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Bioinspired Soft Materials: From “Bio-like” Properties to “Life-like” Functions

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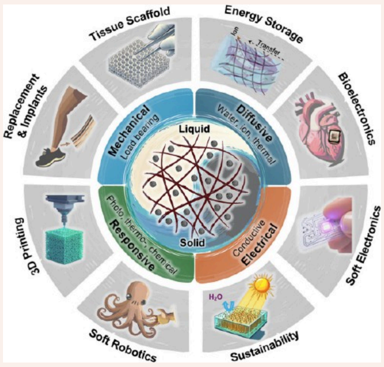

CONSPECTUS: Soft materials are central to technologies that must safely interface to, and move with, living systems—from autonomous soft robots to long-term bioelectronic implants. Yet key challenges persist across multiple scales. At the materials level, combining high mechanical robustness with large, reversible deformation, achieving fast responses without external controllers, and integrating efficient transport while preserving biocompatibility remain difficult. At the system level, long-term stability in extreme environments and scalable, simple manufacturing processes are essential for practical applications.

Here we outline design strategies that act directly on polymer−solvent interactions and on feedback pathways within the material, so that function emerges from the network itself rather than from add-on hardware. First, we harness ion-specific effects and solvent-mediated aggregation to build hierarchical hydrogel architectures with tunable stiffness, toughness, and transport. Simple, scalable processes—e.g., Hofmeister effect-guided salting-out and cononsolvency—allow us to dial in properties across orders of magnitude and to impose anisotropy when needed. These systems achieve high mechanical robustness, fatigue resistance, and rapid mass transport enabled by open-cell porous networks. Second, we engineer responsive behaviors and further augment them with built-in feedback to approach life-like functions with advanced physical intelligence. Coordination-based networks store and release elastic energy on demand for high-power actuation, while local, reversible viscosity/pinning control enables reprogrammable magnetic orientation of embedded particles under modest fields, producing diverse and rapid shape changes. Furthermore, light-driven architectures exploit self-shadowing to generate sustained oscillations without external circuitry, exemplifying built-in feedback enabled autonomy at the materials level.

Together these ideas provide a practical route from “bio-like properties” to “life-like functions”: materials that are tough yet compliant, fast yet stable, and electrically compatible with tissues and devices. We highlight application vignettes in soft robotics, human−machine interfaces, and energy storage systems, and we discuss limits and opportunities for scaling. By emphasizing process-simple routes to hierarchical structure and built-in feedback, this Account offers design principles for integrating multiple properties and functions within a single soft platform and points to future directions where soft matter can autonomously sense, decide, and act.

1. INTRODUCTION

Soft materials have emerged as critical enablers for the seamless integration of synthetic systems with biological organisms, revolutionizing how we mimic, interface with, and interact with living systems.[1−3] Their unique combination of flexibility, stretchability, and biocompatibility has enabled next-generation devices that bridge artificial and biological domains—from wearable electronics that conformally interface with skin for continuous health monitoring, to energy storage devices that power portable and mobile systems, and soft robotics that enable adaptive human−machine interaction and assistive technologies.[4] In interactive systems, soft robotics has transformed machine−human collaboration, enabling applications from surgical assistance to adaptive prosthetics.[5] Yet the very properties that make soft materials attractive—their compliance and adaptability—also present fundamental challenges. Many soft polymers and hydrogels fail to meet durability requirements for load-bearing applications while maintaining desired flexibility,[6,7] and their poor electrical conductivity limits integration with electronic components in smart devices.[1,3] Moreover, insufficient responsiveness to external stimuli (*e.g.*, temperature, light, magnetic fields) further constrains their potential as truly adaptive systems. While material systems such as double-network architectures and conductive polymer composites have demonstrated promising properties,[6,8] conventional approaches to enhance their performance typically rely on complex molecular design and chemical modification of polymer networks, limiting their

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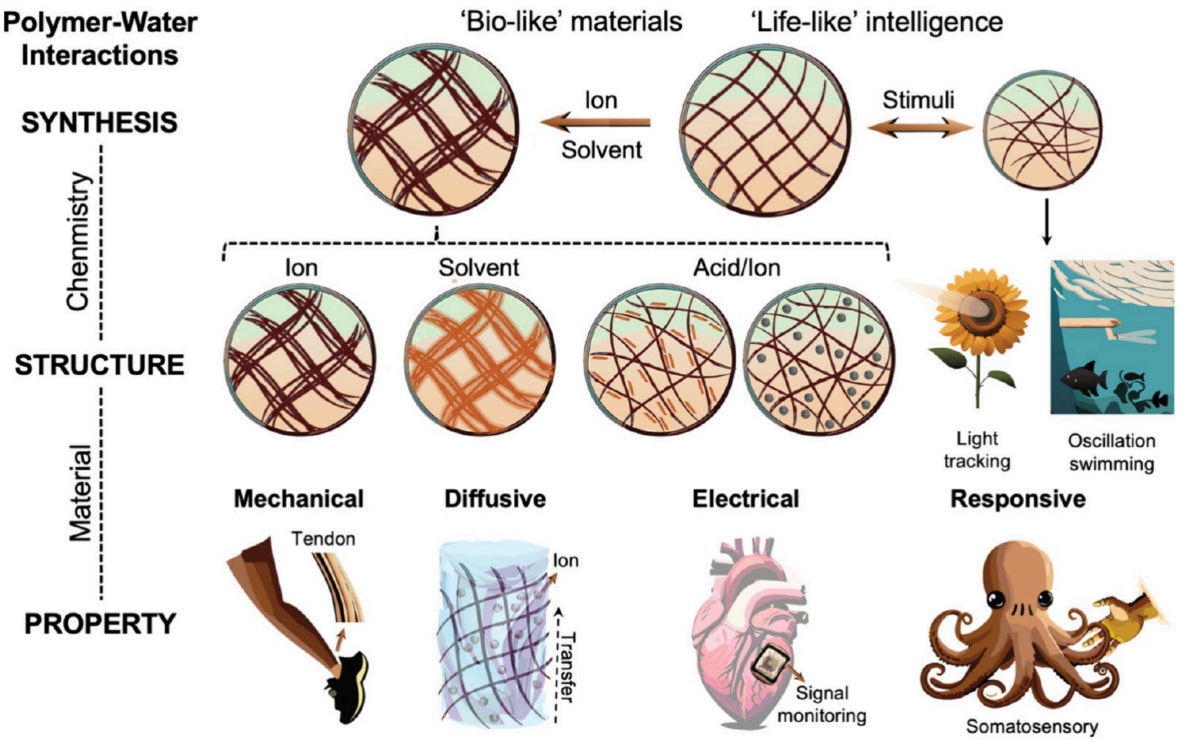

Figure 1. Development of soft materials for human-material integration through controlled polymer−water interactions. Schematic representation of the processing approach from molecular-level manipulation of polymer−solvent interactions to the synthesis of materials with bioinspired structures and functionalities.

versatility and scalability. Moreover, these methods often trade off other essential properties, such as sacrificing mechanical robustness for electrical conductivity.

Building upon these advancements, soft materials are increasingly being developed to create systems that more closely mimic living organisms. This evolution toward truly intelligent materials demonstrates the potential to revolutionize fields such as soft robotics, where materials could embody physical algorithms for real-time decision-making, and wearable bioelectronics, where they could serve as conformal, responsive interfaces for continuous health monitoring and human−machine interaction.[9−12] Realizing this vision requires a deeper understanding of structure−property relationships across multiple scales and the development of innovative processing techniques. It also necessitates a multidisciplinary approach, integrating insights from materials science, chemistry, biology, and engineering to create soft materials with hierarchical structures and multifunctional properties that emulate natural systems. Ultimately, the goal is to develop materials that simultaneously exhibit high mechanical robustness, excellent electrical properties, multistimuli responsiveness, and biocompatibility, while maintaining simplicity in synthesis and scalability in production.

In this Account, we demonstrate our approach to engineering soft materials that progress from "bio-like properties" to "life-like functions"—from materials that mimic the passive mechanical and transport characteristics of biological tissues, to systems that exhibit autonomous, adaptive behaviors through built-in feedback mechanisms. Our work spans three interconnected areas: (1) structure−property engineering by controlling polymer aggregation via simple manipulation of polymer−solvent interactions (Hofmeister effect, cononsolvency, directional freezing) rather than redesigning polymer networks through complex synthesis. This approach precisely controls hydrogel architecture from molecular to macroscopic scales, achieving unprecedented combinations of mechanical strength, mass transport, and electrical conductivity; (2) development of responsive and autonomous systems that incorporate programmed responses (optical, mechanical, magnetic) and built-in feedback mechanisms, enabling materials that can sense, process, and respond autonomously - from adaptive coloration to self-oscillating actuators; and (3) advanced manufacturing and integration methods including 4D printing, room-temperature metal deposition, and liquid metal patterning that enable rapid prototyping and seamless integration of multiple functionalities. This bottom-up approach - from fundamental polymer−water interactions to complex functional systems - creates soft materials capable of mimicking biological tissues for implants, forming conformal interfaces for wearable electronics and energy storage, and interacting dynamically with humans through soft robotics (Figure 1). We not only share fundamental insights into how synthesis parameters govern structure and ultimately determine properties, but also demonstrate how these advancements translate into practical applications across biomedical devices, human−machine interfaces, and autonomous systems. By identifying current limitations and proposing strategies to overcome them, we provide a roadmap for creating soft materials that truly integrate with and enhance human capabilities, pushing the boundaries of what is possible at the intersection of synthetic materials and biological systems.

