Supporting Information

3D Printed Modular Strain Sensors Based on Imidazolium Ionic Liquids

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Supplementary Information

General Reagent Information

All purchased reagents were used as received without further purification unless otherwise specified. 1-vinylimidazole (>98.0%), 1-bromobutane (>99.0%), 1-bromoethane (>98.0), 2isocyanoethyl methacrylate (>98.0%, stabilized with BHT), dibutyltin dilaurate (>95.0%), triethylamine (>99.0%), furan (stabilized with BHT, >99.0+%), maleic anhydride (>99.0%), Propargyl amine (>97.0%), 1,1,1,3,3,3-hexamethyldisilazane (>96.0%), CuBr (>98.0%), N,N,N',N'',N''-Pentamethyldiethylenetriamine (PMDETA, >99.0%), and 4-methoxyphenol (MEHQ, >99.0%) were purchased from TCI America. Lithium bis(trifluoromethanesulfonyl)imide salt (99.95%), 4-dimethylaminopyridine (>99.0%), ptoluenesulfonyl chloride (>98.0%), N,N-dimethylformamide (anhydrous, 99.8%), toluene (anhydrous, 99.8%), sodium azide (>99.5%), furfuryl alcohol (98%), methacrylic anhydride (contains 2,000 ppm topanol A as inhibitor, ≥94%), methanol (>99.9%), (+)-sodium L-ascorbate (>99.0%), aluminum oxide (activated, neutral, Brockmann I) and ethyl acetate (>99.7%) were purchased from Sigma Aldrich. PEG-8000, diethyl ether (anhydrous, stabilized with BHT, >99.0%), acetonitrile, tetrahydrofuran (>99.8%), and dichloromethane (>99.9%) were purchased from Fisher Scientific. Dry tetrahydrofuran and dichloromethane were obtained from a Pure Process Technology purification system.

General Analytical Information

¹H NMR spectra were recorded on a Bruker 300 or 500 MHz instrument. Rheology measurements were performed on a TA Discovery HR-2 Hybrid Rheometer. Mechanical stress/strain measurements were performed on a Test Resources Universal Testing Machine. 3D printed structures were printed using a Form Labs Form 2 SLA printer. Mass Spec measurements were performed on a Bruker Esquire LC - Ion Trap Mass Spectrometer. DSC measurements were performed on a TA Discovery DSC 2500 instrument. Dynamical Mechanical Thermal Analysis (DMTA) was recorded on a TA Instruments DMA 850. Electrical impedance spectroscopy (EIS) measurements were performed with a Keithley 2400 source measure unit (SMU). Cyclic tensile experiments were conducted on a TA Q5000 thermogravimetric analyzer.



Scheme S1. Overview of synthetic route to DA-PEG-1.

Synthesis of exo-3,6-epoxy-1,2,3,6-tetrahydrophtalic anhydride (maleic anhydride Diels-Alder adduct):



Scheme S2. Synthetic route to exo-3,6-epoxy-1,2,3,6-tetrahydrophtalic anhydride

A Teflon stir bar, 10 g of maleic anhydride (0.102 mol), 9.86 mL of furan (0.136 mol), and 30 mL of ethyl acetate were added to a 250 mL round bottom flask. The reaction mixture was left to stir at room temperature for 24 h under N₂. The product was collected via vacuum filtration and dried at room temperature under high vacuum using a Schlenk manifold (Yield: 11.79 g, 69.60%). ¹H NMR (300 MHz, DMSO- d_6): δ 6.60 (d, 2H), 5.36 (dd, 2H), 3.33 (d, 2H).



Figure S1. ¹H NMR spectrum of exo-3,6-epoxy-1,2,3,6-tetrahydrophtalic anhydride in DMSO-*d*₆.

Synthesis of 3a, *4*, *7*, *7a*-*Tetrahydro-2-(2-propyn-1-yl)-4*, *7-epoxy-1H-isoindole-1*, *3(2H)-dione (alkyne maleimide):*



Scheme S3. Synthetic route to alkyne maleimide.

