Supplementary Information (SI) for Journal of Materials Chemistry A.

Supporting Information For

Hydrophobic, Ionically Conductive, Self-adhesive and Fully Recyclable Eutectogels for Stretchable Wearable Sensors and Triboelectric Nanogenerators

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Table S1. Detailed component formulations for the preparation of eutectogels

Sample	TA (g)	EGPEA (g)	1-Nap (g)	Cou (g)	MCC (g)	PIL (g)	Compared to pure SEG
SEG	6.18	1.92	1.08	0.37	0	0	Pure SEG exhibits weak mechanical properties and electrical insulation
SEG-M0.1	6.18	1.92	1.08	0.37	0.01	0	Few significant enhancements and changes. Electrical insulation
SEG-M0.4	6.18	1.92	1.08	0.37	0.04	0	
SEG-M0.7	6.18	1.92	1.08	0.37	0.07	0	
SEG-M1.0	6.18	1.92	1.08	0.37	0.10	0	Difficult to disperse in mixed solutions
SEG-PIL10	6.18	1.92	1.08	0.37	0	0.96	Subtle enhancements and changes. Ionically conductive.
SEG-PIL20	6.18	1.92	1.08	0.37	0	1.92	
SEG-PIL30	6.18	1.92	1.08	0.37	0	2.88	Excess PIL monomer leads to incomplete polymerization and liquid leakage during storage of eutectogel
SEG-PIL20M0.1	6.18	1.92	1.08	0.37	0.01	1.92	Substantial increase in mechanical strength while obtaining ionic conductivity
SEG-PIL20M0.4	6.18	1.92	1.08	0.37	0.04	1.92	
SEG-PIL20M0.7	6.18	1.92	1.08	0.37	0.07	1.92	

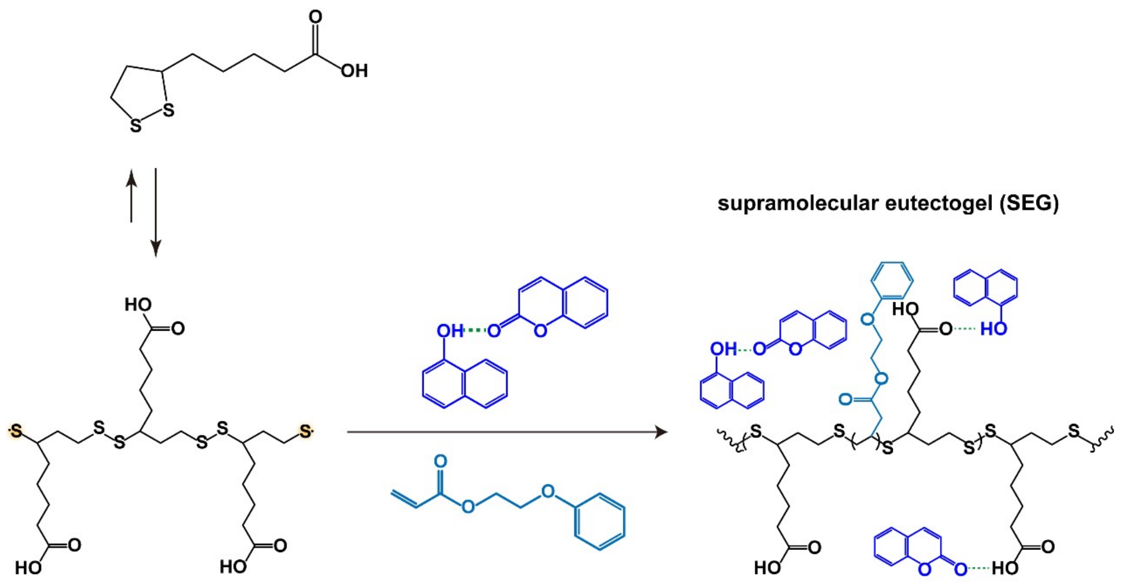

Figure S1. Schematic illustration of the formation of SEG.

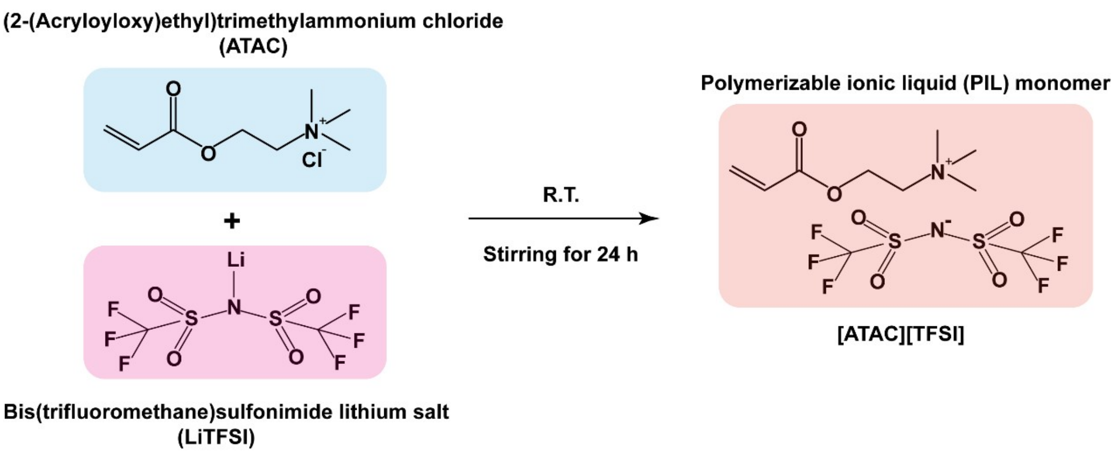

Figure S2. Synthesis of polymerizable [ATAC][TFSI] ionic liquid monomer.