2. TAILORING HYDROGEL STRUCTURE VIA POLYMER−SOLVENT INTERACTIONS

Designing high-performance hydrogels requires precise control over both their molecular and macroscopic structures.

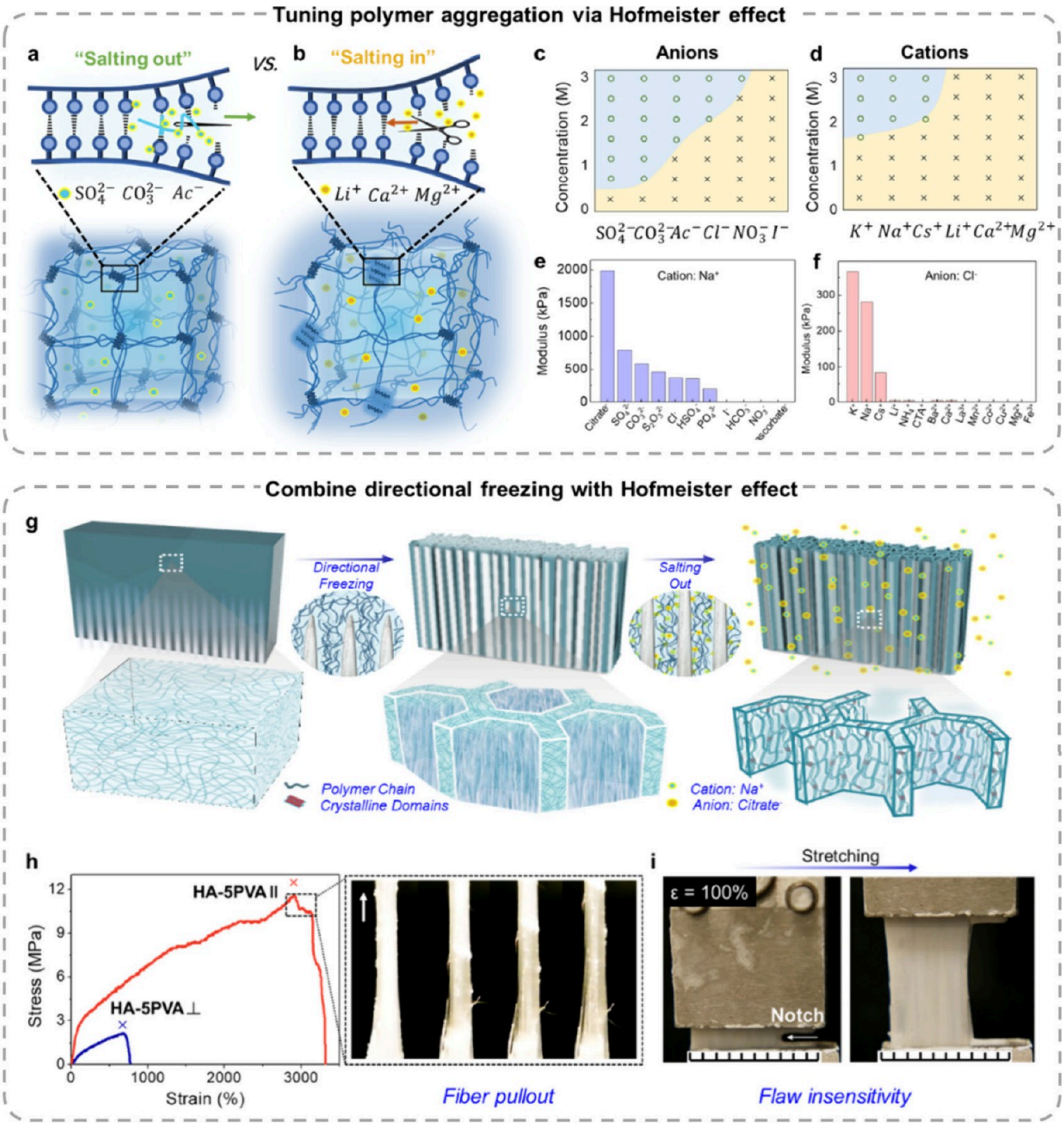

Figure 2. Strategic control of polymer aggregation using the Hofmeister effect and directional freezing. **(a–b)** Schematic figure illustrating how anions and cations influence polymer chain interactions through "salting out" (a) and "salting in" (b) effects. **(c–d)** The Hofmeister series for tuning hydrogel properties, showing how anions (c) and cations (d) influence hydrogel formation across concentration ranges. **(e–f)** Quantify the impact of specific cations (e) and anions (f) on hydrogel modulus. Adapted with permission from ref 27. Copyright 2021 Wiley-VCH GmbH. **(g)** Synergistic approach combining freezing-assisted structuring with salting-out to create hierarchical and anisotropic hydrogel architectures. **(h)** Mechanical anisotropy resulting from the hierarchical structure, evidenced by directional tensile behavior. **(i)** Enhanced fatigue resistance achieved through the multiscale structural control strategy. Adapted with permission from ref 28. Copyright 2021 Springer Nature.

Strategies to improve network topology have produced hydrogels with enhanced mechanical properties through uniform stress distribution. Tetra-PEG architectures with near-ideal network homogeneity achieve tensile strengths up to 27 MPa,[13,14] while slide-ring polyrotaxane systems exploiting pulley effects demonstrate remarkable stretchability exceeding 2500%.[15,16] **Double-network (DN) and dual-cross-linked systems** incorporate sacrificial bonds for energy dissipation. Gong's PAMPS/PAAm DN gels demonstrate fracture energies exceeding 1000 J/m^2,[17] while Suo's alginate/polyacrylamide ionic–covalent hybrids achieve notch-insensitivity with 2300% elongation.[18] Viscoelastic hydrogels based on metal-coordination networks (Fe^{3+}-carboxyl achieving 0.5–18 MPa strength),[19] polyampholyte systems with microphase separation (fatigue thresholds of 1–4 kJ/m^2),[20] and polyprotein cross-linkers (low hysteresis with 126 J/m^2 fatigue threshold)[9] have demonstrated self-healing capabilities and fatigue resistance.

Hierarchical architectures harness multiscale structural features for synergistic performance. Flow-induced aligned nanofibrils produce PVA hydrogels with 14 MPa strength, 1573% elongation, and 153 kJ/m^2 fracture energy.[21] Freeze-casting and 3D printing strategies create anisotropic structures with directional reinforcement,11 with ceramic-reinforced composite hydrogels achieving 6.9 MPa strength, 17.5 MJ/m^3 toughness, and 31.1 kJ/m^2 fatigue resistance.[22] Conductive

