

Architectural engineering of asymmetric phase connectivity in ionogels for tough, transparent, and highly conductive strain sensors

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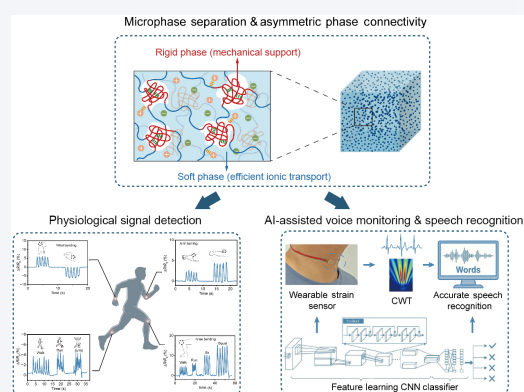
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ABSTRACT: Achieving mechanical robustness and efficient ionic transport simultaneously remains a major challenge for ionogels, as mechanical reinforcement often restricts ion mobility. Here we reported an architectural strategy that resolves this trade-off through composition-programmed asymmetric phase connectivity. By tuning the incompatibility among acrylic acid (AA), poly(ethylene glycol) methacrylate (PEGMA), and the ionic liquid 1-ethyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide ([EMIM][TFSI]), the polymer network spontaneously forms a microphase-separated structure in which a soft ion-conductive phase maintains network continuity, while a rigid reinforcing phase remains confined to dispersed microdomains. This topology enables efficient energy dissipation without disrupting ionic transport or optical transparency.

The resulting ionogels exhibit a balanced combination of high stretchability, enhanced toughness, high ionic conductivity, and optical clarity. In contrast, increasing connectivity of the rigid phase produces bicontinuous morphologies that lead to stiff and opaque gels. The optimized ionogel demonstrates excellent strain-sensing performance with high sensitivity, wide strain detection range, and outstanding durability under cyclic deformation. These results highlight phase connectivity as a key structural parameter governing ionogel performance and establish asymmetric phase architecture as a general design principle for multifunctional soft ionic conductors.

KEYWORDS: ionogel, microphase separation, ionic transport, human-machine interfaces, strain sensing



1 Introduction

Ionogels are emerging functional polymer materials that combine the mechanical support of polymer networks with the

electrochemical stability, non-volatility, and high ionic mobility of ionic liquids (ILs) [1–3]. Unlike conventional hydrogels or organogels, ionogels contain no volatile solvents, avoiding performance degradation caused by liquid evaporation. Compared to solid electrolytes, ionogels achieve higher ionic conductivity by incorporating mobile ionic liquids, offering excellent overall performance. Their stability and versatility make them well-suited for applications in transparent flexible sensors [4, 5], artificial skin [6–8], and energy storage systems [9–12]. Additionally, their mild preparation conditions and strong interfacial adhesion [13–16]

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enable potential use in wearable devices [17–21] and biomedical technologies [22–25]. Their favorable interfacial compatibility allows for seamless integration with biological tissues, further highlighting their critical role in emerging biomedical and biointegrated electronic systems [26–28].

To meet the growing demands of next-generation flexible ionotronics, recent research has yielded multi-functional ionogels exhibiting exceptional mechanical robustness, autonomous self-healing capabilities, and advanced macroscopic processability [29–32]. Despite these inspiring achievements, achieving an optimal balance between mechanical toughness and intrinsic ionic conductivity remains a fundamental challenge. This trade-off arises because high ionic conductivity typically requires a large concentration of ionic liquid, which inevitably plasticizes the polymer network and compromises its mechanical integrity. Conversely, strengthening the network through increased crosslinking or the introduction of rigid segments heavily restricts ion mobility, thereby severely reducing overall conductivity.

To mitigate this trade-off, a variety of material design strategies have been explored. One approach involves introducing lithium salts to increase charge carrier concentration while forming reversible physical crosslinks through coordination with functional groups, thereby improving strength and toughness [33–36]. However, lithium-based systems are highly sensitive to moisture and are limited by the relatively low mobility of lithium ions, which restricts their applicability in flexible electronics. Alternative strategies include the construction of double-network structures [37–39] or strengthening interactions between the ionic liquid and polymer matrix [40, 41]. Although effective in certain cases, these approaches often rely on complex multistep processing or dense network architectures that compromise ion transport by limiting chain mobility.

More recently, phase separation engineering has emerged as a promising strategy to address this challenge [42–45]. By exploiting differences in compatibility between polymer segments and ionic liquids, distinct rigid and soft domains can be generated, in which rigid regions provide mechanical reinforcement, while soft and ion-rich domains preserve efficient ionic conduction [46–48]. However, simply introducing phase separation does not necessarily lead to optimal performance. In many cases, excessive growth or connectivity of rigid domains produces stiff, brittle, or optically opaque materials, limiting their suitability for flexible electronic applications.

We reason that the connectivity and topology of the separated phases may play a more important role than phase separation itself. An ideal ionogel architecture should allow the ion-conductive soft phase to remain sufficiently continuous to support ionic transport and large deformation, while the mechanically reinforcing phase provides energy dissipation without forming a rigid load-bearing network that compromises flexibility or transparency. To corroborate this concept, we developed a copolymer ionogel system based on acrylic acid (AA), poly(ethylene glycol) methyl ether methacrylate (PEGMA), and the ionic liquid 1-ethyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide ([EMIM][TFSI]). In this system, AA exhibits limited compatibility with the ionic liquid and tends to associate into polymer-rich domains that enhance modulus and mechanical strength, whereas PEGMA shows strong affinity toward the ionic liquid, giving rise to ion-rich soft domains that facilitate ionic transport. Random copolymerization of AA and PEGMA integrates these components into a microphase-separated architecture, enabling mechanical

reinforcement and ionic conductivity to be optimized in parallel. Moreover, this architecture suppresses macroscopic phase separation while maintaining environmental stability, optical transparency, and processability, illustrating how compatibility-driven microphase separation can be leveraged for the rational design of high-performance ionogels.

2 Materials and methods

2.1 Materials

AA (99%, containing 180–200 ppm 4-methoxyphenol (MEHQ) as an inhibitor) and phenyl bis(2,4,6-trimethylbenzoyl)-phosphine oxide (PBPO, 97%) were purchased from Shanghai Aladdin Biochemical Technology Co., Ltd. PEGMA (average molecular weight of 475, containing 130 ppm MEHQ and 200 ppm butylated hydroxytoluene (BHT) as inhibitors) and [EMIM][TFSI] (98%) were obtained from Shanghai Bide Pharmaceutical Technology Co., Ltd. All reagents were used as received without further purification.

2.2 Preparation of ionogels

All ionogels were prepared via photoinitiated free radical polymerization. Briefly, predetermined amounts of AA, PEGMA, and ionic liquid [EMIM][TFSI] were mixed and stirred to obtain a homogeneous and transparent solution. PBPO initiator (0.1 wt.%) was then added, followed by ultrasonic treatment until complete dissolution. The solution was deoxygenated by bubbling with argon, and further sonicated for 15 min to eliminate residual oxygen. This deoxygenation process was repeated three times. The resulting precursor solution was transferred into a custom-made mold consisting of two fluorinated ethylene propylene (FEP) films separated by a silicone spacer to control membrane thickness. The samples were irradiated with ultraviolet (UV) light (wavelength of 365 nm and output power of ~ 20 W) for 1 h to induce gelation. The obtained ionogels were further heated at 80 °C in a vacuum oven for 12 h to remove unreacted monomers.

2.3 Mechanical characterization and numerical calculation of ionogels

All mechanical tests were performed at room temperature using an Instron-5944 universal testing system equipped with a 2 kN load cell. For tensile tests, the initial gauge length of the specimen was set to 4 mm, and unless otherwise specified, the extension rate was 10 mm·min⁻¹. During tensile loading-unloading cycles, both loading and unloading rates were set at 10 mm·min⁻¹ with no interval between consecutive cycles. For compression loading-unloading tests, specimens with a height of 1 mm were used, and both loading and unloading rates were set at 6 mm·min⁻¹, with no interval between cycles. The Young's modulus E of the ionogels was calculated by linear fitting of the initial 5% segment of the stress-strain curve, using Eq. (1)

$$E = \sigma / \varepsilon \quad (1)$$

where σ is the nominal stress and ε is the corresponding strain. The work of rupture E_r was determined as the area under the stress-strain curve up to the maximum strain according to Eq. (2)

$$E_r = \int_0^{\varepsilon_{\max}} \sigma(\varepsilon) d\varepsilon \quad (2)$$

where $\varepsilon_{\max}$ represents the strain at fracture.

2.4 Visible light transmittance measurement of ionogels

The optical transparency of the ionogel films (thickness of 1 mm) was evaluated using a UV–visible (UV–Vis) spectrophotometer (Lambda 750, Perkin Elmer) over a wavelength range of 400 to 800 nm with a sampling interval of 1 nm.

2.5 Fourier transform infrared (FTIR) spectroscopy of ionogels

FTIR spectra of the ionogels were obtained with a Nicolet NEXUS870 spectrometer in attenuated total reflectance (ATR) mode. Prior to measurement, samples were heated at 80 °C in a vacuum oven for 1 h and stored in a vacuum-dried environment to prevent moisture adsorption. Spectra were collected in the range of 400 to 4000 cm^{-1} with a resolution of 4 cm^{-1} .

2.6 Differential scanning calorimetry (DSC) analysis of ionogels

DSC measurements were performed on a 204F1 Phoenix NETZSCH differential scanning calorimeter under nitrogen atmosphere. Samples were scanned from –100 to 100 °C at a heating rate of 10 °C·min^{–1}.

2.7 Electrochemical impedance spectroscopy of ionogels

Electrochemical impedance measurements were conducted using a Novocontrol Concept 80 electrochemical workstation. The ionogel samples were sandwiched between two stainless steel electrodes to form a symmetric two-electrode cell. Impedance spectra were recorded over a frequency range of 0.01 Hz to 1 MHz with an alternating current (AC) perturbation amplitude of 10 mV, and all measurements were performed at open circuit potential after the system reached equilibrium. The bulk resistance R was obtained from the intercept of the high-frequency region of the Nyquist plot with the real axis. The ionic conductivity σ of the ionogels was calculated according to Eq. (3)

$$\sigma = \frac{L}{R \cdot A} \quad (3)$$

where L is the thickness of the sample, and A is the area of the electrodes.

