

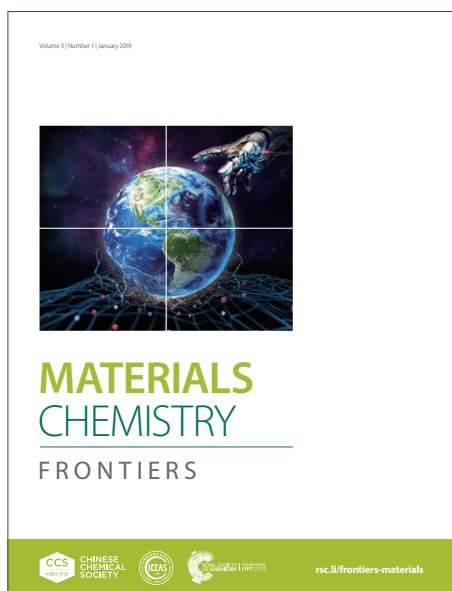
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Hydrogel Ionic Sensory Systems

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Abstract

When interacting with the dynamic world, creatures always outdo their electronic counterparts because of their sophisticated sensory systems. From direct perception of the world via soft tissue to intelligent information processing, immensely varied biological movements of organisms mostly rely on accurate ion transport. Imitating the flexible structure of biological systems and the functional mechanism based on ion transport would essentially solve the unmet needs of the modern sensing technology. Simultaneously, the establishment of the ionic sensor systems would effectively narrow the gap between the biological systems and the artificial equipment to promote seamless interaction of both sides, which could bring a revolution in advanced operations and medical rehabilitation. The outstanding properties of hydrogels enable them to be used as ion conductors like biological tissues for the development of ionic sensory systems. In this review, we introduce the basic principles of ion transport in hydrogels, and discuss the five key elements of hydrogel ionic sensory systems: ionic sensors, ionic transporters, ionic processors, ionic effectors, and ionic power sources, from their structure, working mechanism to applications. Finally, we suggest future directions and potential challenges of hydrogel-based ionic sensory systems.



1. Introduction

Perception connects creatures with the real world. In the process of continuous evolution, organisms have gradually mastered more efficient ways to sense and modulate signals. By using multiple sensors, humans receive external stimuli and convert them into ionic electrical signals. The ionic signals propagate along afferent neurons to central nerves for information processing and interpretation, and then output nerves feedback the signals to effectors to complete a perception-response loop to the external stimulus. The coordinated and sophisticated sensor systems help humans interact harmoniously with the colorful world. The precise migration of ions is the basis for most biological processes including the complete sensing process. In contrast, the sensory systems in modern equipment are mainly based on electronic transport.¹⁻³ At present, machine sensory systems rely on rigid and bulky electronic sensing devices to obtain information from the outside world, which cannot collect high-quality data on irregularly curved and dynamic surfaces while providing timely motion feedback.^{4,5} Simultaneously, electronic based information processing technology has been subject to the physical limitations predicted by Moore's Law, and it is difficult to further reduce the size on the basis of nanoscale devices to improve performance.^{6,7} Therefore, machines that rely on electronic microprocessors for signal processing face the problems of high power consumption and insufficient processing capabilities with the explosive growth of data. This is a far cry from the performance of an accurate, efficient, and low-energy (a power consumption on the order of several fJ/spike) biosensing system.⁸⁻¹²

Inspired by the powerful information acquisition and processing capabilities of organisms, researchers have invested enormous effort in the development of bionic electronic devices to break the barriers to the development of electronic equipment.¹³⁻³⁰ However, according to the current general consensus, the development of technology is difficult to help electronic products achieve advanced information transmission,⁶ because there are fundamental differences in both working principles and composition between traditional electronic devices and biological systems.³¹⁻³⁴ Electronics has a constant architecture in terms of information acquisition and processing. In contrast, creatures use abundant sensors and soft physiological structures sensitively feel environmental stimuli and provide timely action feedback. The transmission of the signals is completed by stimulus-induced ion transport across cell membranes. The transport of ions across membranes involves passive transport by ion channels or active transport by ion pumps, ion pumps and ion channels with the ability to selectively transport ions, which make ion migration more controllable in terms of ion species, direction, and speed.^{35,36} Simultaneously, through bidirectional communication between pre-synaptic and post-synaptic neurons, synapses dynamically modulate weight to realize the adaptive decision-making of organisms to the environment.⁸ Its superior perception and information processing ability are the inherent ascendancy of adaptive, plastic network of sensory neurons. Hence, simulating biological processes from the level of sensing structure and mechanism would fundamentally realize biological-like perception.



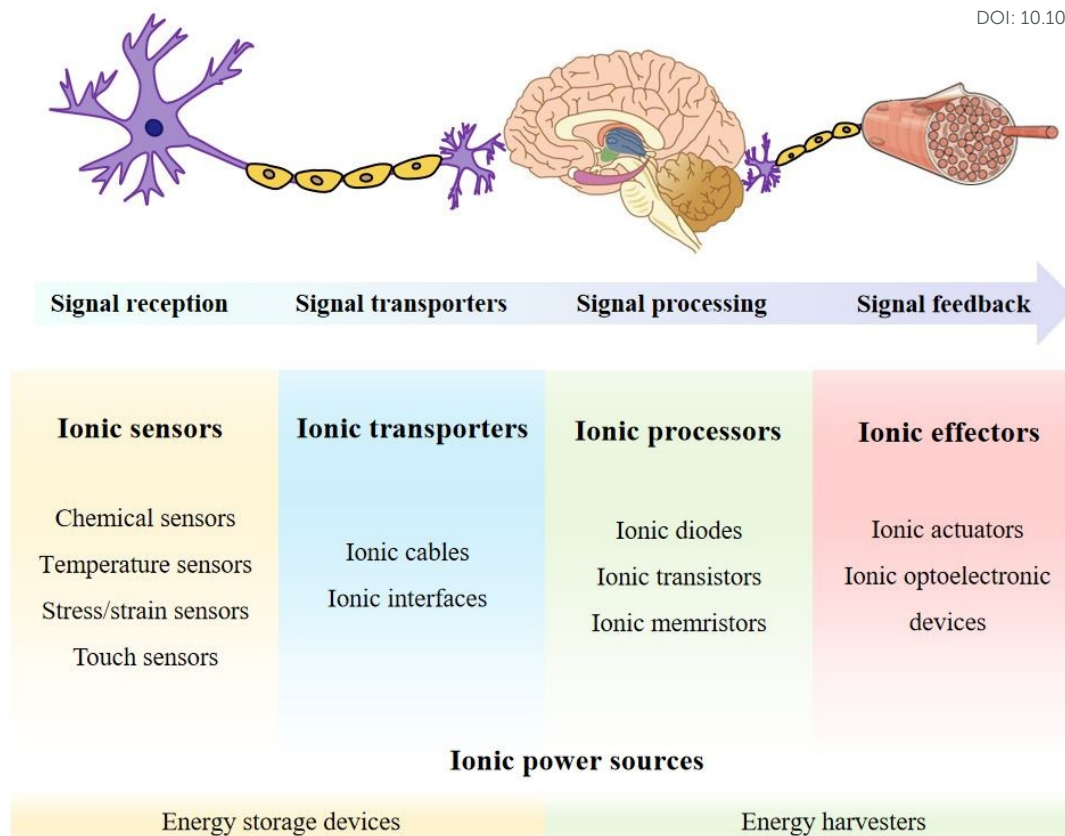
1 A hydrogel is a polymer containing plentiful water, and its high similarity with
2 biological tissue makes it an ideal material for preparing ionic sensory systems.^{37,38}
3 Hydrogel ionic sensory systems realize their functions through ionic motion and
4 arrangement^{39,40}. The high water content of a hydrogel enables it to carry mobile ions
5 for signal transmission. Moreover, contained ions can significantly increase the
6 conductivity of a hydrogel to ~10 S/m for sensing and signal transmission.^{41,42} Since
7 there are two types of ions with opposite charges in nature, the ions selective migration
8 and local accumulation of anions and cations can be achieved by adjusting the
9 electrostatic groups carried by the hydrogels. This means that the computing power of
10 ion hydrogel-based information processors may be unparalleled.

11 An elastic polymer network offers the possibility of making flexible, stretchable
12 ionic conductor equipment. The elastic modulus of hydrogel is 1-100kPa, which is close
13 to the softness of human tissue.^{43,44} At the same time, it can withstand large
14 deformations.⁴⁵ However, changes in the configuration of a polymer network and water
15 molecules rarely affect ionic conductivity.⁴⁶ Hence, hydrogel-based sensing elements
16 can tightly combine with the target and work normally in a stretched state, such as a
17 hydrogel sensor can compliantly attach to dynamic and irregular objects to obtain high-
18 fidelity data acquisition. On the other hand, the transmission and distribution of ions
19 sensitively respond to changes in the geometry of polymer network and environment,
20 thus giving the ionic hydrogel a powerful sensing function.^{47,48} At the same time, the
21 configuration of the polymer would also be affected by the transmission and
22 distribution of ions under the action of electric field, which endows the hydrogel with
23 electrical actuation function.