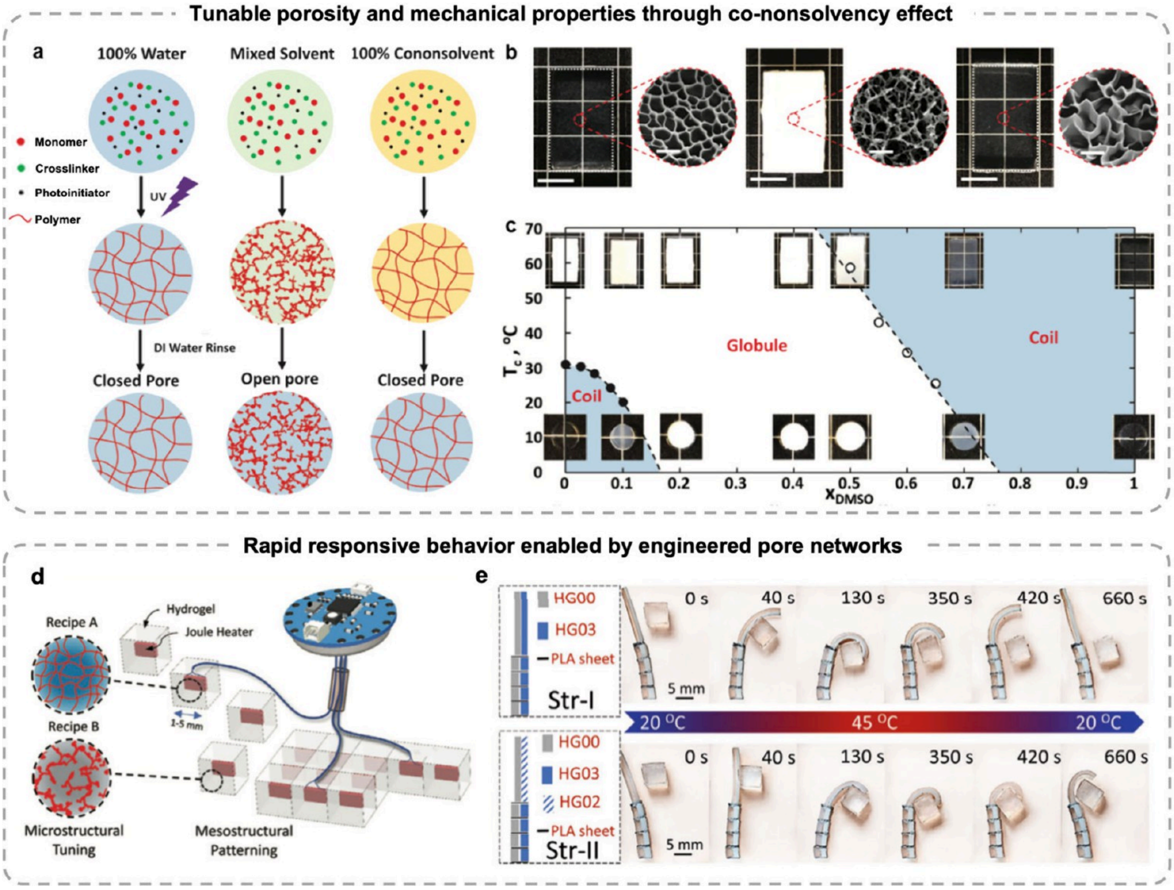

Figure 3. Tunable porosity and responsive behavior of hydrogels via cononsolvency effect. (a) Schematic illustration of hydrogel formation under various water/DMSO ratios, demonstrating the transition from closed pore to open pore structures. (b) SEM images revealing the evolution of microstructures from nonporous (0% DMSO) to highly interconnected porous networks (80% DMSO). (c) Phase diagram showing the relationship between the lower critical solution temperature (LCST) and DMSO mole fraction. Adapted with permission from ref 29. Copyright 2021 Wiley-VCH GmbH. (d) Schematic illustration of mismatching responsive behavior in engineered pore networks. (e) Time-lapse images demonstrating the shape-morphing capabilities of the bilayer hydrogels in response to temperature changes. Adapted with permission from ref 30. Copyright 2021 Wiley-VCH GmbH.

hydrogels based on PEDOT:PSS have achieved conductivities of 20−40 S/cm in pure hydrogel formulations,[23] with 3D-printable systems reaching >11 S/cm while maintaining mechanical compliance and long-term in vivo stability.[24,25]

2.1. Controlling Polymer Aggregation for Tunable Mechanics

Our approach demonstrates how hierarchical structures can be created through simple physicochemical processing of polymer−solvent interactions. By leveraging the Hofmeister effect, cononsolvency, and directional freezing, we achieve control over hydrogel architecture from molecular to macroscopic scales without complex chemical synthesis.[26] This approach addresses inherent hydrogel limitations, including their typically low tensile strength (often below 1 MPa) and poor fatigue resistance, which have hindered their use in load-bearing applications.

We exploited the Hofmeister effect as a powerful tool for tuning the mechanical properties of poly(vinyl alcohol) (PVA) hydrogels.[27] As shown in Figure 2**a-b**, various ions affect PVA chain interactions differently, either by polarizing hydration water molecules, increasing surface tension around hydrophobic segments, or directly binding to PVA chains. Through systematic study, we discovered that the mechanical properties of PVA hydrogels follow the Hofmeister series for both anions and cations (Figure 2**c-f**). This relationship allows precise control over hydrogel stiffness through simple manipulation of salt type and concentration. For instance, by changing the salt from NaCl to Na_2SO_4, we observed a remarkable increase in Young's modulus from 24 ± 2 kPa to 2500 ± 140 kPa, demonstrating a 100-fold enhancement in stiffness. To further enhance performance and introduce hierarchical structures mimicking natural tissues, we combined the Hofmeister effect with directional freezing (Figure 2g).[28] This synergistic approach simultaneously addresses multiple scales of material structure: directional freezing forms an aligned, porous network at the microscale, while subsequent salting-out treatment with sodium citrate promotes polymer aggregation and crystallization at the nanoscale.

The resulting PVA hydrogels exhibit remarkable mechanical properties, including tensile strengths up to 15 ± 1 MPa, elongation at break of 2100 ± 300%, and toughness of 150 ± 20 MJ m^{-3}. Notably, these hydrogels demonstrate fatigue thresholds of 10.5 ± 1.3 kJ/m^2 - eight times higher than previous tough hydrogels (Figure 2**h and i**). This exceptional fatigue resistance is particularly significant, as it addresses a common limitation in hydrogel materials. Moreover, the uniqueness of our approach lies in its simplicity, versatility, and the unprecedented level of control over hydrogel

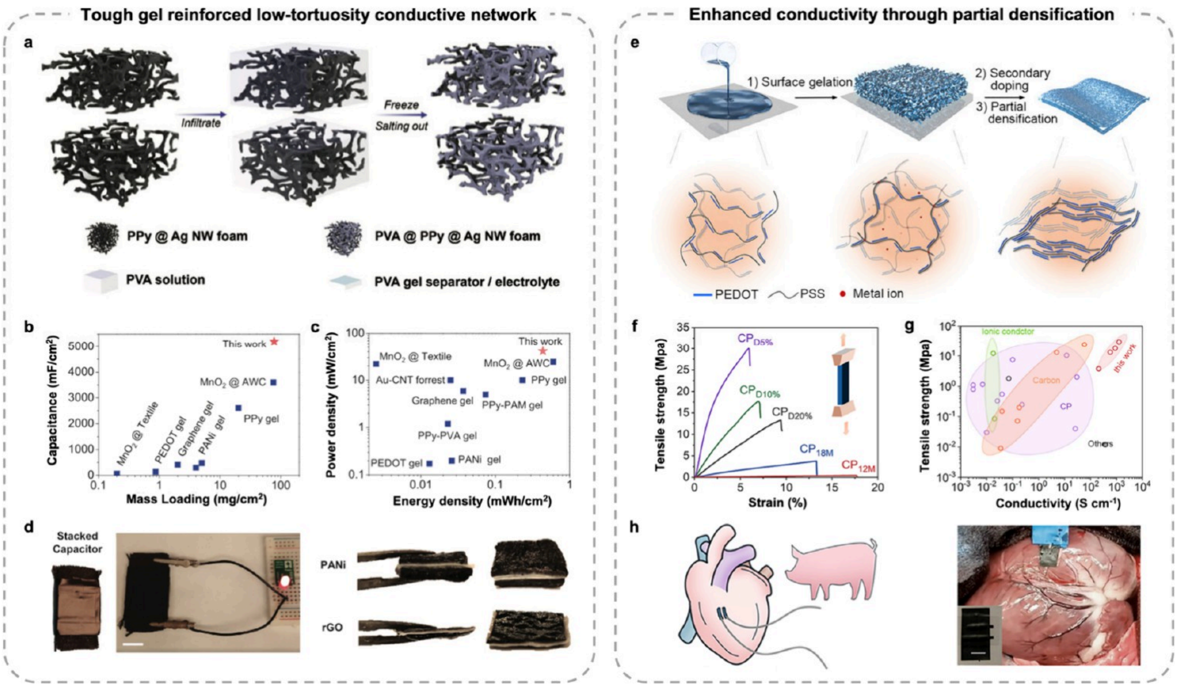

Figure 4. Enhancing conductivity in polymer networks through structural engineering. (a−d) Tough gel reinforced low-tortuosity conductive network: (a) Schematic illustrating the fabrication process of the conductive hydrogel network. (b) Performance comparison of capacitance retention versus mass loading. (c) Ragone plot comparing power density and energy density of various materials. (d) Applications in flexible capacitor. Adapted with permission from ref 36. Copyright 2021 Wiley-VCH GmbH. **(e−h)** High-performance PEDOT:PSS electrodes through controlled densification: (e) Process schematic depicting the partial densification technique. (f) Mechanical properties comparison. (g) Conductivity comparison across different materials and processing methods. (h) Hydrogel electrode applied to cardiac tissue, demonstrating the material's potential in biomedical applications. Adapted with permission from ref 34. Copyright 2022 Elsevier Inc.

properties. Unlike methods that rely on specific chemical modifications or additional components, our strategy is applicable to a wide range of polymer systems and can be easily integrated into existing manufacturing processes.

2.2. Optimizing Mass Transport via Engineered Pore Structures

Mass transport in hydrogels is crucial for many applications, yet enhancing diffusivity often compromises mechanical integrity. We have developed strategies to optimize mass transport while simultaneously improving mechanical properties through engineered pore structures. Our approach leverages the cononsolvency effect, where a polymer becomes insoluble in a mixture of two good solvents (Figure 3a).[29] By polymerizing hydrogels in a water-DMSO mixture, we created unique open-cell porous structures with interconnected microchannels (Figure 3**b and c**). This structure significantly enhances mass transport while maintaining mechanical robustness. For instance, poly(*N*-isopropylacrylamide) (PNIPAAM) hydrogels prepared using this method exhibited a volumetric swelling rate of 138% min^{-1}, nearly 6 times faster than conventional hydrogels (24% min^{-1}). Remarkably, this diffusivity enhancement was accompanied by a 100% increase in Young's modulus and a 120% increase in toughness.

