



Ionic Gels and Their Applications in Stretchable Electronics

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Ionic gels represent a novel class of stretchable materials where ionic conducting liquid is immobilized in a polymer matrix. This review focuses on the design of ionic gel materials and device fabrication of ionic-gel-based stretchable electronics. In particular, recent progress in ionic-gel-based electronic skin (pressure/strain sensors, electric double-layer transistors, etc.), flexible displays, energy storage devices, and soft actuators are summarized, followed by a discussion of challenges in developing ionic-gel-based electronics and suggestions for future research directions that might overcome those challenges.

1. Introduction

Stretchable electronics are mechanically bendable, foldable, and twistable, and can conform to nonplanar surfaces. They can be fabricated via two different approaches. One approach, pioneered by Zhenan Bao's research group at Stanford University, is compositing with intrinsically stretchable materials such as conductive polymers,^[1] carbon nanotubes (CNTs),^[2] graphene,^[3] silver (Ag) nanowires,^[4] and metal meshes.^[5] For instance, percolated networks of Ag nanowires have been widely used to fabricate flexible transparent electrodes, which exhibit fairly good tolerance to strain. Spray-deposited films of single-walled CNTs were successfully employed to make stretchable, skin-like tactile sensors.^[2] The other approach to achieve the stretchability of electronics, pioneered by John Rogers at UIUC and Yonggang Huang at Northwestern University, is making smart design of electrode geometries or structures.^[6] By fabricating wavy or buckled single-crystal silicon ribbons on an elastomeric substrate, the devices can be reversibly stretched and compressed without damaging the silicon ribbons, because the wavy or buckled silicon structure can accommodate strains by changing their shapes including wave amplitudes, wavelengths, and 3D layout. By integrating such wavy and buckled layout of conductors and semiconductors with nonstretchable electronic components, a new class of stretchable electronics was developed.^[7]

The rapid advancement of stretchable electronics facilitates the integration of stretchable devices into complex systems,^[8]

such as electronic skins (E-skin), wearable electronics, soft robotics, and implantable devices. In particular, E-skin, which replicates the sensory capabilities and mechanical stretchability of human skin, has attracted great academic interest in the last decade.^[9] As the largest organ in human body, skin consists of an integrated network of sensors that relay information about tactile and thermal stimuli to the brain. To realize the functionality of human skin, E-skin must be mechanically soft and stretchable, and be capable of translating physical information such as

pressure, temperature, humidity, and physiological messages into digital signals,^[10] mimicking the process of generating nerve signals from human touch.

Service robots, unlike industrial robots, need to be equipped with E-skins so that they can gain a "sense of touch" to feel the objects they handle, to understand the environment they are in, and to interact with humans safely.^[11] To date, various types of tactile sensors and motion-control sensors have been developed.^[12] Takao Someya and coworkers prepared a large-area E-skin by integrating pixel organic field-effect transistors (OFETs) and pressure sensors on PET substrates.^[12e] Ignoring the inadequate resolution, this integration proposed an ideal solution to realize a practical artificial skin by the active matrix readout electronics. Rogers et al. presented a camouflage system made of cephalopod-inspired flexible E-skin, which can spontaneously detect and match the coloration of surrounding areas.^[12f] Bao's group realized the highly sensitive flexible pressure sensor by using microstructured PDMS films, which exhibited unprecedented sensitivity as well as fast response and recovery times.^[12g] Thereafter, the strategies of delicate structural design, advanced material application, and novel assembly of components have been widely used for various E-skin devices.

Currently existing stretchable electronics mostly comprise intrinsically electrical conducting materials with insulating elastomeric substrates. However, the Young's modulus mismatch between hard electronic materials and soft substrates poses great challenges in applying stretchable electronics at large strains. To improve the performance of stretchable electronics, tremendous efforts have been devoted to developing new functional materials.^[13] The recent progresses of stretchable electronics have been reviewed in terms of materials, devices, and applications. For example, Bao et al. reviewed the developments of organic electronic materials in artificial skin.^[14] Lee et al. focused on the applications of intrinsically stretchable components in monitoring human activity and personal healthcare.^[15] Pei et al. concluded the applications of stretchable materials for soft sensors and artificial muscles.^[16] In contrast to the

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conventional sensing material, intrinsically soft, transparent, sensing materials are highly desired.^[17]

Ionic gels have polymer networks swollen with electrolyte solutions or ionic liquids. They take the conducting solid form, but with great stretchability and transparency. Although ionic gels are ionic conducting and electronic insulating, they present a great potential in stretchable devices. Electric double layers (EDL) are formed at contact interfaces between electrode and ionic gels, which can be considered as nanometer-thick capacitors with a capacitance above $1 \mu\text{F cm}^{-2}$.^[18] On application of a low direct current (DC) voltage ($\approx 1 \text{ V}$) to a hybrid circuit of ionic and electronic conductors in series, a sustained electrical current do not occur because ions and electrons do not across the electrode and gel interfaces in the absence of electrochemical reaction. For a hybrid circuit of ionic gels–dielectrics–metals in series, EDL capacitor has a large capacitance compared to the dielectric capacitor. Consequently, the additional voltage crossing interfaces of metal electrode and ionic gel (EDL) is much smaller than the dielectric layer. As long as voltage of EDL is within a certain range (1 V for water), the hybrid circuit is able to transmit electrical signals even under voltages and frequencies much higher than the electrochemical window.

Although electrical resistance of ionic gels is larger than conventional electronic materials resulting from the heavier weights and slower migration rates of ions than electrons, they present less change in electrical resistance when being stretched. Very uniquely, under large strains, the resistance change of ionic gels is much lower than traditional electronic conductors.^[19] Therefore, ionic gels stand as alternative functional materials for stretchable devices, and have demonstrated successful applications in E-skins,^[20] energy storages,^[21] light-emitting devices,^[22] actuators,^[23] and all sorts of sensors^[24] (Figure 1).

In this review, we introduce the advantageous characteristics of ionic gels as superior materials for stretchable electronics, and describe the preparation of ionic gels via chemical and physical approaches. We then summarize the recent progress of ionic-gel-based stretchable devices, including pressure/strain sensors, EDL transistors, displays, energy storage devices, and actuators. Finally, we summarize the challenges in developing ionic-gel-based devices and outlook future research directions that might overcome those challenges.

2. Ionic Gel Materials

Ionic gels, consisted of polymer networks and electrolyte solutions, are attracting considerable attention by virtue of their high stretchability, mechanical conformality, good transparency, and biocompatibility. According to the types of solvents in polymer networks, ionic gels can be categorized into aqueous ionic gels and nonaqueous ionic gels.

2.1. Aqueous Ionic Gels

Aqueous ionic gels, such as hydrogels, can be realized through polymerization of vinyl monomers in electrolyte solutions in the presence of crosslinkers and initiators.^[34] Hydrogels



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have been extensively researched with improved mechanical behavior and employed in various applications. One of the most prominent examples is polyacrylamide (PAM)-based hydrogel, which is formed by in situ polymerization of acrylamide (AAM) monomer in electrolyte solutions (such as NaCl, KCl, and LiCl). Past researches have demonstrated that the PAM-based hydrogels can be applied in transparent loudspeakers,^[19] skin-like and self-healable tactile sensors,^[35] actuators,^[36] and touch panels.^[37] However, evaporation of water from the hydrogels deteriorates the stability and lifetime of devices, hence limiting their applications.

Since the pioneering work on double-network hydrogels by Jianping Gong,^[34d] researches on high mechanical strength hydrogels have made tremendous breakthrough. In 2012, Zhigang Suo reported highly stretchable and tough hydrogels fabricated via combining weak and strong crosslinks.^[34a] They designed a hybrid of alginate and PAM based on ionically

