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Structure Design, Performance Regulation and Multifunctional Applications of Ionogels

Yuhang Li

South China University of Technology, Guangzhou, China
2029909118@qq.com

Abstract. Ionogels have emerged as a class of advanced functional materials for flexible electronics owing to their unique integration of high ionic conductivity, wide electrochemical windows, and excellent mechanical flexibility. This review provides a systematic overview of recent progress in ionogels, covering material design principles, fabrication strategies, and performance regulation mechanisms. Particular emphasis is placed on the selection of ionic liquid/polymer systems, network structure engineering, and nanocomposite reinforcement approaches to achieve optimized conductivity, mechanical robustness, and environmental stability. The intrinsic relationships between microstructure and macroscopic properties are analyzed to elucidate ion transport mechanisms, mechanical enhancement strategies, and stability improvement methods. In addition, the diverse applications of ionogels in flexible sensors, energy storage devices, and biomedical systems are comprehensively discussed, highlighting their multifunctionality and adaptability. Despite these advances, several challenges remain, including high material cost, limited large-scale manufacturability, and long-term stability under harsh conditions. Future research directions are proposed, focusing on green synthesis, low-cost material design, and intelligent optimization assisted by data-driven approaches. This review aims to provide a comprehensive framework for the rational design and practical application of high-performance ionogels.

Keywords: Ionogels, Structure regulation, Flexible electronics, Energy storage

1. Introduction

Flexible electronics show revolutionary potential in health monitoring, human–machine interfaces and wearable devices, but large-scale industrialization is hindered by core material bottlenecks. Traditional conductive materials like metals have high conductivity but inherent rigidity, limiting dynamic deformation applicability. Carbon-based nanomaterials offer better flexibility but face challenges in controllable fabrication, interfacial stability and property tunability, restricting device reliability and lifespan. To address these issues, ionogels, the specific ionic liquid conductive gels formed by confining ionic liquids in polymer 3D networks via physical or chemical means and synergistically possessing ionic liquids' excellent ionic transport performance and polymers' mechanical deformability, have emerged as advanced flexible conductive materials. They combine ionic liquids' high ionic conductivity and wide electrochemical window with polymer networks' stretchability and mechanical robustness. Notably, ionogels are highly designable-their electrical, mechanical and interfacial properties can be tailored by tuning ionic liquid structures, polymer network parameters and incorporating functional nanofillers, meeting diverse application needs.

Despite significant progress, most studies focus on specific material systems or single applications, lacking systematic integration from material design to device applications. This review establishes a framework linking materials, properties and applications, surveys current ionogel research, clarifies microstructure-performance correlations and highlights future directions like green synthesis and intelligent design, providing theoretical and practical guidance for high-performance ionogel development.

2. Fabrication of ionogels

The performance of ionogels largely relies on rational material system selection and fabrication strategy optimization. This section focuses on ionic liquid/polymer matrix selection, network construction strategies, and emerging preparation technologies, to clarify the fundamental principles for ionogel design and fabrication. The main fabrication strategies and their corresponding characteristics are summarized in Table 1.

Table 1. Main types of ionic liquid gels categorized by fabrication strategies

Fabrication strategy	Representative material systems	Main characteristics	Advantages
Chemical methods	PVA/ionic liquid gels, electrolyte gels, ionic liquid–silica gels	Three-dimensional networks formed mainly through noncovalent interactions (e.g., hydrogen bonding, electrostatic interactions, hydrophobic interactions, and π–π stacking), typically without chemical crosslinkers	(1) Simple fabrication procedures; (2) high reversibility; (3) preservation of ionic liquid integrity; (4) tunable functionalities
Physical methods	Polyacrylamide/ionic liquid gels, UV-crosslinked PAA–IL gels, γ-ray–crosslinked PVA–IL gels	Three-dimensional networks formed via covalent crosslinking or in situ polymerization, in which ionic liquids are immobilized within chemically crosslinked networks	(1) High structural stability; (2) suppressed leakage of ionic liquids; (3) excellent mechanical performance; (4) high designability

2.1. Selection of ionic liquids and polymer systems

Ionogel electrochemical performance depends on ionic liquid (IL) conductivity and polymer compatibility. Imidazolium-based ILs suit high-performance gels for high conductivity; pyrrolidinium-based ILs with wider electrochemical windows and better thermal stability are ideal for high-voltage energy storage.