2.8 Resistance testing of ionogels

The resistance of the ionogels was measured using a Keithley DMM6500 digital multimeter, employing a two-wire resistance measurement method. When testing the resistance variation with strain rate, the multimeter was integrated with the mechanical testing machine.

2.9 Process of machine learning-enabled speech recognition

The acquired raw resistance signals initially underwent a filtering pretreatment designed to effectively suppress environmental noise and signal drift. Subsequently, a continuous wavelet transform (CWT) technique based on the Morlet wavelet was employed to map the one-dimensional time-domain signals into two-dimensional (2D) time-frequency spectrograms, thereby facilitating the efficient extraction of time-frequency features correlated with speech dynamics. These CWT feature spectrograms were then input into an improved residual convolutional neural network (CNN) to perform classification tasks. The network architecture

comprised three integral components: an initial convolution block responsible for low-level feature encoding, followed by three cascaded residual blocks for hierarchical feature extraction, and finally, a fully connected layer linked via a channel concatenation module to execute the final classification, establishing the model's robust learning capability regarding subtle speech patterns [49].

3 Results and discussion

3.1 Design strategies for high strength and conductive ionogels

Rational and ingenious molecular design is essential to achieving targeted material functionalities. Fundamentally, AA undergoes negligible ionization within conventional ionic liquids due to their intrinsically low dielectric constants and weak solvation capabilities. Instead of dissociating, AA molecules predominantly associate via strong intermolecular hydrogen bonds, resulting in severe thermodynamic incompatibility and a strong tendency for phase separation within the ionic liquid matrix. Inspired by this phenomenon, a high-performance ionogel structure based on the principle of microphase separation was developed in this study to simultaneously enhance both mechanical properties and ionic conductivity. As illustrated, the formulated system is composed of PEGMA, AA, and the ionic liquid [EMIM][TFSI]. The core of this design strategy lies in utilizing the distinct incompatibility of AA to induce controlled microphase separation during polymerization. Furthermore, as the AA content increases, the system exhibits a morphological transition from a mono-continuous to a bicontinuous phase. This tailored evolution successfully constructs a phase-separated structure with distinct hard and soft domains within the ionogel, achieving synergistic structure–function integration (Fig. 1(a)).

The microstructural evolution of the polymer network fundamentally dictates its macroscopic electromechanical performance. As a foundational component, PEGMA possesses an amphiphilic nature and polyethylene glycol side chains that endow it with a strong affinity for the [EMIM][TFSI] matrix [50]. This characteristic promotes robust interactions between the ionic liquid and the polymer network, facilitating uniform dispersion. In the absence of AA, the resulting PEGMA-based network is isotropic, governed primarily by weak ion–dipole interactions. Consequently, the material exhibits a limited yield zone during stretching, resulting in inherently weak mechanical strength (Fig. 1(b)). Upon the introduction of AA, microphase separation is triggered, forming a mono-continuous phase architecture (Fig. 1(c)). While the aggregation of AA establishes a distinct hard phase that introduces a crucial energy dissipation mechanism, PEGMA continuously drives the formation of stable and soft ionic liquid-rich domains. Crucially, this specific phase-separated structure enables the poly(acrylic acid) chains within the hard domains to maintain a substantial “hidden length”, thereby preserving a high degree of intrinsic extensibility. During deformation, the effective synergistic interaction between these soft and hard phases significantly enlarges the yield zone, thereby dramatically enhancing the mechanical robustness. Notably, this hard-phase integration does not obstruct the continuous ion transport pathways within the PEGMA-stabilized soft domains, successfully achieving the decoupling of mechanical strength and ionic conductivity. However, as the AA concentration further increases, the morphology transitions into a

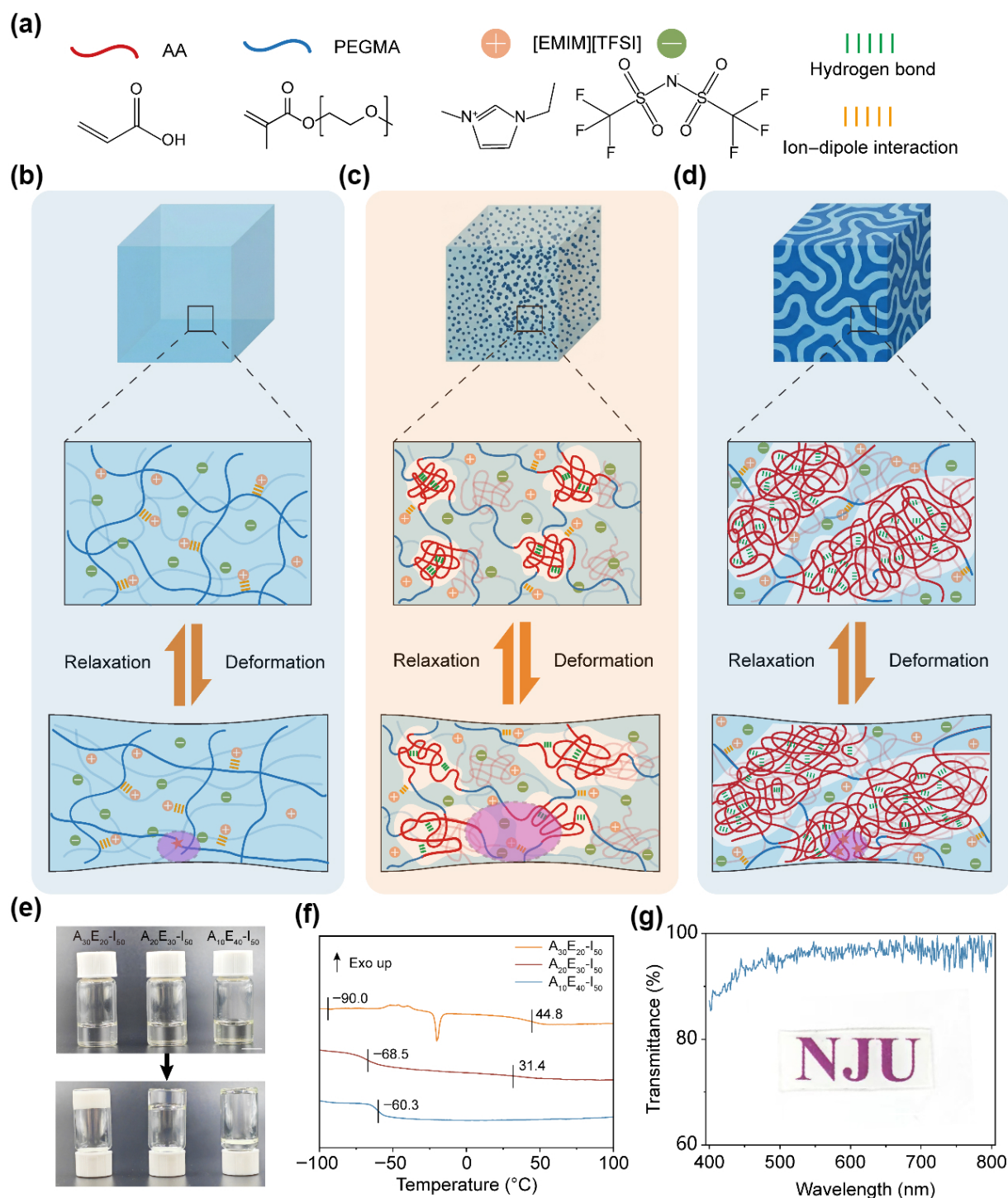


Figure 1 Schematic illustration of the microphase-separated iongel. (a) Chemical structures of the components. ((b)–(d)) Network evolution with increasing AA, showing isotropic-to-microphase separation transition and tensile response. (b) Isotropic network: limited energy dissipation and small yield zone (weak strength). (c) Single-continuous phase: synergistic energy dissipation expands the yield zone, decoupling strength from conductivity. (d) Bicontinuous phase: hard-phase dominance reduces synergy, contracting the yield zone and weakening the gel. Purple areas denote yield zones, and stars denote bond ruptures. (e) Ionogel before and after polymerization with different AA contents. The scale bar is 1 cm. (f) DSC curves of ionogels with varying AA content. (g) Visible light transmittance of the iongel over the wavenumber range of 400–800 nm. The inset shows a photograph of an “NJU” pattern clearly visible through the iongel layer.

bicontinuous phase (Fig. 1(d)). In this state, the hard phase becomes overly dominant, and excessive chain entanglement within these expansive hard domains drastically diminishes the available hidden length. This critical loss of extensibility effectively disrupts the previous phase synergy. This structural shift restricts overall

polymer chain mobility, causing the yield region to shrink during stretching, which ultimately leads to a decline in macroscopic mechanical strength.

Using only a single monomer in the preparation of ionogels inevitably results in significant shortcomings in certain

performance aspects. We found that before polymerization, AA and [EMIM][TFSI], PEGMA and [EMIM][TFSI], as well as their ternary mixture, all formed homogeneous and transparent precursor solutions. However, after polymerization, these samples showed pronounced macroscopic differences. The ionogel prepared solely from AA exhibited macroscopic white phase separation due to extensive hydrogen bonding and aggregation among AA chains (Fig. 1(e)). In contrast, the PEGMA-based ionogel, lacking sufficient hydrogen bonding or other strong interactions, displayed inadequate mechanical support and underwent plastic flow under gravity. In comparison, the ionogels formed by copolymerizing AA and PEGMA combined transparency with mechanical robustness, yielding the shape-stable ionogel with high optical transmittance. For clarity, the prepared ionogels were designated as $A_xE_yI_z$, where A, E, and I represent AA, PEGMA, and ionic liquid, respectively, and x , y , and z denote their corresponding mass fractions relative to the total mass, subject to the relationship $x + y + z = 100$. To verify that the internal phase separation was attributed to hydrogen bonding interactions, DSC was employed to characterize gels with distinct compositional ratios (Fig. 1(f)). In $A_{30}E_{20}I_{50}$, the high AA content led to the formation of larger polymer-rich domains, resulting in a distinct endothermic peak and two glass transition temperatures, assigned to the polymer phase (44.8 °C) and the solvent phase (−90 °C), respectively. For $A_{20}E_{30}I_{50}$, the reduced polymer-rich phase and increased polymer content in the solvent

phase shifted the corresponding glass transition temperature lower and higher, respectively. In $A_{10}E_{40}I_{50}$, phase separation was insignificant, leading to only a single observed glass transition. Additionally, under appropriate composition conditions, the ionogels exhibited excellent visible light transmittance, which could be maintained above 90% (Fig. 1(g)).