A Teflon stir bar and 11.80 g of exo-3,6-epoxy-1,2,3,6-tetrahydrophtalic anhydride (0.071 mol) was dissolved in 150 mL of methanol and 150 mL of THF in a single-neck 500 mL round-bottom flask under N₂ and subsequently cooled to 0 °C. 4.55 mL of propargylamine (0.071 mol), 25 mL of methanol, and 25 mL of THF were added to a separate 100 mL single-neck round-bottom flaskround-bottom flask and the contents of the 100 mL round-bottom flask were poured into the addition funnel. The contents of the addition funnel were added dropwise to the 500 mL roundbottom flask and left to stir for an additional 30 minutes at 0 °C upon completion. 17.76 mL of hexamethyldisilizane (0.085 mol) was added to the round-bottom flask and the reaction mixture was left to stir at 65 °C for 3 days under N₂. The reaction mixture is then cooled to room temperature and the remaining liquid is removed with a rotary evaporator from the orange product. The product is redissolved in minimal DCM and washed twice with a saturated sodium bicarbonate solution, twice with 2 M hydrochloric acid. The product is then dried using magnesium sulfate and vacuum filtered to remove solid impurities. Product was put on rotary evaporator at 30 °C to obtain off-white residue. The product is then recrystallized in acetone, collected via vacuum filtration, and dried at room temperature under high vacuum using a Schlenk manifold (Yield 5.19 g, 36.0%). ¹H NMR (300 MHz, CDCl₃): δ 6.52 (d, 2H), 5.30 (dd, 2H), 4.23 (s, 2H), 2.90 (s, 2H), 2.19 (s, 1H).



Figure S2. ¹H NMR spectrum of 3a,4,7,7a-Tetrahydro-2-(2-propyn-1-yl)-4,7-epoxy-1H-isoindole-1,3(2H)-dione in CDCl₃.



Scheme S4. Synthetic route to furfuryl methacrylate

A Teflon stir bar, 1.41 g of 4-dimethylaminopyridine (0.0116 mol), 43.38 mL of triethylamine (0.347 mol), 20 mL of furfuryl alcohol (0.231 mol), and 20 mL of ethyl acetate were added to a 500 mL single-neck round-bottom flask and left to stir for 5 minutes under N₂. 36.19 mL of methacrylic anhydride (0.243 mol) and 30 mL of ethyl acetate were added to a separate 100 mL single-neck round-bottom flask. A 100 mL addition funnel was attached to the 500 mL roundbottom flask and the contents of the 100 mL round-bottom flask were poured into the addition funnel. The contents of the addition funnel were added dropwise to the 500 mL round-bottom flask. Upon completion, the reaction mixture was left to stir at 55 °C for 24 hours under N₂. Any precipitated salts were removed via vacuum filtration and the remaining reaction mixture was diluted with ethyl acetate. The reaction mixture was washed twice with deionized water, twice with 1 M hydrochloric acid, twice with a saturated sodium bicarbonate solution and twice with a saturated brine solution. The product is then dried using magnesium sulfate and vacuum filtered to remove solid impurities. The product was then run through a neutral alumina column with a 10:1 mixture of hexanes: ethyl acetate as the eluent. The first analyte was collected and put on the rotary evaporator at 35 °C to obtain the product. Minimal MEHQ is added to the product to prevent crosslinking. ¹H NMR (300 MHz CDCl₃): δ 7.43 (d, 1H), 6.42 (dd, 1H), 6.36 (d, 1H), 6.13 (s, 1H), 5.57 (s, 1H), 5.14 (s, 2H), 1.95 (s, 3H).



Figure S3. ¹H NMR spectrum of furfuryl methacrylate in CDCl₃.

Scheme S5. Synthetic route to PEG-tosylate.