Results

			Size (d.nm):	% Volume:	St Dev (d.nm):
Z-Average (d.nm):	2456	Peak 1:	173.2	100.0	24.71
PdI:	0.863	Peak 2:	0.000	0.0	0.000
Intercept:	0.576	Peak 3:	0.000	0.0	0.000
Result quality	Refer to quality report				

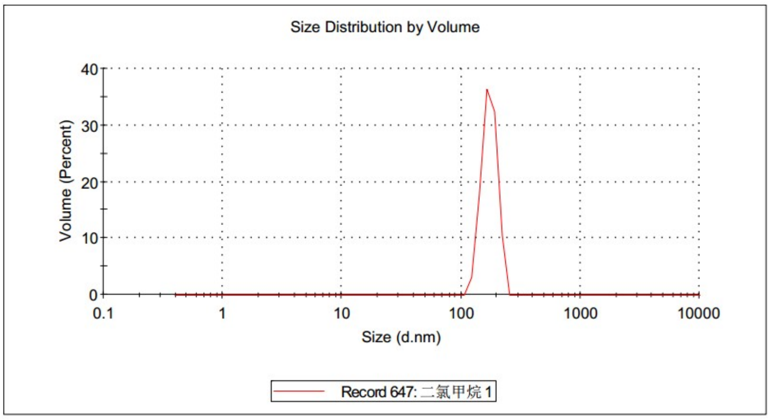

Figure S3. The particle size parameters of the used MCC

(a) (b)

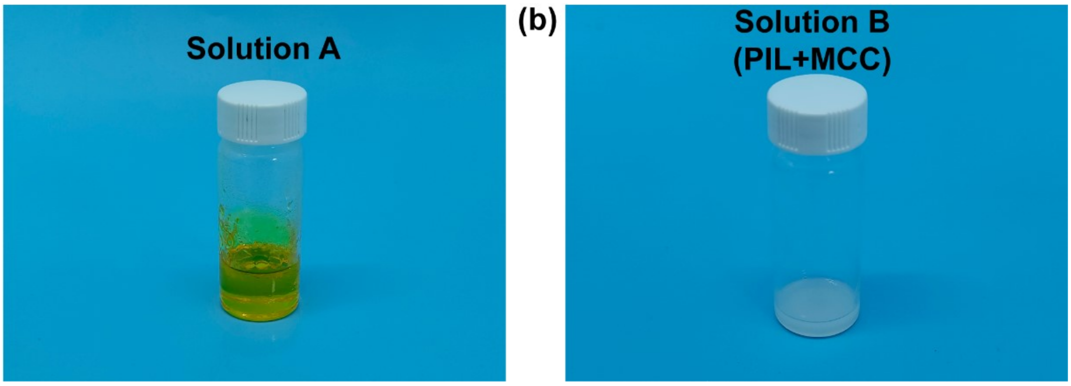

Figure S4. Optical photographs of solution A and solution B

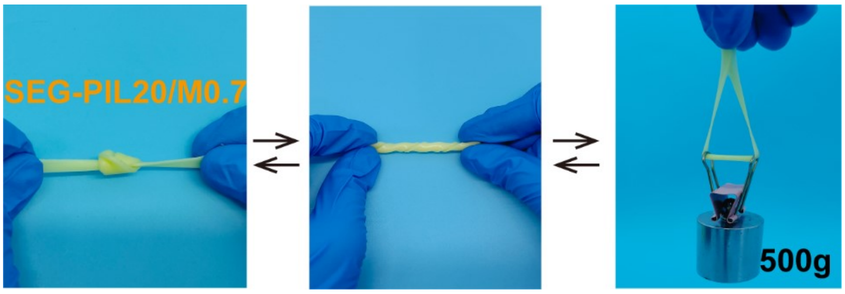

Figure S5. SEG-PIL20/M0.7 has significantly improved mechanical properties and can be easily knotted, twisted and lifted to a load of ~500g.

