} or ΔH_{m2} relative to the equilibrium heat of fusion of PE.[5]

6.2. DSC analyses of ionomer and I-B materials

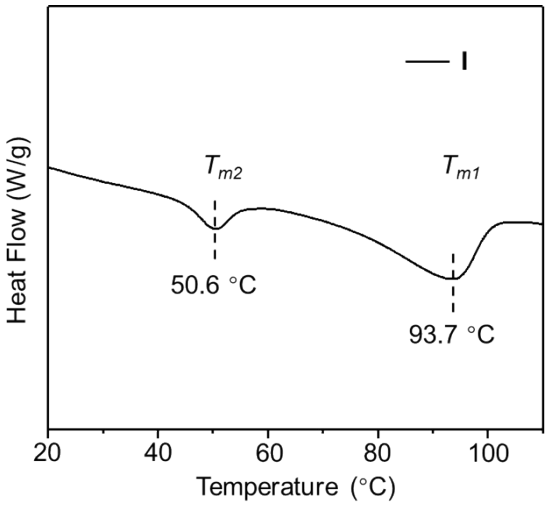


Figure S10. DSC curve of ionomer.

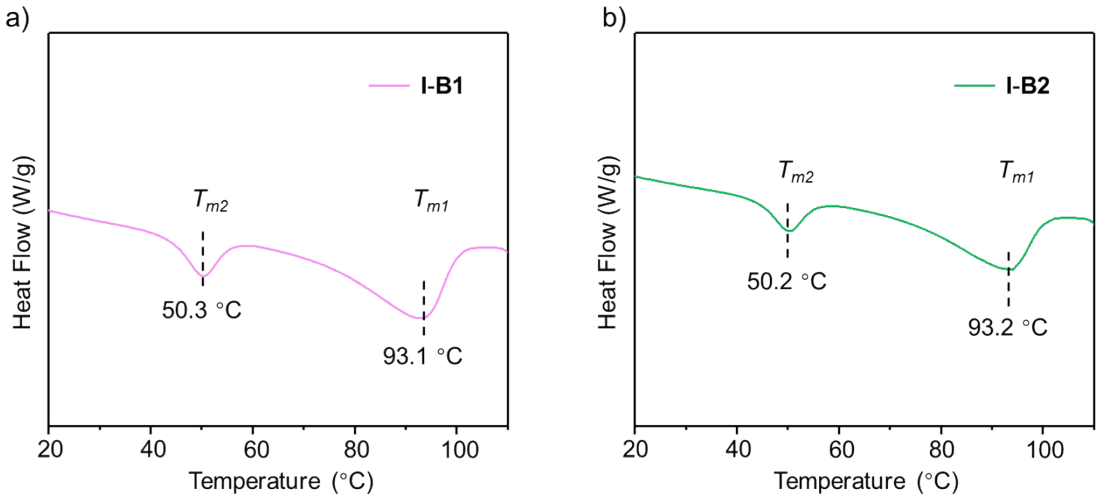


Figure S11. DSC curves of a) **I-B1** and b) **I-B2.**

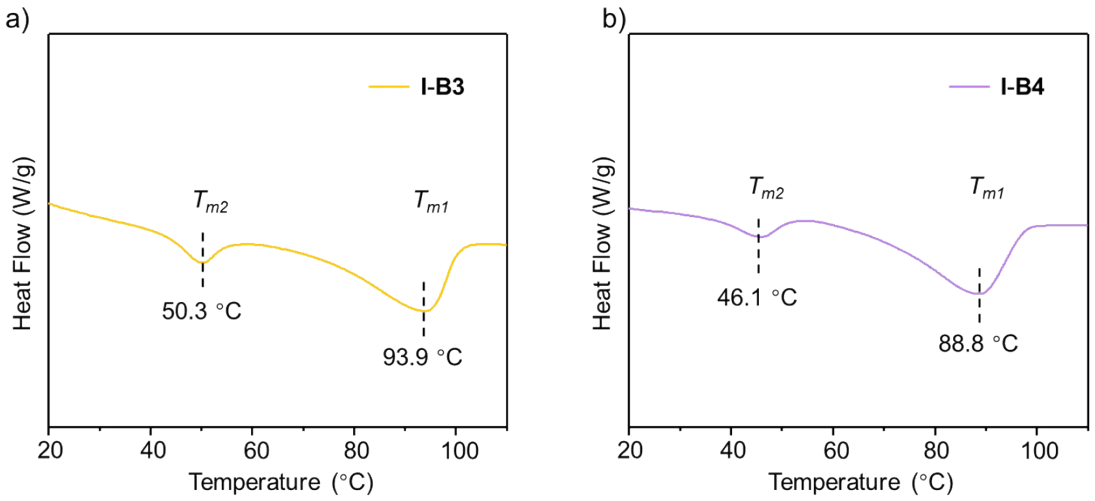


Figure S12. DSC curves of a) **I-B3** and b) **I-B4.**

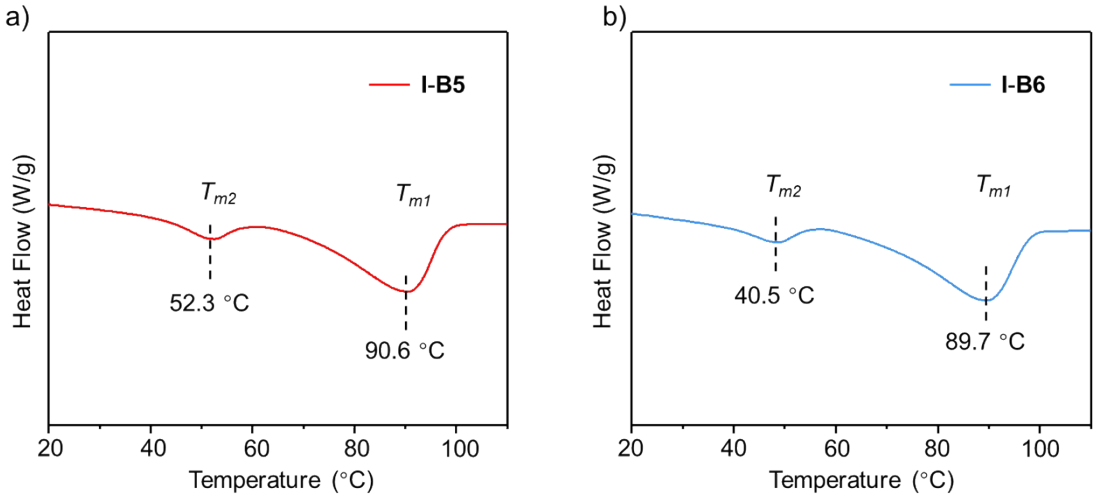


Figure S13. DSC curves of a) **I-B5** and b) **I-B6.**

7. Other supplementary information.

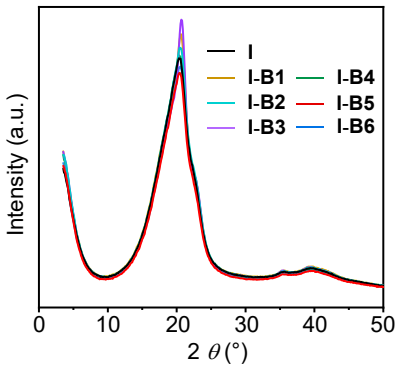


Figure S14. WAXS curves of the ionomer and samples from **I-B1** to **I-B6**.