A Teflon stir bar and 10 g of PEG-8000 (0.00125 mol) was dissolved in 100 mL of anhydrous DCM in a single-neck 250 mL round-bottom flask under N₂ and cooled to 0 °C. Maintaining positive N₂ flow, 2.38 g of tosyl chloride (0.0125 mol) and 0.100 g of 4-dimethylaminopyridine (0.00075 mol) were added portion-wise to the round-bottom flask, followed by 1.74 mL of triethylamine (0.0125 mol) added dropwise with stirring at 0 °C. The reaction mixture was stirred for an additional hour at 0 °C and then left to stir at room temperature for 24 hours under nitrogen. The reaction mixture is then washed twice with deionized water, twice with saturated sodium bicarbonate solution and twice with a saturated brine solution. The product is then dried using magnesium sulfate and vacuum filtered to remove solid impurities and then precipitated twice in diethyl ether. The product was collected via vacuum filtration and dried at room temperature under high vacuum using a Schlenk manifold (Yield: 6.74 g, 64.6%). ¹H NMR (500MHz CDCl₃): δ 7.82 (d, 4H), 7.36 (d, 4H), 4.19 (t, 4H), 2.48 (s, 6H), protons from PEG appear at 3.67 ppm.



Figure S4. ¹H NMR spectrum of PEG-tosylate in CDCl₃.



Scheme S6. Synthetic route to PEG-azide.

A Teflon stir bar and 5.98 g of PEG-tosylate (0.00072 mol) was dissolved in 100 mL of DMF in a single-neck 250 mL round-bottom flask under N₂. Maintaining positive N₂ flow, 0.470 g of sodium azide (0.0072 mol) was added to the round-bottom flask. The reaction mixture is then left to stir at 80 °C for 2 days under N₂. DMF is then removed using a rotary evaporator. The product is then redissolved in minimal DCM and washed twice with deionized water and then twice with a saturated brine solution. The product is then dried using magnesium sulfate and vacuum filtered to remove solid impurities. The product is then precipitated twice in diethyl ether, collected via vacuum filtration, and dried at room temperature under high vacuum using a Schlenk manifold (Yield: 4.41 g, 76.1%). ¹H NMR (500 MHz CDCl₃): δ 3.36 (t, 4H), protons from PEG appear at 3.62 ppm.



Scheme S7. Synthetic route to PEG-maleimide

A Teflon stir bar and 0.17 g of copper (I) bromide (0.00123 mol) were added to a 250 mL singleneck round-bottom flask and pump purged with N₂. 36 mL of methanol, 4 mL of water, 0.257 mL of N,N,N',N'',N''-pentamethyldiethylenetriamine (0.00123 mol), and 0.6 of sodium ascorbate (0.00303 mol) were added to the round-bottom flask and allowed to stir under N₂ for 24 hours. 10 g of PEG-azide (0.00123 mol) and 2.5 g of alkyne maleimide (0.0123 mol) were added to the round-bottom flask and allowed to stir under N₂ for 24 hours. The reaction mixture was then filtered through neutral alumina and put on the rotary evaporator initially at 35 °C to remove DCM, then 50 °C to remove methanol and water. The product is then precipitated twice in diethyl ether, collected via vacuum filtration, and dried at room temperature under high vacuum using a Schlenk manifold (Yield: 7.8 g, 74.7%). ¹H NMR (500 MHz CDCl₃): δ 7.65 (s, 2H), 6.51 (d, 4H), 5.28 (dd, 4H), 4.78 (s, 4H), 4.48 (t, 4H), 2.88 (d, 4H), protons from PEG appear at 3.63 ppm.



Figure S6. ¹H NMR spectrum of PEG-maleimide in CDCl₃.

Deprotection of PEG-maleimide:



Scheme S8. Synthetic route to deprotect PEG-maleimide.

A Teflon stir bar, 1.7 g of PEG-maleimide (0.000200 mol), and 50 mL of toluene were added to a 100 mL single-neck round-bottom flask. The flask was left to stir and refluxed for 24 hours at 140 °C under N₂. The product mixture was placed on the rotary evaporator at 55 °C to remove toluene and then redissolved in DCM. The product is then precipitated twice in diethyl ether, collected via vacuum filtration, and dried at room temperature under high vacuum using a Schlenk manifold (Yield: 1.49 g, 88.9%). ¹H NMR (500 MHz CDCl₃): δ 7.71 (s, 2H), 6.73 (s, 4H), 4.82 (s, 4H), 4.50 (t, 4H), protons from PEG appear at 3.64 ppm.



Figure S7. ¹H NMR spectrum of deprotected PEG-maleimide in CDCl₃. *Synthesis of DA-PEG-1*:



Scheme S9. Synthetic route to DA-PEG-1.