Long alkyl chain ILs phase-separate from nonpolar polymers but blend with polar matrices. Crosslinking mode matters: physical crosslinking (e.g., hydrogen bonding) enables self-healing/injectability but lower strength, chemical crosslinking offers enhanced robustness via covalent bonds.

2.2. Fabrication strategies for ionogels

Selecting suitable preparation methods for ionogels is critical to ensure uniform ionic liquid dispersion, regulate network structures, and enable device integration. These methods are diverse, primarily categorized into physical, chemical, and other approaches based on bonding nature and construction strategies.

Physical methods form ionogel networks via noncovalent interactions. Solvent exchange is simple but limited in polymer choice; in situ phase separation boosts performance without extra treatment. DES gels use low-cost, low-toxic solvents for stable, flexible low-temperature gels. Chemical methods build stable covalent networks: in situ polymerization yields uniform, mechanically favorable networks; radiation crosslinking is initiator-free, high-purity and controllable. The two-step method enables tailored functional ionogels. Block copolymer self-assembly adapts to 3D printing, while lithium salt-induced microphase separation balances mechanical strength, stretchability and ionic conductivity.

2.3. Performance regulation and mechanistic studies

Conduction mechanisms: Ionogel conductivity primarily stems from cation and anion migration in ionic liquids, categorized into two main mechanisms based on charge carrier nature.

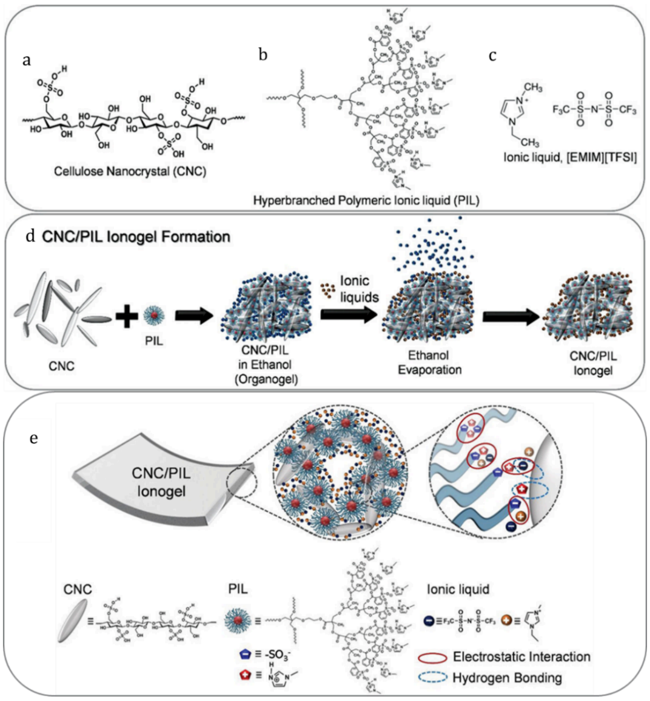

Figure 1. a–c) Structures of CNC, hyperbranched PIL, and [EMIM][TFSI]. d) Schematic of CNC/PIL ionogel formation. e) Formation mechanism via interactions among CNC, PIL, and ionic liquid

Ion migration-dominated mechanism: The ion migration-dominated mechanism is the most common and fundamental. Conductivity depends on free ion concentration and mobility in ionic liquids, following the Arrhenius or Vogel-Fulcher-Tammann equation and correlating closely with temperature. As illustrated in Figure 1, Georgia Tech researchers constructed a network from cellulose nanocrystals (CNCs) and polymerized ionic liquids (PILs), confining 95 wt% of [EMIM]

[TFSI]. Despite ionic liquid immobilization in the high-strength nanonetwork, continuous ion transport pathways were retained, yielding an ionogel with high conductivity (≈7.8 mS·cm-1, comparable to neat ionic liquid) and a compressive modulus of 5.6 MPa. Such conductivity is mainly governed by the ionic liquid's intrinsic properties and effective diffusion pathways within the gel [1].