The versatility of the cononsolvency effect extends beyond PNIPAAM to other systems, including poly(vinyl alcohol) (PVA) and poly(*N*-*tert*-butylacrylamide-*co*-acrylamide) (PNTBAM-*co*-PAM). By tuning solvent composition and reaction temperature, we precisely control pore structure and mechanical properties. This approach enables the creation of hydrogels with tailored properties for specific applications. For instance, we exploited the differential swelling rates resulting from controlled pore sizes in rapid-response actuators. By creating a bilayer structure with different pore sizes (e.g., 10 and 50 μm), we achieved rapid and directional bending upon exposure to water, with response times as fast as 0.3 s (Figure 3d).[30] In filtration applications, we utilized the interconnected pore network to enhance permeability while maintaining selectivity. By adjusting the DMSO/water ratio from 0.2 to 0.8, we could tune the average pore size from 5 to 100 μm, allowing for precise control over filtration cutoff sizes. Furthermore, we developed spatiotemporally reconfigurable hydrogel structures using addressable and tunable voxels. This technique combined cononsolvency principles with advanced fabrication methods to produce hydrogels with programmable local properties. By selectively triggering phase transitions in individual voxels through localized solvent composition changes, we could dynamically alter the hydrogel's overall structure and function, enabling adaptive behaviors like shape-morphing and self-folding (Figure 3e).

The synergy between enhanced mass transport and improved mechanical properties opens new avenues for hydrogel applications, particularly in extreme environments.[31] We demonstrated the fabrication of ultrastretchable, antifreezing conductive gels using the cononsolvency effect. These gels maintain high ionic conductivity (1.65 S/m at −50 °C) and mechanical flexibility (stretchable up to 1000% strain) even at temperatures as low as −50 °C. The antifreezing mechanism relies on the formation of a DMSO/water eutectic mixture within the hydrogel pores. This eutectic composition has a significantly lower freezing point (−73 °C) than either pure water or DMSO. This combination of properties makes these gels promising candidates for flexible electronics and energy

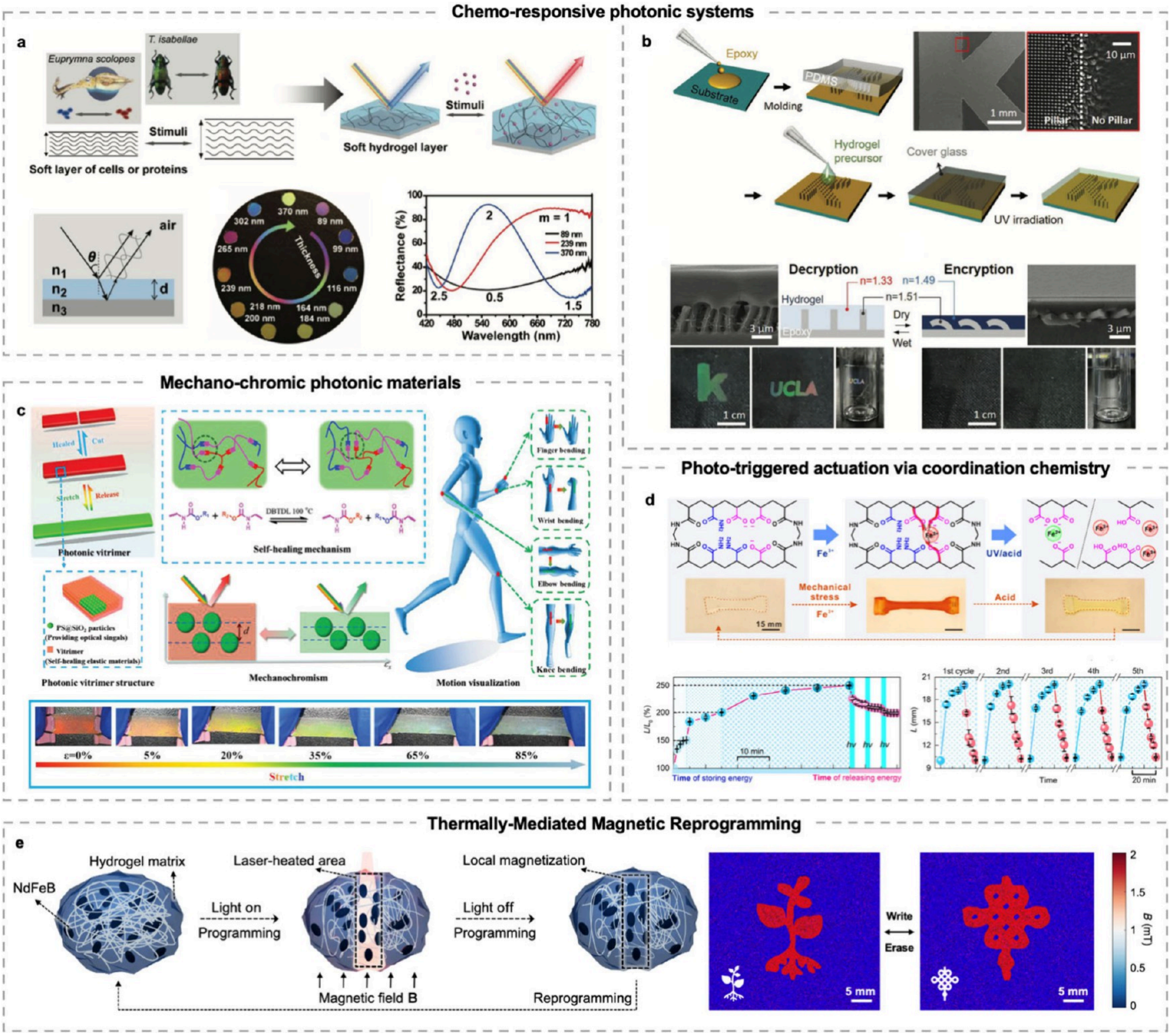

Figure 5. Engineered responsive behaviors in hydrogel-based systems. (a) Chemo-responsive photonic systems: bioinspired hydrogel interferometer for adaptive coloration, illustrating the biomimetic design inspired by cephalopod skin and the principle of color change through thickness-dependent light interference. Adapted with permission from ref 38. Copyright 2018 Wiley-VCH GmbH. (b) Chemo-responsive photonic systems: water-responsive photonic encryption surface, demonstrating the fabrication process and the encryption/decryption mechanism based on hydrogel swelling. Adapted with permission from ref 43. Copyright 2020 Wiley-VCH GmbH. (c) Mechano-chromic photonic materials: mechanochromic photonic vitrimers for motion visualization, showing the self-healing mechanism, mechanochromic response, and color changes under various strains. Adapted with permission from ref 44. Copyright 2020 Wiley-VCH GmbH. (d) Phototriggered actuation via coordination chemistry: programmable elastic recoil for high-power actuation, depicting the molecular mechanism of the Fe^{3+} cross-linked hydrogel network and its mechanical performance during rapid contraction. Adapted with permission from ref 45. Copyright 2020 AAAS. (e) Thermally mediated magnetic reprogramming: magnetically reprogrammable hydrogel actuator with laser-mediated viscoelasticity modulation for reversible magnetic domain patterning and shape morphing. Adapted with permission from ref 46. Copyright 2025 AAAS.

storage devices in extreme environments. Our work on engineered pore structures represents a significant advancement in hydrogel design, overcoming the traditional trade-off between diffusivity and mechanical strength.

2.3. Advancing Electrical Properties in Hydrogel Networks

Enhancing the electrical properties of hydrogels while maintaining their mechanical integrity presents a significant challenge in soft materials. Traditional hydrogels suffer from poor electrical conductivity and low capacitance, limiting their applications in flexible electronics and bioelectronics. Building upon our understanding of controlled molecular aggregation and engineered pore structures, we developed approaches to enhance hydrogel electrical properties.[32−37] Our first approach creates tough-gel reinforced open-porous foam (TROF) by compositing tough PVA hydrogel onto microporous conducting polymer electrodes (Figure 4a).[36] The fabrication involves creating open-porous PPy foam using silver nanowires, infiltrating with PVA solution, then applying freezing and salting out to form a tough hydrogel layer. This combines PPy's high conductivity with PVA's mechanical robustness.

The resulting TROF supercapacitors demonstrate superior areal capacitance (5.25 F/cm^2) and energy density (0.46 mWh/cm^2), along with high power density (41.28 mW/cm^2) due to the open porous structure (Figure 4**b and c**). This scalable method supports modular device architectures, allowing multiple supercapacitor units to be connected in series for enhanced voltage output (Figure 4d).

Furthermore, we advanced hydrogel electrode performance through partial densification of PEDOT:PSS hydrogels for efficient electron−ion transduction (Figure 4e).[34] This process yields exceptional properties: electrical conductivity of 1254 S/cm, tensile strength of 30 MPa, and volumetric capacitance of 73.7 F/cm^3 (Figure 4**f and g**). When applied in supercapacitors, these electrodes deliver outstanding performance with areal capacitance of 2097 mF/cm^2, energy density of 186 μWh/cm^2, and power density of 24000 μW/cm^2. The combination of high conductivity, mechanical compliance, and biocompatibility makes them ideal for bioelectronic interfaces (Figure 4h). This research progression—from enhancing mechanical performance through controlled aggregation, to optimizing mass transport via engineered pores, and finally to advancing electrical properties through hierarchical conductive networks—demonstrates a comprehensive strategy for developing hydrogels with unprecedented combinations of mechanical robustness, efficient mass transport, and high electrical conductivity, bridging the gap between soft biocompatible materials and high-performance functional materials.