3.2 Mechanism study of phase-separated structure in ionogels

To investigate the compositional dependence of phase separation behavior, we initiated our study with the PEGMA/AA gel network system. A controlled ionic liquid incorporation threshold (50 wt.% [EMIM][TFSI]) was strategically selected to enable systematic observation of phase separation initiation while maintaining structural integrity (Fig. 2(a)). A pronounced correlation between AA content and phase separation intensity was observed, with incremental AA incorporation progressively amplifying phase separation. Crucially, a microphase-separated regime was identified at a critical mass ratio of AA to PEGMA of 2:3, representing a metastable transition state between homogeneous and macrophase-separated domains. This compositionally tuned pseudo-equilibrium condition creates an ideal platform for investigating [EMIM][TFSI] ionic liquid-mediated phase separation dynamics while maintaining interfacial stability essential for characterization. These results

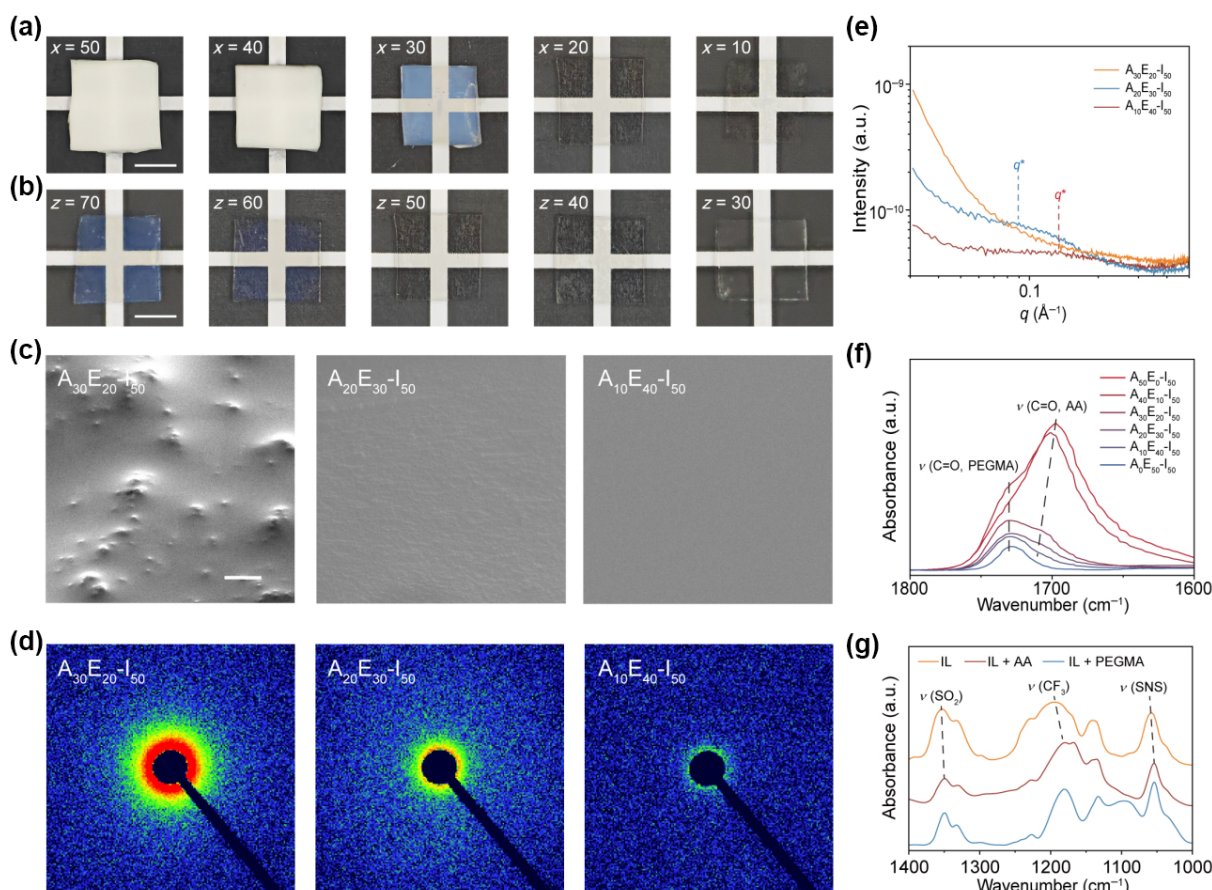


Figure 2 Morphological and microstructural characterization of ionogels with different compositions. (a) Photographs showing the morphological changes of ionogel with a fixed [EMIM][TFSI] content of 50 wt.% and varying AA content. The scale bar is 1 cm. (b) Photographs showing the morphological changes of ionogels with a fixed AA to PEGMA mass ratio of 2:3 and varying [EMIM][TFSI] content. (c) SEM images. The scale bar is 10 μ m. (d) 2D SAXS patterns. (e) Scattering intensity profiles of $A_{30}E_{20}I_{50}$, $A_{20}E_{30}I_{50}$ and $A_{10}E_{40}I_{50}$ ionogels. (f) FTIR spectra showing the effect of [EMIM][TFSI] on the hydrogen bonding between AA molecules. (g) FTIR spectra showing the effect of AA and PEGMA on [EMIM][TFSI].

demonstrate that the phase separation in ionogels originates from the incompatibility between PAA and [EMIM][TFSI]. Based on this analysis, we systematically probed the concentration-dependent phase separation behavior through controlled modulation of ionic liquid content from 30% to 70% (Fig. 2(b)). This parametric investigation revealed threshold ionic liquid loadings that govern the transition between microphase separation and macroscopic phase segregation, providing mechanistic insights into plasticization effects on polymer-ion matrix compatibility.

In order to characterize macroscopic phase separation characteristics, scanning electron microscopy (SEM) was employed to analyze morphological evolution across ionogel formulations with varying acrylic acid content. As shown in Fig. 2(c), A₁₀E₄₀-I₅₀ composition exhibited a homogeneous and smooth morphology, consistent with its absence of phase separation. With the increase of acrylic acid concentration, the A₂₀E₃₀-I₅₀ system developed a slightly corrugated surface texture, indicative of microphase separation initiation at the critical concentration threshold. Further elevation of acrylic acid content in A₃₀E₂₀-I₅₀ resulted in distinct light-scattering domains, corresponding to well-defined macroscopic phase separation (Fig. S1 in the Electronic Supplementary Material (ESM)).

Due to the ultra-soft and highly adhesive nature of the liquid-rich ionogel, which inevitably induces severe probe contamination and scanning artifacts in direct surface visualization like atomic force microscopy (AFM), small-angle X-ray scattering (SAXS) was employed as a robust bulk alternative to reliably evaluate the nanoscale compositional heterogeneity. The two-dimensional SAXS patterns show isotropic rings, indicating the absence of long-range structural orientation (Fig. 2(d)). The corresponding one-dimensional profiles exhibit a broad scattering peak for A₂₀E₃₀-I₅₀ (Fig. 2(e) and Fig. S2 in the ESM), with a characteristic position at $q^* \approx 0.090 \text{ \AA}^{-1}$ ($q = 0.040\text{--}0.35 \text{ \AA}^{-1}$), corresponding to nanoscale density fluctuations with a characteristic length scale of 2–15 nm ($d = 2\pi/q$). For A₁₀E₄₀-I₅₀, the scattering peak shifts to higher q values ($q^* \approx 0.16 \text{ \AA}^{-1}$), indicating smaller characteristic length scales (1–10 nm) and suppressed nanoscale heterogeneity at lower AA content. In contrast, no distinct SAXS peak is observed for A₃₀E₂₀-I₅₀, consistent with phase-separated domains exceeding the detectable length scale of SAXS and transitioning toward mesoscale or macroscopic segregation. Consequently, this structural evolution reveals that the microphase separation is primarily driven by the localized aggregation of short and distinct segments, such as the strongly hydrogen-bonded AA sequences, rather than the macroscopic phase segregation of entire polymer chains.

To further investigate the microscopic interactions within the AE-I ionogels, infrared spectroscopy was performed on samples with different compositions. Infrared spectroscopy results elucidate the mechanistic interactions between AA and PEGMA under coexistence conditions, revealing synergistic hydrogen bonding that governs polymer-polymer association kinetics (Fig. 2(f)). The bands at 1698 and 1728 cm^{-1} are assigned to the C=O stretching vibrations of AA and PEGMA, respectively [51–53]. On the other hand, the peaks at 1352, 1194, and 1058 cm^{-1} correspond to the O=S=O, –CF₃, and S–N–S vibrations of [EMIM][TFSI], respectively (Fig. 2(g)), corroborated by the results of previous studies [54]. Upon introduction of a polymer network consisting of AA or PEGMA, these vibrational peaks all shift to lower wavenumbers, specifically to 1349, 1181, and 1054 cm^{-1} , indicating strong interactions between [EMIM][TFSI] and both AA and PEGMA.

This interaction contributes to the structural stability of the ionogel system. At fixed [EMIM][TFSI] concentration, modulation of the AA/PEGMA ratio exerts negligible influence on the C=O vibrational mode of PEGMA, while the corresponding AA carbonyl stretching exhibits a marked blue shift with increasing PEGMA content. This spectral evolution signifies progressive weakening of carboxyl-carboxyl interactions in AA domains.