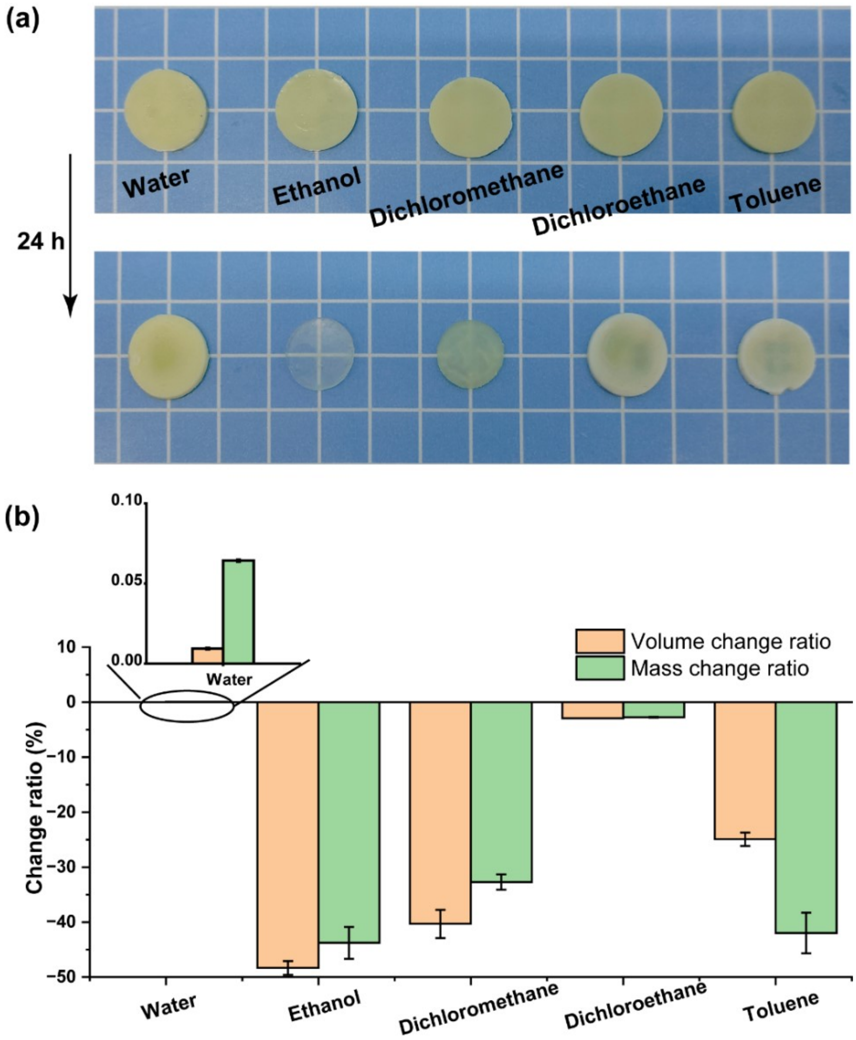

Figure S6. (a) Optical photographs of SEG-PIL20/M0.7 sample after 24 hours immersion in different solvents and (b) changes in mass and volume.

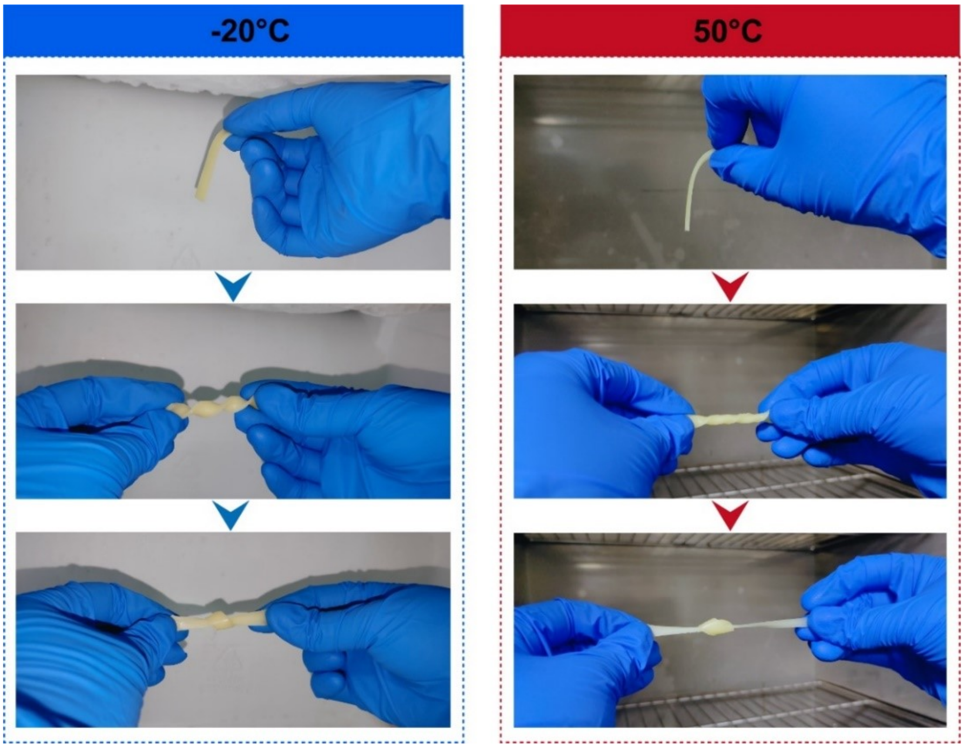

Figure S7. Optical photographs of SEG-PIL20/M0.7 sample at low (-20°C) and high (50°C) temperatures.