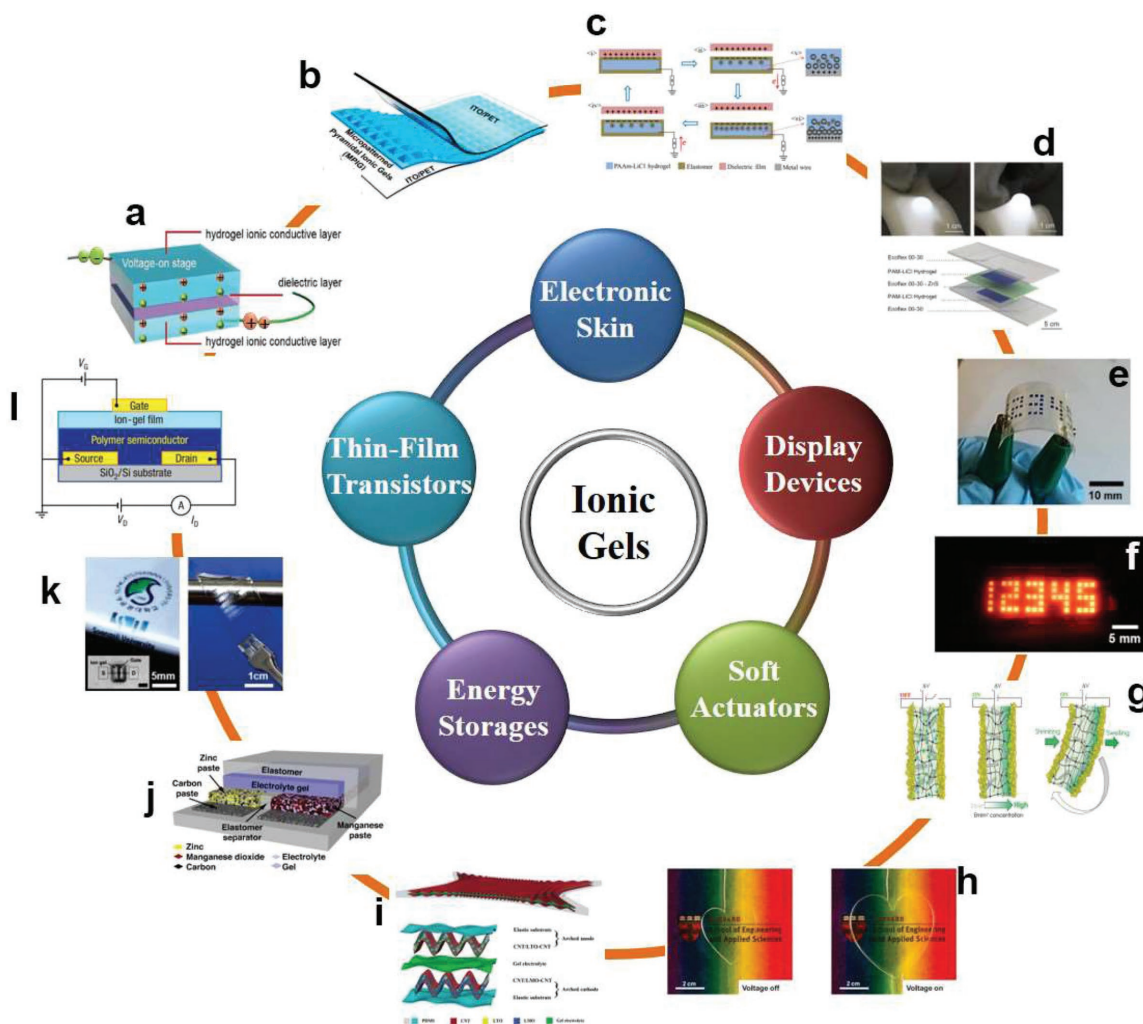


Figure 1. Perspective applications of ionic-gel-based self-healing sensors. Reproduced with permission.^[25] Copyright 2017, Wiley-VCH. **b**) Micropatterned ionic-gel-based sensors. Reproduced with permission.^[24] Copyright 2017, American Chemical Society. **c**) Soft tactile sensors of triboelectric nanogenerator. Reproduced with permission.^[26] Copyright 2017, AAAS. **d**) Ionic-gel-based light-emitting capacitive touch sensor and displays. Reproduced with permission.^[27] Copyright 2016, AAAS. **e**) Flexible electrochromic devices. Reproduced with permission.^[28] Copyright 2015, American Chemical Society. **f**) Flexible electrochemiluminescent devices. Reproduced with permission.^[22] Copyright 2014, American Chemical Society. **g**) Ionic-gel-based electroactive soft actuators. Reproduced with permission.^[29] Copyright 2017, Wiley-VCH. **h**) Ionic-gel-based dielectric elastomer actuator. Reproduced with permission.^[19] Copyright 2013, AAAS. **i**) A super-stretchy battery. Reproduced with permission.^[30] Copyright 2015, Wiley-VCH. **j**) Ultracompliant electrochemical dry cells. Reproduced with permission.^[31] Copyright 2010, Wiley-VCH. **k**) Stretchable graphene transistors. Reproduced with permission.^[32] Copyright 2011, American Chemical Society. **l**) Ionic-gel-gated transistors. Reproduced with permission.^[33] Copyright 2008, Macmillan Publishers Limited.

and covalently crosslinked polymer networks (**Figure 2**). With a remarkably giant fracture energy of about 9000 J m^{-2} , the hydrogels exhibited high stretchability ($>2000\%$) and extremely notch insensitivity (stretch of 1700% with a notch). They further developed a two-step method to synthesize alginate/PAM hydrogels crosslinked by multivalent cations (e.g., Zn^{2+} , Al^{3+} , Fe^{3+}), leading to improved mechanical toughness and stretchability.^[38] In the hybrid-component hydrogels, both ionic and covalent bonds are essential to dissipate energy under stretching. The effective energy dissipation can be attributed to the hybrid distribution of bonding strengths: the covalent bonds serve as strong and permanent crosslinks to ensure high modulus, whereas the ionic bonds behave as reversible sacrificial bonds to ensure high energy dissipation.^[39]

To achieve mechanically tough ionic gels and address the issues of poor stretchability and recyclability of ionic gels, mechanisms to dissipate mechanical energy mainly contain fracture of polymer chain, reversible crosslinking of polymer chains, transformation of configurations in polymer chains.^[40] The reversible crosslinking is driven by physically or chemically dynamic interactions, such as hydrogen bonds, ionic interaction, metal–ligand interactions, hydrophobic interactions, and reversible covalent bonds. Zheng et al. developed a stiff and tough hydrogel with randomly dispersed acrylamide and acrylic acid repeating units, which was further physically crosslinked by coordination bonds between ferric ions and carboxylic groups of the copolymers.^[41] The reversible bonds of metal–ligand interactions as well as the dynamic hydrogen

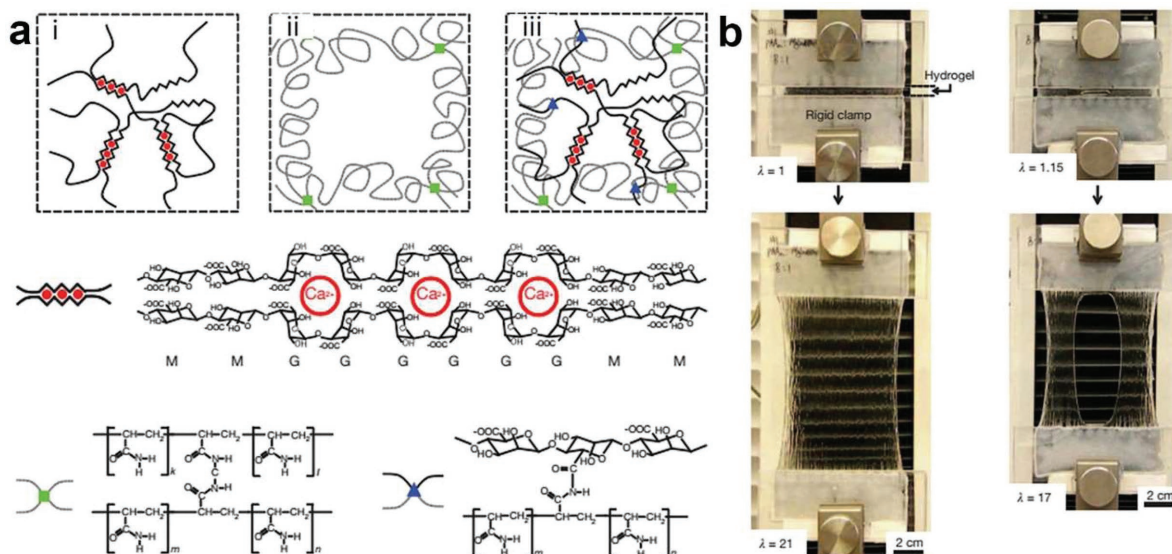


Figure 2. Highly stretchable and tough hydrogels. a) Schematics of intertwined hybrid hydrogel networks (iii) ionically crosslinked alginate networks (i) and covalently crosslinked polyacrylamide networks (ii); b) Hybrid hydrogels exhibited highly stretchable (stretch to 21 times its initial length) and notch-insensitive (stretch to 17 times its initial length). Reproduced with permission.^[34a] Copyright 2012, Macmillan Publishers Limited.

bonding between carboxyl and amino groups provided recoverable energy dissipation networks, which endowed the hydrogels with high toughness and good recyclability. By varying the water content, concentration of metallic ions, and pH values of the solutions, the energy dissipation networks can be tuned to obtain various mechanical performance of hydrogels.

Proteins represent the other type of polymer, which have large amounts of functional ligand groups for reversible coordination with metal ions. The protein-based hydrogels primarily employ the interactions of structured domains, such as coiled coils, titin-like motifs, and folded fibrin, to dissipate mechanical energy, so they exhibit extremely high mechanical strength and toughness.^[39] Recently, hybrid polymer networks formed by both covalent crosslinkable protein and metal crosslinkable protein have been developed to facilitate effective energy dissipation. Hydrogels containing unstructured proteins were reported to obtain extraordinary mechanical properties with extremely high strength (>2.5 MPa), toughness (>1300 J m⁻²), excellent stretchability (>500%), and good self-adhesion.^[42]

To achieve tough hydrogels with both high strength and deformability, new approaches have been developed to obtain highly effective energy dissipation networks, such as hybrid networks, nanocomposites, and dual crosslink networks. Hu et al. designed a hybrid network hydrogel with superb mechanical properties.^[43] The copolymerization of *N*, *N*-dimethylacrylamide and methacrylic acid leads to the formation of hybrid structures, which contain polymer-rich hydrogen-bonded networks and relatively loose and dilute polymer networks (Figure 3). The hybrid structures were mediated by the multiple intermolecular hydrogen bonding and hydrophobic interactions, respectively. Based on energy dissipation networks such as loose chemical covalent bonds network and dense clusters of sacrificial hydrogen bonds networks, the resulting hydrogels showed excellent mechanical performances with high modulus of 28 MPa and high extensibility of 800%. Compositing with

nanomaterial fillers is another strategy to enhance the mechanical property of polymer hydrogels.^[44] Wu et al. utilized biomineralization process to obtain bioinspired ionic gels, which showed excellent self-healability, mechanical compliance, and biocompatibility.^[25] The ionic gels were prepared by physically crosslinking calcium carbonate nanoparticles with networks of polyacrylic acid (PAA) and alginate. The slightly dissolved Ca²⁺ from calcium carbonate served as ion carriers, and no other salts were added in the process.

Ionic gels are one of the most promising soft conductors for E-skins, implantable and wearable biosensors in human health monitoring due to their biocompatibility, biodegradability, and mechanical similarity with skin. Hybrid with solid substrates has further extended the application of ionic gels in stretchable bioelectronics, microfluidics, soft robotics and biomedical devices. Briefly speaking, tough bonding between hydrogels and solid substrates (metals, glass, and polymers) is essential for their integration in devices and systems. Although tough and conductive hydrogels have been developed, the weak interfacial bindings between hydrogels and other materials severely limit their integration and multifunctionality. Zhao's group reported a strategy to anchor the polymer-network hydrogels to solid surfaces based on covalent chemical anchorage (Figure 4).^[45] Robust hybrid circuit of ion/electron conductors were prepared by sandwiching tough PAM hydrogel chemically anchored between two titanium electrodes. The interface between ionic gels and metal electrodes is conductive enough to power an LED light. They also reported a versatile method to integrate hydrogels and elastomers into hybrid structures,^[46] which exhibited extremely tough interfacial bonding (1000 J m⁻²) and functional microstructures. This simple yet versatile method is composed of three steps: pre-shaping elastomers and hydrogels, elastomer surface modification for hydrogel grafting, and covalent crosslinking of hydrogel and elastomer. Recently, Mooney group reported the design of two-layer tough adhesives, which