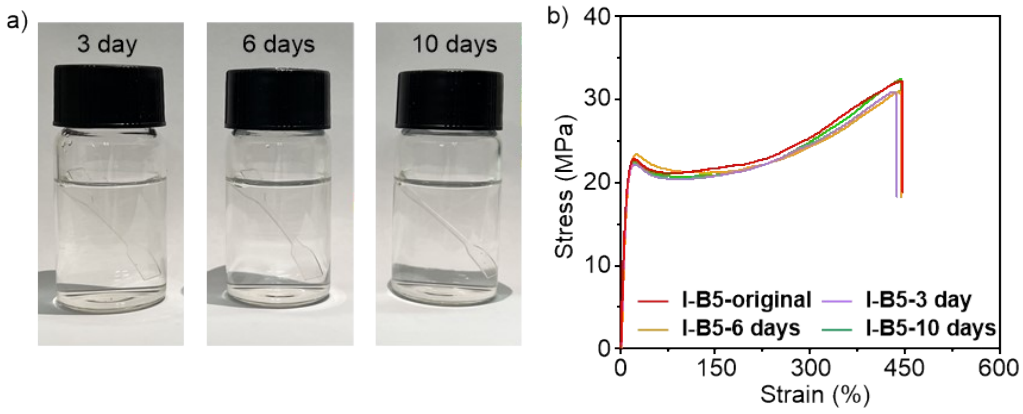


Figure S15. WAXS curves of the ionomer and samples from **I-B1** to **I-B6**.

Table S3. Swelling behavior of **I-B5** immersed in DCM, Hexane and Toluene.

Sample	Solvent	Time (h)	Temperature (°C)	W_1 (g)	W_0 (g)	Q_e (%)
I-B5	DCM	72	25	0.99	0.97	2.06
I-B5	Hexane	72	25	0.98	0.97	1.03
I-B5	Toluene	72	25	1.00	0.98	2.04

The equilibrium swelling ratio (Q_e) was calculated via Equation S2:

$$Q_e = (W_1 - W_0)/W_0 \quad \textbf{Equation S2}$$

Where Q_e is the equilibrium swelling ratio, W_1 and W_0 are the weights of swollen and dry **I-B5** samples.