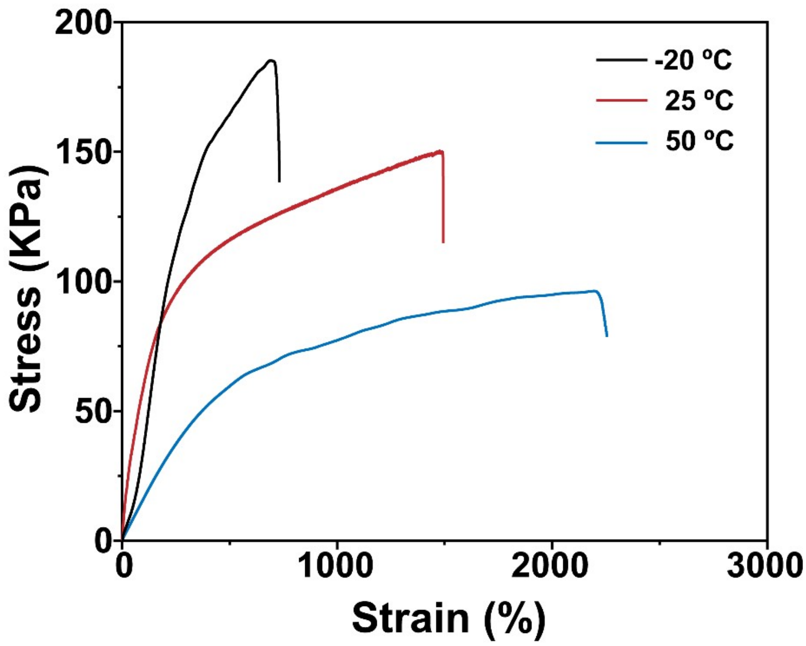

Figure S8. Stress-strain curves of SEG-PIL20M0.7 at different temperatures

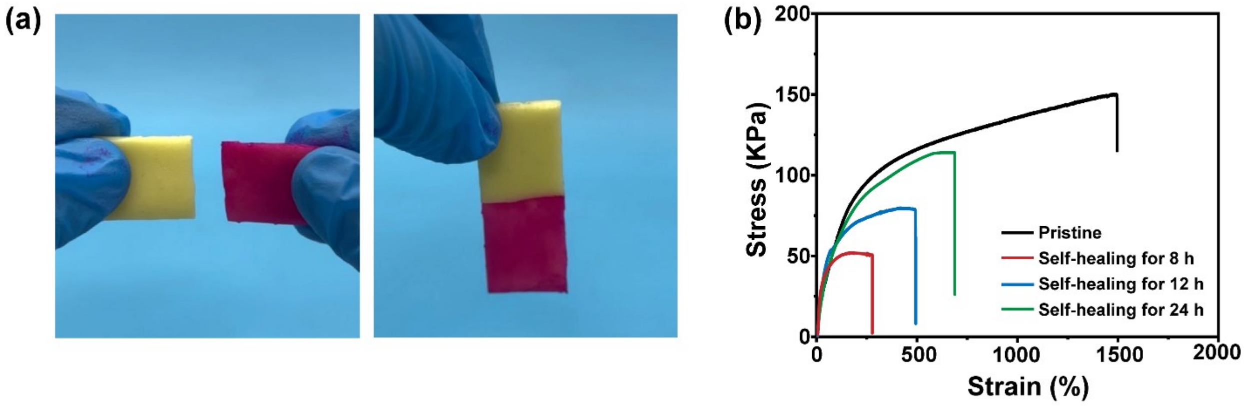

Figure S9. (a) The two cut pieces of SEG-PIL20M0.7 could be rejoined, demonstrating good self-healing capability. One piece was stained with Rhodamine B to better distinguish the healing interface. (b) Stress-strain curves of SEG-PIL20M0.7 after healing for different times.

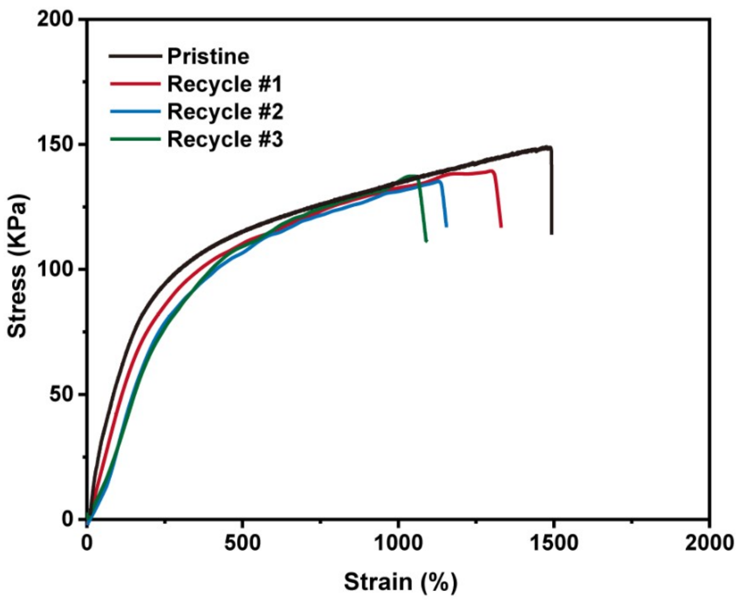

Figure S10. Stress-strain curve of SEG-PIL20/M0.7 sample after hot melt recovery.