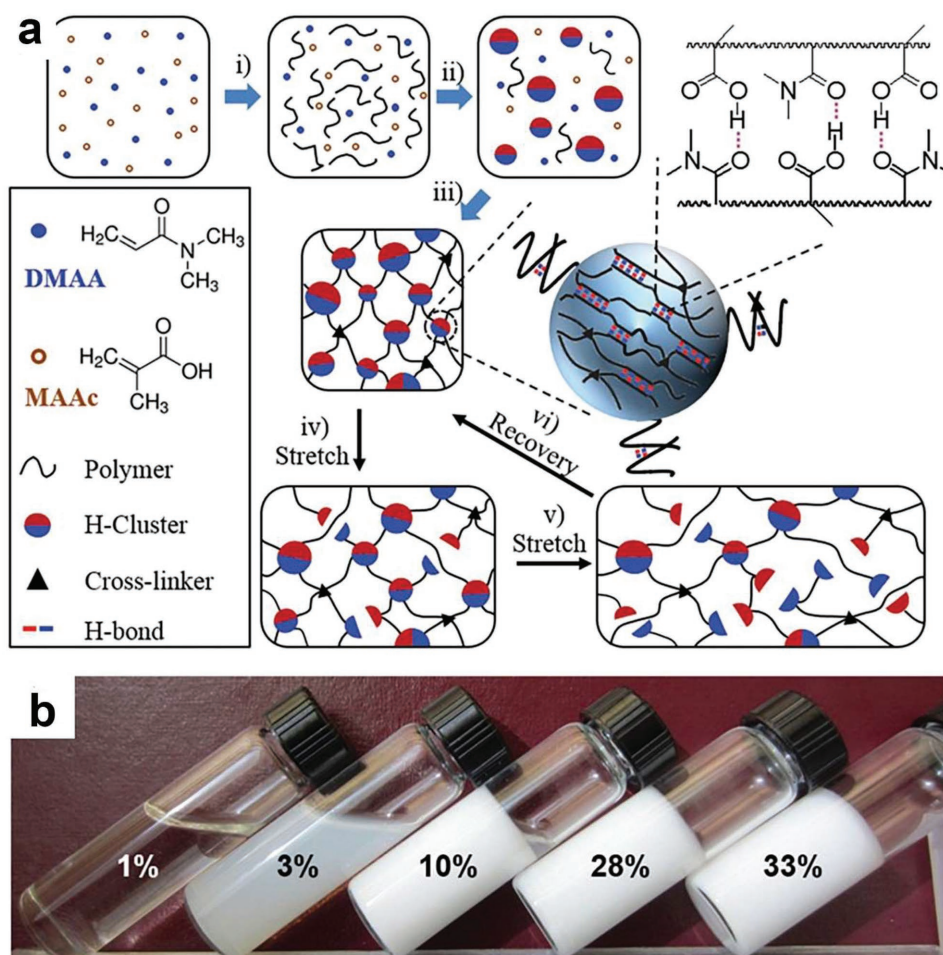


Figure 3. Hybrid network hydrogel. a) Schematic illustration of hydrogels by copolymerization of *N,N*-dimethylacrylamide and methacrylic acid (i–iii) and a proposed molecular mechanism of the stretching and recovery processes based on the proper balance between loose chemical covalent bonds networks and the multiple sacrificial hydrogen bonding networks (iv–vi). b) Photographs of the hydrogels with different total monomer weight concentration. Reproduced with permission.^[43] Copyright 2015, Wiley-VCH.

consisted of a positively charged polymer adhesive surface and a tough energy dissipative matrix.^[47] The adhesive surface exhibited tough bonding to the wet substrate through electrostatic interactions, covalent bonds, and physical interpenetration, whereas the tough matrix dissipates energy through hysteresis under deformation. This exciting research presented a promising route to achieve ionic gel hybrids via tough bonding between ionic gels and solid substrates.

2.2. Nonaqueous Ionic Gels

Nonaqueous ionic gels are usually prepared by swelling polymer networks in organic electrolyte solutions and have physical interactions in polymer networks.^[48] The earliest ionic gels were mainly made from the composites of poly (ethylene oxide) (PEO) and lithium salts.^[49] However, they had poor ionic conductivity (10^{-5} S cm^{-1}) attributed to the low mobility of lithium ions at room temperatures. As an alternative, the physically assembled ionic gels with ionic liquids have been

widely investigated.^[50] Ionic liquids, a class of room temperature molten salts, have a wide range of appealing properties, including negligible volatility; nonflammability; exceptionally thermal, chemical, and electrochemical stability; and excellent ionic conductivity. Therefore, nonaqueous ionic gels with ionic liquids have good ionic conductivity, high mechanical strength, optical transparency, and high flexibility. These properties make ionic gels viable for applications such as electrolyte-gated transistors,^[51] gas separation membranes,^[52] and energy storage.^[21] Besides, polyelectrolytes, comprising of immobile covalently crosslinked charged polymer networks and mobile counter ions, represent a unique type of ionic gels.^[53] Watanabe et al. reported a series of polymer electrolytes based on in situ free radical polymerization of compatible vinyl monomers at room temperatures.^[54] Nonaqueous ionic gels can be processed via various techniques such as dip-casting, spin-coating, inkjet printing, and transfer printing. However, preparation of these ionic gels needs a large amount of harmful organic solvents, which is not favored in biomedical applications.

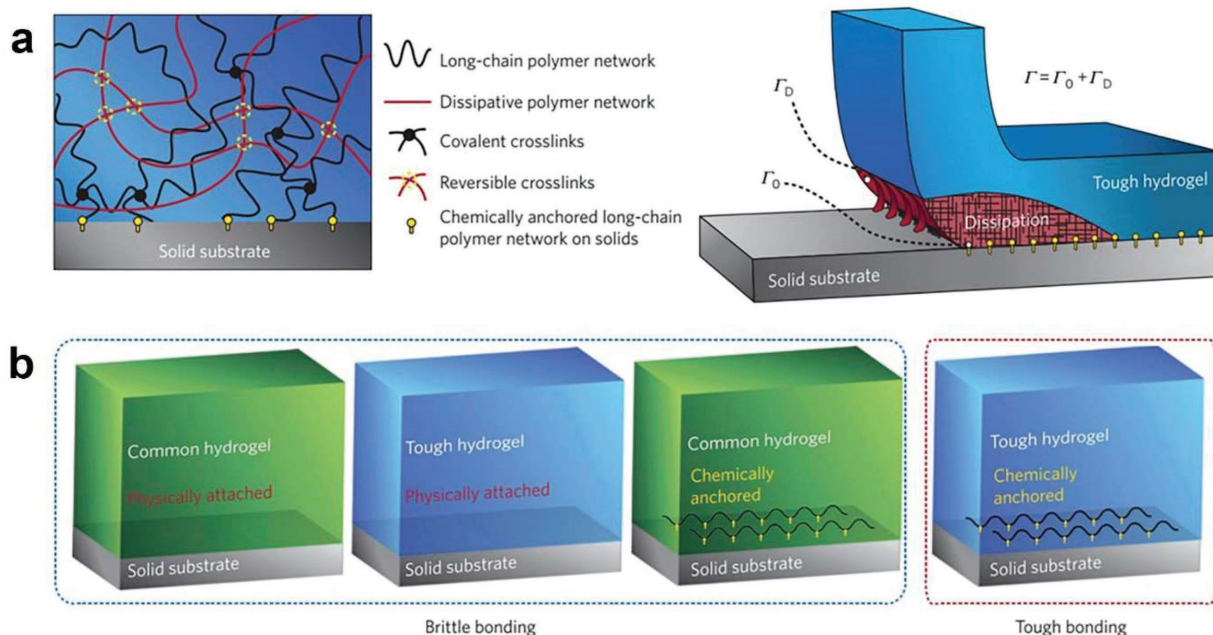


Figure 4. Design strategy for anchoring the polymer-network hydrogels to solid surfaces. a) Schematic illustration of the covalently chemical anchors. b) The design of tough hydrogel–solid bonding interfacial toughness. Reproduced with permission.^[45] Copyright 2016, Macmillan Publishers Limited.

Nonaqueous ionic gels are formed by gelation of polymer network in ionic liquids with noncovalent associations such as hydrogen bonding, phase separation, and supramolecular interaction. Unlike the solid polymer electrolytes, nonaqueous ionic gels possess favorable ionic conductivity ($\approx 10^{-3}$ S cm⁻¹), good chemical and electrochemical stability. As the basic materials of self-assembly ionic gels, many kinds of copolymers have been reported, including poly(styrene-*b*-ethylene oxide-*b*-styrene) (SOS),^[50] poly(styrene-*b*-methyl methacrylate-*b*-styrene) (SMS),^[33] polystyrene-*b*-poly(ethyl acrylate)-*b*-polystyrene (SEAS),^[48] triblock copolymers and copolymer of vinylidene fluoride and hexafluoropropylene (P(VDF-co-HFP)).^[55] Wang et al. designed self-healing ionic conductors by employing the dynamic bonds of ion–dipole interactions between poly(vinylidene fluoride-co-hexafluoropropylene) (PVDF-co-HFP) polar polymer network and imidazolium salts (Figure 5a).^[56] The property of ionic gels was extremely affected by the polymer segment units and ionic liquids. The transparent and stretchable ionic gels were achieved by mixing specially appointed ionic liquids (1-ethyl-3-methylimidazolium trifluoromethanesulfonate (EMITf)) as electrolytes and PVDF-co-HFP with high HFP unit (>45%; Figure 5b). The ionic gels exhibited a Young's modulus of 0.1 MPa and an ionic conductivity of 5.66×10^{-5} S cm⁻¹ at 43 wt% EMITf. The ionic gels completely restored all mechanical properties within 24 h at room temperature without any external stimulus (Figure 5c). Besides, the ionic gels showed high stretchability (strains exceeding 5000%) and transparency (average transmittance of 92%). Lee et al. utilized poly(methyl methacrylate) (PMMA) as the polymer network and lithium perchlorate (LiClO₄) as electrolyte to prepare ionic gels.^[57] PMMA and LiClO₄ were fully dissolved in a mixture of acetonitrile

and propylene carbonate to obtain a viscous, clear, and gel-like solution, and then acetonitrile was fully evaporated to get solid ionic gels. The gels can sustain extremely large mechanical deformations up to 700% stretching strain and close to 100% transmittance. Resistance change of the ionic gels closely matched with the $R/R_0 = \lambda^2$ curve, meaning that their resistivity was independent of their stretch ratio. Moreover, the ionic gels maintained constant resistance even after 1000 cycles' repetitive stretching to 700%.

Self-healing ability is considered as the main merit of E-skin. The repeatable self-healing ability has been demonstrated to be enabled by dynamic covalent bonds, supramolecular interactions, ionic bonding, and hydrogen bonding.^[58] Self-healing hydrogels are advantageous in lifetime, mechanical robustness, and reliability.^[59] Supramolecular polymer gels exhibited a superior self-healing ability due to their dynamically reversible noncovalent interactions.^[60] Wu et al. reported the first example of autonomously self-healable E-skin made from supramolecular mineral hydrogel.^[25] Wang's group reported an ionic gel with high mechanical toughness and self-recovering ability due to their noncovalent crosslinking interaction.^[61] The tough ionic gels comprised of physical gelation of the hydrogen bonding interactions between 1-ethyl-3-methylimidazolium chloride, hydroxyethyl methacrylate, and chitosan. Charged polymer networks represent a new avenue of highly tough and self-healing ionic gels due to their reversible sacrificial bonds of electrostatic interaction.^[62] Luo et al. developed a new class of oppositely charged polyionic complexes with a wide distribution of bond strengths of dynamic crosslinking. The polyionic gels presented a fracture strength of several MPa and tearing fracture energy up to 10 000 J m⁻². In addition, the polyionic gels can even heal in saline solutions.