3. FROM PROGRAMMED RESPONSES TO AUTONOMOUS BEHAVIORS

The hydrogel engineering strategies described in above—from controlling polymer aggregation for tunable mechanics, to engineering pore structures for enhanced mass transport, to advancing electrical properties—have established materials with unprecedented combinations of mechanical robustness, diffusivity, and conductivity. These foundational properties now enable the next critical step: transforming static, high-performance materials into materials with dynamic, autonomous behaviors. As hydrogel engineering advances, the focus is shifting from optimizing individual properties to designing multifunctional systems capable of intelligently responding to their environment. In this section, we explore how the unique characteristics of hydrogels can be leveraged to develop materials that not only react to stimuli, but do so in ways that mimic the adaptive behaviors found in living organisms. By integrating responsive mechanisms, feedback loops, and programmable features—built upon the structural and functional foundations established previously—hydrogels are being transformed from simple responsive materials into materials with autonomous behaviors.

3.1. Engineering Responsive Behaviors for Smart Adaptations

Our research into responsive hydrogels has evolved from mimicking basic biological functions to creating sophisticated, multifunctional materials that adapt to various stimuli. This progression reflects the increasing sophistication in controlling hydrogel properties across multiple scales, from molecular interactions to macroscopic structures. By leveraging the principles of controlled molecular aggregation and engineered pore structures explored in our earlier work, we have developed a series of increasingly complex responsive systems that showcase the versatility of hydrogels in applications ranging from adaptive optics to high-performance actuators.

Our exploration of responsive hydrogels began with chemo-responsive photonic systems. We first developed a bioinspired hydrogel interferometer for adaptive coloration.[38−42] Drawing inspiration from the color-changing abilities of cephalopods, we created a system consisting of a thin hydrogel layer covalently bonded to a reflective substrate (Figure 5a).[38] As external stimuli cause the hydrogel to swell or shrink, its thickness changes, altering the interference pattern of reflected light. This mechanism allows for dynamic color changes with a response time of less than 1 s and a wide color range spanning the entire visible spectrum. The precise control over hydrogel thickness, achieved through our advanced processing techniques, enables us to produce distinct reflectance spectra with peak wavelength shifts of up to 200 nm. Building on this concept, we developed a water-responsive photonic encryption surface, or "Hydrocipher" system (Figure 5b).[43] The system consists of an array of micropillars (diameter ∼ 1 μm, height ∼ 2 μm) coated with a thin layer (∼100 nm) of responsive hydrogel. In its hydrated state, the swollen hydrogel creates a refractive index mismatch with the pillars, resulting in visible diffraction colors. Upon dehydration, the hydrogel shrinks and matches the refractive index of the pillars, effectively "hiding" the structural color. This reversible process occurs within 5 s, providing a rapid and efficient encryption mechanism.

Beyond chemical stimuli, we extended responsive optical properties to mechanical deformation. We developed mechanochromic photonic vitrimers for motion visualization (Figure 5c).[44] This work represents a significant step toward multifunctional adaptive materials, combining the self-healing capabilities we explored in our tough hydrogels with responsive optical properties. The photonic vitrimer's network structure, based on dynamic covalent bonds, allows for the rearrangement of chemical bonds under specific conditions (e.g., temperatures above 100 °C), enabling self-healing within 30 min. Simultaneously, the incorporation of photonic crystal structures imparts structural color that changes in response to mechanical deformation, with a sensitivity of up to 5 nm wavelength shift per 1% strain.

Transitioning from optical responses to mechanical actuation, we developed phototriggered actuators utilizing Fe^{3+}-carboxyl coordination chemistry. Our first system features programmable elastic recoil for high-power actuation (Figure 5d),[45] where Fe^{3+} ions form reversible coordination bonds that allow mechanical programming into metastable states for elastic energy storage. UV light triggers bond dissociation and rapid contraction, achieving forces up to 8 kPa and power densities of 570 W/kg—surpassing mammalian skeletal muscles. More recently, we achieved thermally mediated magnetic reprogramming by using laser heating to transiently modulate the network's viscoelasticity (Figure 5e).[46] The key innovation lies in using thermal stimuli as a control lever: laser-induced heating of the Fe^{3+}-carboxyl network temporarily reduces pinning forces on embedded NdFeB particles, unlocking their magnetic domains for reorientation under modest magnetic fields (80 mT). Upon cooling, the network recross-links, locking in the new magnetic pattern. This approach allows repeated reprogramming of magnetic patterns in a single hydrogel sheet, producing diverse deformation modes with subsecond response times (0.7−0.8 s). Together, these Fe^{3+}-based systems demonstrate how coordination chemistry can enable both high-power mechanical actuation

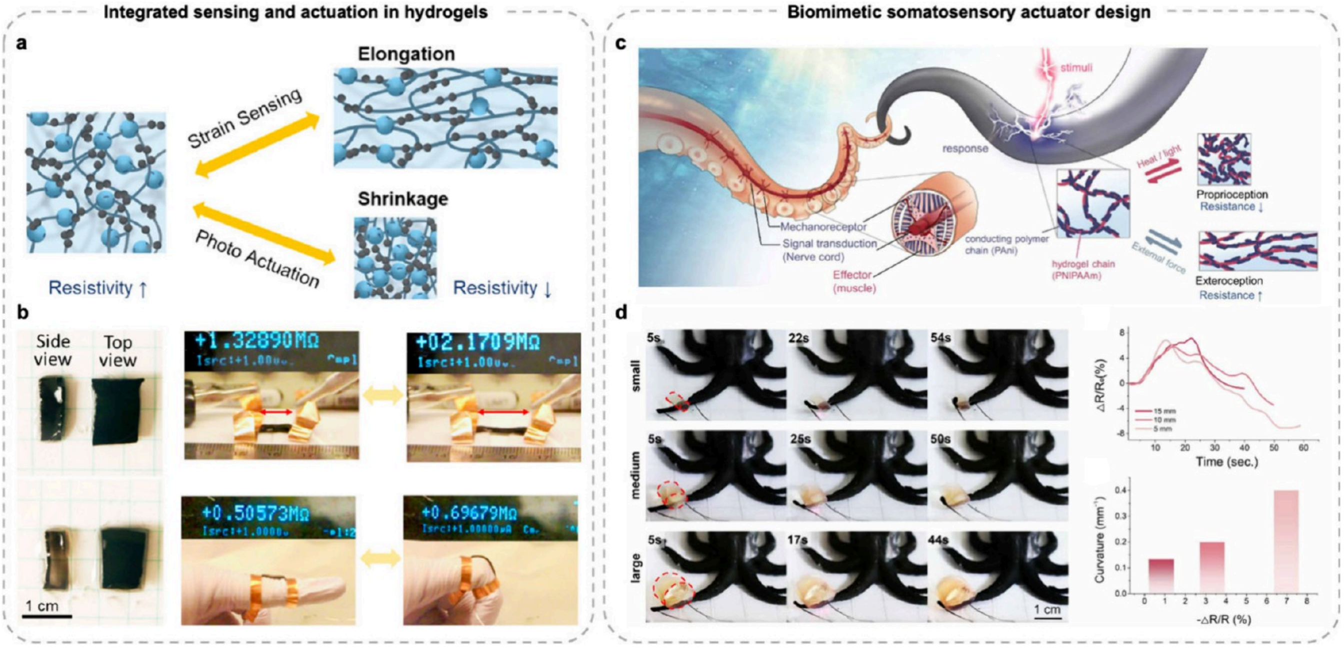

Figure 6. Self-regulating hydrogel combining sensing and actuation capabilities. (a) Schematic illustration of the integrated sensing and actuation mechanisms in the conductive photothermally responsive hydrogel, showing strain sensing through resistivity changes and photoactuation through light-induced shrinkage. (b) Demonstration of the hydrogel's strain sensing ability under different deformations, including elongation and bending, with corresponding resistance measurements. Adapted with permission from ref 47. Copyright 2021 Elsevier Inc. (c) Biomimetic design comparison between the somatosensory actuator and biological sensory neurons. (d) Time-lapse images demonstrating the hydrogel actuator's capability to grasp and lift an object in response to light stimulation. Adapted with permission from ref 48. Copyright 2021 AAAS.

and magnetically reconfigurable responses, representing a significant advance toward adaptive soft materials with on-demand functionalities.

3.2. Self-Regulating Materials: Combining Sensing and Actuation

The integration of sensing and actuation capabilities within a single material system represents a significant advancement toward creating truly intelligent and adaptive systems. Building upon our work in conductive and responsive hydrogels, we developed a stretchable, conductive, photothermally responsive hydrogel that serves as a self-sensing actuator, effectively mimicking biological somatosensory functions.[47,48] This material combines a thermally responsive PNIPAAm network with a conductive PAni network, creating a single homogeneous system capable of both sensing and actuation (Figure 6a).[47] The PNIPAAm provides thermal responsiveness and mechanical flexibility, while PAni functions as both a photothermal converter and a piezoresistive sensing element. The hydrogel responds to stimuli analogously to biological systems: mechanical deformation causes polyaniline (Pani) network rearrangement, changing electrical resistance and enabling strain sensing with a gauge factor of 1.4 over 0−107% strain (Figure 6b). Simultaneously, light exposure triggers photothermal response, causing PNIPAAm phase transition and contraction with up to 40% strain within 13 s under near-infrared stimulation. This approach creates self-regulating materials with potential applications in soft robotics, wearable electronics, and biomedical devices.