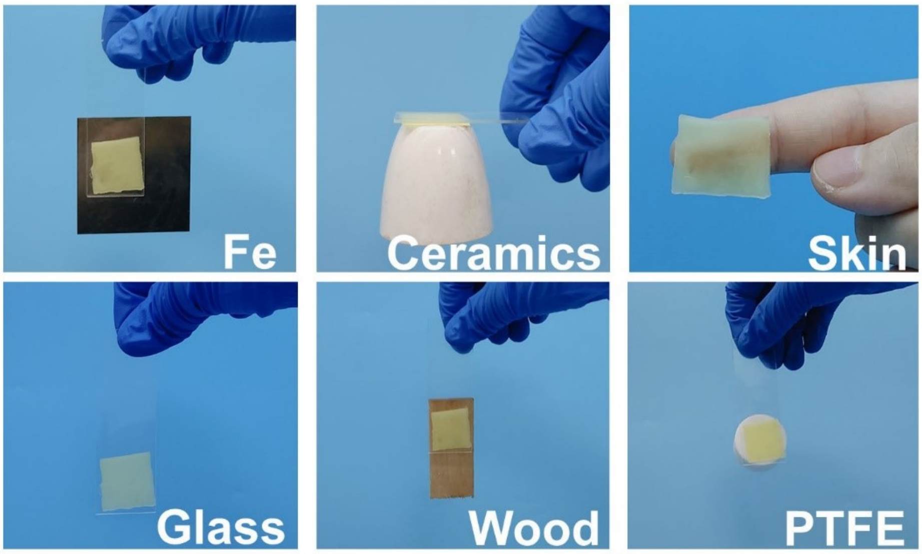

Figure S11. Optical photographs of SEG-PIL20/M0.7 adhered to different substrates in the air.

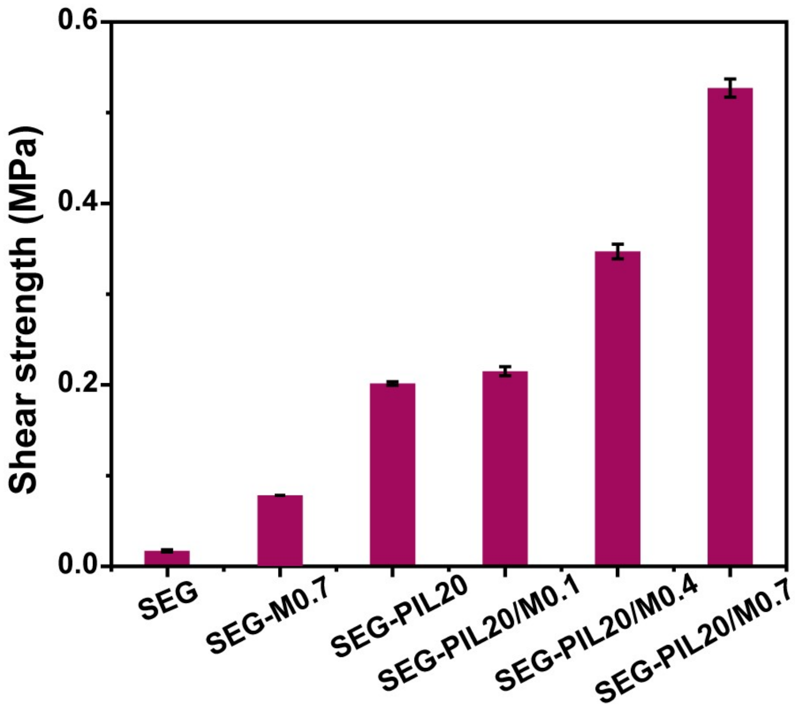

Figure S12. Adhesion strength of different eutectogels on Fe.

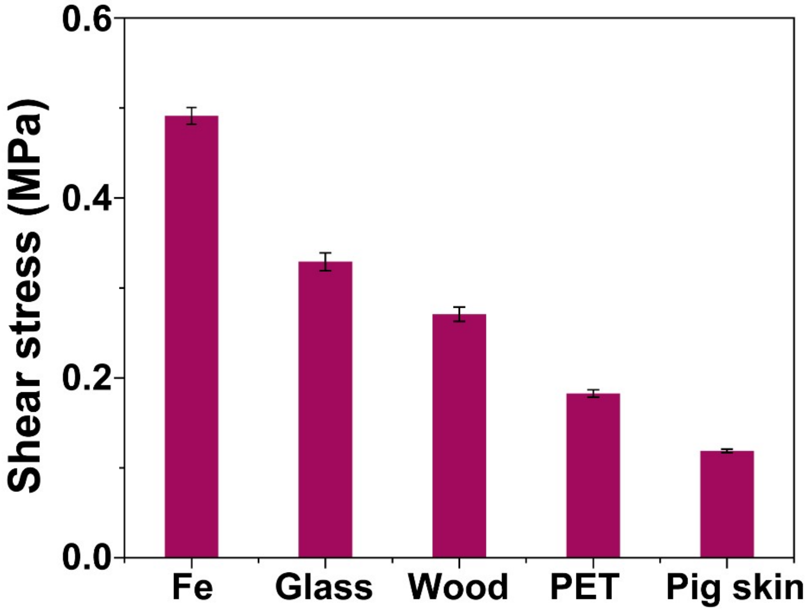

Figure S13. Adhesion strength of SEG-PIL20M0.7 after 24 h of underwater immersion