Electron/ion synergistic mechanism: To For higher conductivity or added functionalities (e.g., electrochromism, sensing), conductive fillers (carbon nanotubes, graphene, PEDOT:PSS, etc.) are incorporated to form hybrid electron–ion transport pathways. As shown in Figure 2, a ternary composite electrode (graphene/ionogel/PEDOT:PSS) utilized electrostatic interactions between ionogel anions and PEDOT chains, plus π-π interactions between PEDOT and graphene-synergistically reducing sheet resistance to 30 Ω·sq-1 while retaining 88% optical transparency at 550 nm. Here, ions transport through the gel matrix and electrons through the filler network, with interfacial coupling enhancing overall electrical performance [2].

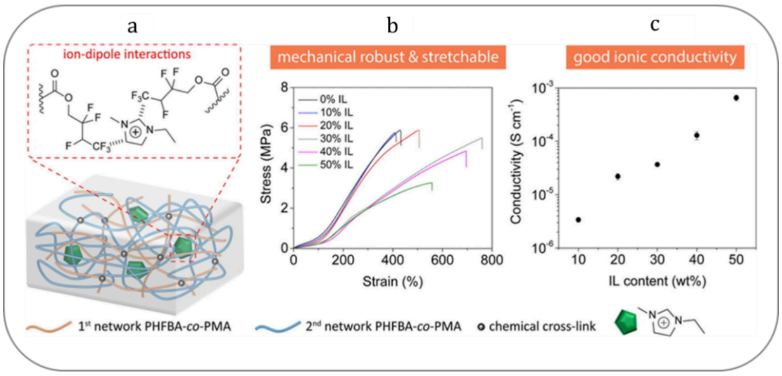

Figure 2. a) Schematic of DN ionogel structure and ion–dipole interactions between P(MA-co-HFBA) and imidazolium cations. b) Mechanical properties: tensile stress–strain curves at varying EMITFSI content. c) Ionic conductivity versus IL content

Optimization of Mechanical Properties: High toughness and high ionic conductivity are often contradictory-high ionic liquid content plasticizes polymer networks, degrading mechanical performance. Various reinforcement strategies have been developed to address this.

Network structure design: Dynamic or reversible physical and chemical crosslinking points can be introduced into polymer networks as sacrificial bonds. These bonds break preferentially under external force to dissipate energy, which significantly improves gel toughness and fracture energy. The main network remains intact and can recover after the stress is removed. Zhu and coworkers synergistically strengthened ionogels by double network structure and ion-dipole interactions. Strong ion-dipole interactions were constructed between -CF_3 dipoles on polymer side chains and imidazolium cations in EMITFSI ionic liquid. These interactions acted as physical crosslinks and connected the two network layers, endowing ionogels with high stretchability, high strength and stable conductivity. Dynamic covalent bonds including boronate ester bonds and disulfide bonds also provide ionogels with outstanding self-healing performance [2].

Nanocomposite reinforcement: Introducing nanoscale reinforcements (cellulose nanocrystals, clay, silica) enhances gel modulus and strength through large specific surface area and high rigidity. Georgia Tech developed interconnected nanocomposite networks using cellulose nanocrystals and hyperbranched polymeric ionic liquids, stabilizing ionic liquids and achieving a 5.6 MPa compressive modulus. As demonstrated in Figure 3, Yan et al. prepared physically crosslinked ionogels via metal–organic framework coordination, which exhibited exceptional mechanical performance with 11 000% strain, 58 MPa modulus, 25 MJ·m-3 toughness and 125 kJ·m-2 fracture energy [3].