24 The polymer network does not scatter light, a hydrogel with high water content
25 (60-90 wt%) usually shows a refractive index (1.333) and transmittance as high as ~99%
26 similar to water.^{49,50} More importantly, because devices of hydrogel ionic sensory
27 systems normally consist of non-toxic hydrogels, those applications are much less
28 restricted in terms of biocompatibility, biodegradability, and ecological requirements.
29 These fascinating properties make the development of hydrogel ionic sensory systems
30 highly anticipated.

31 Herein, we discuss five essential components of hydrogel ionic sensory systems:
32 ionic sensors for signal reception, ionic transporters for signal transmission, ionic
33 processors for signal interpretation and processing, ionic effectors for signal feedback,
34 and ionic power sources for energy supply (**Figure 1**). We focus on device structures,
35 working mechanisms, functions, and potential applications. Prior to this, the transport
36 mode, modulation mode, the mechanism of transport of ions in hydrogels, and the
37 method to improve the charge transport in the hydrogel were elaborated to better
38 understand the working principle of hydrogel-based sensory system components. In
39 addition, future directions and key challenges related to hydrogel-based devices are
40 proposed.





1
2 **Figure 1.** Overview of perception-response loop of hydrogel ionic sensory systems.

3 **2. Transport of ions in hydrogels**

4 In biological systems, transmission of information is closely linked to accurate ion
5 migration. Based on the understanding of the physicochemical properties of both ions
6 and hydrogels, and the mechanisms of ion transport in hydrogels to carefully design
7 components, ion migration in the hydrogel ionic sensor systems would be more
8 controllable in terms of direction, velocity, and type.

9 **2.1 Mode of ion transport**

10 According to the types of ions that can move freely in the electric field, ion
11 transport is divided into two forms: the transport mode in which both anions and cations
12 can migrate in the electric field is called non-selective ion transport. Under the action
13 of an electric field, cations move to the negative electrode, and anions move to the
14 positive electrode (**Figure 2a**). But in selective ion transport, only one type of ions can
15 pass through ion-selective materials. The selective transport of ions can be achieved by
16 polyelectrolyte hydrogels, which are composed of fixed polymer chains with charges
17 and counterions with opposite charges (**Figure 2b**).⁵¹ When the electric field is applied,
18 counterions migrate directionally, and the polymer chains hinder the entry of ions with
19 the same charges as them. More complex information processing and sensing functions
20 can be achieved through accurate ion transportation.

21 **2.2 Mode of ion modulation**

22 There are two mechanisms involved in modulating ions in hydrogels: capacitive
23 process and faradaic process. In the capacitance model, ions and electrons accumulate



1 at the interface between hydrogels and metal electrodes to form an electric double layer
2 (EDL) for signal transmission and detection (**Figure 2c**). The faradaic process involves
3 redox reactions in which electrons are transferred between different substances (**Figure**
4 **2d**).⁵²

5 **2.3 Ion conductivity**

6 The ion transport capacity of hydrogels is quantified by ion conductivity, which is
7 the core performance of a hydrogel as an ion conductor. Ion conductivity σ reflects the
8 solvation/dissociation and migration of ions, in terms of the free ion number n_i and the
9 ionic mobility μ_i :⁵³

$$10 \quad \sigma = \sum n_i \mu_i Z_i e$$

11 where Z_i is the valence order of the ion, and e is the unit charge of electrons. The
12 concentration and mobility of carriers are related to the concentration of salts, type of
13 salts, and the properties of the polymer. We will discuss methods to improve the ion
14 conductivity of hydrogels from the following aspects:

15 *Salt concentration*

16 The reasonable increase in concentration of electrolyte salts would bring more
17 charge carriers to increase the conductivity. However, after a certain salt concentration,
18 the system exhibits a high viscosity that is harmful to ion migration, resulting in a
19 decrease in ion conductivity.^{54,55} Further increasing the salts content would cause the
20 ions to be unable to dissolve completely and even show the nature of the solid.

21 *Ionic types*

22 An ion exists in the form of a hydrate ion in the aqueous electrolyte, not form of
23 single ion.^{56,57} Reducing the hydrated water molecules around ions to increase the
24 concentration and migration speed of carriers is an effective strategy to improve ion
25 conductivity. Metal cations with higher charge density tend to attract more water
26 molecules, thereby reducing ion mobility.^{58,59} For example, the charge density of a
27 monovalent cation decreases with increasing ionic radii and its ionic conductivity is
28 directly proportional to the charge density.⁶⁰ The same pattern also exhibits in anions.
29 Another example, bulky-anions can effectively reduce the number of water molecules
30 around the cations, thereby promoting the generation and transportation of cations.⁶¹
31 For example, compared with ZnSO_4 , $\text{Zn}(\text{CF}_3\text{SO}_3)_2$ is more beneficial to Zn^{2+}
32 conduction.

33 *Medium of ion transport*

34 Ions are transported through polymer network and water in a hydrogel (**Figure**
35 **2e**). The hydrophilic functional groups of the polymer can bond with water molecules
36 through non-covalent interactions (such as hydrogen bonds and electrostatic
37 interactions). According to the strength of the interaction with the polymer chains,
38 water can be divided into three types, namely bound water, intermediate water and free
39 water.^{62,63} The interaction with polymer chains of free water is negligible. The mesh of
40 the polymer network is much larger than water molecules, so that the water can
41 maintain similar properties to bulk water. The high dielectric constant ($\epsilon=80$) and the
42 low viscosity of water (1 cP at 20 °C) facilitate the solvation of ions and increase ionic
43 mobility.

44 The polymer network provides an elastic backbone for a hydrogel, but it is an



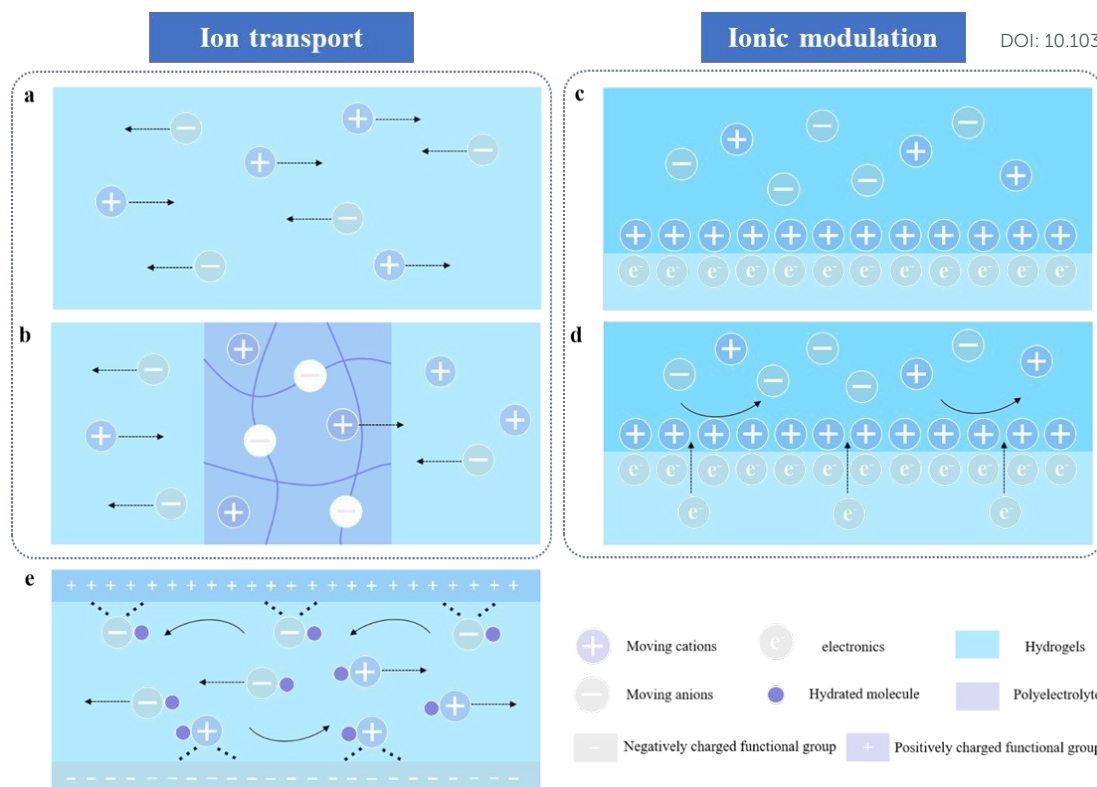
insulator. Compared with liquid electrolytes, the ionic conductivity of a hydrogel containing electrolytes would decrease significantly. Therefore, the hindrance of the polymer to ion migration should be reduced. For instance, reducing the mass fraction of the polymer can effectively improve conductivities.⁶⁴⁻⁶⁶ On the other hand, hydrogels have a porous structure. These pores of tens of nanometers allow water molecules to move freely and provide abundant channels for ion transport. So, enlarging the pore size of hydrogels is beneficial to ions transmission.^{67,68} As an example, a nanofibrillated cellulose/polyacrylamide hydrogel with enlarged pore sizes via nanofibrillated cellulose as a synthesis template has significantly improved ionic conductivity. As pore sizes are enlarged from 20-40 μm to 60-80 μm , the ionic conductivity also increases from 16.9 to 22.8 mS cm^{-1} .⁶⁷ Another example, using hydroxypropyl cellulose to expand the pore size and significantly increased the conductivity of the hydroxypropyl cellulose / polyvinyl alcohol hydrogel from 1.7 to 3.4 S m^{-1} .⁶⁸ It should be noted that although increasing the pore size can promote ion transport, this comes at the cost of sacrificing the mechanical integrity and structural stability of the hydrogel network.⁶⁹ Hydrogels with high porosity usually have poor mechanical durability and are prone to collapse due to dehydration, which limits their long-term applications.⁷⁰