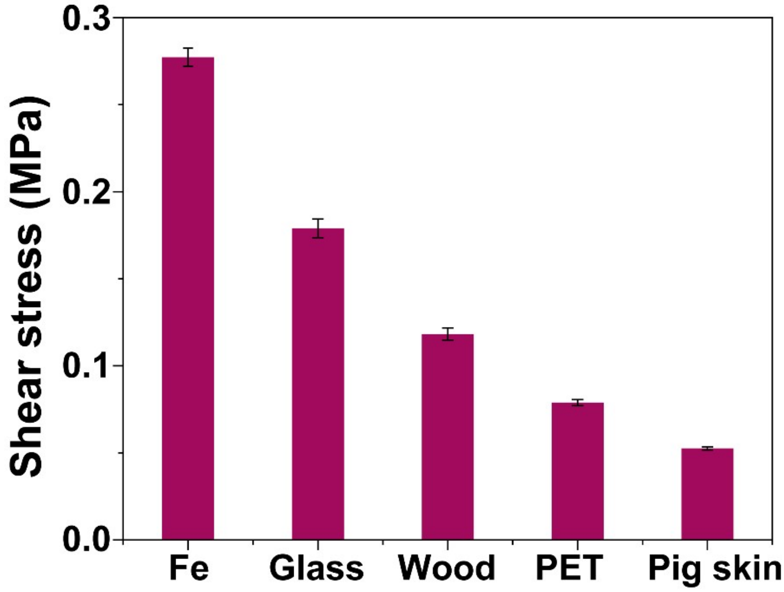

Figure S14. Adhesion strength of recycled SEG-PIL20M0.7

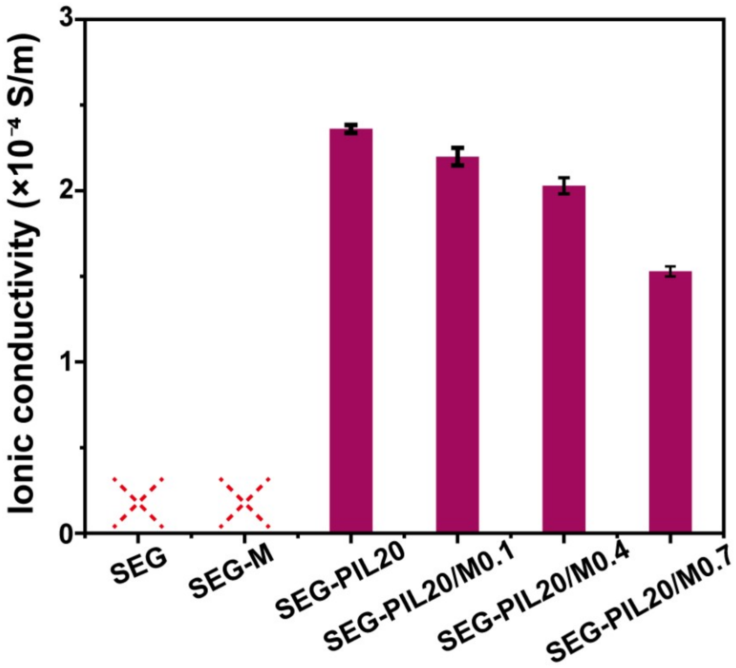

Figure S15. Ionic conductivity of different samples. Note: SEG and SEG-M are electrically insulating.

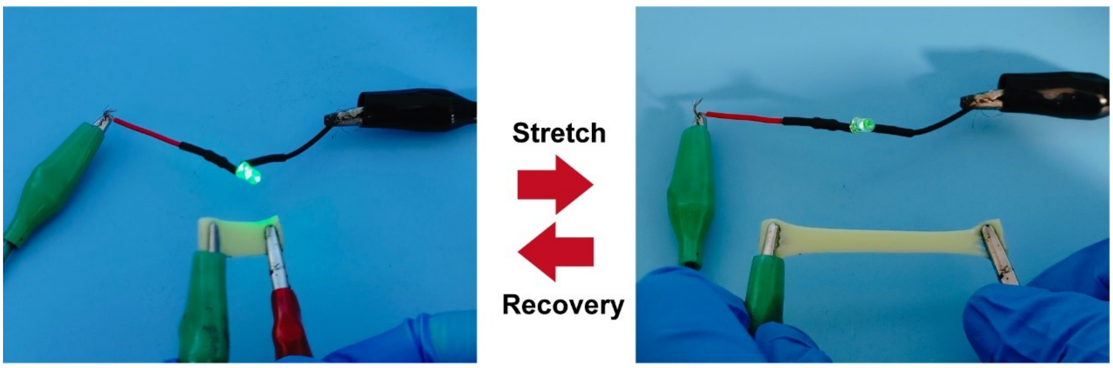

Figure S16. By connecting the SEG-PIL20/M0.7 sample in series with a small LED bulb in a circuit, it can keep the bulb lit during deformation.