A Teflon stir bar and 4.78 g of deprotected PEG-maleimide (0.000578 mol) were added to a 100 mL single-neck round-bottom flask under N2 and heated to 70 °C. Once the polymer was fully melted, 1.77 mL of furfuryl methacrylate (0.0115 mol) was added to the round-bottom flask. The reaction was left to stir at 70 °C for 24 hours under N₂. The product was dissolved in minimal DCM then precipitated twice in diethyl ether, collected via vacuum filtration, and dried at room temperature under high vacuum using a Schlenk manifold (Yield: 4.40 g, 88.5%). ¹H NMR (500 MHz, CDCl₃): δ 7.64 (s, 2H), 6.54 (d, 2H), 6.43 (dd, 2H), 6.12 (dd, 2H), 5.55 (s, 2H), 5.25 (s, 2H), 4.89 (s, 4H), 4.58 (d, 4H), 4.46 (t, 4H), 3.38 (s, 2H), 3.00 (d, 1H), 2.92 (d, 1H), 1.91 (s, 6H).



Figure S8. ¹H NMR spectrum of methacrylated PEG-maleimide in CDCl₃.



A Teflon stir bar and 60 g of PEG-8000 (0.0075 mol) were dried under vacuum for 12 hours in a single-neck 1 L round-bottom flask. The flask was then backfilled with dry nitrogen. Maintaining positive N₂ flow, 550 mL of anhydrous DCM was added to the round-bottom flask and stir at 30 °C until PEG is fully dissolved. Add 0.89 mL (0.0015 mL) dibutyltin dilaurate to the PEG solution. Attach a 50 mL addition funnel to the 1 L round-bottom flask. In a separate 100 mL round-bottom flask, add 50 mL of anhydrous DCM along with 5.3 mL (0.0375 mol) of 2-isocyanatoethyl methacrylate and swirl to mix. Pour the isocyanate/DCM mixture into the 50 mL addition funnel. Adjust the flow rate of the addition funnel so that the isocyanate is added *slowly* (about 1 drop per second) to the reaction mixture. This can take up to 2 hours. Following the complete addition of isocyanate, remove the addition funnel and reattach the vacuum adapter to the round-bottom flask. Stir reaction mixture at 30 °C for 2 days under nitrogen. The reaction mixture is then concentrated on a rotary evaporator at 30 °C until viscous. The product is precipitated twice in diethyl ether. The product was collected by vacuum filtration and dried at room temperature under high vacuum using a Schlenk manifold (Yield: 19.48 g, 93.7%). ¹H NMR (500 MHz, CDCl₃): δ 6.04 (s, 2H), 5.52 (t, 2H), 5.18 (t, 1H) 1.87 (s, 6H), protons from PEG appear at 4.14 ppm and 3.57 ppm. 13 C NMR (300 MHz, CDCl₃): δ 70.59. IR (ATR-FTIR, ν_{max} , cm⁻¹): 2880, 1720, 1468, 1340, 1280, 1092, 960, 840.



Synthesis of 1-vinyl-3-butyl imidazolium bromide ([BVIM]Br):



Scheme S11. Synthetic route to [BVIM]Br

200 mL (2.2 mol) of 1-vinyl imidazole and 285 mL (2.65 mol) of 1-bromobutane were stirred magnetically at 45 °C for 24 hours. The viscous solution was allowed to cool before washing 3-4 times with excessive diethyl ether until the liquid solidifies. The product was collected by vacuum filtration and dried at room temperature under high vacuum using a Schlenk manifold. The product appears as a white solid (Yield: 456.23 g, 90.1%). ¹H NMR (500 MHz, DMSO-D6): δ 9.61 (s, 1H), 8.24 (t, 1H), 7.97 (t, 1H), 7.33 (m, 1H), 6.00, 5.43 (d of d, 2H), 4.22 (t, 2H), 1.81 (q, 2H), 1.30 (m, 2H), 0.91 (t, 3H). ¹³C NMR (300 MHz, CDCl₃): δ 135.22, 128.74, 123.17, 119.17, 108.57, 48.81, 30.98, 18.68, 13.19. IR (ATR-FTIR, v_{max}, cm⁻¹): 3092, 3040, 2956, 1652, 1568, 1540, 1456, 1160, 984, 928, 880, 804, 740, 724, 660, 636, 604.