The functional groups carried by the polymer also can affect the migration behavior of the ions by interacting with the ions. As an important example, a zwitterionic electrolyte hydrogel composed of repeating units of anion and cation charged groups shows unique advantages in this regard. The rich charged groups not only can be highly hydrated with water molecules to induce the hydrogel strong water retention ability, but also provide hopping migration channels to greatly improve the ion conductivity.⁷¹⁻⁷³ In the meantime, the cations and anions charge on the same molecule can induce the dissociation of the introduced salt through electrostatic interaction, thereby increasing the number of free ions to improve conductivity.⁷⁴ The energy storage devices based on the outstanding advantages of the polyelectrolyte hydrogel electrolyte exhibit a remarkably impressive capacitance and rate capacity.^{71,75,76}

Weakening the interaction between polymers and ions is also an effective way to improve ion conductivity.^{77,78} For instance, decreasing the oxygen content in the polymer can reduce the coordination effect between Li^+ and the polymer, which leads to enhance Li^+ conduction.⁷⁸ However, polymer-ion interactions present a delicate balance. Weakening coordination between polymer chains and ions improves ionic mobility, but excessively weak interactions may reduce ion selectivity and lead to poor signal fidelity.⁷⁹ In contrast, strong electrostatic interactions can enhance ion selectivity and dissociation, yet may introduce slower response due to ion trapping.⁸⁰

Besides, the Vogel-Tamman-Fulcher (VTF) relationship reveals the connection between T_g and ionic conductivity of a polymer: a polymer with a lower T_g has a higher ionic conductivity.⁸¹⁻⁸⁴ The amorphous feature of a hydrogel with lower crystallinity can effectively enhance the ionic conductivity due to the flexibility of chain segmental motion.⁸⁵ For example, introduced additives or polymer modifications have been used to reduce the crystallinity of hydrogels to increase ion conductivity.^{65,86-89}





1 **Figure 2.** Transport of ions in hydrogels. a-b) Schematic illustration of two modes of ion transport:
 2 a) In non-selective ion transport, both anions and cations can move directionally through an electric
 3 field; b) Schematic diagram of selective ion transport, such as a cation selective system, only cations
 4 can be transported through a selective membrane. c-d) Two types of ionic modulation: c) Schematic
 5 illustration of the electrical double layer (EDL) formation. Cations and electrons accumulate on both
 6 sides of the interface between the hydrogel and the electrode, respectively. d) Schematic diagram of
 7 the faraday process. Electrons can pass through the interface between the hydrogel and the electrode
 8 to participate in the redox reaction. e) Schematic illustration of the transport process of ions in the
 9 hydrogel. The transport process involves solvation of ion and polymer-ion interaction.

11 3. Hydrogel ionic sensors

12 Using sensors to receive external stimuli is the beginning of a sensing process. A sensor
 13 is a machine that converts detected signals into electric signals.^{90,91} The human body
 14 harbors numerous sensors that enable us to perceive five delicate sensations, including
 15 hearing, sight, touch, taste, and smell.⁴⁹ Similarly, through careful design, hydrogel
 16 ionic sensors can be used to effectively monitor a wide range of signals and convert
 17 them into the desired signal output. Hydrogel ionic sensors rely on the high compliance
 18 of the hydrogels, changes in the spatial distribution and the movable speed of ions to
 19 work.

Functional materials	Sensor types	Sensitivity	Sensing range	Stability (cycles)	Refs.
Hydrogels	Pressure sensors	3.06 kPa ⁻¹	0 ~ 380 kPa	2000	Zhuo et al. ⁹²
	Strain Sensor	GF = 4.9	0-800%	3000	Cui et al. ⁹³
	Pressure sensors	0.013 kPa ⁻¹	1.3 kPa ~ 70 kPa	20000	Pu et al. ⁹⁴



Nano materials	Strain Sensor	GF = 2.26	1% ~ 300%	500	Wang et al. ⁹⁵
	Gas sensor	0.24	1 ~ 40 ppm	>1000	Chen et al. ⁹⁶
	Strain sensor	GF ≥ 14000	0 ~ 900%	2000	Yang et al. ⁹⁷
	Pressure sensor	2048 kPa ⁻¹	100 kPa	1000	Tian et al. ⁹⁸
	Temperature sensor	0.142% °C ⁻¹	25 ~ 50 °C	5000	Gandla et al. ⁹⁹
Liquid metal	Strain sensor	GF = 4.91	320%	500	Chen et al. ¹⁰⁰
	Tactile sensor	(2~20) × 10 ⁻³ kPa ⁻¹	2 ~ 400 kPa	500	Yeo et al. ¹⁰¹
	Pressure sensor	39% kPa ⁻¹	25 kPa	6000	Zhang et al. ¹⁰²
	Strain sensor	GF = 8.35	0 ~ 120%	10000	Yao et al. ¹⁰³

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Table 1. Comparison of the performance of different ionic sensors based on hydrogels, nano materials and liquid metal.

3.1 Chemical sensors

Hydrogel-based chemical sensors convert external chemical stimuli into visual electrical signals. Chemical sensors can be divided into two categories according to whether they react with the detected substances.

Chemical sensors that react with substances affect ion conductivity by changing the structure of the pores and the interaction with ions of the polymer. Inspired by the tongue, the astringency sensor was constructed by using polyacrylamide containing mucin and lithium chloride (LiCl).⁷⁷ When exposed to astringent compounds, the microporous hydrogel is converted into micro/nanoporous structures with hydrophobic aggregates due to the binding of the contained mucin to incoming molecules (**Figure 3a-b**). This is conducive to shortening diffusion paths and frictionless sliding of ions, thus improving ion conductivity to detect astringent substances.

For chemical sensors that do not react with the detection substances, ionic hydrogels can respond to changes in humidity and gas composition, because these changes are closely related to the hindering effect of ion transport. This mechanism makes it possible to use ionic hydrogels as gas or humidity sensors. For example, NH₃ and NO₂ can form hydrogen bonds with the oxygenated functional groups of the polymer network and be effectively absorbed by the hydrogel. These absorbed gases impede the movement of ions, resulting in an increase in resistance (**Figure 3c**).¹⁰⁴ Hydrogels can also effectively absorb moisture in the air, and the increase in humidity can help hydrogels absorb more water and cause swelling. This reduces the density of the polymer network to alleviate the hindrance of ion migration, resulting in a decrease in the resistivity of the hydrogel (**Figure 3d**).¹⁰⁵ The above-mentioned mechanism provides inspiration for the preparation of ionic hydrogel-based gas sensors and humidity sensors for air quality detection. As another innovative chemical sensor, parallel electrode arrays can use the capacitance response to distinguish different liquid molecules (**Figure 3e-f**).¹⁰⁶ The liquid to be detected forms a bridging electrode, which leads to an increase in the capacitance signal. The strength of the increasing capacitance is closely related to the polarity of the liquid molecules, while the duration of increasing capacitance is controlled by volatility and wettability. Consequently, different liquids can be identified according to the strength and duration of the increased capacitance.