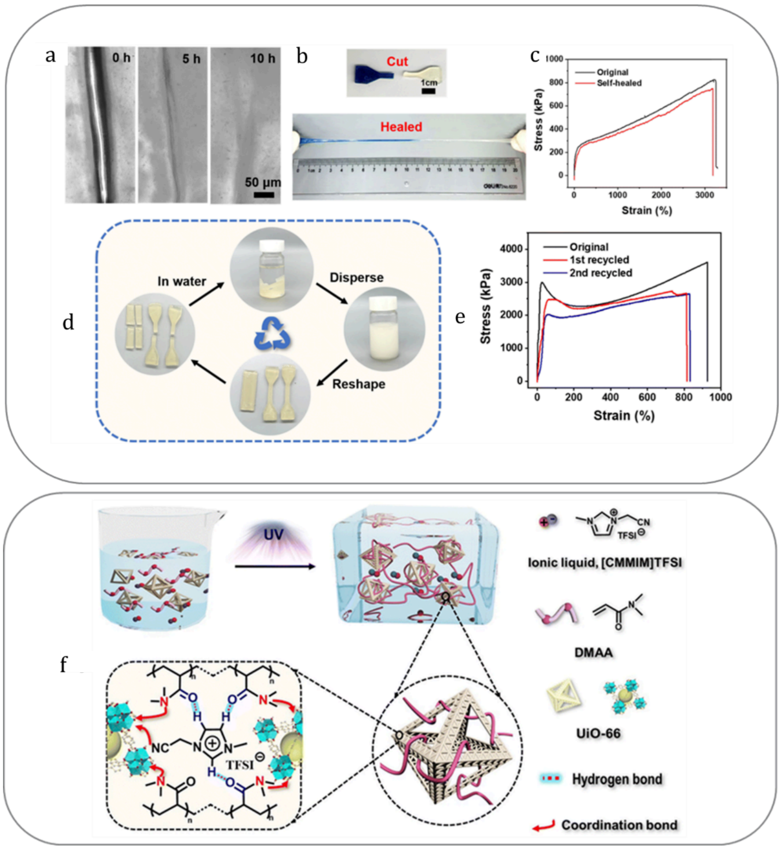

Figure 3. Self-healing and recyclability of UiO-66 ionogels. a) Optical images of cut samples during healing at different times. b) Demonstration of stretchability. c) Stress–strain curves of various samples. d) Schematic of recyclability. e) Uniaxial tensile curves after different recycling cycles. f) Schematic of UiO-66 ionogel preparation

Enhancement of Environmental Stability: In practical applications, the long-term stability of ionogels in complex environments determines their potential for large-scale industrial use. Hydrophobic modification and self-healing design are important strategies to improve environmental stability. The environmental stability of ionogels under various conditions is summarized in Figure 4.