To validate the theoretical feasibility of our rational microphase separation design, density functional theory (DFT) calculations combined with electrostatic potential (ESP) mapping were conducted on simplified molecular models (Figs. S3 and S4 in the ESM). These computational results provide molecular-level insight into the interaction asymmetry that intrinsically drives the phase separation. The calculated binding energies indicate that AA–AA self-association is energetically more favorable than AA–PEGMA or PEGMA–PEGMA interactions, suggesting an intrinsic tendency for AA segments to associate at the molecular level. In addition, ESP mapping illustrates pronounced charge complementarity between AA and the [TFSI] anion, supporting the possibility of favorable electrostatic interactions between carboxyl groups and ionic liquid components.

While small-molecule DFT and ESP analyses provide a qualitative thermodynamic rationale for the preferential AA–AA association, they inherently omit bulk collective effects. In the actual ionogel, intrinsic polymer architectural parameters ultimately dictate the microdomain size and spatial distribution. Specifically, the highly random copolymer sequence generated during free-radical polymerization restricts extensive AA aggregation to the nanoscale; conversely, a blockier sequence would thermodynamically drive the formation of macroscopic rigid domains. Additionally, the molecular weight of the PEGMA side chains directly influences domain spacing. Higher molecular weight side chains introduce greater steric hindrance and enhance localized swelling by the ionic liquid, thereby expanding the spatial distance between the isolated AA microdomains. Together, these architectural factors ensure the stable coexistence of dispersed rigid domains within a continuous and ion-conductive soft matrix.

3.3 Compositional and environmental dependencies of ionogel properties

To further optimize and explore the physicochemical properties of the ionogels, we systematically characterized the mechanical performance across various AA/PEGMA ratios via tensile testing, aiming to elucidate the governing effect of the polymer network architecture on the material's overall behavior. Characteristic stress-strain curves profiled in Fig. 3(a) reveal composition-driven mechanical behavior transitions. With the increase of AA content, the critical strength rised from 0.052 to 8.90 MPa, while Young's modulus at 5% strain rised from 0.029 to 55.04 MPa. Elongation at break follows a non-monotonic trend, peaking at 30% strain (Fig. 3(b)). The mechanical enhancements originate from AA's carboxyl groups forming hydrogen-bonding domains that amplify network rigidity and crosslinking density. Notably, the A₂₀E₃₀-I₅₀ formulation with optimized microphase separation demonstrated exceptional work of rupture up to 2.33 MJ·m^{−3} (Fig. 3(c)), attributable to its balanced architecture: Hard-phase domains provide physical crosslinks while soft-phase regions enable chain extensibility, maintaining network homogeneity for optimal strength-ductility synergy.

Systematic modulation of ionic liquid concentration

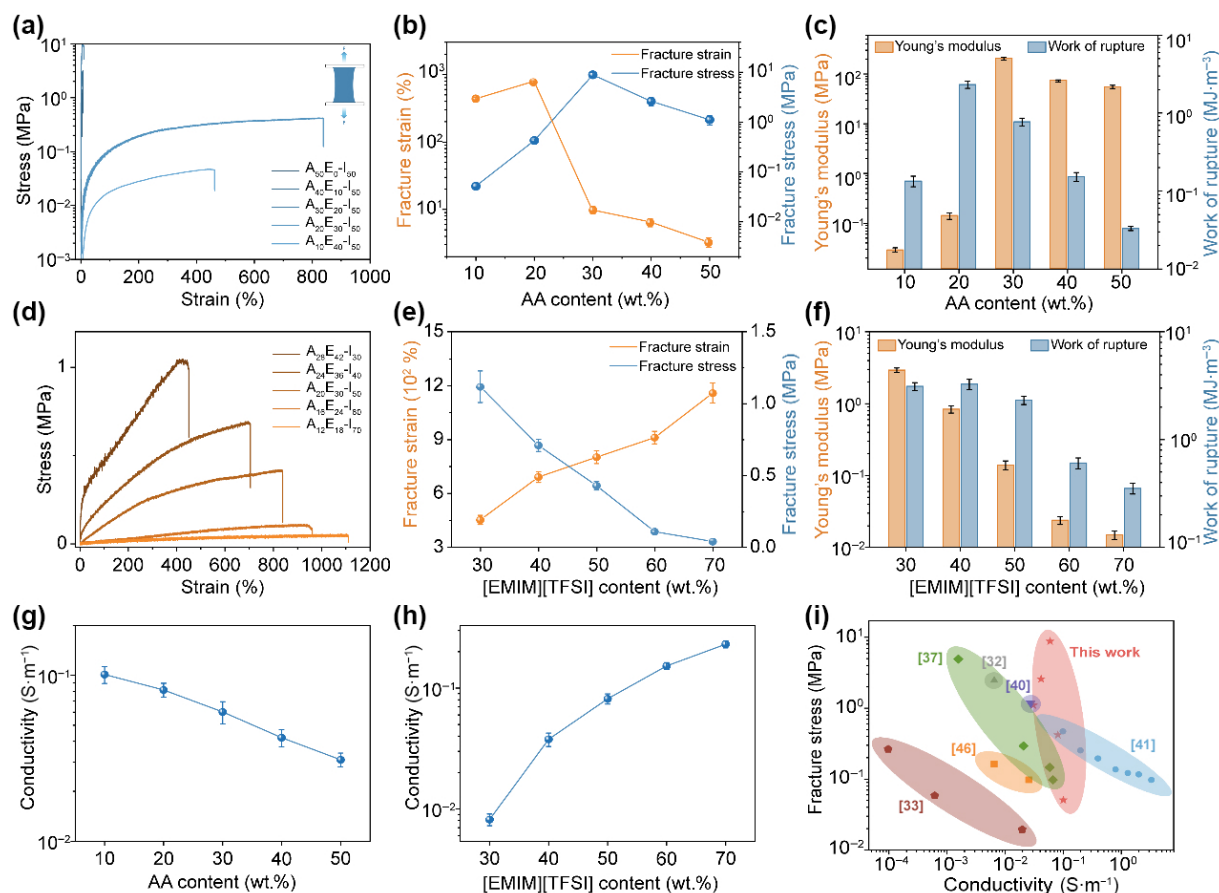


Figure 3 Mechanical properties and conductivity of ionogels with different compositions. For ionogels with a fixed [EMIM][TFSI] content of 50 wt.% and varying AA to PEGMA mass ratios: (a) stress–strain curves, (b) critical strain and stress, and (c) Young's modulus and work of rupture. For ionogels with a fixed AA to PEGMA mass ratio of 2:3 and varying [EMIM][TFSI] content: (d) stress–strain curves, (e) fracture length and ultimate stress, and (f) Young's modulus and work of rupture. (g) Ionic conductivity of ionogels with a fixed [EMIM][TFSI] content of 50 wt.% and different AA content. (h) Ionic conductivity of ionogels with a fixed AA to PEGMA mass ratio of 2:3 and varying [EMIM][TFSI] content. The sample size for all tests was $n = 3$. (i) Comparative benchmarking of fracture stress and ionic conductivity against state-of-the-art soft sensing materials. The red stars denote the current work, showcasing a significant decoupling of mechanical and electrical properties.

demonstrated an inverse relationship between [EMIM][TFSI] content and mechanical robustness. When maintaining the optimal AA:PEGMA ratio at 2:3, incremental elevation of ionic liquid from 30% to 70% induced a three-order-of-magnitude reduction in critical stress from 1.12 to 0.041 MPa, while the Young's modulus from 2.93 to 0.015 MPa at 5% strain (Fig. 3(d)). The compositional adjustment conversely enhanced material extensibility, achieving over 250% improvement in critical strain from initial 454% to 1160%, as quantitatively corroborated in corresponding mechanical datasets (Fig. 3(e)). The work of rupture exhibited concurrent attenuation from 3.12 to 0.35 MJ·m⁻³, reflecting compromised network integrity (Fig. 3(f)). Mechanistic analysis attributed these transitions to ionic liquid-induced suppression of chain entanglement through competitive solvation effects and dilution of physical crosslinks, preferentially enhancing deformability while sacrificing load-bearing capacity. Considering the combined impact of ionic liquid content on both mechanical and electrical properties, A₂₀E₃₀-I₅₀ was used for all subsequent measurements unless otherwise specified.

The operational reliability of flexible sensing systems fundamentally depends on the cyclic mechanical resilience and spontaneous recovery capability of constituent materials under dynamic loading conditions. Cyclic tensile testing revealed characteristic hysteresis loops in stress–strain profiles (Fig. S5(a)

in the ESM), with hysteresis area progression demonstrating strain-amplified energy dissipation. Quantitative analysis shows the A₂₀E₃₀-I₅₀ formulation achieves maximum tensile dissipation energy of 0.205 MJ·m⁻³ at 500% strain (Fig. S5(b) in the ESM), attributable to progressive dissociation of physical crosslinks within the polymer network under mechanical excitation. Parallel compression testing exhibited analogous energy dissipation behavior, recording 0.763 MJ·m⁻³ dissipation at 80% compressive strain while maintaining complete structural rebound capability without residual deformation (Figs. S5(c) and S5(d) in the ESM), confirming exceptional pressure resilience essential for repeated sensing operations.

Notably, the ionogel exhibits a characteristic Mullins effect during the initial loading cycles, where macroscopic deformation ruptures a portion of the sacrificial physical crosslinks (primarily the hydrogen bonds confined within the AA-rich rigid domains) to efficiently dissipate energy (Fig. S6 in the ESM). Crucially, the rapid and reversible re-association of these dynamic non-covalent interactions primarily governs the material's excellent anti-fatigue properties. Once the mechanical load is removed, the dissociated hydrogen bonds swiftly reform, preventing the accumulation of microscopic damage and enabling the polymer network to fully recover its initial mechanical state without significant residual deformation.

Superior interfacial adaptability stems from the ionogel's multi-modal adhesion mechanism. Synergistic interfacial interactions, including ion–dipole coupling, electrostatic attraction, and van der Waals forces, enable rapid conformal contact with diverse substrates, such as plastic, glass, and metal, while permitting controlled detachment through reversible bond reconfiguration (Fig. S7(a) in the ESM). Lap shear testing quantified interfacial adhesion strengths spanning 38 kPa for aluminum to 100 kPa for wood (Figs. S7(b) and S7(c) in the ESM). Energy dissipation during interfacial decohesion mechanisms effectively attenuated stress concentration at bonded interfaces. This dynamic adhesion equilibrium ensures reliable sensor–substrate integration without interfacial failure during cyclic deformation.