The biomimetic nature of this somatosensory actuator is evident in its ability to mimic the functions of biological sensory neurons (Figure 6c).[48] Similar to sensory neurons that detect and transmit information about contact, pressure, and temperature, our hydrogels can sense mechanical deformation and respond to photothermal stimuli. This design enables the creation of artificial systems that closely mimic the complex behaviors of living organisms. To demonstrate the dynamic capabilities of the somatosensory actuator, we showcased its ability to perform complex tasks such as object manipulation. The hydrogel actuator can grasp and lift objects weighing up to 5 g in response to light stimulation, with the ability to sense its own deformation during the process, allowing for precise control and feedback (Figure 6d). By combining sensing and actuation within a single material system, we have developed a self-regulating hydrogel that represents a significant step toward materials with autonomous behaviors. This approach opens up new possibilities for autonomous soft robots, adaptive wearable devices, and advanced biomedical implants that can sense their environment and respond accordingly, bridging the gap between artificial systems and living organisms.

3.3. Self-Oscillating Materials Driven for Autonomous Behaviors

Building upon our foundational work in responsive hydrogels and actuators, we have developed self-oscillating materials that represent a significant leap toward truly autonomous systems capable of complex, life-like behaviors.[49−56] This section explores three innovative approaches to self-oscillating materials, each demonstrating increasing levels of complexity and autonomy.

Our initial self-oscillating systems leveraged light-responsive materials to achieve autonomous behaviors through self-shadowing feedback mechanisms. Inspired by plant phototropism, we first developed a bilayer structure (Figure 7a)[51] with active PNIPAAm containing light-absorbing particles

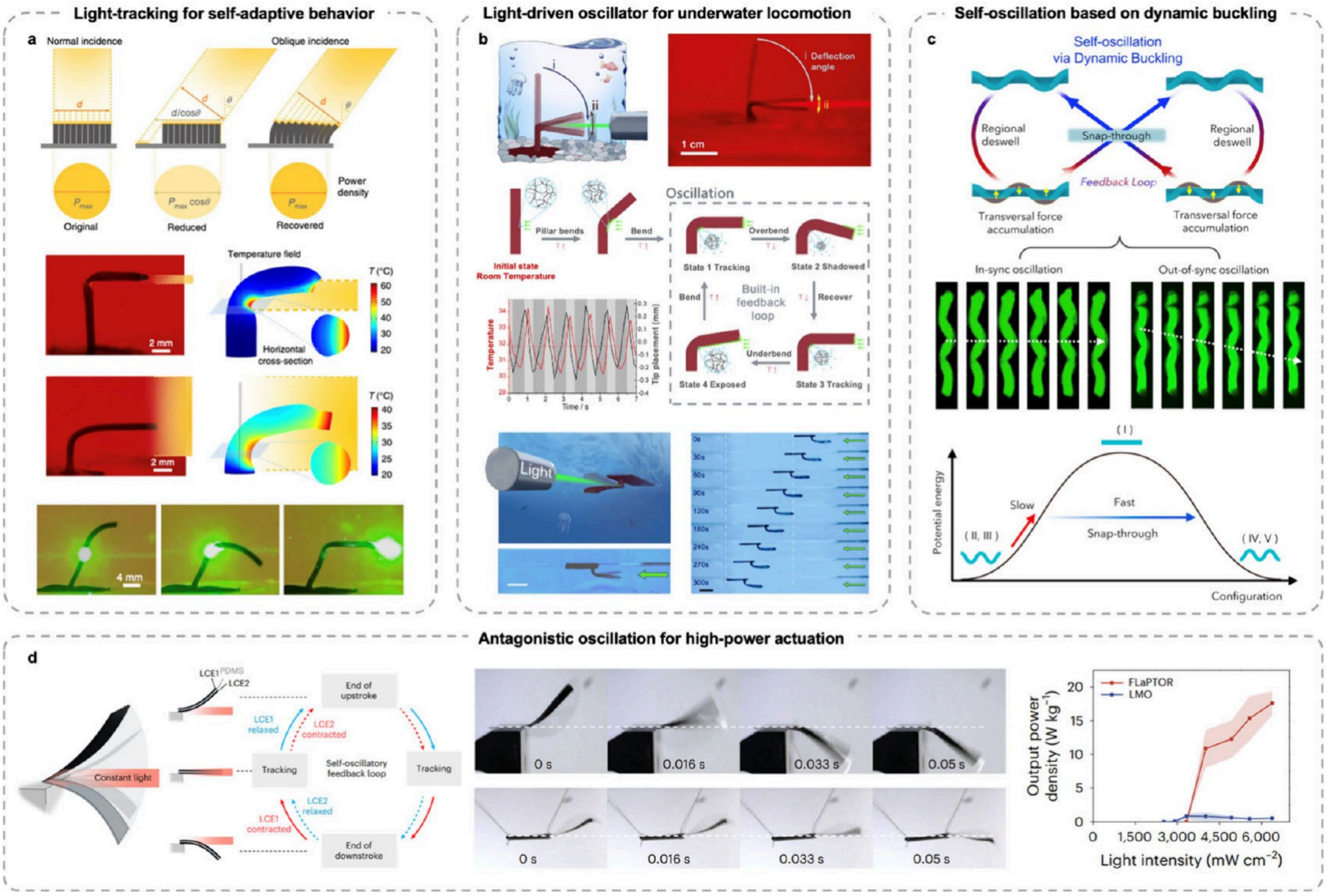

Figure 7. Self-oscillating materials driven by diverse stimuli for autonomous behaviors. (a) Light-tracking system based on PNIPAAm, illustrating the bending mechanism, temperature distribution during actuation, and omnidirectional light tracking capabilities. Adapted with permission from ref 51. Copyright 2019 Springer Nature. (b) OsciBot: A light-driven oscillator for underwater locomotion, showing the experimental setup, oscillation mechanism, and underwater navigation capabilities. Adapted with permission from ref 49. Copyright 2019 AAAS. (c) Self-oscillating hydrogel based on dynamic buckling, depicting the buckling mechanism, feedback loop, and oscillatory motion in response to chemical gradients. Adapted with permission from ref 52. Copyright 2021 Elsevier Inc. (d) Antagonistic photo-oscillator with trilayer architecture, antagonistic contraction mechanism between dual LCE layers, and self-shadowing-induced feedback for autonomous flapping actuation. Adapted from ref 53. Copyright 2025 Springer Nature.

(gold nanoparticles or graphene oxide) and a passive substrate. Upon light exposure, localized heating causes asymmetric contraction and bending toward the light source with <0.1 s response time. This bending creates self-shadowing that establishes a feedback loop, enabling the system to track moving light sources at speeds up to 0.3 rad/s across intensities from 0.2 to 8 W/cm^2. Building on this principle, we created OsciBot (Figure 7b)[49]—a soft phototactic swimmer that achieves underwater locomotion through self-sustained oscillations under constant illumination. The system consists of a cylindrical PNIPAAm hydrogel pillar (diameter ~ 0.5 mm) containing light-absorbing gold nanoparticles. The autonomous oscillation mechanism relies on a self-shadowing negative feedback loop coupled with the lower critical solution temperature (LCST) behavior of PNIPAAm (~32 °C): when light illuminates one side of the pillar, photothermal heating raises the local temperature above the LCST, triggering rapid dehydration and contraction of the illuminated surface. This asymmetric strain causes the pillar to bend toward the light source with curvature changes up to 1.6 cm^{-1} within 4 s. Crucially, structural inertia drives the pillar to overbend beyond the equilibrium position, creating self-shadowing that blocks further light absorption. In the shadow, the hydrogel cools below the LCST, allowing water re-entry and reswelling, which pushes the pillar back into the illuminated region to restart the cycle. This chemo-mechanical oscillation occurs at frequencies up to 4 Hz without any pulsed input, converting steady light energy into periodic motion through far-from-equilibrium dynamics. When configured as a swimming paddle, the asymmetric stroke—power stroke during contraction and recovery stroke during expansion—generates net propulsion at 1.15 body lengths per minute.