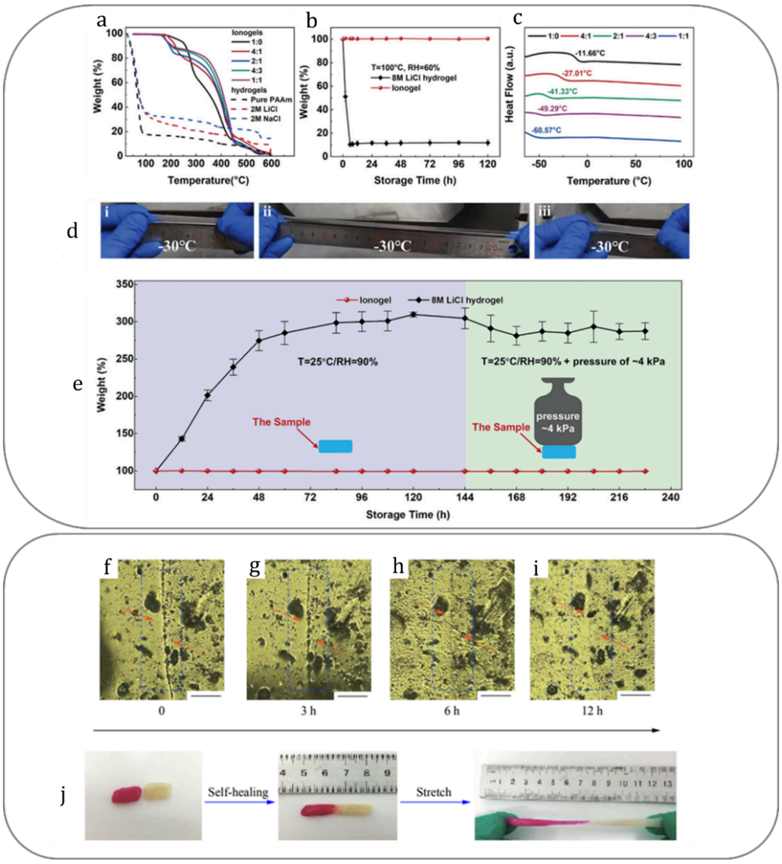

Figure 4. Stability of the ionogels. a) TGA curves at different IL contents, showing decomposition temperatures >200 °C. b) High-temperature tolerance. c) DSC curves; Tg decreases from −11.66 to −60.57 °C with increasing IL content (1:0 to 1:1). d) Ionogel (IL 4:3) remains elastic at −30 °C: (i) original, (ii) stretched ≈4×, (iii) recovery. e) Ambient and mechanical stability; stable weight at 90% RH under ≈4 kPa compression. f–i) Optical images of healing for ionic/DCB gel at 30 °C over time (scale bar: 50 μm). j) Photographs of self-healing behavior

Hydrophobic modification to suppress water evaporation: Conventional hydrogels and hydrophilic ionogels are easily affected by environmental humidity. They tend to dry out or swell with moisture, resulting in fast performance degradation. Hydrophobic modification of gel components is an effective method to enhance environmental stability. Jia et al. synthesized ionogels using hydrophobic ionic liquid $[C_2mim][NTf_2]$ and acrylate copolymer networks including P(MEA-co-IBA) [4]. The fully hydrophobic character enables the ionogel to absorb negligible water at 90% RH and maintain stable weight over time, while conventional hydrogels suffer obvious swelling or dehydration. Hydrogen bonds between polymer chains and ionic liquids lock the ionic liquid tightly in the matrix, avoiding leakage under long-term mechanical loading and ensuring outstanding mechanical stability. Fluorinated ionic liquids or polymers can further strengthen hydrophobicity.

Self-healing functional design: Self-healing capability can greatly extend material service life and enhance device reliability and durability. This function is usually realized by introducing dynamic chemical bonds into gel networks. Ionogels based on dynamic covalent bonds and electrostatic interactions have been widely reported. Dynamic bonds in these gels can dissociate and reorganize reversibly after damage, allowing autonomous recovery of mechanical and electrical properties. The self-healing design suppresses micro-damage during service and serves as an effective way to improve long-term stabilit [5].

Through Table 2, the specific differences in type and performance of representative Ionogels can be compared, providing a reference for understanding design rules or selecting candidate material systems for specific applications.

Table 2. Key properties and structural design of representative ionogels

Ionic Liquid Type	Polymer Network Construction Strategy	Key Performance Indicator	Stability Indicators
[EMIM] [TFSI]	CNC + hyperbranched PIL, multiple non-covalent crosslinking	Ionic conductivity: 5.4~7.8 $mS\cdot cm^{-1}$ at 30°C; Compressive strain: 75%; Compressive elastic modulus: 0.6~5.6 MPa; Compressive yield strength: 55~260 kPa	Thermal stability: Initial decomposition ≈370°C, operating temperature range 163~383 K; Leakage resistance: No obvious IL leakage
[EMIM] [TFSI]	MA/HFBA as monomers, EGDMA as crosslinker, photo-polymerization to construct double network	Ionic conductivity: 10^{-4}~10^{-5} $S\cdot cm^{-1}$; Tensile strain: Up to 784% (30 wt% IL); Tensile strength: ≈5.6 MPa;	Thermal stability: Stable below 300°C; Long-term stability: No mass loss for 1 month; Leakage resistance: No obvious IL leakage
[CMMIM] [TFSI]	DMAA as monomer, UiO-66 as crosslinking point, photo-polymerization to form physically crosslinked network	Tensile strain: Up to 11000% (90 wt% IL); Young's modulus: Up to 58 MPa (50 wt% IL); Fracture energy: 125 $kJ\cdot m^{-2}$	Thermal stability: Stable below 250°C; Cycle stability: Maintains elasticity after 20000 compression cycles; Self-healing property: Healing efficiency ≈99% in 12 h at room temperature; Recyclability: Remoldable after aqueous dispersion
$[C_2mim]$ $[NTf_2]$	MEA/IBA copolymerization to form single network, PEGDA as crosslinker, prepared by photo-polymerizatio	Ionic conductivity: 1.69×10^{-5}~4.27×10^{-4} $S\cdot cm^{-1}$; Tensile strain: >2000% (maximum 2580%); Young's modulus: 20~102 kPa; Tensile strength: 125~752 kPa	Thermal stability: Stable below 200°C in air and 250°C in nitrogen Environmental stability: No mass change for 144 h at 90% RH; Mechanical stability: No IL leakage under 4 kPa pressure; Cycle stability: Maintains functionality after 200 tensile cycles
DDPMIMCl	Copolymerization of 4-VPBA and AMPSL	Ionic conductivity: 0.59 S/m at room temperature; Tensile strain: ≈570%; Tensile strength: ≈99.4 kPa; Self-healing efficiency: 77.4% at 30°C, 84.1% at 60°C	Thermal stability: Mass remains basically unchanged at 100~200°C; Self-healing property: Cracks heal in 12 h at room temperature, healing efficiency improves at 60°C;