The electrochemical characteristics and stimuli-responsive behavior of the ionogels were systematically investigated to evaluate their efficacy as matrix materials for flexible sensing. By deliberately manipulating the monomeric stoichiometry and ionic phase fraction, we successfully engineered percolated “ionic highways” that facilitate efficient charge transport without compromising structural coherence or interfacial stability. As demonstrated by compositional analysis, the ionic conductivity exhibits a stoichiometry-governed evolution, increasing from 0.031 to 0.101 S·m^{−1} as the PEGMA content is raised from 0 wt.% to 40 wt.% (Fig. 3(g)). This 225% enhancement is attributed to microphase-separation-induced pathway optimization; specifically, beyond a 30% PEGMA loading threshold, the formation of side chain polyethylene oxide (PEO) microdomains establishes continuous charge transport corridors that activate an efficient ion-hopping conduction mechanism.

Furthermore, concentration studies identify a conductivity maximum of 0.231 S·m^{−1} at 70 wt.% [EMIM][TFSI] loading, where expanded free volume and elevated charge carrier density collectively enhance ionic dissociation efficiency (Fig. 3(h) and Fig. S7 in the ESM). However, since IL concentrations exceeding 50 wt.% induce a plasticization effect that reduces crosslinking density by 68% and precipitates mechanical degradation, a strategic formulation balance is necessitated to decouple high ionic functionality from structural robustness.

The balanced physicochemical properties enabled by the microphase-separated architecture indicate a promising potential for these ionogels in flexible sensing applications. A notable aspect of this design is the apparent decoupling of mechanical support from ionic transport. As suggested by the relationship between crosslinking density and mechanical reinforcement (Fig. 3(i) and Fig. S8 in the ESM), the polymer network effectively maintains structural integrity while preserving its electrochemical functionality. This synergy between robustness and conductivity provides a foundation for high-performance sensing, the compositional and environmental dependencies of which are discussed in the following section.

3.4 Environmental and sensing characteristics of AE-I ionogels

Building upon the structural design that effectively decouples mechanical support from ionic transport, the ionogels demonstrate a compelling synergy between robust physical integrity and responsive electrochemical behavior. While the microphase-separated architecture ensures structural stability, the ionic phase remains highly sensitive to thermal stimuli. Notably, the thermal activation of charge carriers not only drives a substantial increase in

conductivity but also aligns the material's optimal transport dynamics with the human physiological temperature range. This alignment suggests a high degree of compatibility for wearable applications, the specific compositional and environmental dependencies of which were investigated in the following section.

The charge transfer process was further amplified by thermal activation, which drives a 20-fold conductivity increase, from 0.011 S·m^{−1} at −10 °C to 0.230 S·m^{−1} at 50 °C, owing to accelerated ionic mobility and enhanced polymer chain relaxation dynamics (Fig. S9 in the ESM).

To comprehensively assess environmental adaptability, the ionogel's mass and conductivity were tracked over a seven-day period under multiple environmental stresses. Under extreme thermal environments (80 and −20 °C) and vacuum exposure, the sensor exhibited negligible variations, with a mass change of less than 0.5% and a conductivity fluctuation of more than 2%. Exposure to a higher ambient humidity of 60–70 %RH (Figs. 4(a)–4(c) and Fig. S9 in the ESM) induced a controlled hydration effect, resulting in a transient 3.6% mass gain within the first 24 h that subsequently stabilized with a 12% conductivity enhancement due to moisture plasticization. Importantly, further evaluation of the sensor's baseline stability under normal room temperature and typical indoor humidity (Fig. S10 in the ESM) revealed negligible signal interference. This robust overall environmental tolerance confirms the material's reliability for prolonged wearable device operation, providing a solid foundation for the subsequent systematic investigation of its strain-sensing behavior.

Sensitivity is a critical metric for evaluating sensing performance, typically quantified by the gauge factor (GF), which is defined as the slope of the relative resistance change $\Delta R/R_0$ versus applied strain ε

$$GF = (\Delta R/R_0)/\varepsilon \quad (4)$$

where ΔR is the change in resistance, R_0 is the initial resistance, and ε is the applied strain. Remarkably, the AE-I ionogel demonstrates an extraordinarily linear response ($R^2 = 0.997$) over a broad strain range of 0%–500%, maintaining a stable GF of 0.339 (Fig. 4(d)). The fundamental physical origin of this strict linearity lies in the synergistic decoupling within the microphase-separated architecture. Specifically, the soft and PEGMA-rich phase acts as continuous “ionic highways”. During macroscopic deformation, this liquid-rich soft phase remains highly continuous, uniformly accommodating the applied strain without experiencing topological disconnections or sudden disruptions in the ion-transport pathways. Concurrently, the isolated rigid AA-rich domains act as robust anchoring points that effectively dissipate localized stresses, preventing structural necking or chaotic yielding. This structural synergy ensures that the geometric elongation of the conductive channels scales strictly proportionally with the macroscopic stretching, thereby translating into the highly stable and strictly linear resistance–strain response.

Compared to sensors with narrow linear regions, this ultra-wide linear operational range endows the AE-I ionogel with superior sensing stability and predictability. It not only eliminates the need for complex non-linear calibration algorithms during signal processing but also ensures consistent electromechanical reliability across both subtle and severe deformations, making it an ideal candidate for diverse human–machine interface (HMI) applications.

Continuous tensile tests were performed on AE-I ionogel-based

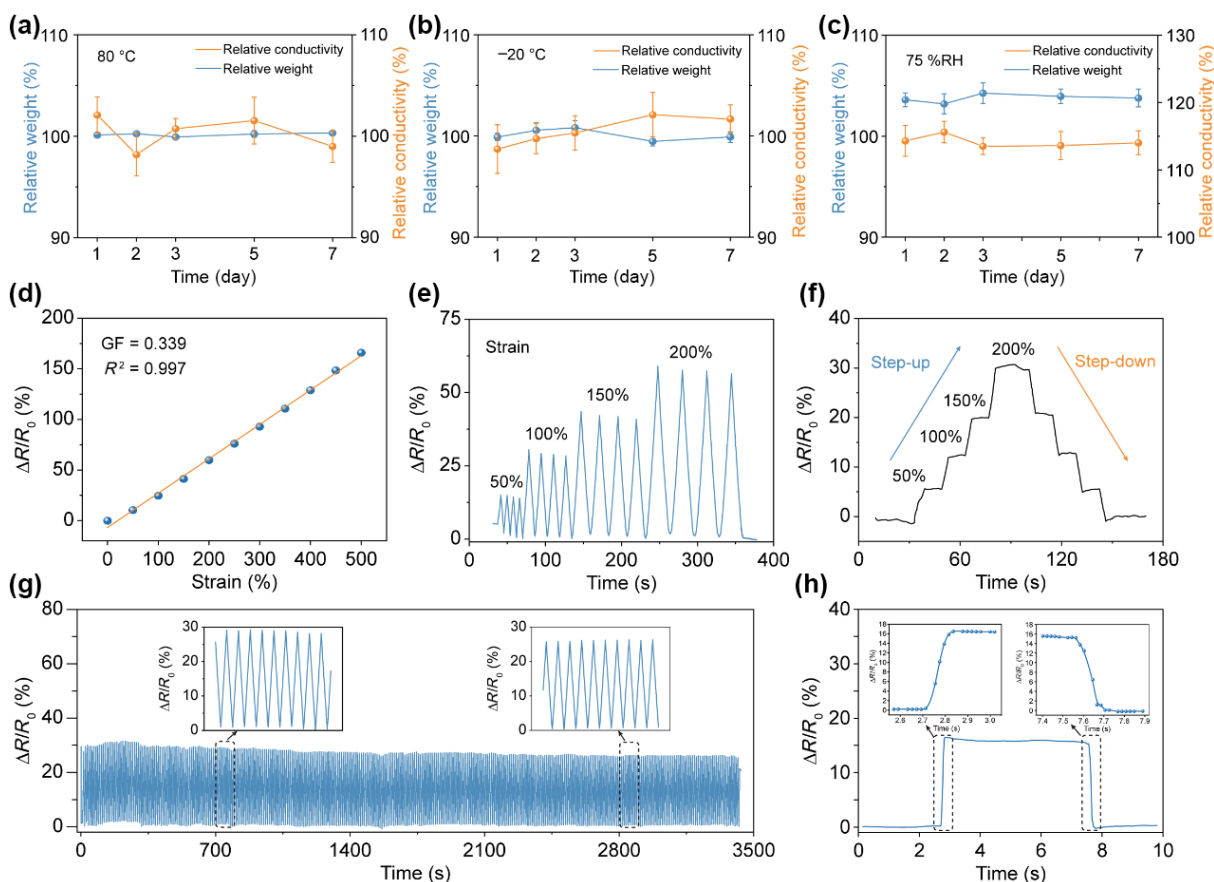


Figure 4 Electrical properties and environmental stability of the ionogels. ((a)–(c)) For the $A_{30}E_{30}I_{50}$ ionogel, changes in ionic conductivity and mass under (a) high temperature, (b) low temperature, and (c) 75 %RH. (d) Relative resistance change of the $A_{30}E_{30}I_{50}$ ionogel as a function of strain. (e) Relative resistance change during dynamic cyclic testing at different strains. (f) Cyclic strain sensing performance of the ionogel from 0% to 200% strain. (g) Relative resistance change of the ionogel over 3500 s of cycling at 100% strain, cycle number $n = 340$. (h) Response and recovery time of the ionogel sensor.

strain sensors at large strains ranging from 100% to 400%. The results demonstrate that the relative resistance changes correspond closely to the applied strain, and the resistance increases with increasing strain, indicating excellent strain-sensing capability (Fig. 4(e)). Furthermore, the loading–unloading experiments confirmed the outstanding stability and repeatability of the AE-I ionogel sensor (Fig. 4(f)). Even after 300 continuous loading–unloading cycles at 100% strain over nearly an hour, the sensor maintains a relatively stable resistance signal, highlighting its exceptional anti-fatigue properties and mechanical durability (Fig. 4(g) and Fig. S11 in the ESM). In addition, the AE-I ionogel sensor demonstrates exceptional dynamic responsiveness, achieving rapid response and recovery times of 140 ms under a 50% applied strain (Fig. 4(h)). Such nearly instantaneous tracking of mechanical loading and unloading indicates minimal viscoelastic hysteresis within the material network. Consequently, the sensor is exceptionally well-suited for capturing highly dynamic and high-frequency signals without distortion. These experimental results comprehensively verify that the combined high sensitivity, rapid response, and robust fatigue resistance of the gel fulfill all essential operational performance criteria.