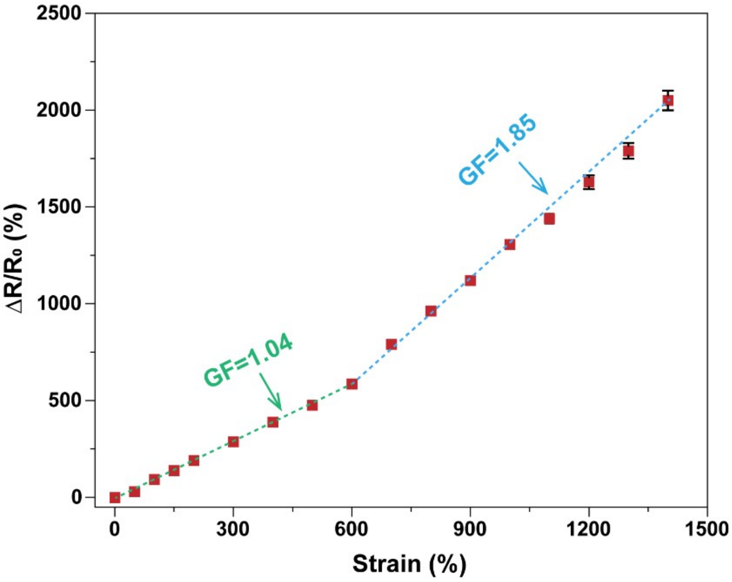

Figure S17. The dependence of relative resistance changes of SEG-PIL20M0.7 on the strain

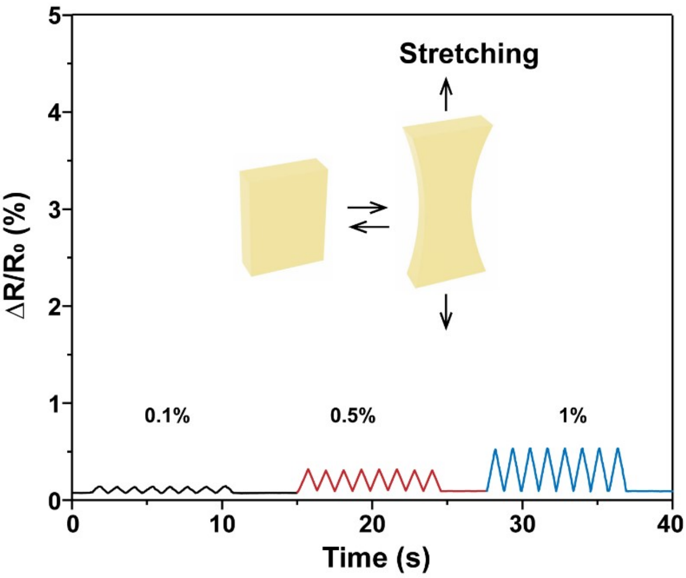

Figure S18. Plots of resistance change of the SEG-PIL20M0.7 as a function of time for the applied strain in the range of 0.1%–1%

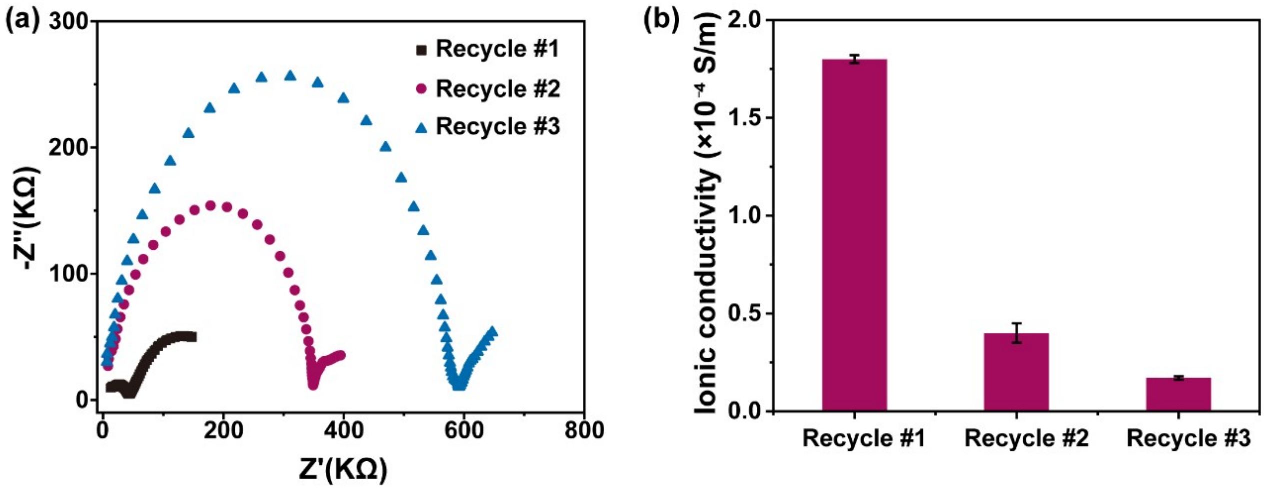

Figure S19. (a-b) Electrochemical impedance spectra of recovered eutectogels with their corresponding calculated ionic conductivities.