3. Applications of ionogels

3.1. Flexible electronic devices

In flexible electronics, ionogels are mainly used in strain sensors and flexible circuits. As strain sensors, their key advantages include high sensitivity, wide strain range and fast response. The Shaanxi University of Science and Technology team developed a PAM/PBA-IL/CNF hydrogel sensor. Enabled by dynamic boronate ester bonds and hydrogen-bond networks, it exhibits excellent sensing performance: gauge factor up to 8.36, strain range up to 1810%, response time ~10 ms, and detection of tiny strains as low as 0.2%, enabling precise monitoring of human pulse to joint bending [6]. For flexible circuits, ionogels act as stretchable conductors and transparent electrodes. Graphene/ionogel@PEDOT:PSS composite electrodes show low sheet resistance (30 Ω·sq-1), high optical transmittance (88% at 550 nm) and good mechanical flexibility, making them ideal for high-performance flexible transparent circuits [7].

3.2. Energy storage devices

In energy storage, ionogels as electrolytes boost the performance of supercapacitors and solid-state batteries. For supercapacitors, they offer wide voltage windows and excellent cycling stability. A study reported a zwitterionic ionogel electrolyte (GPE-MPC) via solvent-free UV crosslinking. 2-methacryloyloxyethyl phosphorylcholine (MPC) promoted EMIMTFSI ionic liquid dissociation, achieving 1.66 mS/cm ionic conductivity. Solid-state supercapacitors with this electrolyte delivered 3.0 V operating voltage, 30.4 Wh/kg energy density and stable performance over 10,000 cycles. This solves solid electrolytes' low ionic conductivity with an eco-friendly solvent-free fabrication [8].

3.3. Biomedical applications

In the biomedical field, ionogels are ideal for bioelectrodes and wearable health-monitoring devices due to biocompatibility and electrical performance. A study developed a highly conductive underwater-stable ionic skin with polyurethane and [EMIM][TFSI]. Fluorine–cation interactions between polyurethane's fluorine-rich segments and the ionic liquid gave it high conductivity, good mechanical properties and long-term underwater stability. Electrodes based on this material stably monitor epidermal bioelectrical signals underwater and daily environments, offering a reliable solution for all-weather multi-scenario wearable medical monitoring [9].

3.4. Other applications

Ionogels have wider applications in many frontier interdisciplinary fields. In electromagnetic wave management, their absorption properties can be tuned in real time by stretching the gel to adjust internal structures, showing great potential for smart stealth and compatible electronics. In advanced manufacturing and robotics, ionogels can be 3D-printed into complex structures and used as artificial muscles to actuate soft robots. Their light weight and high strength also make them promising candidates for aerospace and robotic applications.

4. Conclusion and outlook

Ionic conductive gels are key materials for flexible electronics with high conductivity, wide electrochemical windows and good mechanical tunability. They still face two major challenges: cost and stability. High production cost limits large-scale application, which can be relieved by biomass-based synthesis and low-content high-performance designs. These materials degrade easily under extreme temperatures and humidity. Related solutions include optimizing network interactions, selecting suitable polymers and using hydrophobic components. Future development will focus on green synthesis and intelligent design. Green synthesis uses renewable materials to reduce environmental impact. Intelligent design applies AI to predict performance and optimize formulas. With multidisciplinary cooperation, ionic conductive gels will move from lab research to industrial application

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