The combination of robust environmental stability and exceptional electromechanical performance underscores the potential of the AE-I ionogels as a high-fidelity sensing platform (Fig. S12 in the ESM). By maintaining linear responsiveness, rapid temporal dynamics, and consistent signal integrity over extended

cycling, the sensor effectively overcomes the common limitations of signal drift and mechanical fatigue in soft conductors. These attributes, rooted in the unique decoupling of mechanical and electrical properties, ensure the acquisition of clean, repeatable, and high-quality biomechanical data. Such hardware excellence establishes a reliable foundation for transitioning from basic motion monitoring to more complex tasks, such as the interpretation of subtle physiological signals via intelligent machine-learning frameworks.

3.5 Versatile sensing and intelligent human–machine interaction

The AE-I ionogel-based strain sensor, characterized by its remarkable sensitivity, mechanical durability, and stable interfacial adhesion, demonstrates exceptional capability in real-time monitoring of both large-scale and subtle human motions. When adhered to a finger, the sensor's high fidelity allows for the precise distinction of varying bending angles (Fig. 5(a)). Its superior mechanical toughness further enables the tracking of diverse body movements, ranging from joint-level flexion, such as the knee (Fig. 5(b)), wrist (Fig. S13(a) in the ESM), and arm (Fig. S13(b) in the ESM), to high-impact activities like jumping and running when mounted on a shoe sole (Fig. 5(c)). Furthermore, the sensor's low detection limit facilitates the acquisition of subtle physiological signals, including tactile pressure (Fig. S13(c) in the ESM) and neck muscle contractions (Fig. S13(d) in the ESM), confirming the

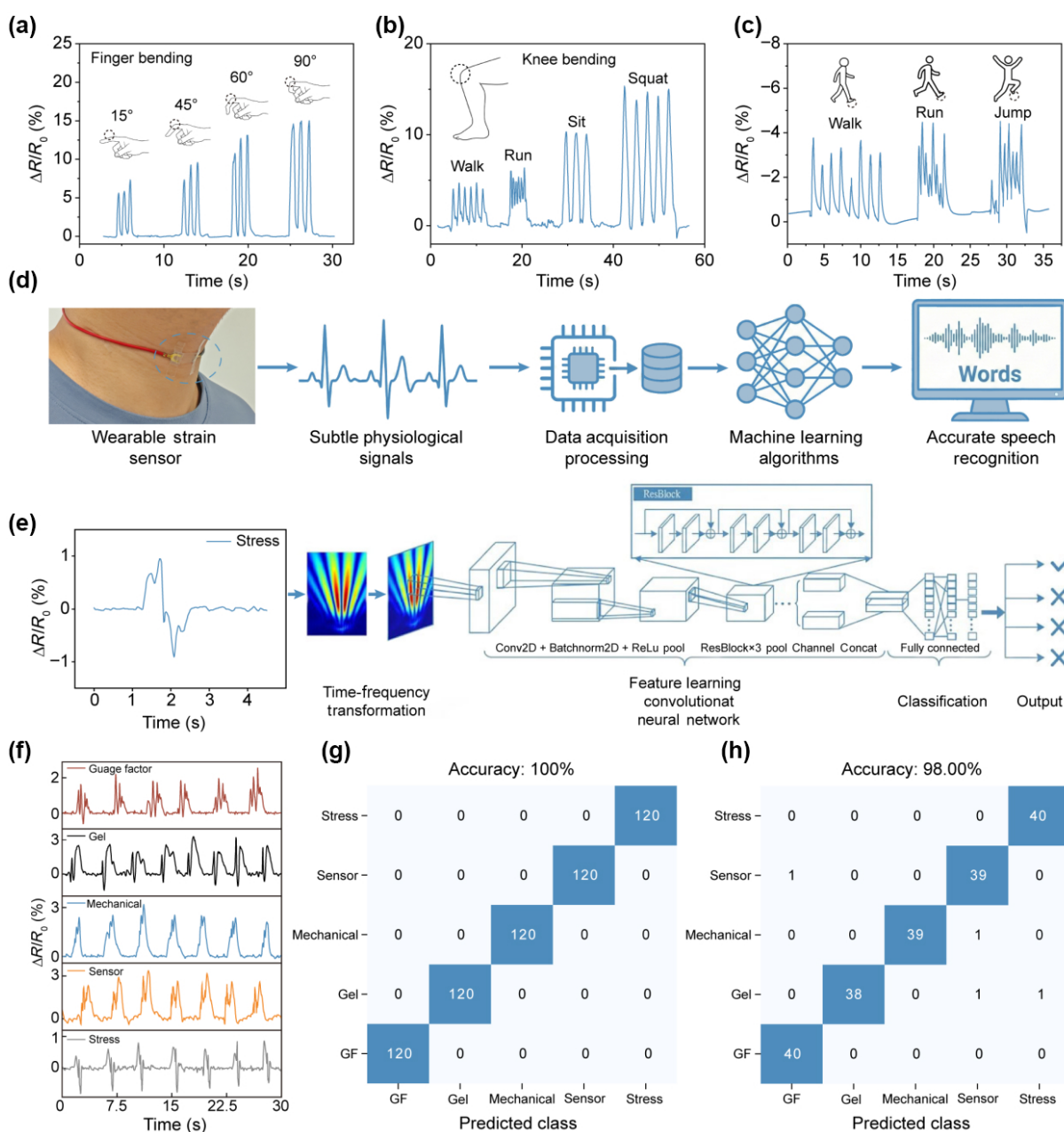


Figure 5 Application of the ionogel sensor for traditional human motion monitoring and machine learning-enabled speech recognition. ((a)–(c)) Relative resistance response of the ionogel sensor to (a) finger bending, (b) knee flexion, and (c) foot movements. (d) Schematic illustration of the machine learning-enabled human–machine interaction system. (e) Schematic of the speech-recognition pipeline. Raw resistance signals recorded during speech are converted into time-frequency representations using CWT, and the resulting spectrograms are classified using a residual CNN comprising Conv2D, BatchNorm, ReLU activation, pooling layers, and residual blocks. (f) Representative resistance response profiles of the ionogel corresponding to five English words: “Guage factor”, “Gel”, “Mechanical”, “Sensor”, and “Stress”. (g) Confusion matrix for the training set, showing 100% classification accuracy, number of training set samples $n = 600$ (120 per class). (h) Confusion matrix for the testing set, demonstrating robust generalization with an accuracy of 98.00%, number of test set samples $n = 200$ (40 per class).

versatility of this microphase-separated network in capturing a broad spectrum of biomechanical data.

Unlike conventional soft sensors that often struggle with signal noise under dynamic conditions, the AE-I ionogel features exceptional broadband stability coupled with high sensitivity. This unique capability empowers the material to accurately track high-frequency and minute voiceprint signals, successfully overcoming the traditional challenges posed by the subtle amplitude of laryngeal muscle movements. Therefore, the AE-I ionogel exhibits significant potential value in the realm of human-machine interaction [55–57].

Leveraging this high-fidelity signal acquisition, we constructed an intelligent speech recognition system utilizing deep learning algorithms as an efficient backend interpretation module.

As illustrated in the intelligent speech interface, the throat-mounted ionogel sensor acquires high-quality resistance signals originating from intricate laryngeal muscle vibrations (Fig. 5(d)). These raw signals exhibit unique and repeatable patterns corresponding to specific English words (Fig. 5(f)), providing a reliable and consistent data foundation for subsequent machine learning processing. To demonstrate the intelligent

interaction potential, the acquired signals were transformed into spectrogram representations and classified via a residual CNN architecture (Fig. 5(e)).

To ensure reliable performance in complex scenarios (e.g., heavy sweating or drastic environmental changes), an artificial intelligence (AI)-assisted signal processing framework was developed. By decoupling slow and low-frequency baseline drifts from rapid physiological signals, this deep learning algorithm automatically removes moisture-induced artifacts, guaranteeing high-fidelity data acquisition for real-time human-machine interaction. Building on this robust signal foundation, a dataset of 800 resistance samples across five spoken words was collected for speech recognition, which was partitioned into a training set of 600 samples (120 per class) and a test set of 200 samples (40 per class).

Owing to the superior stability and high recognition accuracy afforded by the ionogel's enhanced mechanical and electrical properties, the model achieved a remarkable training accuracy of 100% and a testing accuracy of 98% (Figs. 5(g) and 5(h), and Fig. S14 in the ESM). Compared to previously reported systems, the AE-I ionogel-based sensor exhibits enhanced consistency in signal acquisition, significantly reducing the learning burden on the AI module and highlighting its potential as a robust hardware platform for silent speech interfaces and advanced intelligent interaction.

4 Conclusions

In summary, we report an architectural strategy for ionogels based on composition-programmed asymmetric phase connectivity. By regulating the incompatibility among AA, PEGMA, and the ionic liquid [EMIM][TFSI], the polymer network spontaneously forms a microphase-separated morphology in which a soft ion-conductive phase remains continuous while the rigid reinforcing phase is confined to dispersed microdomains. This asymmetric topology effectively reconciles mechanical reinforcement with ionic transport and optical transparency. The resulting ionogels exhibit a unique combination of high stretchability, enhanced toughness, good ionic conductivity, and optical clarity. Importantly, we show that excessive connectivity of the rigid phase leads to bicontinuous structures that produce stiff and opaque gels, highlighting that phase connectivity rather than phase separation alone determines material performance. Flexible strain sensors fabricated from this ionogel deliver stable electrical signals under large deformations, excellent cyclic durability, and high strain detection resolution. These sensors are capable of monitoring a variety of human physiological activities with rapid response and high reliability, demonstrating strong potential for applications in wearable electronics and soft sensing systems. Overall, this work establishes asymmetric phase architecture as a general design principle for multifunctional ionogels, providing new insights for engineering soft ionic materials in flexible electronics, soft robotics, and biointegrated devices.