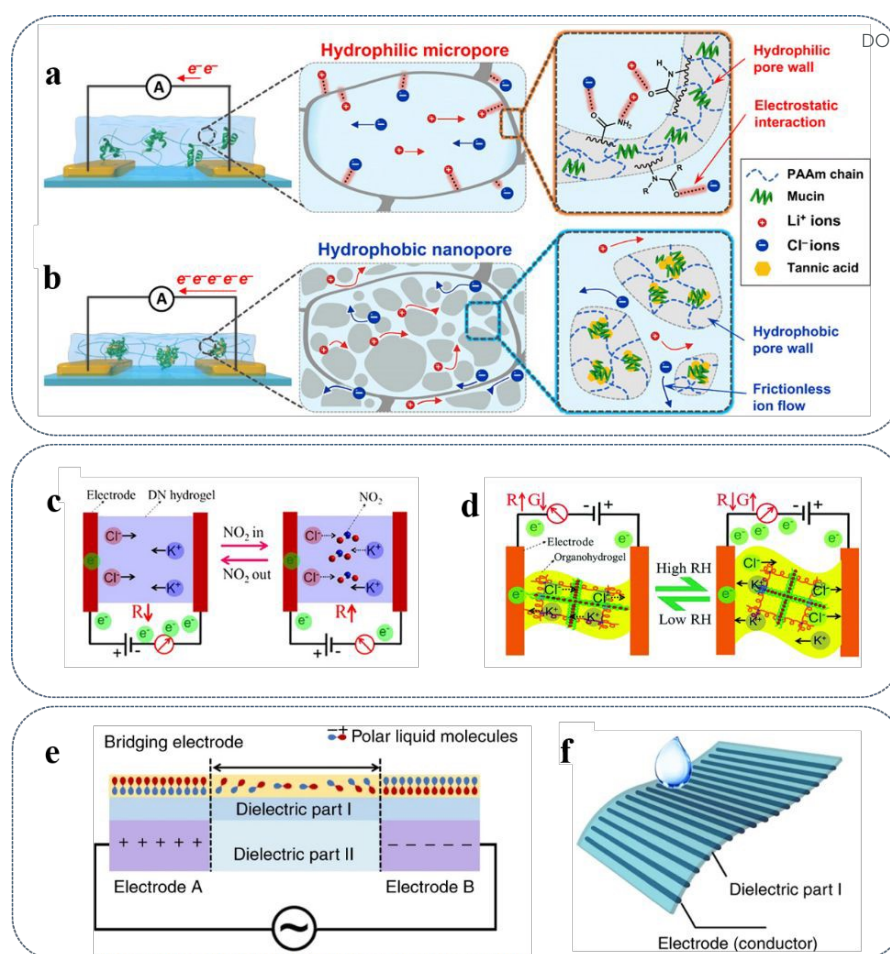


Figure 3. Hydrogel ionic chemical sensors. a-b) Schematic illustration of the working principle of an astringency sensor. a) Before contacting astringent substances, the hydrogel has hydrophilic micropores and electrostatically charged pore walls that can limit ion transport.⁷⁷ b) After contacting with astringent substances, the hydrogel exhibits hierarchical micro/nanopore that can shorten diffusion paths, and hydrophobic pore walls that can facilitate ion sliding.⁷⁷ c) Scheme depicting the working mechanism of a NO₂ sensor. The NO₂ molecules absorbed by a hydrogel hinder the movement of ions and increase the resistance.¹⁰⁴ d) Schematic illustrating of humidity sensing mechanism. The moisture in the air absorbed by a hydrogel reduces the density of the polymer network, thereby facilitating ion migration.¹⁰⁵ e) Schematic illustration of a capacitive sensors for liquid recognition. Liquid acting as a bridging electrode would increase capacitance. The characteristics of the increased capacitance signals reflect the properties of the liquid.¹⁰⁶ f) A 3D-printing capacitive liquid recognition sensor.¹⁰⁶

3.2 Temperature sensors

The electrical signals of the hydrogel-based temperature sensors respond to changes of temperature. Changes of temperature would affect the migration rate of ions and the volume of temperature-sensitive hydrogels. Therefore, temperature sensors are mainly divided into two types: resistive temperature sensors based on the temperature-conductivity relationship and capacitive temperature sensors based on the temperature-volume relationship.¹⁰⁷

The resistance of an ionic hydrogel is sensitive to temperature, as the increase in temperature would accelerate the migration of ions while favouring their



dissociation.¹⁰⁸ Therefore, the resistance of a hydrogel decreases with increasing temperature. On this basis, the variations in temperature can be detected by measuring the resistance of a hydrogel (**Figure 4a**).¹⁰⁹⁻¹¹¹ Studies have shown that increasing the initial resistance of ionic hydrogels is beneficial to improve the temperature sensitivity.¹¹² Because the larger initial resistance means that the migration of ions suffers more barrier. At elevated temperatures, ions can overcome the obstacles of the network and move freely. Consequently, the effect of resistance reduction will be relatively more evident. On the other hand, the introduction of organic solvents with smaller specific heat capacity is also an effective strategy to enhance the temperature sensitivity of hydrogels.^{112,113} When absorbing the same amount of heat, the hydrogel containing a solvent with a lower specific heat capacity has a higher temperature rise, resulting in more noticeable changes of resistance. For example, glycerin is introduced into the hydrogel by solvent replacement method to improve temperature sensitivity.¹¹² Compared with hydrogels, the thermal sensitivity of glycerin organohydrogels has increased significantly from 2.95%/°C to 19.6%/°C. Notably, the organohydrogel temperature sensor exhibited an apparent response to the gentle touch of hot objects and non-contact sensation (**Figure 4b**).

A capacitive temperature sensor is formed by sandwiching a piece of dielectric by two pieces of thermo-responsive hydrogel (**Figure 4c**).¹¹⁴ When the temperature reaches the critical temperature, the volumetric variations of the thermo-responsive hydrogels influence the distance of electrodes and contact area of hydrogels and a dielectric elastomer, thus altering the capacitance (**Figure 4d**).

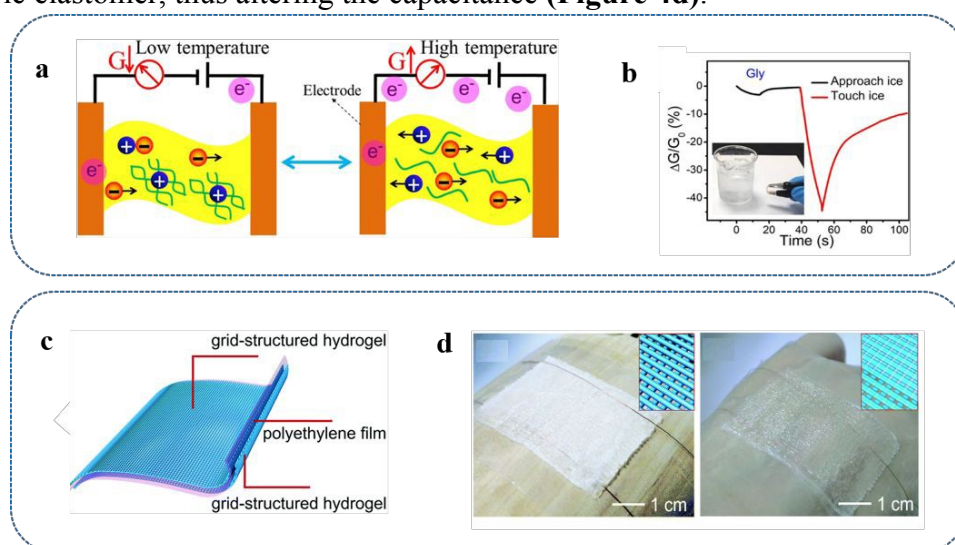


Figure 4. Hydrogel ionic temperature sensors. a) Schematic illustration of the working principle of a resistive temperature sensor. The increase in temperature facilitates to improve migration rate of ions and the dissociation of ions, thereby reducing resistance.¹¹² b) Real-time response of temperature sensor to approach and touch with ice water.¹¹² c) Schematic diagram showing a capacitive temperature sensor based on thermally responsive hydrogel electrode.¹¹⁴ d) When the critical temperature is reached, the capacitance of the sensor would increase due to the expansion of the hydrogel.¹¹⁴

3.3 Stress/strain sensors

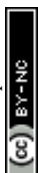
Ion-conducting hydrogel-based stress/strain sensors convert mechanical stimuli into



1 electrical signals. By monitoring the applied stress and strain, the operating state of the
2 target can be judged to ensure its normal operation. From a theoretical perspective, such
3 electromechanical responses originate from coupled ion transport and interfacial
4 electrochemical processes. Specifically, the redistribution of mobile ions under
5 deformation and applied electric fields can be described by the Poisson–Nernst–Planck
6 (PNP) framework, which governs ion flux driven by concentration gradients and
7 electric potential.¹¹⁵ In addition, the formation of electrical double layers (EDLs) at
8 hydrogel–electrode interfaces plays a dominant role in determining the effective
9 capacitance and signal output.¹¹⁶ According to the working mechanism, hydrogel
10 stress/strain sensors can be divided into two categories: capacitance mode and
11 resistance mode.¹¹⁷

12 A dielectric elastomer is sandwiched between two ion-conducting hydrogel layers
13 to form a capacitive sensor. The capacitance of EDL formed at the interface of a
14 hydrogel and an electrode is much larger than that of a dielectric elastomer. Therefore,
15 the equivalent circuit is close to the capacitance of the dielectric elastomer. The
16 capacitance value is determined by $C = \epsilon S/d$, in which ϵ , S and d are the permittivity,
17 contact area, and thickness of the elastomer, respectively. If pressure is applied or the
18 sensor is stretched, the thickness of the sensor decreases and the area increases,
19 increasing the capacitance.¹ Thus, when the sensor deforms under an external force,
20 stress and strain can be measured by the change of capacitance. The relationship
21 between deformation and capacitance is predictable. When it is stretched by λ times,
22 the original capacitance C_0 of the capacitive strain sensor and the capacitance after
23 stretching C follow the linear relationship of $C = C_0 \lambda$.¹¹¹

24 For resistive sensors, a hydrogel can be regarded as a resistor, varying the
25 configuration of polymer network and water molecules would cause changes in the
26 resistance of a hydrogel (**Figure 5a**). Because the resistance of the hydrogel is $R =$
27 $\rho l/A$,¹¹⁸ which is determined by the resistivity (ρ) and geometry (cross-sectional area A ,
28 the length l). Deformation has negligible effect on the ion conductivity of the
29 hydrogel.^{43,46} Therefore, the resistance of the hydrogel can respond to the deformation.
30 By connecting the ion-conducting hydrogel to the resistance tester, stress and strain can
31 be measured by testing the change in resistance. The change of resistance after
32 deformation of a resistive sensor is also predictable: the resistance R scales as $R = R_0 \lambda^2$
33 (R_0 is the initial resistance).¹¹⁸ The performance of resistive sensors is mainly attributed
34 to the properties of the hydrogel, so its relatively simple structure allows the advantages
35 of the hydrogel to be directly demonstrated. For instance, the high stretchability of
36 hydrogel allows resistive sensors to measure large deformations and a self-healing
37 hydrogel can effectively improve the durability of a sensor without being limited to the
38 properties of other composite materials (**Figure 5b**).¹¹⁹ Some resistive sensors are
39 capable of exhibiting bidirectional piezoresistive effects. For instance, a vitrimer
40 heterogel (VHG) with bicontinuous structure demonstrates switchable hardness-gated
41 ion transport pathways.¹²⁰ This behavior is closely associated with its two primary,
42 mechanically tunable states, namely the stiff and soft states (**Figure 5c**). A wide
43 pressure response range and a tunable detection limit are critical for the performance of
44 resistive sensors. For example, multiphase ionogels constructed by elastic ionogel



1 frameworks and switchable poly(stearyl methacrylate) (PSMA) micro-inclusions
2 exhibit pronounced stiffness-memory behavior.⁹² As a result, resistive sensors derived
3 from these ionogels demonstrate programmable pressure-response characteristics and
4 wide pressure range (**Figure 5d**), making them well suited for operation across diverse
5 application scenarios.