Figure S10. ¹H NMR spectrum of [BVIM]Br in DMSO- d_6 .

Synthesis of 1-vinyl-3-butyl imidazolium bis(trifluoromethanesulfonyl)imide ([BVIM]TFSI):



Scheme S12. Synthetic route to [BVIM]TFSI

96.34 g (0.42 mol) of [BVIM]Br and 122.6 g (0.427 mol) of Lithium bis(trifluoromethanesulfonyl) imide salt were dissolved in 160 mL of DI water and stirred at room temperature for 48 hours. The mixture was then poured into a separatory funnel and the ionic liquid layer was allowed to separate. The ionic liquid was separated from the water layer, then passed through a column of neutral alumina. The product appears as a clear liquid. (Yield: 152.12 g, 84%). ¹H NMR (500 MHz, DMSO-D6): δ 9.46 (s, 1H), 8.18 (t, 1H), 7.91 (t, 1H), 7.28 (m, 1H), 5.96, 5.42 (d of d, 2H), 4.19 (t, 2H), 1.80 (q, 2H), 1.27 (m, 2H), 0.91 (t, 3H). ¹³C NMR (300 MHz, CDCl₃): δ 135.28, 128.85, 123.21, 119.16, 108.58, 48.97, 31.03, 18.77, 13.15. IR (ATR-FTIR, v_{max}, cm⁻¹): 3156, 2972, 1660, 1552, 1348, 1180, 1132, 1052, 956, 920, 740, 612, 568, 508.



Figure S11. ¹H NMR spectrum of [BVIM]TFSI in DMSO-*d*₆.

Ion Gel Resin Preparation:

Example of a typical procedure to prepare 20 g of 7.5-7.5-BVIM resin for 3D printing: 7.5 wt% (1.5 g) of PEG-BUM and 7.5 wt% (1.5 g) of **DA-PEG-1** is added to an amber vial. 0.75 wt% (0.15 g) of BAPO was added, followed by 0.025 wt% (0.005 g) of Sudan 1. 16.85 mL of [BVIM]TFSI (~85 wt%) is then added and the mixture is vortexed for 10-15 mins before being stored at 4 °C overnight. Solution is vortexed again if the PEG is not fully dissolved. The amount of each component used to make the fusible ion gels are summarized in Table S1.

Table 51. Formulations for Fusible for Gets (20 g search					
Resin	PEG-BUM	DA-PEG-1	BAPO [g]	Sudan 1 [g]	[BVIM]TFSI
Formulation	[g]	[g]			[mL]
10-BVIM	0	2.0	0.15	0.005	17.85
15-BVIM	0	3.0	0.15	0.005	16.85
20-BVIM	0	4.0	0.15	0.005	15.85
7.5-7.5-	1.5	1.5	0.15	0.005	16.85
BVIM					

Table S1. Formulations for Fusible Ion Gels (20 g scale)

General process for 3D Printing Ion Gel Objects Using a Stereolithographic Apparatus (SLA) 3D Printer.

The desired resin was poured into the resin tray and the print was initiated on a Form Labs Form 2 SLA printer in open mode. The z-axis step size was 0.100 mm. CAD files used for building blocks were downloaded from Thingiverse.com (credit to mathgrrl and Rob65). Upon print completion, the structure was removed from the build plate carefully using a metal scraper and residual resin was removed by gently rolling the structure in kimwipes until minimal resin remained. If the shape of the structure permits, the printed structures may be centrifuged at 1000 RPM for one minute to help remove residual resin. The structure was then cured using 405 nm light for 30 mins on one side, flipped, then cured for an additional 30 mins on the other side.

CreatingModular3DPrintedIonGelSensors.3D printed blocks were stuck together in the desired configuration and placed in a parafilmcovered glass petri dish. Samples were then heated in a vacuum oven. 10-BVIM, 15-BVIM, and20-BVIM samples were heated overnight at 60 °C under ambient pressure. 7.5-7.5-BVIM sampleswere heated at 85 °C for 4 hours under ambient pressure. Fused structures were then removed fromthe oven and allowed to reach room temperature.

Rheology.