Electronic Supplementary Material: Supplementary material (confocal microscopic evaluation of morphology, SAXS data analysis, theoretical modeling of binding interactions, energy dissipation and fatigue resistance, multi-substrate interfacial adhesion, impedance and ionic transport, environmental stability and durability, wearable sensing for human motion monitoring, and deep learning-enhanced speech recognition of AE-I ionogels) is available in the online version of this article at <https://doi.org/10.26599/NR.2026.94908880>.

Data availability

All data needed to support the conclusions in the paper are presented in the manuscript and the Electronic Supplementary Material. Additional data related to this paper may be requested from the corresponding author upon request.

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Declaration of competing interest

All the contributing authors report no conflict of interests in this work.

Author contribution statement

J. J. G.: Conceptualization, methodology, investigation, experimental design, data curation, formal analysis, visualization, writing – original draft. L. F. L.: Methodology, data curation, project administration, writing – original draft. T. C. L.: Methodology, data curation, project administration, writing – original draft. X. Y. H.: Methodology, data curation, formal analysis, visualization. Z. X. Z.: Methodology, formal analysis, software. H. Y. L.: Methodology, data curation, formal analysis, visualization. Y. Z.: Methodology, data curation, formal analysis, visualization. Y. Q. C.: Methodology, data curation, formal analysis, visualization. W. X. S.: Methodology, experimental design, formal analysis, software. B. X.: Methodology, supervision, resources, project administration, funding acquisition. Y. L.: Methodology, supervision, resources, project administration. L. D.: Conceptualization, methodology, investigation, data curation, formal analysis, writing – original draft, supervision, resources, project administration. Y. C.: Methodology, supervision, resources, funding acquisition. All the authors have approved the final manuscript.

Use of AI statement

None.

References

- [1] Zhu, Y. Q.; Romain, C.; Williams, C. K. Sustainable polymers from renewable resources. *Nature* **2016**, *540*, 354–362.
- [2] Kaltenbrunner, M.; Sekitani, T.; Reeder, J.; Yokota, T.; Kuribara, K.; Tokuhara, T.; Drack, M.; Schwödiauer, R.; Graz, I.; Bauer-Gogonea, S. et al. An ultra-lightweight design for imperceptible plastic electronics. *Nature* **2013**, *499*, 458–463.
- [3] Fang, Y. L.; Bai, Z. X.; Xu, W. M.; Xiong, X. H.; Wei, J. J.; Hu, Q.; Wang, H. B.; Cui, J. X. Mechanical design principles of conductive gels applied for flexible electronics. *Adv. Funct. Mater.* **2025**, *35*, 2416398.
- [4] Zheng, Y. P.; Cui, T. Y.; Wang, J. W.; Ge, H.; Gui, Z. Unveiling innovative design of customizable adhesive flexible devices from self-healing ionogels with robust adhesion and sustainability. *Chem. Eng. J.* **2023**, *471*, 144617.

- [5] Xu, L. G.; Huang, Z. K.; Deng, Z. S.; Du, Z. K.; Sun, T. L.; Guo, Z. H.; Yue, K. A transparent, highly stretchable, solvent-resistant, recyclable multifunctional ionogel with underwater self-healing and adhesion for reliable strain sensors. *Adv. Mater.* **2021**, *33*, 2105306.
- [6] Zhang, M. W.; Zhao, L. Y.; Tian, F.; Zhao, X. J.; Zhang, Y.; Yang, X.; Huang, W.; Yu, R. Bionic artificial skin based on self-healable ionogel composites with tailored mechanics and robust interfaces. *Adv. Mater.* **2024**, *36*, 2405776.
- [7] Xu, J. H.; Wang, H.; Du, X. S.; Cheng, X.; Du, Z. L.; Wang, H. B. Self-healing, anti-freezing and highly stretchable polyurethane ionogel as ionic skin for wireless strain sensing. *Chem. Eng. J.* **2021**, *426*, 130724.
- [8] Li, T. Q.; Wang, Y. T.; Li, S. H.; Liu, X. K.; Sun, J. Q. Mechanically robust, elastic, and healable ionogels for highly sensitive ultra-durable ionic skins. *Adv. Mater.* **2020**, *32*, 2002706.
- [9] Zhao, W.; Lei, Z. Y.; Wu, P. Y. Mechanically adaptative and environmentally stable ionogels for energy harvest. *Adv. Sci.* **2023**, *10*, 2300253.
- [10] Chen, N.; Zhang, H. Q.; Li, L.; Chen, R. J.; Guo, S. J. Ionogel electrolytes for high-performance lithium batteries: A review. *Adv. Energy Mater.* **2018**, *8*, 1702675.
- [11] Wu, J. T.; Huang, L.; Wang, S. P.; Li, X. R.; Wen, L.; Li, X. M.; Feng, T. T.; Li, P. Y.; Fang, Z. X.; Wu, M. Q. et al. Ionogel electrolyte with dynamic metal-ligand interactions enabled self-healable supercapacitor with high energy density. *Energy Storage Mater.* **2023**, *57*, 549–556.
- [12] Guo, J. N.; Sun, Z.; Zhou, Y. J.; Yan, F. Poly(ionic liquid)-based energy and electronic devices. *Chin. J. Chem.* **2022**, *40*, 1099–1108.
- [13] Shi, Y. K.; Wu, B. H.; Sun, S. T.; Wu, P. Y. Peeling-stiffening self-adhesive ionogel with superhigh interfacial toughness. *Adv. Mater.* **2024**, *36*, 2310576.
- [14] Ge, G.; Zhang, Y. Z.; Xiao, X.; Gong, Y. T.; Liu, C.; Lyu, C.; Ong, W. L.; Ho, G. W.; Yang, Z.; Huang, W. Rapidly gelling, highly adhesive, and mechanically robust ionogels for stretchable and wireless electronics. *Adv. Funct. Mater.* **2024**, *34*, 2310963.
- [15] Yang, X. C.; Wang, X. X.; Wang, C. Y.; Yin, M.; Ma, K.; Li, Y.; Chen, K. Z.; Qiao, S. L. Janus-adhesive ionogels for wearable real-time monitoring and prevention of infantile meningitis. *Chem. Eng. J.* **2024**, *499*, 156197.
- [16] Wang, S. X.; Li, J. J.; Li, S.; Wu, X. C.; Guo, C.; Yu, L. M.; Murto, P.; Wang, Z. H.; Xu, X. F. Self-contained underwater adhesion and informational labeling enabled by arene-functionalized polymeric ionogels. *Adv. Funct. Mater.* **2023**, *33*, 2306814.
- [17] Chen, Y. X.; Chen, Y. Y.; Gao, R. Z.; Yu, X. P.; Lu, C. Reversible molecule interactions enable ultrastretchable and recyclable ionogels for wearable piezoelectric sensors. *ACS Appl. Mater. Interfaces* **2024**, *16*, 50027–50035.
- [18] Yan, X. R.; Zhao, R. R.; Lin, H. J.; Zhao, Z. D.; Song, S. S.; Wang, Y. F. Nucleobase-driven wearable ionogel electronics for long-term human motion detection and electrophysiological signal monitoring. *Adv. Funct. Mater.* **2025**, *35*, 2412244.
- [19] Le, K.; Sun, X.; Chen, J. J.; John, J. V.; Servati, A.; Heidari, H.; Khademhosseini, A.; Ko, F.; Jiang, F.; Servati, P. Stretchable, self-healing, biocompatible, and durable ionogel for continuous wearable strain and physiological signal monitoring. *Chem. Eng. J.* **2023**, *471*, 144675.
- [20] He, Q. S.; Zhong, Q. Y.; Sun, Z.; Zhang, H. R.; Zhao, Z. J.; Shi, Z. Y.; Liu, X. F.; Zhao, Z. F.; Lu, J.; Ye, Y. Z. et al. Highly stretchable, repeatable, and easy-to-prepare ionogel based on polyvinyl chloride for wearable strain sensors. *Nano Energy* **2023**, *113*, 108535.
- [21] Zhu, Q. Q.; Liao, W. Q.; Sun, C.; Qin, X.; Zhang, F. J.; Ji, H. F.; Li, Y. Q.; Wen, Z.; Sun, X. H. Highly stretchable, conductive, and wide-operating temperature ionogel based wearable triboelectric nanogenerator. *Nano Res.* **2023**, *16*, 11638–11645.
- [22] Bhar, B.; Singh, R.; Ramesh, V. K.; Dey, S.; Nandi, S. K.; Paily, R.; Mandal, B. B. Wearable e-bandage with antimicrobial ionogel as an integrated electroceutical device for accelerated wound healing. *ACS Mater. Lett.* **2024**, *6*, 3453–3461.
- [23] Kuddushi, M.; Xu, B. B.; Malek, N.; Zhang, X. H. Review of ionic liquid and ionogel-based biomaterials for advanced drug delivery. *Adv. Colloid Interface Sci.* **2024**, *331*, 103244.
- [24] Kim, J. S.; Kim, J.; Lim, J. W.; Kim, D. J.; Lee, J. I.; Choi, H.; Kweon, H.; Lee, J.; Yee, H.; Kim, J. H. et al. Implantable multi-cross-linked membrane-ionogel assembly for reversible non-Faradaic neurostimulation. *ACS Nano* **2023**, *17*, 14706–14717.
- [25] Fan, X. T.; Liu, S. Q.; Jia, Z. H.; Koh, J. J.; Yeo, J. C. C.; Wang, C. G.; Suratman, N. E.; Loh, X. J.; Le Bideau, J.; He, C. B. et al. Ionogels: Recent advances in design, material properties and emerging biomedical applications. *Chem. Soc. Rev.* **2023**, *52*, 2497–2527.
- [26] Zhang, P.; Yang, Y. F.; Li, Z. B.; Xue, Y.; Wang, F. C.; Shan, L. J.; Wang, Y. F.; Shi, X. T.; Wu, K.; Liu, J. Conducting hydrogel-based neural biointerfacing technologies. *Adv. Funct. Mater.* **2025**, *35*, 2422869.
- [27] Shan, L. J.; Xue, Y.; Chen, X. M.; Wang, Y. F.; Feng, Y. H.; Dong, L.; Wang, C.; Zhang, P.; Wang, F. C.; Guo, L. N. et al. Mechanically compliant and impedance matching hydrogel bioelectronics for low-voltage peripheral neuromodulation. *Adv. Mater.* **2026**, *38*, e11014.
- [28] Lee, J.; Miri, S.; Bayro, A.; Kim, M.; Jeong, H.; Yeo, W. H. Biosignal-integrated robotic systems with emerging trends in visual interfaces: A systematic review. *Biophys. Rev.* **2024**, *5*, 011301.
- [29] Yang, L.; Sun, L. J.; Huang, H. F.; Neisiany, R. E.; You, Z. W. An interfacial robust and entire self-healing ionogel-elastomer hybrid for elastic electronics enables discretionary assembly and reconfiguration. *Sci. China Chem.* **2024**, *67*, 1316–1323.
- [30] Sun, L. J.; Huang, H. F.; Zhang, L. Z.; Neisiany, R. E.; Ma, X. P.; Tan, H.; You, Z. W. Spider-silk-inspired tough, self-healing, and melt-spinnable ionogels. *Adv. Sci.* **2024**, *11*, 2305697.
- [31] Tan, H.; Sun, L. J.; Huang, H. F.; Zhang, L. Z.; Neisiany, R. E.; Ma, X. P.; You, Z. W. Continuous melt spinning of adaptable covalently cross-linked self-healing ionogel fibers for multi-functional ionotronics. *Adv. Mater.* **2024**, *36*, 2310020.
- [32] Yang, L.; Sun, L. J.; Huang, H. F.; Zhu, W. F.; Wang, Y. H.; Wu, Z. K.; Neisiany, R. E.; Gu, S. J.; You, Z. W. Mechanically robust and room temperature self-healing ionogel based on ionic liquid inhibited reversible reaction of disulfide bonds. *Adv. Sci.* **2023**, *10*, 2207527.
- [33] Wang, Y.; Wei, Z. X.; Ji, T. T.; Bai, R. B.; Zhu, H. L. Highly ionic conductive, stretchable, and tough ionogel for flexible solid-state supercapacitor. *Small* **2024**, *20*, 2307019.
- [34] Yu, L.; Liu, Q.; Wang, L. B.; Guo, S. T.; Hu, Q. M.; Li, Y. Q.; Lan, X. W.; Liu, Z. F.; Hu, X. L. Boosting lithium batteries under harsh operating conditions by a resilient ionogel with liquid-like ionic conductivity. *J. Energy Chem.* **2021**, *62*, 408–414.
- [35] Wang, S.; Jiang, Y. J.; Hu, X. L. Ionogel-based membranes for safe lithium/sodium batteries. *Adv. Mater.* **2022**, *34*, 2200945.
- [36] Zhang, M. W.; Yu, R.; Tao, X. L.; He, Y. Y.; Li, X. P.; Tian, F.; Chen, X. Y.; Huang, W. Mechanically robust and highly conductive ionogels for soft ionotronics. *Adv. Funct. Mater.* **2023**, *33*, 2208083.
- [37] Qiu, W. L.; Chen, G. Q.; Zhu, H.; Zhang, Q.; Zhu, S. P. Enhanced stretchability and robustness towards flexible ionotronics via double-network structure and ion-dipole interactions. *Chem. Eng. J.* **2022**, *434*, 134752.
- [38] Zhao, X. J.; Xu, J. H.; Zhang, J. Y.; Guo, M. R.; Wu, Z. L.; Li, Y. Y.; Xu, C.; Yin, H. Z.; Wang, X. L. Fluorescent double network ionogels with fast self-healability and high resilience for reliable human motion detection. *Mater. Horiz.* **2023**, *10*, 646–656.
- [39] Dinh Xuan, H.; Timothy, B.; Park, H. Y.; Lam, T. N.; Kim, D.; Go, Y.; Kim, J.; Lee, Y.; Ahn, S. I.; Jin, S. H. et al. Super stretchable and durable electroluminescent devices based on double-network ionogels. *Adv. Mater.* **2021**, *33*, 2008849.
- [40] Wang, M. X.; Xiao, X.; Siddika, S.; Shamsi, M.; Frey, E.; Qian, W.; Bai, W. B.; O'Connor, B. T.; Dickey, M. D. Glassy gels toughened by solvent. *Nature* **2024**, *631*, 313–318.