6 Conducting resistance detection on a piece of hydrogel in a classic structure of
7 parallel-plate capacitive sensor can achieve simultaneous detection of capacitance and
8 resistance (**Figure 5e**). The capacitance sensor is only sensitive to deformation, and the
9 resistive sensor is sensitive to temperature and deformation at the same time, so a sensor
10 system that can sense multiple stimuli is constructed (**Figure 5f,g**).¹¹¹

11 Resistive sensors typically exhibit higher sensitivity owing to the strong
12 dependence of electrical resistance on ion mobility and the deformation of conductive
13 pathways within the hydrogel network.¹²¹ However, this mechanism also makes them
14 more susceptible to environmental factors such as temperature, humidity, and ion
15 concentration, leading to signal cross-sensitivity and reduced stability.¹²²

16 In contrast, capacitive sensors primarily rely on changes in geometric
17 configuration and dielectric properties, making their signal output less sensitive to
18 fluctuations in ionic transport. This inherent decoupling from ion mobility variations
19 contributes to improved signal stability and reproducibility, particularly in complex or
20 dynamic environments.¹²³ Nevertheless, their sensitivity is typically lower, particularly
21 under small deformations, due to the relatively weak dependence of capacitance on
22 structural changes.

23 From a materials design perspective, resistive sensors typically require highly
24 conductive and deformable ionic networks to maximize sensitivity, whereas capacitive
25 sensors depend on mechanically robust dielectric layers and well-defined interfacial
26 structures to ensure stable signal output.¹²⁴ Consequently, the choice between resistive
27 and capacitive sensing mechanisms involves an inherent trade-off between sensitivity
28 and signal stability, which should be carefully optimized according to specific
29 application requirements.¹²²

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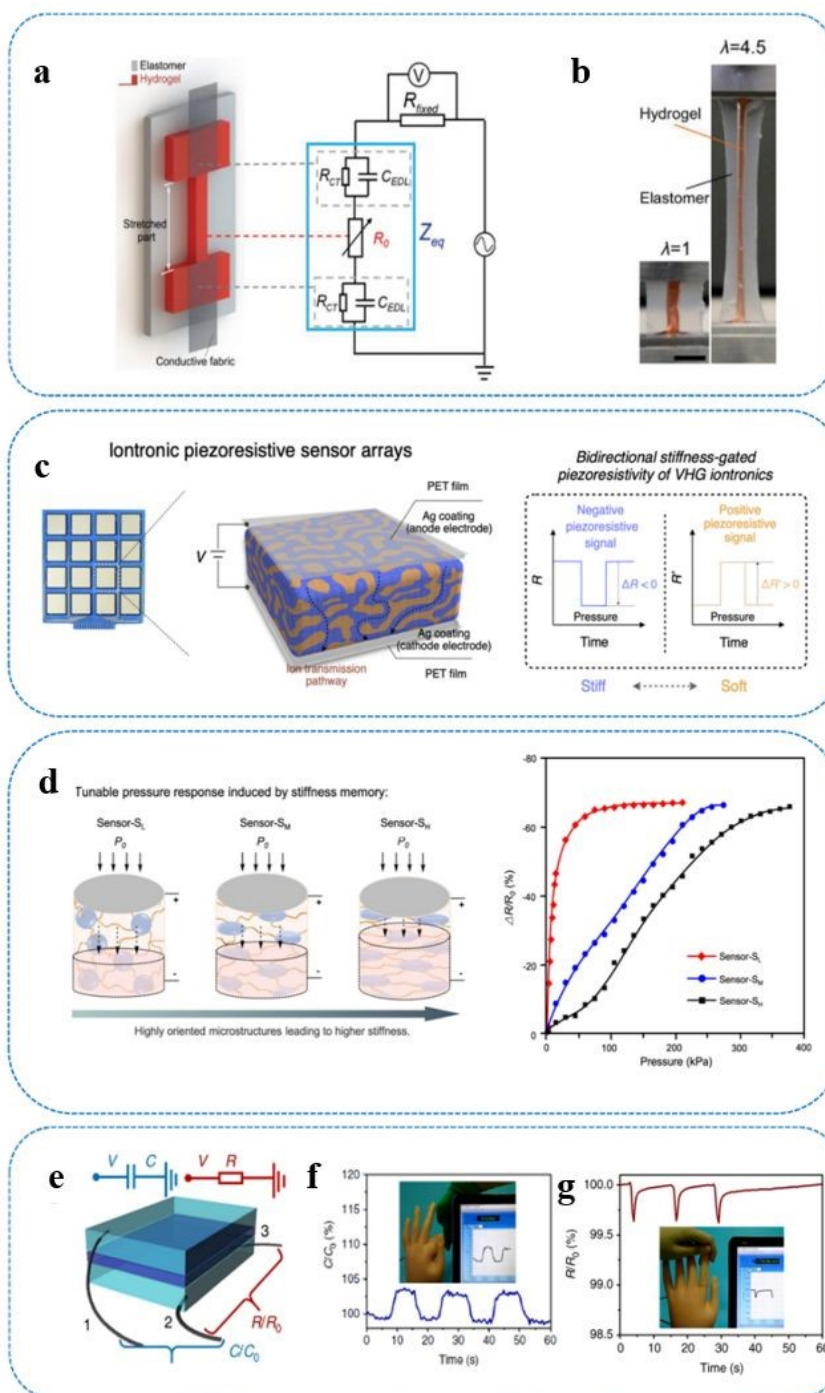
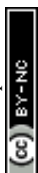


Figure 5. Hydrogel-based ionic stress/strain sensors. a) Schematic of a resistive stress/strain sensor. The resistance of the hydrogel increases with stretching.¹¹⁸ b) A resistive stress/strain sensor can withstand large deformations.¹¹⁸ c) Schematic diagram of VHG iontronic piezoresistive sensor arrays that had the capability of the bidirectional stiffness-gated piezoresistivity.¹²⁰ d) Schematic diagram showing the pressure-response change with varied stiffness of the heterophasic ionogels.⁹² e) Schematic diagram of a sensor that monitors both capacitance and resistance.¹¹¹ f) The capacitive signals respond to changes in strain. When a sensor monitors the bending of the finger.¹¹¹ g) The resistive signals respond to changes in temperature when a finger touches the sensor.¹¹¹

3.4 Touch sensors

Hydrogel-based touch sensors are used to detect physical contact, and they can



1 determine the occurrence, location, and even proximity of objects. The sensing
2 mechanism can be further understood from an interfacial electrochemical perspective.
3 Mechanical stimuli induce spatial redistribution of ions and modulation of the electrical
4 double layer structure, which directly affects interfacial capacitance and impedance.¹²⁵
5 Such processes are governed by ion transport dynamics and interfacial charge screening
6 effects, highlighting that the sensing behavior is intrinsically controlled by
7 electrochemical–mechanical coupling rather than purely geometric deformation.¹²⁶
8 Based on ionic hydrogels, next-generation stretchable and biocompatible touch sensors
9 can be developed. This system can easily and intuitively realize human-machine
10 interaction by combining a hydrogel touch sensor and a display. Hydrogel-based touch
11 sensors can be fallen into two categories: capacitive sensing and electrostatic induction
12 sensing.¹²⁷

13 A touch sensor adopts a surface capacitive system, in which the same voltage is
14 applied to all corners.¹²⁸ When a grounded conductor touches the surface of the
15 hydrogel, a potential difference is generated between the electrode and the touch point
16 (**Figure 6a**). Current is generated between the electrode and the grounded conductor.
17 The closer to the contact point, the current will be greater. The location of the touch
18 point is determined by comparing the magnitude of the current. Thanks to the high
19 stretchability, high transparency and low modulus of hydrogels, touch sensors can be
20 comfortably attached to the human body to provide a visual man-machine interface
21 (**Figure 6b**). The touch sensor is further endowed with pressure-sensitive adhesiveness
22 and self-healing properties to improve the practical performance via an innovative
23 hydrogel.^{129,130}