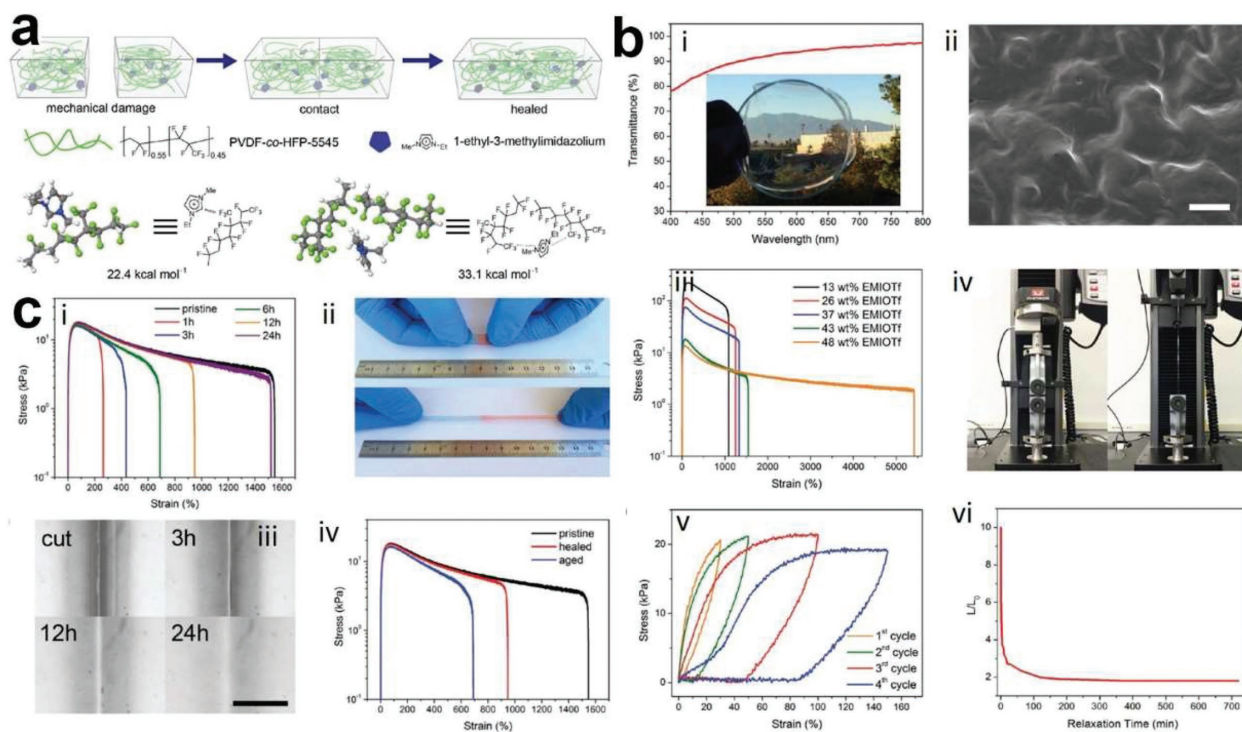


Figure 5. Transparent, self-healing, highly stretchable ionic gels. a) Schematic illustration of healing process and chemical structure of polymer and imidazolium cation. b) Physical and mechanical properties of gels: i) transmittance spectrum; ii) SEM image, scale bar, 10 μm ; iii) tensile strain tests of ionic gels with different amounts of ionic liquid; iv) photograph of the extreme stretchability of ionic gels; v) stress–strain cycling tests of ionic gels; and vi) a tensile set test. c) Self-healing properties of ionic gels. Reproduced with permission.^[56] Copyright 2017, Wiley-VCH.

3. Ionic-Gel-Based Stretchable Devices

Whitesides' group confirmed that the ionic gels comprised of PAM hydrogel and NaCl showed a high frequency (10 kHz) and voltage (10 kV) operation capability.^[19] Even though ionic gels have higher resistivity than most traditional rigid electronic conductors, they own an intrinsic smaller change in sheet resistance than existing electronic conductors under extension. Furthermore, ionic gels exhibit an advantage of high transparency (a transmittance of 99.99% for a 100-mm-thick hydrogel),^[19] good stretchability (about 500%)^[27] and biocompatibility. All these characteristics of ionic gels present a great promise in the fields of wearable electronics, flexible displays, and E-skins.

3.1. Transparent and Stretchable E-Skin Sensors

Ionic gels change shape when deformed, and the corresponding change in resistance can be utilized for pressure and strain sensing. Sun et al. developed a stretchable and transparent skin-like pressure sensor using PAM hydrogel as the ionic conductor (Figure 6).^[20] The capacitive ionic-gel-based electronic skin had a simple structure with a stretchable and transparent dielectric acrylic elastomer sandwiched between two ionic conductors. By connecting the ionic conductors to metal conductors, a hybrid ionic–electronic circuit was formed, and can transduce ionic skin signals without electrochemical reactions (Figure 6a). The capacitance of ionic skin increased as

the dielectric deformed under external forces, so the ionic skins were capable of detecting large deformations (Figure 6b), such as bending of fingers. Moreover, the good stretchability enabled it to detect stimuli in a wide dynamic range (1–500%). In addition, a distributed sensor sheet, attached on the back of a hand, demonstrated the capability of detecting the location and pressure level of touch (Figure 6c,d).

Pan's group employed ionic gel matrix to realize interfacial capacitive pressure sensing (called iontronic film sensor; Figure 7a).^[63] The sensors were prepared by integrating two flexible polymeric surfaces (with thin film of ionic gel coated on the surface) in a parallel-plate configuration. Based on the highly capacitive interface at the ionic–electronic contact, the sensors exhibited high device sensitivity (3.1 nF kPa⁻¹) and rapid response (submillisecond range). Lei et al. achieved a novel type of mechanically adaptable sensor by using a bio-inspired supramolecular mineral hydrogel (Figure 7b).^[25] The capacitive sensors were constructed by integrating two hydrogel films with a polyethylene dielectric layer, and showed a high pressure and strain sensitivity (0.17 kPa⁻¹), good reliability, and biocompatibility. Owing to its unique viscoelasticity and mechanical compliance, the hydrogel film can be dynamically self-adapted to a prosthetic finger and accommodated to the finger movements. Furthermore, the self-healable ionic-gel-based electronic skin was able to fully recover the capacitive sensitivity at room temperature. Cho et al. reported capacitive pressure sensors with topographically patterned arrays of pyramidal ionic gels (Figure 7c).^[24] The micropatterned pyramidal ionic gel's diamond tips consisting of polymer networkers

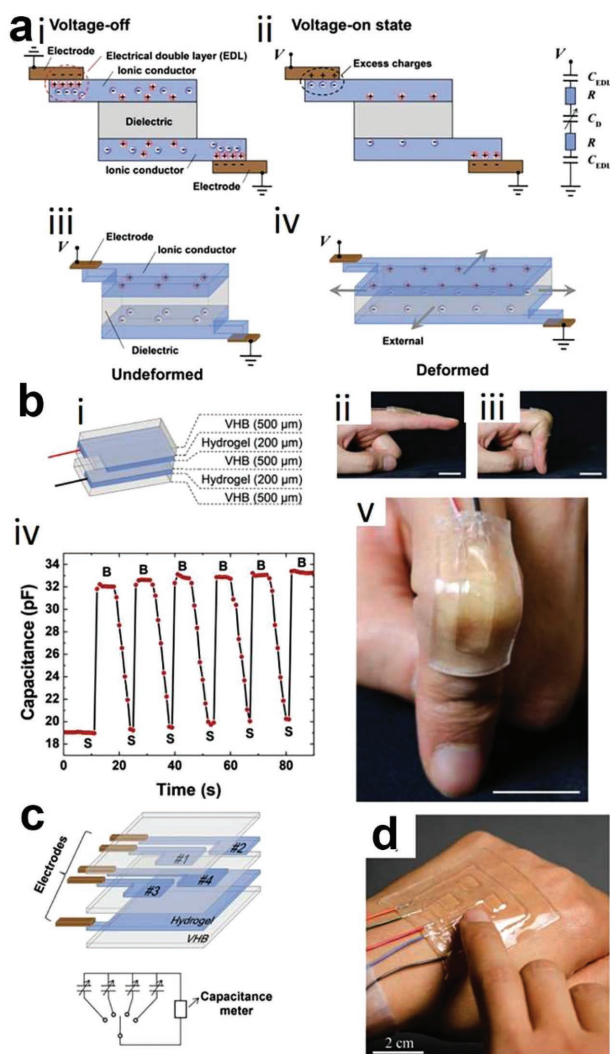


Figure 6. Capacitive ionic skins using PAM hydrogel as the ionic conductor. a) The working mechanism of hybrid ionic–electronic circuit; stretchable and transparent dielectric acrylic elastomer were sandwiched between two ionic conductors. b) Strain sensor of ionic skin: i) structures of ionic skin; ii) strain sensor attached to a straight finger; iii) the bending of finger stretched the sensor; iv) the capacitance of strain sensor under bending cycles; and v) fully transparent strain sensor. c) Structure of pressure sensor arrays. d) The sensor array was attached on the back of a hand. Panels (a–d) reproduced with permission.^[20] Copyright 2014, Wiley-VCH.

of P(VDF-HFP) and ionic conductor of 1-ethyl-3-methylimidazolium bis(trifluoromethylsulfonyl)amide ([EMI][TFSA]) exhibit significant capacitance variation as a function of pressure, resulting in unprecedented sensor sensitivities ($\approx 41 \text{ kPa}^{-1}$) and a broad sensing range (0.5–5 kPa). Wang’s group recently reported a soft E-skin of triboelectric nanogenerator (STENG) exhibiting ultra-high stretchability (strain of 1160%), transparency (up to 96.2%), and biocompatibility (Figure 7d).^[26] The PAAm-LiCl hydrogel electrode enabled the assembled device an exquisite tactile sensing ability. Xu et al. demonstrated an environmentally friendly hydrogel-based triboelectric nanogenerator with polyvinyl alcohol hydrogel as substrate.^[64] Tian

et al. developed a simple 3D extrusion printing approach to fabricate a stretchable resistive strain sensor by integrating ionically conductive PAM hydrogel and PDMS.^[65] The fabricated strain sensor has a gauge factor of 0.84, and is capable of detecting finger motions. Compared with the conventional manual assembling procedure, the 3D extrusion printing can realize multiple material printing with high speed, high resolution, and low costs.