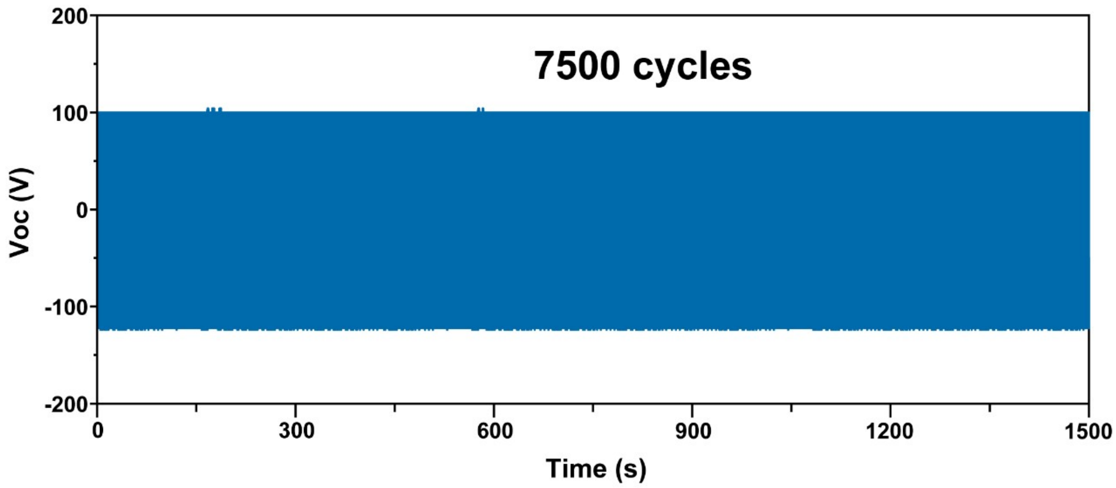

Figure S20. Variation of SEG-PIL20/M0.7 TENG output voltage after 7500 continuous cycles.

Table S2 Comparison of our prepared SEG-PIL/M with existing reported eutectogels

Entry	Hydrophilic or hydrophobic	Self-adhesion underwater	Dependence on chemical initiators	Recyclability	Ionic conductivity	Strain-sensitivity (GF)	Ref.
Zn^{2+}/PAA/Cellulose	Hydrophilic	N/A	Yes	No	0.721 mS/cm	N/A	1
ChCl/Gly/IA/AESO	Hydrophilic	N/A	Yes	No	2.5 to 3.8 × 10^{-5} S/cm	N/A	2
(PAA-ChCl)/PVA/DCNC	Hydrophilic	N/A	Yes	No	1.14 × 10^{-4} S/m to 4 × 10^{-5} S/m	1.43	3
PVA/ChCl/EG	Hydrophilic	No	No	N/A	0.28 S/m	1.2	4
GMA/LA/ChCl/Lignin	Hydrophilic	No	No	No	4.92 mS/cm	N/A	5
PVA/ChCl/PA	Hydrophilic	No	No	N/A	0.0656 S/m	1.593	6
$ZnCl_2$/EG/PAA/HPC	Hydrophilic	No	Yes	No	36.6 mS m^{-1}	N/A	7
Betaine-EG/PHEAA–gelatin–MXene	Hydrophilic	No	Yes	No	0.56 mS m^{-1}	2.7 (0-220%), 5.1 (220−580%), and 13.7 (580−740%)	8
PDES/CMFs	Hydrophilic	No	Yes	No	0.09 S m^{-1},	1.46 (0-300%), 2.59 (300-800%) and 3.71 (800-1300%)	9
Poly(IBA-co-EGPEA)/TEBAC/Thy	Hydrophobic	Yes	Yes	Yes	5.28 × 10^{-3} S/m	N/A	10
SEG-PIL/M	Hydrophobic	Yes	No	Yes	2.48×10^{-4} S/m	1.04 (0-600%), 1.85 (600-1400%)	This work

References

1. J. Zhu, C. Shao, S. Hao, K. Xue, J. Zhang, Z. Sun, L.-P. Xiao, W. Ren, J. Yang, B. Cao and R. Sun, *Chem. Eng. J.*, 2025, **506**, 159636.
2. S. Locatelli, G. C. Luque, R. Ruiz-Mateos Serrano, A. Dominguez-Alfaro, G. Reniero, M. L. Picchio, J. Leiva, L. M. Gugliotta, G. G. Malliaras, D. Mecerreyes, L. I. Ronco

and R. J. Minari, *ACS Appl. Polym. Mater.*, 2025, DOI: 10.1021/acsapm.4c03592.

3. A. Liu, X. Li, W. Xu, X. Duan, J. Shi, X. Li, J. Chu and H. Lei, *Int. J. Biol. Macromol.*, 2025, **284**, 138188.
4. T. H. Vo, P. K. Lam, R.-m. Chuang, F.-K. Shieh, Y.-J. Sheng and H.-K. Tsao, *Chem. Eng. J.*, 2024, **493**, 152877.
5. S. Sun, S. Hao, Y. Liu, S. Sun, Y. Xu, M. Jiang, C. Shao, J. Wen and R. Sun, *ACS Nano*, 2024, DOI: 10.1021/acsnano.4c12130.
6. Y. Shao, C. Dang, H. Qi, Z. Liu, H. Pei, T. Lu and W. Zhai, *Matter*, 2024, **7**, 4076-4098.
7. C. Lu, X. Wang, Y. Shen, S. Xu, C. Huang, C. Wang, H. Xie, J. Wang, Q. Yong and F. Chu, *Adv. Funct. Mater.*, 2024, **34**, 2311502.
8. B. Guo, M. Yao, S. Chen, Q. Yu, L. Liang, C. Yu, M. Liu, H. Hao, H. Zhang, F. Yao and J. Li, *Adv. Funct. Mater.*, 2024, **34**, 2315656.
9. X. Sun, Y. Zhu, J. Zhu, K. Le, P. Servati and F. Jiang, *Adv. Funct. Mater.*, 2022, **32**, 2202533.
10. M. Li, Z. Liu, Y. Hu, R. a. Li and Y. Cao, *Chem. Eng. J.*, 2023, **472**, 145177.