24 The triboelectric nanogenerators (TENGs) rely on the coupling effect of contact
25 electrification and electrostatic induction to convert mechanical motion into changes of
26 electrical signals.^{131,132} TENGs usually have four working modes, namely contact-
27 separation mode, sliding mode, freestanding mode, and single electrode mode.^{133,134}
28 The recently developed the TENG based on an ion-conducting hydrogel has natural
29 flexibility, high transparency and high stretchability, which is a breakthrough for energy
30 harvesters with fixed shapes.⁹⁴ The hydrogel-based TENG adopts single-electrode
31 mode, in which elastomer and hydrogel serve as the electrification layer and electrode.
32 The hydrogel electrode is encapsulated in an elastomer dielectric cell and connected to
33 an external load by metal. As we all know, when two objects touch, electron transfer
34 would occur at the interface. But due to the coincidence of the interface, the two objects
35 show electrical neutrality. When the interface is separated, the two objects is charged
36 with an equal amount of opposite owing to charges the redistribution of electrons. This
37 contact charging phenomenon would exist in the insulator for a relatively long time,
38 thus generating static electricity. When the dielectric contacts TENG's elastomer,
39 electrons are transferred. When the dielectric leaves the elastomer, the static charges
40 can induce the oppositely charged ions in the hydrogel to accumulate at the interface to
41 balance the static charges. At the same time, the EDL is formed at the metal / hydrogel
42 interface. Electrons flow from the metal wire to the ground would form a current until
43 all the static charges in the elastomer are screened. If the moving dielectric returns to
44 the elastomeric state, a current in the opposite direction is generated. By repeating this



1 process, alternating current will be generated (**Figure 6c**). Sensors based on
2 electrostatic induction do not require external power source, which is conducive to its
3 application in mobile devices (**Figure 6d**).

4 Humans obtain the tactile sensation by detecting changes in friction, roughness,
5 hardness, and temperature.¹³⁵ In contrast, the hydrogel-based touch sensors rely on
6 capacitive and electrostatic induction, so it exhibits a proximity-sensing function that
7 surpasses the human body's own tactile sensation. In order to sense the proximity of an
8 object, a capacitive touch sensor is assembled using ionic hydrogel-based electrode
9 array containing disc-shaped and ring-shaped electrodes separated by elastomers.¹³⁶
10 The coupling of the two electrodes generates a vertical electric field. A nearby object
11 can reduce the coupling between the electrodes and be induced. The hydrogel electrodes
12 enable the sensor to detect touch during bending and stretch, and can detect the position
13 of multiple objects (**Figure 6e-f**). Similar to the phenomenon of contact electrification,
14 objects close to the sensor can also cause electrostatic induction to generate induced
15 currents to monitor the presence of objects (**Figure 6g-h**).¹³⁷

16 For hydrogel-based touch sensors, maintaining high signal fidelity is particularly
17 challenging due to their strong coupling with external environments and complex
18 stimuli. Ionic hydrogel systems are inherently sensitive to ambient humidity, which can
19 significantly alter their electrical properties and cause pronounced signal drift and
20 instability in sensing outputs.¹³⁸ Moreover, variations in contact conditions, such as
21 pressure distribution and electrode–hydrogel interface fluctuations, can introduce
22 additional noise and reduce the accuracy of spatial or proximity sensing, as these factors
23 dynamically modulate interfacial impedance and effective contact area, thereby
24 generating signal artefacts and degrading signal-to-noise ratio.¹³⁹ In high-density sensor
25 arrays, undesired signal interference and crosstalk between adjacent sensing units
26 further degrade signal clarity, posing challenges for precise touch localization.
27 Additionally, the capacitive nature of ionic systems can amplify low-frequency noise
28 and slow response dynamics, limiting their ability to faithfully capture rapid or subtle
29 touch events. Consequently, strategies to decouple environmental effects, stabilize
30 interfaces, and suppress signal interference are crucial for advancing high-fidelity ionic
31 touch sensing systems.¹⁴⁰



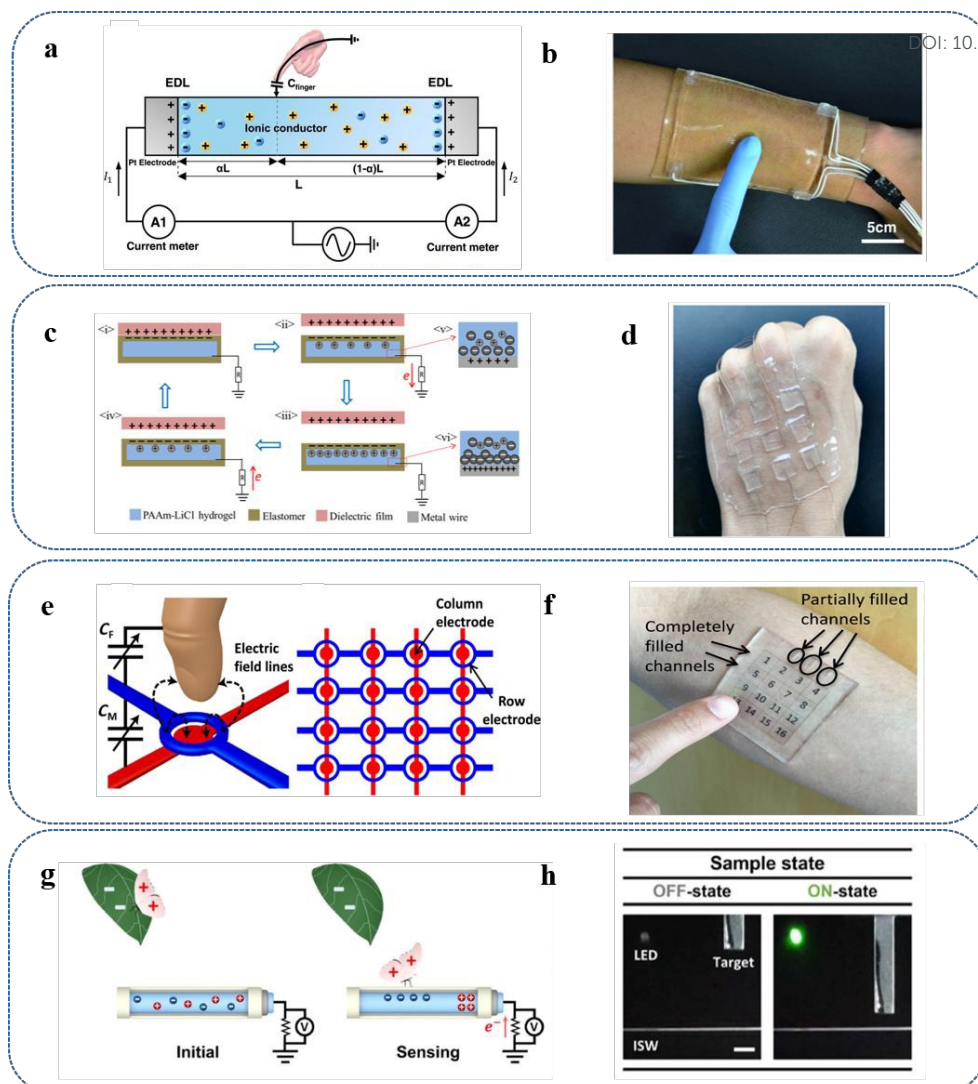


Figure 6. Hydrogel-based ionic touch sensors. a) Schematic diagram showing of a surface-capacitive touch sensor. The touched position of the ground conductor is confirmed by the current caused by the potential difference between the touch point and the electrode.¹²⁸ b) The ionic touch sensor was attached to an arm for operation.¹²⁸ c) Schematic diagram showing the working mechanism of a triboelectric touch sensor. When an object touches the sensor, contact electrification occurs at the interface. Detaching and attaching of a charged object induces voltage across the external load by electrostatic induction.⁹⁴ d) Schematic diagram of a triboelectric touch sensor with 9 pixels attached on a hand.⁹⁴ e) Schematic diagram showing of a capacitive proximity sensor. The coupling disc-shaped and ring-shaped electrodes generate a vertical electric field to detect the approaching of objects.¹³⁶ f) The transparent sensor array combined with a printed number pad is attached to the arm.¹³⁶ g) Schematic illustration of the working principle of a proximity sensor based on electrostatic induction. When an object approaches the sensor, the sensor will generate electrostatic induction to detect the object.¹³⁷ h) Using a pre-programmed circuit, a LED is turned on when the induced voltage generated by the approaching object reaches the threshold.¹³⁷

4. Hydrogel ionic transporters

Humans coordinate the functions of sensing, deciding, and acting in different parts through long-distance and rapid ion transport in nerve fibers. Hydrogel ionic sensory



1 systems rely on ion transporters to realize the interconnection of ionic sensors, ionic
2 processors, and ionic effectors. An ion transporter is a connection element based on ion
3 conduction. It is not only the link among the components of ionic sensory systems, but
4 also the interface between electronic and biological systems.

5 **4.1 Ionic cables**

6 Hydrogel ionic cables enable transport signals over long distances and at high
7 speeds.

8 An ion cable based on electric double layer mimics the function of an axon, which
9 consists of two layers of hydrogel separated by a layer of elastomer (**Figure 7a**).¹⁴¹
10 Based on the advantages of the structure, the migration of ions in the ionic cable only
11 occurs locally and does not require passing through the two electrodes. An increase of
12 16 orders of magnitude in ionic diffusivity enables the ionic cable to transmit an action
13 potential of stable amplitude at a speed over 100 m/s, and can transport a signal up to
14 100 MHz within 10 cm (**Figure 7b**). It is worth noting that, due to the limitation of the
15 electrochemical window of the hydrogel, the ionic cable can only be applied under an
16 AC voltage with an amplitude within 1 V. The ion conductive hydrogel provides high
17 transparency to the ionic cable, while allowing it to function normally when stretched
18 more than eight times. Proportionally reducing the device size will not affect the
19 performance of the ionic cable. These interesting properties pave the way for stretchable
20 ion propagation technology and its miniaturization.