3.2. Flexible Electric Double-Layer Transistors

As ionic conductors, high polarization in ionic gels can be achieved by the reverse motion of cations and anions in an electric field. Ionic gels represent a novel class of high-capacitance gate dielectric materials in flexible EDL transistors. Based on Keplinger’s discussions about the EDLs (Figure 8a),^[19] EDLs can be formed at the interfaces of electronic and ionic conductors, according to the double-layer theory. Charges in EDLs are separated over nanometers and the capacitance is estimated to be on the order of $1\text{--}10 \mu\text{F cm}^{-2}$. While the dielectric capacitor has a small capacitance (on the order of $10^{-6} \mu\text{F cm}^{-2}$) due to the long separation distance. Therefore, voltage across the EDL is quite small, thus electrochemical reaction is prevented.^[19]

Lodge’s and Frisbie’s groups demonstrated that the ionic gels could be successfully used as the gate dielectric in a top-gate polymer thin-film transistors (TFTs) operating at high frequencies (greater than 100 Hz).^[51] These ionic gels, composed of poly(styrene-block-ethyleneoxide-block-styrene) (SOS) triblock copolymer and 1-butyl-3-methylimidazolium hexafluorophosphate ([BMIM][PF₆]) ionic liquid, offers both large specific capacitance ($>10 \mu\text{F cm}^{-2}$) and greatly shortened the polarization response times ($\approx 1 \text{ ms}$). They demonstrated gel-gated organic TFTs fabricated by commercial printing method (Figure 8b).^[33] Owing to their solution processability, ionic gels can be directly printed onto the plastic substrate and integrated into high-performance ion-gel-gated organic TFTs with high operation frequency (10 kHz). As high-capacitance gate dielectric materials, ionic gels have also showed exceptionally high polarizability.

With the extremely large capacitances, good solution processable features, and high polarizability, ionic gel dielectric materials hold promising future for flexible electronics. Lee et al. prepared stretchable and transparent all-graphene-based transistors by printing (Figure 8c).^[32] The graphene patterns realized by photolithography and etching served as both the source/drain electrodes and the semiconducting channels. Ionic gels composing of both triblock copolymer and ionic liquids were printed as the gate dielectric layer, and conductive polymer served as the gate electrode. The fabricated stretchable transistors exhibited high mobility and good stability (5% strain, 1000 cycles). Therefore, with ionic gel dielectrics, printing methods represented a promising route to fabricate high-performance flexible and stretchable electronics. Besides printing methods, ionic gel dielectric layers can also be fabricated by spin-coating^[66] or “cut and stick” method.^[67] Recently, a solid-state electrolyte-gated field-effect transistors (FET) were fabricated by solution-processed method. Blending of high-k polymer and ionic gels achieved remarkable FET performance with large charge-carrier mobilities.^[68]

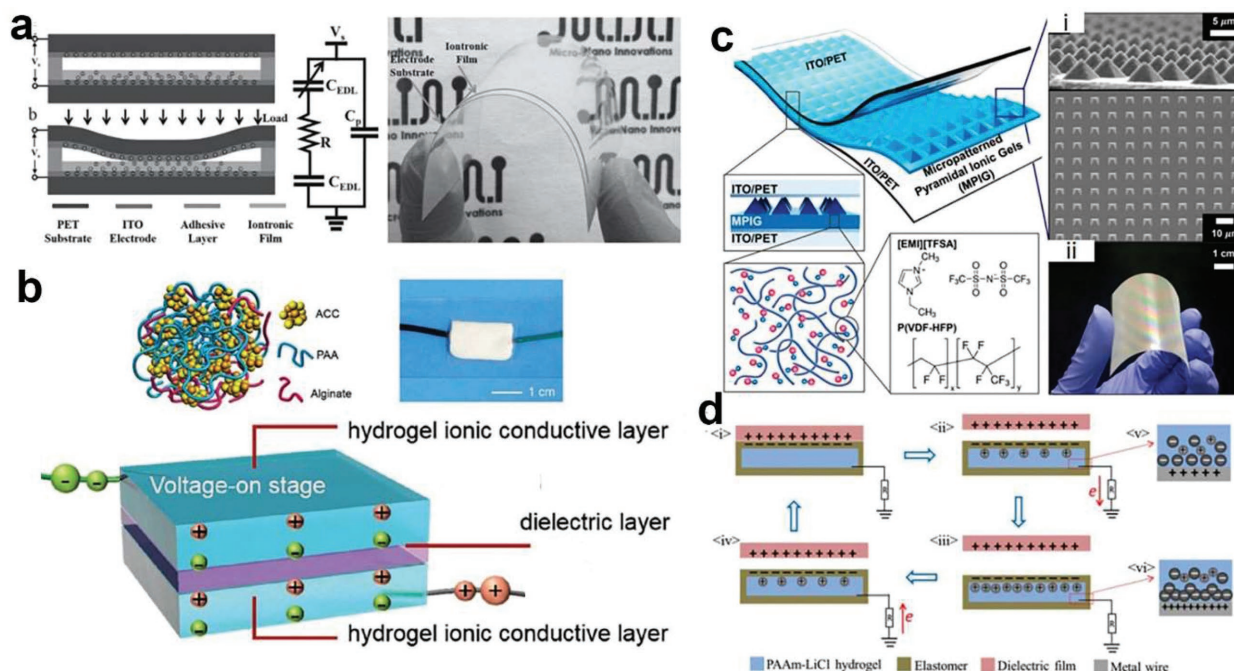


Figure 7. Ionic-gel-based sensors. a) Interfacial capacitive pressure sensor based on the formation of an electrical double layer. Reproduced with permission.^[63] Copyright 2015, Wiley-VCH. b) Capacitive pressure sensor based on bioinspired supramolecular mineral hydrogel. Reproduced with permission.^[25] Copyright 2017, Wiley-VCH. c) Capacitive pressure sensor with micropatterned pyramidal ionic gels diamond tips. Reproduced with permission.^[24] Copyright 2017, American Chemical Society. d) Soft tactile sensor of triboelectric nanogenerator with PAM ionic gels as conductor. Reproduced with permission.^[26] Copyright 2017, AAAS.

Moreover, ionic gel dielectrics present wide applications in various transistor devices. Kim et al. fabricated a graphene FET array on a flexible polymer substrate using a high-capacitance ion gel as gate dielectric,^[69] and the FET showed low operating voltages (<3 V), high mobility and on-current. Takenobu reported molybdenum disulfide (MoS₂) TFTs with ionic gel as the gate dielectric.^[70] And, the FET exhibited high on/off ratios (10⁵), good mobility (12.5 cm²·V⁻¹·s⁻¹), a low operating voltage (0.68 V), and superior mechanical flexibility. Besides, there was no degradation of electrical performance when the TFT was bent at curvature radius of 0.75 mm. Xu et al. developed transistors based on buckled CNT films with ion gel dielectrics,^[71] demonstrating excellent on/off ratio (>10⁴), good mobility (10 cm²·V⁻¹·s⁻¹), low operating voltage (<2 V), and fair performance sustention under strains up to 50%. Jeong's group developed an array of polymer transistors made entirely of stretchable components.^[72] Each transistor consists of stretchable Au nanosheets as electrodes, ionic gels as the gate dielectric, and poly(3-hexylthiophene) (P3HT) nanofibers as the active channel material. The device was stable for over 1500 cycles of stretching at a strain of 0.7.

3.3. Flexible and Stretchable Display Devices

Stretchable display devices are important components in wearable electronics, bioinspired camouflage, and biomedical applications. Various types of stretchable display devices, including organic light-emitting diodes (LEDs),^[73] light-emitting electrochemical cells,^[74] electroluminescent devices,^[75] and

electrochromic devices^[76] have been developed. The biggest challenge in constructing flexible displays is the fabrication of transparent, stretchable, and conductive electrodes. Shepherd's group presented PAM-based ionic gel as stretchable electrodes in a light-emitting capacitive touch sensor (Figure 9).^[27] Relying on its sandwich structure, ionic-gel-based light-emitting displays worked at 700 Hz under a high electric field of ≈25 kV cm⁻¹ (Figure 9b). Ionic gels enable the light-emitting displays to deform with various modes, including stretching, rolling, folding, and wrapping (Figure 9c). In addition, the ionic-gel-based electrodes experience small resistance changes even under stretching. When integrated with soft robots, the hyperelastic light-emitting displays act as camouflage system with the functions of dynamic coloration and sensory feedback. They extended the same method by incorporating ionic gel electrodes to a full-color display at a subcentimeter scale resolution, and realized multipixel color display and multitouch sensing. Owing to its good conductance, high transparency, and excellent stretchability, ionic-gel-based electrodes presented great potential in light-emitting displays. Yang et al. reported an ionotronic luminescence device assembled by sandwiching phosphor particles between two layers of ionic conductors.^[77] The luminescence device can sustain luminescence effect under strain up to 1500%. Lee's group presented a light-emitting display with PMMA-based ionic gel as electrodes, which can endure extreme mechanical deformations up to 700%.^[57]

As aforementioned, ionic gels have been extensively used as electrodes for sensors, and as dielectrics for gel-gated transistors. In addition, ionic gels are instinctive electrolytes comprising of both mobile cations and anions. Owing