Rheometrical characterization was performed on a TA Instruments DHR-2 equipped with an Advanced Peltier Plate system. Experiments were performed using an 8 mm flat stainless steel upper plate. Resin samples were loaded via glass pipette onto the bottom plate. The sample is then trimmed after the upper plate was lowered to the trim gap at 600 μ m. The final geometry gap was then set to 500 μ m, and pre-shear was applied at 5 °C for 10 s before additional sample conditioning at 25 °C for 8 min. Viscosity vs Shear Rate experiments were performed to measure the viscosity of the ion gel resins. The viscosity of resins must remain below 10 Pa*s for resin to flow properly during SLA printing. All resins reported in this manuscript had viscosities below this threshold. Resins were found to increase in viscosity with increasing amount of PEG co-polymer present.



Figure S12. Viscosity vs Shear rate of ionic liquid gel resins

Tensile Tests.

A TestResources Universal Test System 1.1 kN electromechanical actuator single column load frame with a 43 N high accuracy S10 type load cell was used to evaluate the mechanical properties of 3D printed ion gels. For dimensions of all tensile specimens, ISO 527-2 5B specimen specifications were used. The samples were attached to vice grips with diamond grit jaw and subjected to increasing strain at a constant rate of 5 mm/min until mechanical failure. Samples designated "mechanically mended" were cut with a razor at the center of the gauge region and stuck together mechanically by pushing the two ends of the pieces together. The samples were self-adhered prior to loading. Samples designated "thermally mended" were cut with a razor at the center of the gauge region and heated together at 85 °C for 4 h prior to loading.

Dynamic Mechanical Thermal Analysis.

Dynamical Mechanical Thermal Analysis (DMTA) was recorded on a TA Instruments DMA 850 equipped with a liquid nitrogen purge cooler. Specimens with dimensions of 20 mm x 3 mm x 0.6 mm were tested using a tensile dual screw film clamp. The experiment was carried out over a temperature range of -50 °C to 120 °C at a ramp rate of 2 °C/min, with an amplitude of 10 µm and a frequency of 1 Hz. A glass transition temperature (T_g) of 15 °C was determined from the peak in tan (δ).



Figure S13. Dynamic Mechanical Analysis (DMA) thermogram of 7.5-7.5-BVIM sensors, demonstrating the T_g (15 °C) of the gels. Results over a temperature range of -50 °C to 120 °C are shown.

Electrical Impedance Spectroscopy.

Ionic conductivities were determined by electrochemical impedance spectroscopy (EIS) in an Autolab 302N potentiostat galvanostat at various temperatures (20-100 °C) using a temperature control Microcell HC station. Samples were placed between stainless steel electrodes (surface area = 0.5 cm2). The plots were obtained using 10 mV amplitude in the 100 kHz to 1 Hz range.

Stress Relaxation Measurements.

Relaxation modulus and activation energy was determined by stress relaxation experiments performed in an ARES rheometer (Rheometrics) using a film tension fixture and 5 % of strain at temperatures ranging from 75 °C to 105 °C. The samples used for these measurements had a width between 2.5 to 3 mm and thickness between 0.75 to 1.05 mm. Temperature dependent relaxation times are described by the Arrhenious equation: $\tau(T) = e^{(Ea/RT)}$. The activation energy (E_a) of the dynamic bond was determined from the slope of the natural logarithm of τ plotted against 1000/T (E_a = 84 ± 5 kJ/mol).

Gel Fraction.

To determine the amount of unpolymerized ionic liquid in a fully cured structure, cylindrical pucks (d = 10 mm, h = 5 mm) of each resin formulation (10-BVIM, 15-BVIM, 20-BVIM) were printed on a Form 2 SLA printer. After printing, each sample was cured for 1 h with 405 nm light under a flow of nitrogen. Each puck was soaked in 50 mL of methanol for one week, with the solvent being replaced three times over that period. After one week, the samples were removed and placed in a vacuum oven at 50 °C overnight. To determine the amount of polymerized material remaining, the mass of the fully dried sample is subtracted from the original mass of the puck. The mass lost is due to unpolymerized ionic liquid being washed out by methanol. The average results are displayed in the table below. The data reveals that under all conditions, a portion of the ionic liquid remains unpolymerized after printing, leaving the networks swollen.