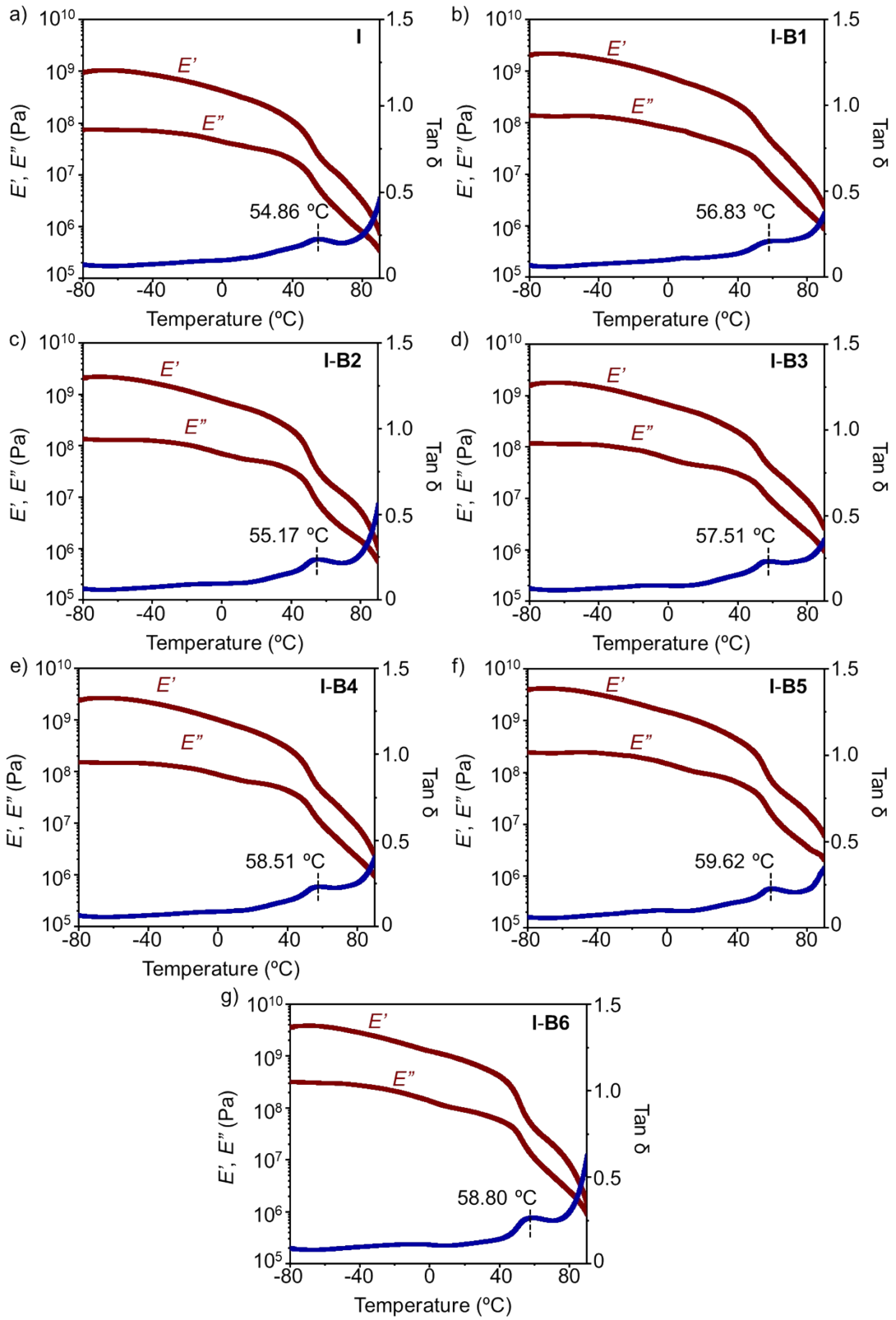


Figure S16. WAXS curves of the ionomer and samples from **I-B1** to **I-B6**.

8. Reference

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5. E. Pregi, D. Kun, A. Wacha and B. Pukanszky, *European Polymer Journal*, 2021, **142**, 110110.

9. Author contributions

P. Wen, Y. Zeng, and L. Zhang prepared samples and conducted characterizations; Y. Zeng, P. Wen and M. Chen conceived the idea; P. Wen conducted calculations; P. Wen, Y. Zeng, T. Wang and M. Chen analysed results; Jeffrey M. Cogen, Colin Li Pi Shan and Yixuan Liu reviewed and edited the manuscript. T. Wang and M. Chen supervised the project and wrote the manuscript.