Beyond light-driven systems, we also developed self-oscillating systems using chemical-mechanical coupling. Our pH-responsive hydrogel system (Figure 7c)[52] consists of a hydrogel strip placed in a microfluidic channel containing spatially separated acid and base solutions. The self-sustained oscillation arises from the interplay between slow chemical response and fast structural instability: when the hydrogel swells in acidic conditions, the constrained volumetric expansion generates compressive stress that accumulates until reaching a critical buckling threshold (0.4 mN). At this point, the strip undergoes rapid snap-through buckling—a bistable geometric transition—that abruptly flips the hydrogel to the opposite side, exposing it to the alkaline environment. In the base, the hydrogel deprotonates and shrinks, releasing the

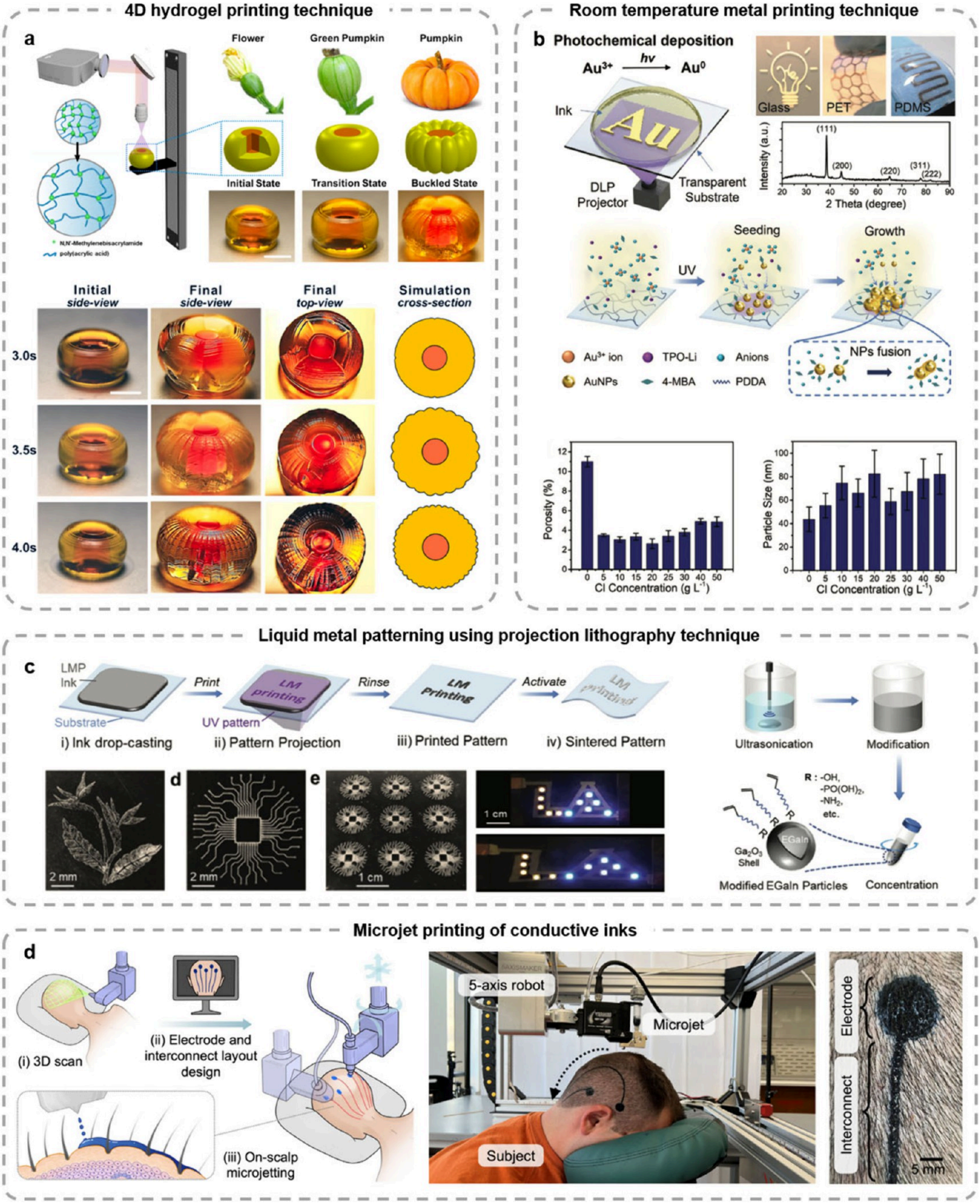

Figure 8. Additive manufacturing methods for functional and intelligent materials. (a) 4D hydrogel printing technique, illustrating the process of creating shape-morphing structures and demonstrating temperature-responsive shape changes in a printed "pumpkin" structure. Adapted with permission from ref 58. Copyright 2019 American Chemical Society. (b) Room temperature metal printing technique, showing the mechanism of anion-assisted photochemical deposition of gold, the printing process, and the influence of chloride ion concentration on conductivity and particle size. Adapted with permission from ref 60. Copyright 2022 Wiley-VCH GmbH. (c) Liquid metal patterning using projection lithography, depicting the process flow, examples of high-resolution patterns, and the mechanism of particle modification and coalescence. Adapted with permission from ref 57. Copyright 2024 Wiley-VCH GmbH. (d) Direct on-scalp printing of PEDOT:PSS e-tattoos via microjet dispensing with personalized sensor layouts and dual-ink system for electrode-interconnect fabrication. Adapted with permission from ref 61. Copyright 2025 Elsevier Inc.

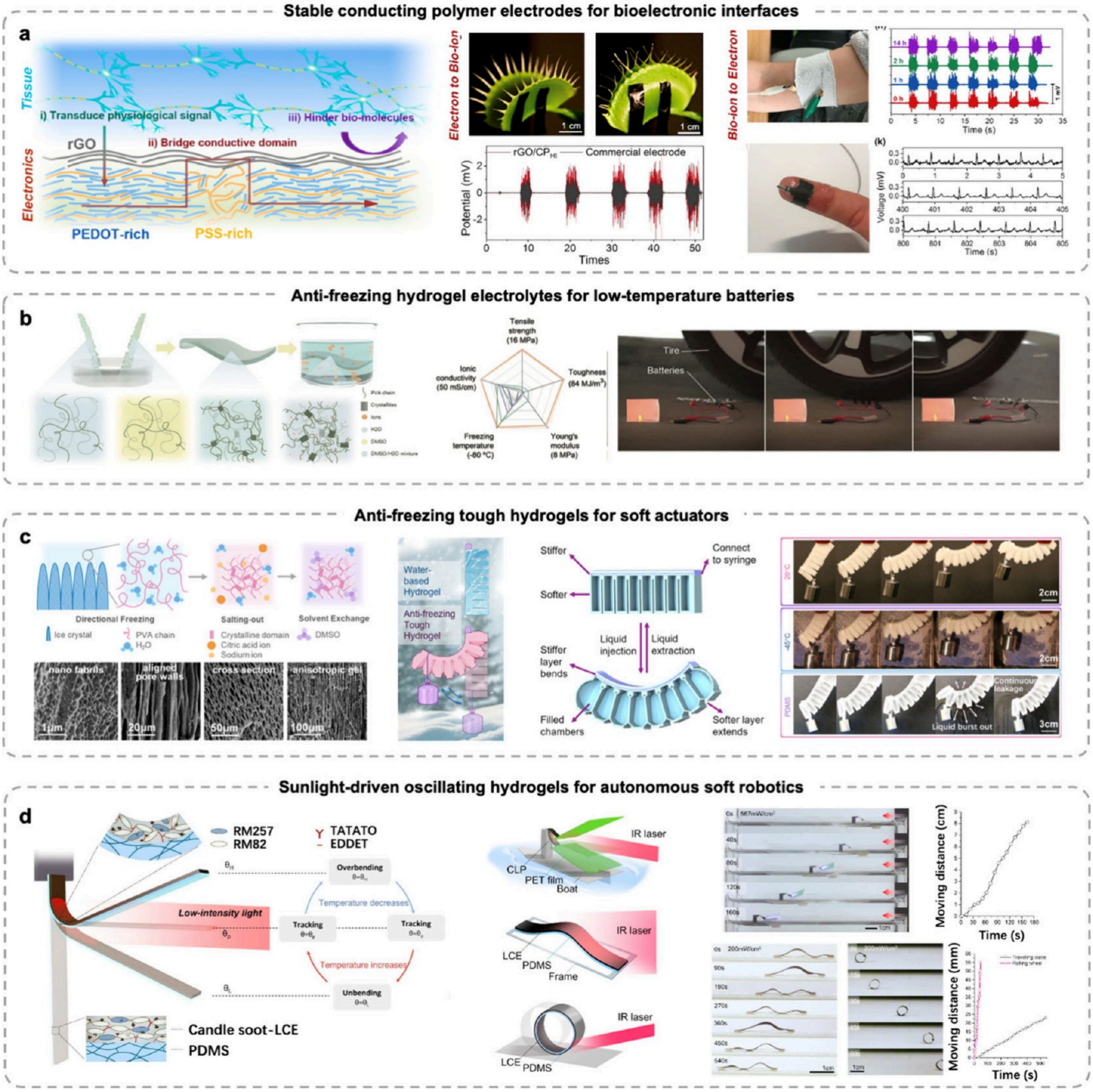

Figure 9. Applications of intelligent soft materials. (a) Stable conducting polymer electrodes for bioelectronic interfaces, showcasing the bilayer structure design principle and its impact on long-term stability in biological environments. Adapted with permission from ref 63. Copyright 2022 Elsevier Inc. (b) Antifreezing hydrogel electrolytes for low-temperature batteries, illustrating how engineered pore structures and ion−polymer interactions enable high performance in extreme conditions. Adapted with permission from ref 64. Copyright 2023 Wiley-VCH GmbH. (c) Antifreezing tough hydrogels for soft actuators, demonstrating how directional freezing and salting out techniques create materials with exceptional low-temperature mechanical properties. Adapted with permission from ref 65. Copyright 2021 Elsevier Inc. (d) Sunlight-driven oscillating hydrogels for autonomous soft robotics, highlighting the integration of photoresponsive and passive materials to achieve self-sustained motion from constant illumination. Adapted with permission from ref 50. Copyright 2023 AAAS.

compressive stress and triggering a reverse snap-through back into the acidic region. This chemo-mechanical coupling, combined with the spatial separation of opposing stimuli, creates a negative feedback loop that sustains autonomous oscillations at 0.5 Hz without external timing control. The key insight is that the slow chemical diffusion acts as an energy pump, while the fast structural instability serves as a mechanical switch, converting steady chemical gradients into periodic mechanical motion.