Name	Initial Mass (g)	Final Mass (g)	% mass lost	Gel Fraction
10-BVIM (1)	0.5878	0.3742	36.33%	63.66%
10-BVIM (2)	0.5816	0.3590	37.26%	62.74%
10-BVIM (3)	0.5722	0.3462	39.49%	60.50%
Averages	0.5805 ± 0.0078	0.3598 ± 0.0140	37.69 ± 1.6239	62.30 ± 1.6252

Table S2. Gel fraction data of 10-BVIM.

Table S3. Gel fraction data of 15-BVIM.

Name	Initial Mass (g)	Final Mass (g)	% mass lost	Gel Fraction
15-BVIM (1)	0.5718	0.3413	40.31%	59.68%
15-BVIM (2)	0.5844	0.3659	37.39%	62.61%
15-BVIM (3)	0.5791	0.3627	37.37%	62.63%
Averages	0.5784 ± 0.0063	0.3566 ± 0.0133	38.35 ± 1.6916	61.64 ± 1.6974

Table S4. Gel fraction data of 20-BVIM.

Name	Initial Mass (g)	Final Mass (g)	% mass lost	Gel Fraction
20-BVIM (1)	0.5668	0.3583	36.78%	63.21%
20-BVIM (2)	0.5606	0.3569	36.33%	63.66%
20-BVIM (3)	0.5751	0.3642	36.67%	63.32%
Averages	0.5675 ± 0.0072	0.3598 ± 0.0038	36.59 ± 0.2345	63.39 ± 0.2345

Name	Initial Mass (g)	Final Mass (g)	% mass lost	Gel Fraction
7.5-7.5-BVIM (1)	0.5814	0.3765	35.24	64.75%
7.5-7.5-BVIM (2)	0.5781	0.3724	35.58	64.41%
7.5-7.5-BVIM (3)	0.5774	0.3767	34.76	65.24%
Averages	$0.5789 \ \pm 0.0021$	0.3752 ± 0.0024	$35.19\% \pm 0.4119$	$64.8\% \pm 0.4172$

Table S5. Gel fraction data of 7.5-7.5-BVIM.

Mechanical Characterization Data Summary.

The table below summarizes modulus, stress, and strain values obtained for the ionogels.

Materials	10-BVIM	15-BVIM	7.5-7.5-BVIM	20-BVIM
Nominal Stress at Break, σ_b (kPa)	1090.51 ± 122.51	468.02 ± 43.31	485.79 ± 24.34	253.51 ± 3.18
Nominal Strain at Break, ε_{tb} (%)	1184.91 ± 121.47	860.59 ± 21.12	897.96 ± 30.20	693.52 ± 33.24
Young's Modulus, E _t (kPa)	739.30 ± 217.89	119.09 ± 18.68	235.67 ± 25.31	67.32 ± 16.17

Table S6. Summary of Mechanical Characterization Data.

Thermogravimetric Analysis.

Thermogravimetric analysis was conducted on a TA Q5000 thermogravimetric analyzer. Samples were examined over a temperature range of 30-500 °C at a rate of 10 °C/ min under N₂ atmosphere. The first derivative (blue) shows the decomposition temperatures of the sample, while the weight loss thermogram (red) demonstrates the amount of mass lost from the sample. Material decomposition began at 300 °C (10% mass lost), followed by complete decomposition at 417 °C, exhibiting good thermal stability. The initial mass loss is likely due to unpolymerized ionic liquid remaining in the gels.



Figure S14. Thermogram of 7.5-7.5-BVIM. In red is the mass loss (%) of sample during heating. In blue is the derivative of mass loss (%/ °C) of samples during heating.

Source Measuring Unit Conductivity Experiments.

Conductivity measurements were performed with a Keithley 2400 source measure unit (SMU). Electrodes were attached to the sensors by wrapping wires around the object or placing copper plating in contact with the sample and wiring. Results from the compression of the cubic sensor are displayed in Figure S15.





Figure S15. SMU graph demonstrating change in conductivity of cubic sensor in response to compression. Also shown is the connection of the sensors to the SMU.