21 Signal transduction in organisms involves ion transport through nanochannels.¹⁴²⁻
22 ¹⁴⁵ Studies have shown that ion transport in a charged channel with a size equivalent to
23 the Debye length exhibits a different nanofluidic transport behavior from the whole.¹⁴⁶
24 In order to simulate the ion migration behavior of biological systems, densified natural
25 materials with surface charges are used to obtain hydrogel ionic cable with
26 nanochannels for ion transport.¹⁴⁷⁻¹⁵¹ The densified narrow nanochannels are conducive
27 to the rapid migration of ions. On the other hand, the charges on the surface of the
28 nanochannels facilitate the enrichment of the conductive ions, thus showing an increase
29 in ion concentration (**Figure 7c**). At low ion concentrations, hydrogel ionic cable with
30 nanochannels exhibits a conductivity that is 90 times higher than bulk solutions (**Figure**
31 **7e**).¹⁴⁸



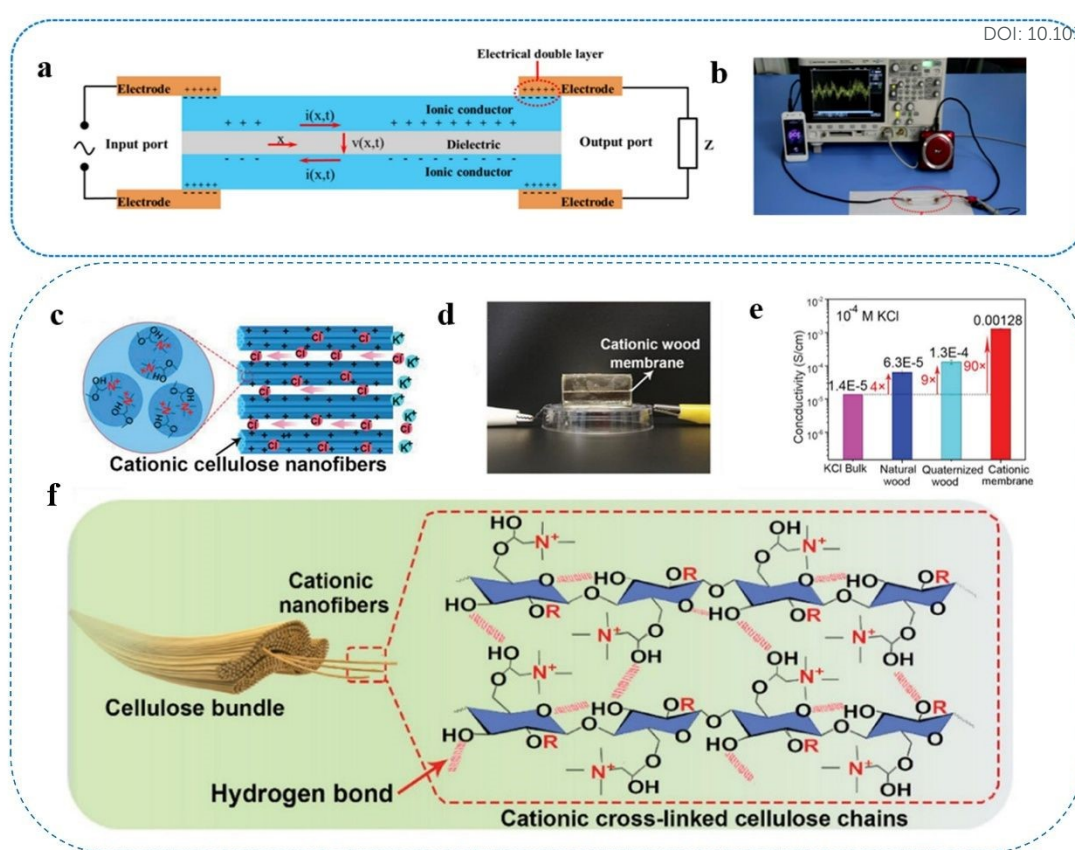
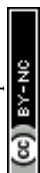


Figure 7. Hydrogel-based ionic cables. a) Schematic diagram of working mechanism of ionic cable. Ions migrate locally in the cable without crossing two electrodes.¹⁴¹ b) Ion cables were used to transport cell phone signals to speakers and oscilloscopes.¹⁴¹ c) Rapid and selective migration of ions in the densified hydrogel cable with nanochannels.¹⁴⁸ d) Device diagram for testing ion conductivity of hydrogel cable with nanochannels.¹⁴⁸ e) The densified hydrogel cable with nanochannels has a significantly improved conductivity.¹⁴⁸ f) Schematic of the hydrogen bonding among the molecular chains of the cationic wood membrane.¹⁴⁸

4.2 Ionic interfaces

An ionic interface is used for ion transport between electronic systems and biological systems. At the fundamental level, ionic–electronic interfaces are governed by interfacial electrochemical physics. When an ionic conductor contacts an electronic conductor, mobile ions accumulate near the interface to form electrical double layers, which act as nanometer-scale capacitors and dominate charge transfer behavior. The structure and dynamics of these EDLs, together with ion migration in the bulk electrolyte, can be described using continuum ion transport theories such as the Poisson–Nernst–Planck model.^{115,116} Utilizing an ionic interface can record various bioelectronic signals, and stimulate the biological systems simultaneously. On the other hand, ionic interfaces provide a reliable and safe connection mechanism that helps patients with sensory dysfunction integrate artificial organs into the human body. Thus, ionic interface technology realizes bidirectional communication between machines and living beings.

Commercial wearable and implantable electronic devices can collect and transmit biological signals in different parts of the human body and apply them to clinical



1 diagnosis and treatment, which has achieved great success.^{152,153} However,
2 conventional electronic devices rely on rigid metal materials that are severely
3 mismatched with the physical and mechanical properties of human tissue. Therefore,
4 they are prone to high interface impedance due to loose contact when used *in vitro*;^{154,155}
5 when they are implanted, cells will wrap the interface and block the signal¹⁵⁶ and even
6 produce an inflammatory response.¹⁵⁷ At the same time, because electronic equipment
7 and the human body have different signal carriers, the threshold voltage required for
8 the conversion of electrons to ionic current at the biological interface often causes pain
9 and damage to biological tissues.^{158,159} Due to the inherent biocompatibility, the
10 modulus that matches the human tissue, and the ideal ionic conductivity, ionic
11 hydrogels effectively eliminate the problems of current conversion and mechanical
12 mismatch between the electrode and the tissue.^{31,160} Hence, they have been widely used
13 in non-invasive applications, such as wearable devices and epidermal electrodes.¹⁶¹⁻¹⁶³
14 However, ionic hydrogels can exchange substances with the internal tissue media of
15 the human body (e.g., ion leakage caused by differences in ion concentration), which
16 limits their application in invasive devices.^{158,164}

17 Recently, the signal monitoring and stimulation of the mouse brain has been
18 achieved via an ion-conducting hydrogel encapsulated by an elastomer (**Figure 8a**).¹⁶⁵
19 The hydrogel contains artificial cerebrospinal fluid (ACSF), which helps the hydrogel
20 to simulate the living environment of neurons chemically, mechanically, and
21 electrically to reduce rejection. The frozen elastomer-sealed hydrogel interface can be
22 easily inserted into the brain of a mouse and collect nerve signals after thawing. Thanks
23 to the high light transmittance of the hydrogel, a single fiber of the hydrogel can
24 simultaneously perform optical stimulation (**Figure 8b**). Neuro-compatible hydrogel
25 interfaces have great potential in clinical applications.