Recently, we achieved a major advance in power output by developing FLaPTOR (Figure 7d)[53]—an antagonistic photo-oscillator inspired by insect flight muscles. This system uses a trilayer structure with two photoactive liquid crystalline elastomer (LCE) layers sandwiching an inactive PDMS layer. Unlike conventional oscillators that only actuate in one direction, FLaPTOR creates bidirectional actuation: when one LCE layer contracts under light, the opposite layer relaxes, producing a push−pull mechanism like biological muscle pairs. Combined with the self-shadowing feedback mechanism, this design generates autonomous oscillations with an output power density of 33 W kg^{-1}—comparable to insect flight muscles and 275 times higher than conventional configurations. The system has achieved a thrust-to-weight ratio of 0.32 in flapping wing applications, representing significant progress toward autonomous aerial microrobots. Together, these self-oscillating systems show how integrating feedback mechanisms within materials enables autonomous behaviors for applications ranging from chemical sensing to aerial robotics.

4. ADDITIVE MANUFACTURING METHODS FOR FUNCTIONAL AND INTELLIGENT MATERIALS

The fundamental material processing strategies we explored previously, such as controlled molecular aggregation and engineered pore structures, have laid the groundwork for advanced additive manufacturing techniques. These methods provide unprecedented control over material properties and structures across multiple scales, facilitating the development of complex, multifunctional devices. In this section, we discuss how these principles extend to 4D hydrogel printing, room temperature metal printing, and liquid metal patterning, showcasing the versatility and broad applicability of our approach.[57−61]

Our 4D hydrogel printing builds directly on our understanding of stimuli-responsive hydrogels and structure−property relationships (Figure 8a).[58,59] This technique combines digital light processing (DLP) 3D printing with the thermoresponsive properties of hydrogels like poly(*N*-isopropylacrylamide) (PNIPAAm). By carefully controlling the composition and cross-linking density of the hydrogel layers, we can program specific deformation behaviors into printed structures. For example, we created a "pumpkin" structure that morphs from a sphere (diameter ∼ 2 cm) to a complex, buckled form when heated from 25°C to 45°C over 30 min. This shape change is driven by the same principles of polymer chain collapse and water expulsion that we explored in our earlier work on responsive hydrogels.

We developed complementary metal printing techniques that leverage UV-triggered processes for flexible electronics. Our room temperature gold printing (Figure 8b)[60,62] uses anion-assisted photochemical deposition, where Au^{3+} ions combine with photoinitiators and anions (e.g., Cl^{-}) to form conductive patterns without high-temperature annealing. By adjusting chloride concentration (0.05−0.3 M), we tune conductivity from 105 to 107 S/m and achieve feature sizes down to 15 μm. For applications requiring extreme deformability, we adapted projection lithography for liquid metals (Figure 8c),[57] suspending gallium-based particles (∼2 μm) in photocurable resin. UV exposure selectively cures the resin to trap particles, which after mechanical sintering form continuous pathways with 20 μm resolution and 700% stretchability. Together, these techniques provide a versatile platform for creating conductive patterns—from high-resolution rigid features to ultrastretchable circuits—all through room-temperature processing compatible with flexible substrates.

Additionally, we developed direct on-scalp printing of PEDOT:PSS e-tattoos using microjet dispensing to overcome the hair barrier that prevents conventional e-tattoo transfer (Figure 8d).[61] High-velocity ink droplets (150 μm nozzle, 6 Hz) penetrate through hair to reach the scalp, while 5-axis robotic control ensures conformal deposition on curved surfaces. By formulating separate inks for electrodes (NaCl-doped, 9 k$\Omega\cdot cm^2$ impedance) and interconnects (DMSO/glycerol-enhanced, 83 S/cm conductivity), we achieved both electrical performance and mechanical compliance (>30% stretchability) in 30 μm thick films. This approach enables personalized EEG array fabrication in 15 min with electrodes maintaining stable impedance below 40 k$\Omega\cdot cm^2$ for over 20 h, eliminating manual placement and substrate transfer steps.

By developing these advanced additive manufacturing methods, we have obtained a comprehensive toolkit for fabricating functional and intelligent materials across multiple length scales and material classes. These techniques not only build upon our fundamental understanding of material processing and structure−property relationships but also enable the design of complex, multifunctional devices that combine the responsive behaviors of hydrogels with the electrical properties of metals.

5. APPLICATIONS OF INTELLIGENT SOFT MATERIALS

The soft materials we have developed through controlled molecular aggregation and engineered structures enable critical applications across bioelectronics, energy storage, and soft robotics. In bioelectronic interfaces (Figure 9a),[63] our bilayer conducting polymer electrodes—combining PEDOT-rich and PSS-rich domains with a reduced graphene oxide (rGO) molecular sieve—achieve efficient charge transport while maintaining exceptional stability in biological environments. These electrodes demonstrate low impedance (1 kΩ at 1 kHz), long-term stability (>1000 h in physiological conditions), and mechanical flexibility (30% strain tolerance without degradation), making them ideal for long-term wearable and implantable devices.

For extreme environment applications, we leveraged our understanding of ion−polymer interactions and structural engineering to develop antifreezing hydrogels that function at temperatures as low as −70 °C. As battery electrolytes (Figure 9b),[64] these materials maintain high ionic conductivity (0.42 mS/cm at −70 °C) through hierarchical pore structures (1−10 μm), enabling stable cycling (>1000 cycles) at −20 °C with 88% capacity retention. For soft robotics (Figure 9c),[65,66] the same antifreezing principle—combined with directional freezing and salting out—yields tough hydrogels with remarkable low-temperature mechanics: 8.5 MPa tensile strength and 1400% elongation at −5 °C. We demonstrated robotic grippers operating at −50 °C, showcasing how these materials enable both energy storage and actuation in environments where traditional soft materials fail.

Beyond static applications, we achieved autonomous behaviors through the sunlight-driven LiLBot system (Figure 9d)[50]—extending our oscillator concept to liquid crystal elastomers. This bilayer design couples photoresponsive LCE with passive elastomer, creating self-sustained oscillations (up to 4 Hz) under one-sun illumination (100 mW/cm^2) via built-in feedback loops. The system enables diverse autonomous motions including walking (2.7 mm/min), rolling, and synchronized wing flapping without external control. Together with our bioelectronic interfaces and extreme-environment materials, these autonomous systems demonstrate how fundamental advances in molecular control and structural engineering translate into transformative technologies across healthcare, environmental monitoring, and robotics.

6. OUTLOOK AND CONCLUSION

The field of intelligent soft materials has seen remarkable progress in recent years, driven by innovative processing strategies and novel material designs. In this Account, we have explored how fundamental insights into material behavior can lead to new technologies through the development of processing strategies including the Hofmeister effect and cononsolvency. These approaches have enabled unique combinations of mechanical strength, diffusion properties, responsiveness, and functionality in hydrogels. By applying these principles across different material systems and length scales, we have progressed from basic hydrogels to self-regulating systems and advanced manufacturing methods. While this journey has been fascinating, we recognize that realizing the full potential of intelligent soft materials remains in its early stages.

Looking forward, two concrete directions emerge from our work. First, extending hierarchical design from mechanical to functional integration: engineering each structural level (molecular−nano−micro) to simultaneously contribute to toughness, transport, and responsiveness. For example, integrating conductive networks at nanoscale while maintaining macroscale toughness via directional freezing could yield materials that are both structural and functional. The challenge is achieving synergy—where signal pathways enhance rather than compromise energy dissipation—mimicking how tendon collagen provides both load-bearing and mechanotransduction. Second, generalizing autonomous behavior from self-oscillation to self-optimization across all properties. Recent review and article related to mechanochemical work[67,68] shows materials can strengthen under repeated loading; future researches can extending this "stimulus history → feedback → reorganization → optimization" logic beyond mechanics. Imagine conductive hydrogels that redistribute electron pathways to minimize hotspots, ion networks that expand high-flux channels, or photonic structures that tune periodicities based on light history—all without external computation. The challenge is designing feedback loops that independently optimize different properties through dynamic molecular bonds. Together, these directions move from static design to materials that continuously improve through use, embedding intelligence within structure itself.

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Author Contributions

C. W. Z. and X. H. wrote and revised the original manuscript, and X. H. supervised this study. The manuscript was written through the contributions of all authors, and all authors have approved the final version of the manuscript.

Notes

The authors declare no competing financial interest.

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