Cyclic Uniaxial Tensile Tests.

Cyclic tensile experiments were conducted on an Instron 5585H load frame with a 50 N load cell. Constant strain of 50% was applied at strain rates ranging from 5 to 100 mm/ min. Additionally, a constant strain rate of 50 mm/ min was applied over strains ranging from 5% to 200%.



Figure S16. Cyclic tensile stress-strain fatigue test a) 2 cycles b) 10 cycles. Also shown are cyclic tensile stress-strain curves at different strain rates c) 2 cycles d) 10 cycles.



Figure S17. Cyclic tensile stress-strain fatigue test at rate of 5 mm/ min at 50% strain.



Figure S18. Cyclic tensile stress-strain fatigue test at rate of 12.5 mm/ min at 50% strain.



Figure S19. Cyclic tensile stress-strain fatigue test at rate of 25 mm/ min at 50% strain.



Figure S20. Cyclic tensile stress-strain fatigue test at rate of 50 mm/ min at 50% strain.



Figure S21. Cyclic tensile stress-strain fatigue test at rate of 100 mm/ min at 50% strain.



Figure S22. Cyclic tensile stress-strain fatigue test at 5% strain at a rate of 50 mm/ min.



Figure S23. Cyclic tensile stress-strain fatigue test at 25% strain at a rate of 50 mm/ min.



Figure S24. Cyclic tensile stress-strain fatigue test at 50% strain at a rate of 50 mm/ min.



Figure S25. Cyclic tensile stress-strain fatigue test at 100% strain at a rate of 50 mm/ min.



Figure S26. Cyclic tensile stress-strain fatigue test at 200% strain at a rate of 50 mm/ min.

Gauge Factor Determination.

The gauge factor of a material is the ratio of change in electrical resistance to applied mechanical $\Delta R/R$

strain (GF = ε). Gauge factor measurements for the ion gel sensors were conducted on a 5585H load frame with a 50 N load cell while connected to a Keithley 2400 SMU. Samples were subjected to tensile strain ranging from 5-200% at a strain rate of 50 mm/ min. The change in resistance was plotted against % strain to determine the gauge factor.



Figure S27. Top Left & Bottom Left: Change in resistance at different strain amounts, ranging from 5-50% strain and 50-200% strain. Top Right & Bottom Right: Change in resistance vs % strain. The slope of the line is the gauge factor.

Lap shear test or Adhesion Tests.

Adhesion tests were performed on the TestResources Universal Test System with a load cell of 1 kN and a speed of 1.5 mm/min, following the ASTM D1002 standard.

These tests were performed on different substrates such as aluminum, glass, Teflon and

aluminum, glass, teflon and cardboard, all of them with the following dimensions: $2.5 \times 10 \times 0.16$ cm (width x length x thickness). Before adhering to the material, all substrates except the cardboard were cleaned with isopropanol.

To prepare the lap joint, $2.5 \ge 2.5 \ge 0.1 \text{ cm}$ (width x length x thickness) squares were printed and adhered to the substrate by applying manual pressure for 3 min. Then, the setup was attached to the grips of load frame and pulled until the sample was detached.

Finally, the adhesive strength was determined by the peak load divided by the overlap area.



Figure S28. Stress vs strain graph demonstrating adhesion of ion gel sensors to various substrates.

Substrate	Stress at Break (MPa)	Strain at Break (%)
Glass	5.45 ± 0.14	10.9 ± 0.56
Reattached Glass	5.37 ± 0.8	10.34 ± 0.21
Aluminum	4.42 ± 0.6	5.62 ± 0.32
Cardboard	2.02 ± 0.35	3.65 ± 1.05
Teflon	3.04 ± 0.42	5.36 ± 0.98

 Table S7. Summary of adhesion data.



Figure S29. Images demonstrating sensor adhesion to glass, aluminum, cardboard, and Teflon.



Figure S30. Image demonstrating adhesive patch (2.5 cm x 2.5 cm) supporting a 500g weight.



Figure S31. 15-BVIM gels, which only contains dynamic crosslinks, experienced a loss of shape fidelity during heating at 60 °C. Heating the dynamic networks above their T_g and under fast Diels-Alder exchange caused the material to reflow under the forces of gravity