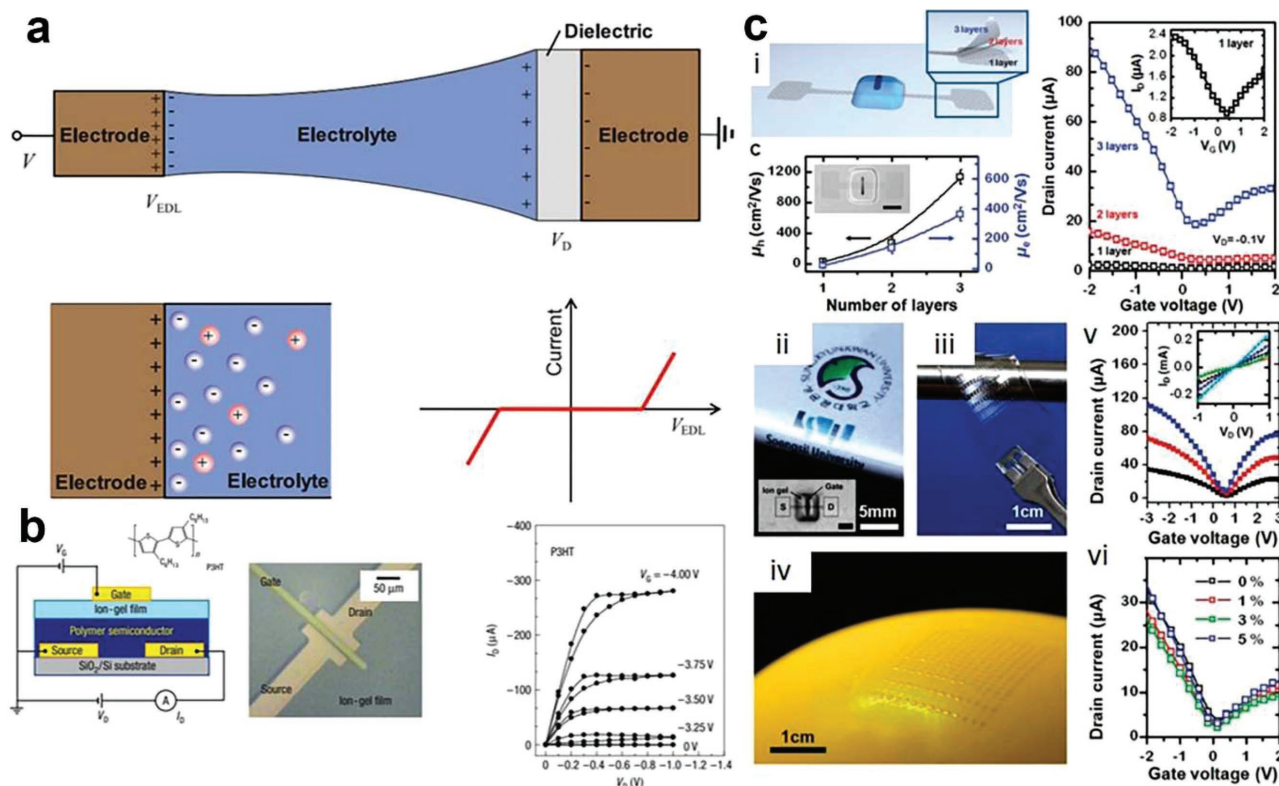


Figure 8. Ionic gel dielectrics and gel-gated transistors. a) The formation of electric double layer in an electronic conductor, an ionic conductor, and an insulator system. Reproduced with permission.^[19] Copyright 2013, AAAS. b) Schematic diagram and optical image of gel-gated OFETs and output characteristics of the P3HT-based GEL-OTFT at five different gate voltages. Reproduced with permission.^[33] Copyright 2008, Macmillan Publishers Limited. c) Stretchable graphene transistors: i) schematic illustration and transfer characteristics of stretchable graphene FETs; ii–iv) images of the ion-gel-gated graphene FETs on: ii) polyethylene terephthalate (PET), iii) PDMS, and iv) balloon (iv); v,vi) transfer characteristics curves. Reproduced with permission.^[32] Copyright 2011, American Chemical Society.

to its outstanding mechanical stretchability and high ionic conductivity, ionic gels provide an attractive electrolyte platform for flexible electrochemical device applications, such as electrochromic (EC) devices (Figure 10)^[28–78] and electrochemiluminescent (ECL) devices (Figure 11).^[22–79]

Similarly, Lodge's and Frisbie's groups demonstrated the multifunctional EC ionic gels by incorporating redox-active species to electrochromic ionic gels.^[28] The multifunctional EC gels were prepared by blending PS-PMMA-PS triblock copolymer with ionic liquids ([EMI][TFSI]), and then incorporated with an active EC material (methyl viologen dication). ECDs were composed of two transparent flexible electrodes (ITO coated PET films) and an electrolyte ionic gel layer (Figure 10a). The EC materials change optical absorbance by means of electrochemical reactions (Figure 10b,c). With the addition of anodic redox species (ferrocene), the ECDs exhibited color changes at low voltage (<1 V). In addition, patterned, multicolored, and flexible display devices based on electrochromic gels were successfully demonstrated (Figure 10d). Therefore, serving as electrolyte medias, ionic gels showed great potential on electrochemical applications. However, the electrochemical devices can only operate under low voltages due to the narrow electrochemical window of ionic gels. They also prepared ECL ionic gels by the addition of electrochemical luminophores (Figure 11a).^[22] Like electrochromic devices, ECL devices were comprised by

sandwiching the ECL ionic gels with two transparent electrodes. By applying AC voltage, both the oxidized and reduced species were generated by the electrochemical reaction on the same electrode, then excited species were created by electron-transfer reaction between the oxidized and reduced species (Figure 11a i–iii). According to the advantages of solution processability, the flexible emissive displays with patterned ionic gel electrolytes layer were fabricated by solution casting. The ECL displays with patterns of numbers worked successfully even after bending (Figure 11b). However, some tricky problems must be considered, such as the high transient ionic currents under high-frequency switching and large resistive voltage drops in the gel because of the limitation of ion mobility.

3.4. Flexible and Stretchable Energy Storage Devices

With the development of soft materials, great interest has been aroused in integral flexible and stretchable electronic system, such as E-skins, rollup displays, and bendable mobile phones.^[80] As the power source for a complete and independent system, most energy storage devices (typically, lithium-ion batteries and supercapacitors) are too heavy, rigid, and bulky to meet the requirements for soft electronic applications.^[81] Therefore, there is great demand for flexible and stretchable

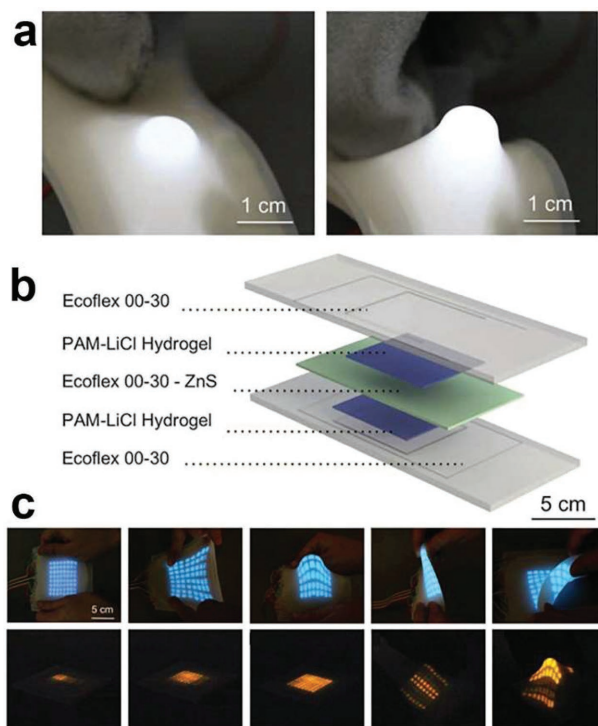


Figure 9. Light-emitting capacitive touch sensor based on ionic gel electrode. a) Images of stretchable light-emitting capacitance. b) Schematic illustration of five-layer structure. c) Multipixel electroluminescent displays in various states of deformation and illumination. Reproduced with permission.^[27] Copyright 2016, AAAS.

energy storage devices with excellent energy and power density retention upon stretching.^[30,82]

Electrochemical devices are mainly composed of two facing electrodes immersed in electrolyte and separated by a separator film.^[83] In recent years, electrolyte and separator have become the severe limiting factor for the stretchable electrochemical energy devices, compared with the mature technology of current collectors and electrodes.^[84] Polymer electrolytes are advantageous over conventional liquid electrolytes for stretchable electrochemical devices because they are leak proof, ease of fabrication into desired shapes and sizes. And, polymer electrolyte can act as both the ionic conductor and the separator.^[85] Thus, high-performance polymer electrolytes with superior ionic conductivity are highly desired for the all-solid-state energy storage devices.^[86] Ionic gel electrolytes, composed of a polymeric framework, solvent (plasticizer), and supporting electrolytic salt, are promising candidates as electrolytes for stretchable electrochemical devices for two reasons.^[87] Firstly, ionic gels have excellent electrochemical and thermal stability, high ionic conductivity, and negligible vapor pressure (ionic-liquid-based gels).^[88] Secondly, owing to the soft nature of polymers, ionic gels offer great flexibility to sustain structural integrity of devices under great elongation and curvature.^[89]

More special ionic gels composed of block copolymers and ionic liquids with toughness, stretchability, and compressibility have been also developed.^[90] Kim's group demonstrated an all-solid-state flexible supercapacitor fabricated with bacterial nanocellulose (BNC), CNTs, and ionic-liquid-based poly(styrene-block-ethylene oxide-block-styrene) (PSPEO-PS) triblock copolymer gel electrolytes.^[91] The hydrophobic polystyrene (PS)

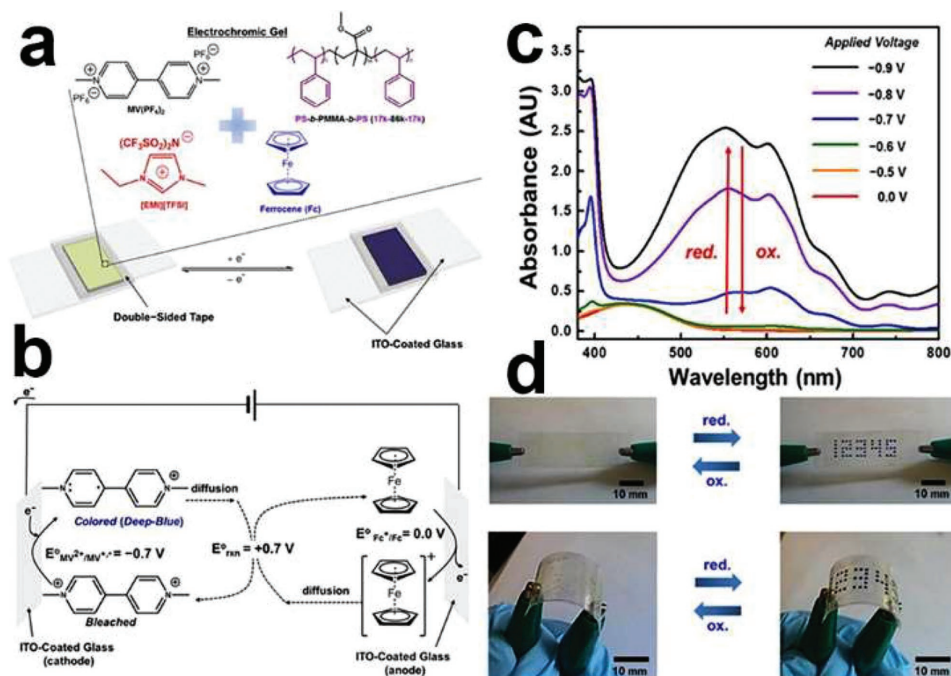


Figure 10. Electrochromic devices. a) Schematic illustration of electrochromic devices (ECDs) based on the EC gel consisting of $MV(PF_6)_2$, Fc, SMS, and $[EMI][TFSI]$. b) Diagram of electrochemical reactions. c) UV-vis absorption spectra at various applied voltages. d) Photographs of the ECDs in the bleached and colored states: before bending (top) and after bending (bottom). Reproduced with permission.^[28] Copyright 2015, American Chemical Society.