- [41] Cao, Z. Q.; Liu, H. L.; Jiang, L. Transparent, mechanically robust, and ultrastable ionogels enabled by hydrogen bonding between elastomers and ionic liquids. *Mater. Horiz.* **2020**, *7*, 912–918.
- [42] Cong, Z. H.; Cui, Z. Y.; Liu, C.; Wang, J. L.; Huo, X. S.; Xu, J. B.; Niu, J. High strength and tough ionogels with bicontinuous phase network structure induced by electrostatic adsorption triggered microphase separation. *Adv. Funct. Mater.* **2024**, *34*, 2410588.
- [43] Tie, J.; Mao, Z. P.; Zhang, L. P.; Zhong, Y.; Xu, H. Strong and ultratough ionogel enabled by ingenious combined ionic liquids induced microphase separation. *Adv. Funct. Mater.* **2023**, *33*, 2307367.
- [44] Lv, D.; Li, X.; Huang, X.; Cao, C. Y.; Ai, L. Q.; Wang, X. J.; Ravi, S. K.; Yao, X. Microphase-separated elastic and ultrastretchable ionogel for reliable ionic skin with multimodal sensation. *Adv. Mater.* **2024**, *36*, 2309821.
- [45] Zhou, R.; Jin, Y.; Li, Y. P.; Jin, H. Y.; Zeng, W. H.; Mei, J. Y.; Liu, Y. Q. *In-situ* phase separation constructing robust hydrophobic ionogels with multifunction. *Chem. Eng. J.* **2023**, *476*, 146840.
- [46] He, X. N.; Zhang, B.; Liu, Q. J.; Chen, H.; Cheng, J. X.; Jian, B. C.; Yin, H. L.; Li, H. G.; Duan, K.; Zhang, J. W. et al. Highly conductive and stretchable nanostructured ionogels for 3D printing capacitive sensors with superior performance. *Nat. Commun.* **2024**, *15*, 6431.
- [47] Wang, M. X.; Zhang, P. Y.; Shamsi, M.; Thelen, J. L.; Qian, W.; Truong, V. K.; Ma, J.; Hu, J.; Dickey, M. D. Tough and stretchable ionogels by *in situ* phase separation. *Nat. Mater.* **2022**, *21*, 359–365.
- [48] Huang, Y. Y.; Ming, X. Q.; Tang, Z.; Chen, G. Q.; Duan, X. Z.; Zhu, H.; Zhu, S. P.; Zhang, Q. Ultra-strong ionogel adhesives via *in situ* microphase separation. *Adv. Funct. Mater.* **2025**, *35*, 2417011.
- [49] Jin, Y. K.; Wen, B.; Gu, Z. X.; Jiang, X. T.; Shu, X. L.; Zeng, Z. P.; Zhang, Y. P.; Guo, Z. N.; Chen, Y.; Zheng, T. T. et al. Deep-learning-enabled mxene-based artificial throat: Toward sound detection and speech recognition. *Adv. Mater. Technol.* **2020**, *5*, 2000262.
- [50] Li, H. L.; Xu, F. C.; Li, Y.; Sun, J. Q. Self-healing ionogel-enabled self-healing and wide-temperature flexible zinc-air batteries with ultra-long cycling lives. *Adv. Sci.* **2024**, *11*, 2402193.
- [51] Wang, P. Z.; Zhang, L.; Hu, Z. Q.; Shang, J. Q.; Zhou, J. Transparent and anti-fingerprint coating prepared with chitin nanofibers and surface modification via vapor deposition. *Prog. Org. Coat.* **2022**, *172*, 107126.
- [52] Liu, M. Q.; Wan, X. Z.; Yang, M.; Wang, Z.; Bao, H.; Dai, B.; Liu, H.; Wang, S. T. Thermo-responsive jamming of nanoparticle dense suspensions towards macroscopic liquid–solid switchable materials. *Angew. Chem., Int. Ed.* **2022**, *61*, e202114602.
- [53] Zhu, X. L.; Fang, Z.; Deng, Q. H.; Zhou, Y.; Fu, X. L.; Wu, L. Z.; Yan, W. W.; Yang, Y. Poly(ionic liquid)@PEGMA block polymer initiated microphase separation architecture in poly(ethylene oxide)-based solid-state polymer electrolyte for flexible and self-healing lithium batteries. *ACS Sustainable Chem. Eng.* **2022**, *10*, 4173–4185.
- [54] Bui, K.; Nguyen, G. T. M.; Vancaeyzeele, C.; Vidal, F.; Hu, X.; Wan, C. Y.; Plesse, C. Stretchable, healable, and weldable vitrimer ionogel for ionotronic applications. *Chem. Eng. J.* **2023**, *474*, 145533.
- [55] Chen, X. J.; Lou, Z. Y.; Gao, X. X.; Yin, L.; Qin, S. Y.; Lin, M. Y.; Zhang, F. G.; Lu, Y.; Ding, S. C.; Liu, R. X. et al. A noise-tolerant human–machine interface based on deep learning-enhanced wearable sensors. *Nat. Sens.* **2025**, *1*, 39–51.
- [56] He, W. Q.; Lin, R. M.; Kong, S. X.; Qiang, M. Y.; Huang, L. Q.; Dai, B.; Yao, X.; Su, L.; Zhang, X. J. Intelligent sensing: The emerging integration of machine learning and soft sensors based on hydrogels and ionogels. *Adv. Sci.* **2025**, *12*, e17851.
- [57] Yang, C. Q.; Zhang, D. Z.; Wang, W. W.; Zhang, H.; Zhou, L. N. Multi-functional MXene/helical multi-walled carbon nanotubes flexible sensor for tire pressure detection and speech recognition enabled by machine learning. *Chem. Eng. J.* **2025**, *505*, 159157.



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