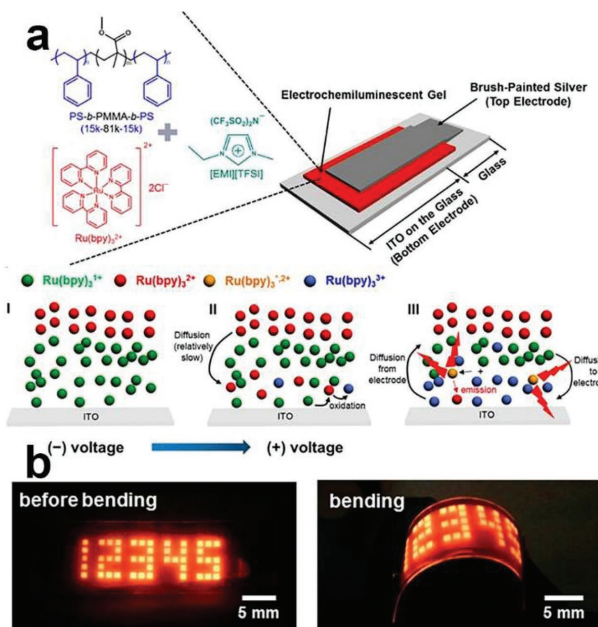


Figure 11. Electrochemiluminescent (ECL) device with an emissive gel layer. a) Schematic illustration of ionic-gel-based ECL and chemical species and reactions near the electrode (I–III). b) Photographs of the ON states for fabricated ECL devices before bending (left image) and after bending (right image). Reproduced with permission.^[22] Copyright 2014, American Chemical Society.

block in triblock copolymer ensures good adhesion between the electrolyte and the CNT electrodes via van der Waals interaction, and the PEO block can effectively retain the hydrophilic

ionic liquid in polymer matrix. Owing to the adhesive chemical property of ionic gels, the electrochemical performances of assembled supercapacitor were well maintained over 200 bending cycles to a radius of 3 mm. Wang et al. synthesized a gel polymer electrolyte (GPE) consisting of a commercial membrane (Celgard) incorporated with a P(EO-co-PO (propylene oxide)) copolymer swelled by liquid electrolyte for lithium-ion batteries.^[92] The P(EO-co-PO) matrices, which are reinforced by blending with diglycidyl ether of bisphenol A (DGEBA), can effectively dissociate the lithium salt and enclose sufficient solvent molecules to facilitate the lithium-ion transport in solvent channels. As a result, the ionic gel electrolyte has an ionic conductivity of $\approx 10^{-3} \text{ S} \cdot \text{cm}^{-1}$ at room temperature, and demonstrates excellent mechanical integrity. Based on the considerations above, Peng's group demonstrated a self-healing lithium-ion battery by pairing the self-healing cathode and anode with the aqueous lithium sulfate/sodium carboxymethylcellulose ($\text{Li}_2\text{SO}_4/\text{CMC}$) as both gel electrolyte and separator between them.^[93] The specific capacitance of the aqueous LIBs could be restored to 61% after five breaking/healing cycles with good maintenance of the mechanical properties of the whole device (Figure 12a,b).

In recent works, Huang and coworkers presented a new polymer electrolyte comprised of PAM hydrogel crosslinked by vinyl hybrid silica nanoparticles (VSNPs).^[94] The fabricated supercapacitors using the PAM-VSNPs gel electrolyte possess enhanced specific capacitance with intrinsic super-stretchability (up to 1000% strain) and good capacitance retention under high compressibility (up to 50% strain; Figure 12c–e). Yang et al. reported a high-performance GO-doped ionic gel (P(VDFHFP)-EMIMBF4-GO gel).^[95] GO as a 3D network distributes homogeneously throughout the gel matrix, and acts

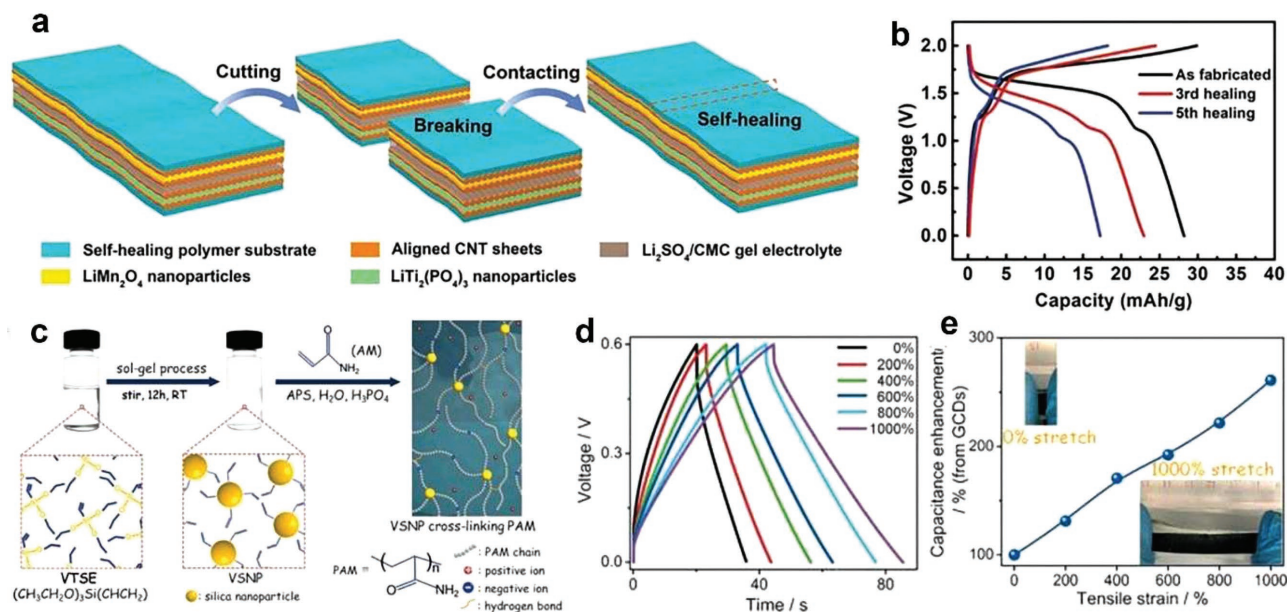


Figure 12. a) Schematic illustration of self-healing aqueous lithium-ion battery. b) Galvanostatic charge–discharge curves of the aqueous lithium-ion battery after different numbers of self-healing cycles at a current density of 0.5 A g^{-1} . Reproduced with permission.^[93] Copyright 2016, Wiley-VCH. c) Preparation of VSNPs from VTSE followed by preparation of the VSNPs-PAM electrolyte. d) Galvanostatic charge/discharge (GCD) curves from 0% to 1000% tensile strain. e) Capacitance enhancement factor obtained from GCD curves as a function of the tensile strain. Panels (c–e) reproduced with permission.^[94] Copyright 2017, Wiley-VCH.

like an ion “highway” to facilitate the ion transport, leading to 260% increase in ionic conductivity compared with that of pure ionic gels. Peng’s group developed a silicon–oxygen battery fiber with high energy density ($512 \text{ Wh} \cdot \text{kg}^{-1}$) and ultra-high flexibility (effectively work after bending for 20 000 cycles), by designing a coaxial architecture with a lithiated silicon/CNT hybrid fiber as inner anode, a mixture of bis(trifluoromethane sulfonimide) (LiTFSI), lithium nitrate (LiNO_3), tetraglyme, poly(vinylidene fluoride-cohexafluoropropylene) (PVDF-HFP), *N*-methyl-2-pyrrolidinone (NMP), 2-hydroxy-2-methyl-1-phenyl-1-propanone (HMPP), and trimethylolpropane ethoxylate triacrylate (TMPET) ionic gels as middle electrolyte and a bare CNT sheet as outer cathode.^[96]

3.5. Ionic-Gel-Based Soft Actuators

Actuators convert electrical energy to mechanical motions.^[97] Ionic gel actuators, comprising a layer of ionic polymer electrolyte sandwiched between flexible electrodes, are an emerging category of electroactive soft actuators, which have exhibited numerous applications in artificial muscles,^[97a] soft robotics.^[97b] When a voltage is applied, electromechanical motion in the ionic polymer electrolyte occurs through the migration of ions toward oppositely charged electrodes. Currently, electroactive polymers (EAP), or ionic polymer–metal composites (IPMCs) have been well developed as actuators, including both ionic and nonionic EAPs such as poly(vinylidene fluoride)-based (PVDF) polymers, conjugated polymers, and perfluorinated ionomers. Ionic gel actuators comparatively utilize ionic polymers or gels, ionic liquids, and deformable electrodes, which can exhibit several exceptional advantages including large displacements in bending motion, fast switching response, low operating voltage, and ease of fabrication at low costs.^[98] Meanwhile, these actuators have wide electrochemical potential windows, high ionic conductivity and low vapor pressure, resulting from the introduction of chemically stable ionic liquids.^[88] Recently developed high-performance polymer actuators composed of ionic liquid, soluble sulfonated polyimide polyelectrolytes, and carbon materials, demonstrated inexpensive processability, favorable mechanical properties and excellent durability of more than 5000 cycles in dry air.^[99]

The effective and rapid electromechanical transduction of ionic gel actuators relies on the migration and spatial redistribution of mobile ions in the polymeric structures, as well as surface or interfacial adhesion between electrodes and polymer layer.^[29,100] At the early stage, to improve interfacial adhesion between electrodes and ionic gel electrolyte during deformation cycles, carbon-based materials such as carbon power film, carbon grease, CNTs, and graphene, were investigated as alternatives to metals for flexible electrodes.^[99a,101] Recently, Kotal et al. successfully developed a high-performance ionic artificial muscle based on sulfur and nitrogen co-doped graphene electrodes, the fabricated actuator possessed a large bending strain up to 0.36% under 1 V at 0.1 Hz, which was 4.5 times higher than PEDOT:PSS electrodes.^[102] It demonstrated good durability (96% of initial strain after 18 000 cycles), while the heteroatom co-doping in reduced graphene oxide resulted in more defect sites, increased charge density, larger surface area

and larger pore sizes. Integrating metal nanocomposites with ionic gel is another strategy to address the problem of electrode cracking and delamination during actuation. For example, Yan et al. fabricated an ionic soft actuator with chemically crosslinked PAA–polyacrylonitrile (PAN) ionic gel and gold nanocomposite electrodes,^[29] as shown in **Figure 13a**, exhibiting high-performance actuation response at low voltages (up to 1.04% net strain at 5 V) and good durability in the frequency response (up to 76 000 cycles at 2 V and 1 Hz). Although very compliant and conformal to the deformation of actuators, such materials showed poor transparency, limited mechanical properties, and high sheet resistances.

Dielectric elastomer actuators offer an alternative operation based on the coulomb force interaction in two flexible electrodes. And, performance of this group of actuators is highly dependent on elastomer’s stiffness, dielectric constant, and breakdown voltage.^[97b] Acrylic elastomers, silicones, polyurethanes, and rubber are typical membrane materials employed in dielectric elastomer actuators. As shown in **Figure 13b**, Keplinger et al. designed a transparent actuator using PAAM-based hydrogel as ionic conductor electrolyte and VHB 4910 tape (3M) as the dielectric elastomer.^[19] The fabricated actuator obtained an area strain of 167%, and actuators made from larger pre-stretching and thinner hydrogels have larger actuated strains. Furthermore, they demonstrated that the transparent actuator can performance as a loudspeaker producing an entire audible range of sound from 20 Hz to 20 kHz. Chen et al. demonstrated an actuator with nonvolatile ionic gels as the soft ionic conductor.^[103] The nonvolatile ionic gel was prepared by polymerization of acrylic acid in an ionic liquid and exhibited low elastic modulus and excellent stretchability. And, the actuators were tested to perform stably after a million cycles of excitation both in a dry oven and in air. Recently, Wang et al. reported an electrically activate transparent artificial muscle using autonomous self-healing ionic gels as conductors.^[56] By harnessing ion–dipole interactions between the fluorinated polymers and the ionic liquids, the ionic gels show outstanding self-healing capabilities with all mechanical properties fully restored within 24 h, leading to increased lifetime of the actuators. In conclusion, development of ionic–gel-based soft actuators opens up a promising direction for future biomedical devices.^[36]

4. Conclusions and Outlook

The developments of stretchable electronics hold attractive prospects for applications in E-skin, wearable devices, implantable devices, and soft robotics. Suo et al. presented in a recently published article that first-generation hydrogel-based devices use hydrogel as stretchable, transparent, ionic conductors.^[104] As stretchable functional materials, ionic gels exhibit not only competitive electrical properties, such as high ionic conductivity, good conductive stability, and short polarization time, but also biocompatibility, good transparency, and mechanical conformance to curved surfaces such as on human organs and robotic body parts. In addition, ionic gels adapt to universal solution processing methods, holding great prospect for larger-scale printable electronics fabricated by jet printing, screen printing, and spin coating. However, there are some limitations

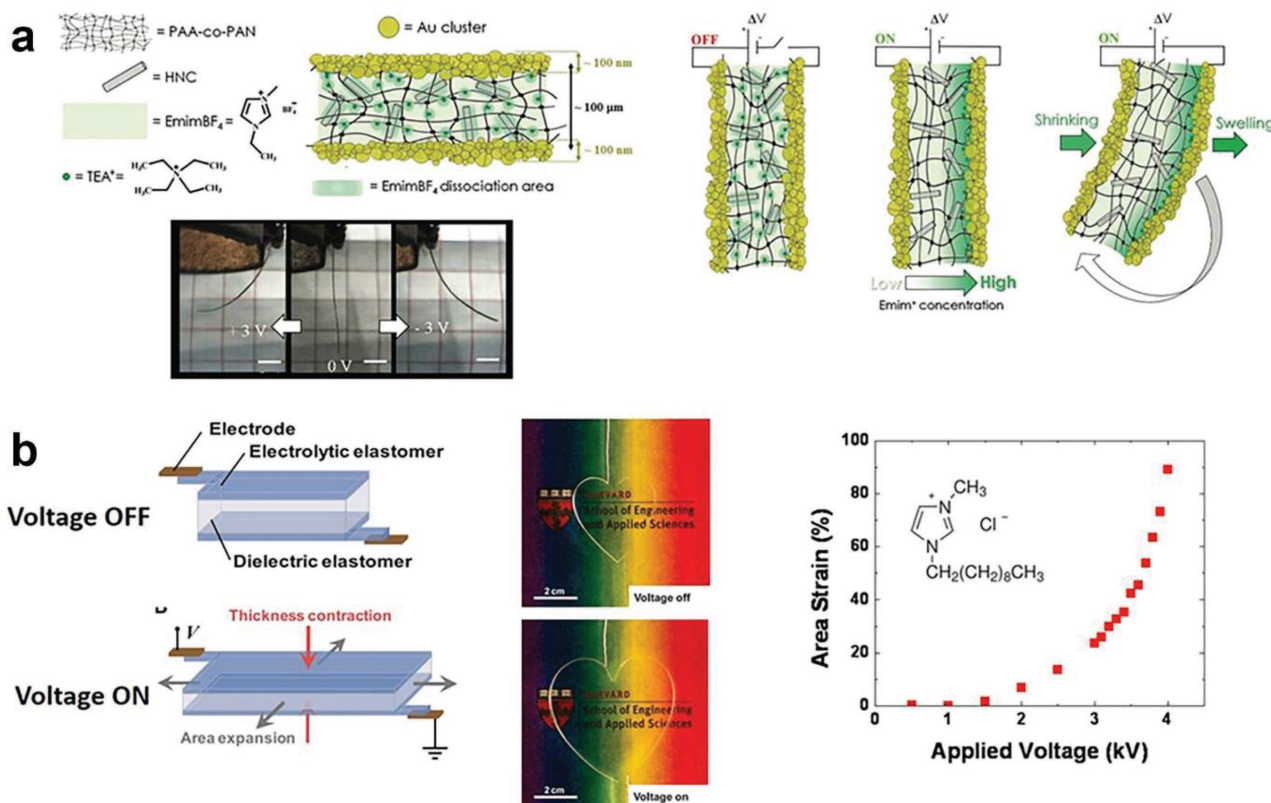


Figure 13. a) Schematic and actuation mechanism of ionic gel actuators composed of chemically crosslinked PAA-co-PAN networks with halloysite nanoclays (HNC) and imidazolium-based ionic liquid (EmimBF₄), gold nanocomposites as flexible electrodes. Bending example is operated at 3 V and 0.02 Hz. Reproduced with permission.^[29] Copyright 2017, Wiley-VCH; b) Schematic of dielectric elastomer actuator in the voltage OFF/ON where the dielectric elastomer is sandwiched between two layers of an electrolyte elastomer, area strain, and recorded loudspeaker performance of the dielectric elastomer actuator using ionic liquid ([C₁₀MIM][Cl]) as conductor as a function of applied voltage. Reproduced with permission.^[19] Copyright 2013, AAAS.

of using ionic gels as conductors, dielectrics, and electrolytes, respectively.

As ionic gels are connected to the positive and negative terminal of the power supply, they can only support low voltage, due to the inevitable electrochemical reactions at high voltage. However, by sandwiching a dielectric layer as a separator, ionic gels demonstrate much better electrode capability for capacitive devices, such as electroluminescence devices and actuators, which are powered by high voltage as high as 10 kV. Nevertheless, owing to the evaporation of solvents (water or organic solvents) and leakage of electrolytes (salts or ionic liquids), ionic-gel-based devices are confronted with tricky problems such as decaying of electrode conductivity and internal short circuit at high voltages. Though proper packaging of ionic-gel-based devices can partially solve the above problems, the weakly bonded interface between ionic gel and packaging elastomer remains problematic to mechanical reliability. Therefore, it is essential to research on novel packaging of ionic-gel-based devices in order to improve the durability and stability of the devices.

Owing to their high polarization features, ionic gels represent a class of high-capacitance dielectric materials. Under electric fields, positive and negative ionic species migrate reversely and accumulate at the electrode–ionic gel interface, leading to the formation of EDLs. EDLs can be considered as parallel-plate

capacitors, which demonstrate high specific capacitance because of its nanometer thickness. Therefore, ionic-gel-gated transistors often have low onset voltages and high channel currents, leading to a potential disadvantage of ion-gel-gated transistors with relatively high off current (source-to-gate current) and leakage current (drain-to-gate leakage), resulting in power waste. Thus, it is essential to optimize the property of ionic gel dielectrics and the architecture of transistor.

As instinctive electrolytes, ionic gels meet the increasing demand for flexible energy devices including Li-ion batteries, supercapacitors, solar cells, and fuel cells. Compared to liquid electrolytes, ionic gel electrolytes are mechanically and thermally stable, and can fulfill multiple roles of electrolyte, separator, and binder. Gel electrolytes with wide potential windows, nonvolatility, and nonflammability offer tremendous advantages in making solid-state supercapacitors and thin-film electronics. Current ionic gel electrolytes have ionic conductivities typically lower than 10 mS cm⁻¹, which is significantly lower than liquid electrolytes. The other issue with ionic gel electrolytes is their poor compatibility with traditional electrode materials. Thus, it is crucial to extend the exploration on intrinsic chemical mechanisms and crosslinking structures of ionic gels to improve their electrochemical performance as electrolytes.

Other than the specific limitations of ionic gels as conductors, dielectrics, and electrolytes, respectively, there are general aspects demanding further exploration for future application of ionic gels in stretchable electronics.

First of all, interfacial adhesion between ionic gels and other components, such as substrates, electrodes, and sensor elements, need to be improved to achieve device stability. As one of the effective strategies to improve surface adhesion, chemical covalent bonding has achieved tough interface bonding of hydrogels to rigid components. However, the chemical bonding involved multistep modification and covalent crosslinking reactions, restricting the rapid assembly of ionic-gel-based devices. Besides, the chemical and electrochemical corrosions at the interface should be carefully considered in order to achieving long lifetime of these devices. In addition, stable interfacial adhesions for wet and dynamic surfaces in implantable biomedical devices are much more challenging and require more vigorous effort.

The second challenge is to develop advanced manufacturing techniques that are specific to ionic-gel-based stretchable electronics. Solution processing methods, such as inkjet printing, screen printing, and spin coating, have been well developed for ionic-gel-based electronics, but limited to fabrication of simple and 2D planar structures. As an emerging technology for fabricating complex architectures from digital models, 3D printing presents its potential to quickly form 3D soft objects for applications in stretchable and flexible sensors, energy devices, and bioinspired structures (such as tissue engineering, artificial organs, and soft actuators). During 3D printing, the ionic gel materials are usually patterned into a functional structure and subsequently fixed by a physical or chemical gelation process. Cost-effective direct ink writing (DIW) 3D printing, which uses ionic gel inks housed in a syringe barrel that are extruded in the form of a fiber through the tip of the syringe, is a burgeoning technique being applied to high-precision ionic gel printing.

Last but not the least, self-healing ionic gels have presented advantages on extending life cycles and performance stability, because of effective healing of mechanical damage. The slow healing speed, low healing efficiency, and incomplete functional recovery of currently developed self-healing ionic gels greatly limit the advancement of self-healing stretchable electronic devices. The exploration of the mechanism of self-healing is crucial in developing rapid and autonomous healable ionic gel materials. Both theoretical simulations at the molecular level and experimental characterizations at the macroscopic level are necessary to understand such self-healing mechanism. In addition, efforts should be devoted to fabrication of advanced self-healing ionic gels to be employed in stable and long-lifetime stretchable devices.

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Conflict of Interest

The authors declare no conflict of interest.

Keywords

electronic skin, ionic gels, soft actuators, stretchable electronics

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