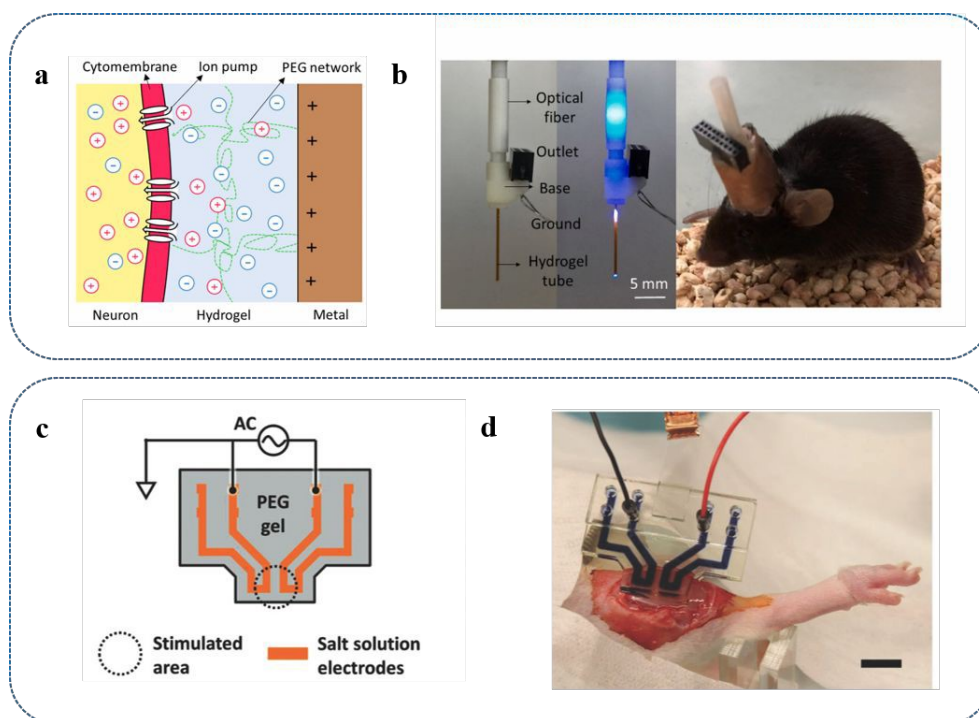
26 On the other hand, the aqueous two-phase system (ATPS) composed of a
27 polyethylene glycol hydrogel and salt solution can improve the stability of ions *in*
28 *vivo*.¹⁵⁸ A polyethylene glycol hydrogel can provide a flexible conductive shell in direct
29 contact with tissues, and the separated salt solution contained in the hydrogel provides
30 high ionic conductivity without ion diffusion into the surrounding tissue media (**Figure**
31 **8c**). When a voltage is applied, the induced ionic current will be transmitted in the
32 channel perfused by the salt solution, and can be transferred to the electrical response
33 component via the polyethylene glycol hydrogel. In long-term applications, the
34 hydrogel of aqueous two-phase system enables effective electrical stimulation of
35 muscles (**Figure 8d**).

36 A key challenge for practical applications lies in the integration of ionic devices
37 with conventional CMOS-based electronic systems. While ionic interfaces have
38 demonstrated excellent performance in bridging biological systems and electronic
39 devices, extending these concepts to standardized electronic platforms remains
40 nontrivial. Unlike electronic circuits that rely on electron transport, ionic systems
41 operate through ion migration,¹⁶⁶ resulting in fundamental mismatches in signal carriers,
42 operating voltages, and response times. These differences originate from the
43 intrinsically slower dynamics of ion transport and the electrochemical nature of ionic
44 conduction, which can be described by coupled ion transport and interfacial models.¹⁶⁷



1 Consequently, efficient ion-to-electron signal transduction and seamless interfacing
 2 with CMOS circuits remain challenging, limiting the development of fully integrated
 3 ionotronic systems.

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4
 5 **Figure 8.** Hydrogel-based ionic interfaces. a) Schematic diagram showing the working principle of
 6 hydrogel neuroelectronic interface. Ions are transported between the organism and the electrode
 7 through the hydrogel neural interface.¹⁶⁵ b) A hydrogel Neural interface was implanted in mouse
 8 for applying laser stimulations and detecting.¹⁶⁵ c) Schematic diagram of Ionic interfaces of ATPS.
 9 The PEG hydrogel acts as a shell to prevent the ionic solution contained in it from spreading.¹⁵⁸ d)
 10 An ionic interface of ATPS was placed on the muscle for electrical stimulation.¹⁵⁸

11 5. Hydrogel ionic processors

12 Biological processors possess a highly dynamic structure and a non-linear operation
 13 mode. Synapses in neuronal networks continuously change formation and stability in
 14 response to stimuli and the migration of ions also shows the characteristics of precise
 15 adjustment.¹⁶⁸ Compared to machines that can only respond to programmed stimuli,
 16 biological decision-making systems allows organisms to dynamically adapt to changes
 17 in the environment and make more intelligent decisions. Based on hydrogels to achieve
 18 precise ion migration, it is expected to develop smart ionic processors that can be used
 19 in modern science and technology. Hydrogel-based ionic processors are mainly divided
 20 into three categories, diodes for rectifying ionic signals, transistors for adjusting and
 21 amplifying ionic signals, and memristors for storing ionic signals.

Ionic diodes					
Type	Commutating ratio	Operating voltage range	Maximum current	Refs.	
DNA hydrogel nanochannel	52.75	-2 V ~ 2 V	30 nA	Wu et al. ¹⁶⁹	



Stretchable hydrogel diode	18.5	-7.5 V ~ 7.5 V	3 mA	Wang et al. ¹⁷⁰ DOI: 10.1039/D6QM00061D	View Article Online
Ionic transistors					
Type	Commutating ratio	Operating voltage range	Maximum current	Refs.	
NPN Ion Bipolar Junction Transistor	35	30	Rise time: 9 s / Fall time: 3 s	Tybrandt et al. ¹⁷¹	
PNP Ion Bipolar Junction Transistor	39	100	Rise time: 12 s / Fall time: 5 s	Tybrandt et al. ¹⁷²	
Ionic memristors					
Type	Commutating ratio	Operating voltage range	Maximum current	Refs.	
Bipolar polyelectrolyte gel memristor	8	4000s	0.7 μ A	Zhang et al. ¹⁷³	
All-soft-matter liquid metal memristor	100	3h	3 μ A	Koo et al. ¹⁷⁴	

Table 2. Comparison of the performance of different hydrogel ionic processors.

5.1 Ionic diodes

Hydrogel ionic diodes can be formed by two pieces of hydrogels containing polyanions and polycations respectively. Ion diodes have obvious rectification performance: under forward bias, polyelectrolyte counter ions move across the interface, thereby forming a conduction current. Under reverse bias, counter ions move to the nearby electrode and cannot pass through the interface to form a current. Hydrogel ionic diode membranes exhibit current density and rectification ratios comparable to or higher than organic-based diodes.¹⁷⁵ Nanopores with surface charge have been applied to ion rectification (**Figure 9a**).^{176,177} It has been reported that nanopore modified by stimuli-responsive DNA hydrogels can achieve high ion flux and adjustable rectification ratio.^{169,178} DNA hydrogels are assembled to the tip of the conical nanopore. Hydrophilic networks of the hydrogel and introduced negative charges effectively contribute to the transport of cations. The adjustable stiffness state and pH stimulation of DNA hydrogels can precisely control the gated states of on/off and the transport direction of cations or anions (**Figure 9b**).

Miniaturized ion diodes and transistors are essential for complex integrated circuits and fast response to external inputs.^{175,179} By irradiating ultraviolet light on the patterned mask, researchers can polymerize polymers at the target position in the microfluidic channel (**Figure 9c**).¹⁸⁰ It is widely used in microfluidic devices based on charge-selective polymers. Based on this technology, a polyelectrolyte hydrogel diode is prepared on the microchip channels of hundreds of micrometers, and the diode showed significant rectification (**Figure 9d**).¹⁷⁵ Integrating multiple hydrogel diodes on a microchip will produce ionic logic gates including AND, OR and NAND. The fluorescent emission on the polyelectrolyte plug visually displays input signals. These studies are valuable explorations of ion diodes in the field of bionic integrated circuits



1 allowing the functional control and real-time display of ionic current. Ionic signal
2 amplification have realized through an open-junction ionic diode and the mechanism of
3 ion-to-ion signal amplification was proposed.^{181,182} At the open junction of the
4 depletion region, an ion solution is injected to temporarily neutralize the depletion
5 region. A breakdown current will generate with a significant increase in signal strength.

6 Stretchable ionic diodes have been investigated as well.^{170,183,184} The
7 polyelectrolyte hydrogel is modified by utilizing methacrylate to prepare a stretchable
8 ionic diode, which exhibits rectification behavior beyond the stretch of 3, and
9 maintained its performance under repeated deformation (**Figure 9e**).¹⁸³ Studies have
10 shown that increasing the stretching ratio has a weakening effect on the rectification
11 ratio, which is because the increasing effect of stretching on the resistance of the
12 hydrogel (**Figure 9f**).¹⁷⁰ Stretchable ion diodes can find potential applications in
13 stretchable ion circuit systems and flexible wearable devices.

14 Despite the promising rectification behavior of hydrogel ionic diodes, their
15 practical application faces significant challenges. One critical issue is the temporal
16 stability of rectification performance under sustained bias. Ionic rectification
17 fundamentally arises from asymmetric ion transport and the formation of ion
18 enrichment and depletion regions at heterojunction interfaces under opposite bias
19 conditions.^{185,186} Continuous ion migration under prolonged bias may lead to persistent
20 concentration polarization, structural rearrangement of the hydrogel network, and
21 potential dehydration effects, all of which can degrade the rectification ratio over time.

22 In addition, the relatively slow ion dynamics, governed by diffusion and migration
23 processes, fundamentally limit the response speed of ionic diodes compared to electron-
24 based devices.¹⁸⁵ Therefore, the design of ionic diodes requires balancing rectification
25 performance, response speed, and long-term stability, which remains a key challenge
26 for their integration into complex iontronic circuits.

27 Recent studies have demonstrated that ferroelectric nanochannels with
28 spontaneous polarization enable field-free unidirectional ion transport, offering a stable
29 rectification platform for low-power ionic circuits. For instance, ionic diodes
30 employing ferroelectric BiFeO₃ (BFO) nanopore membranes maintain stable operation
31 for 50 minutes under alternating ± 1 V bias without observable decay in ionic current.¹⁸⁷



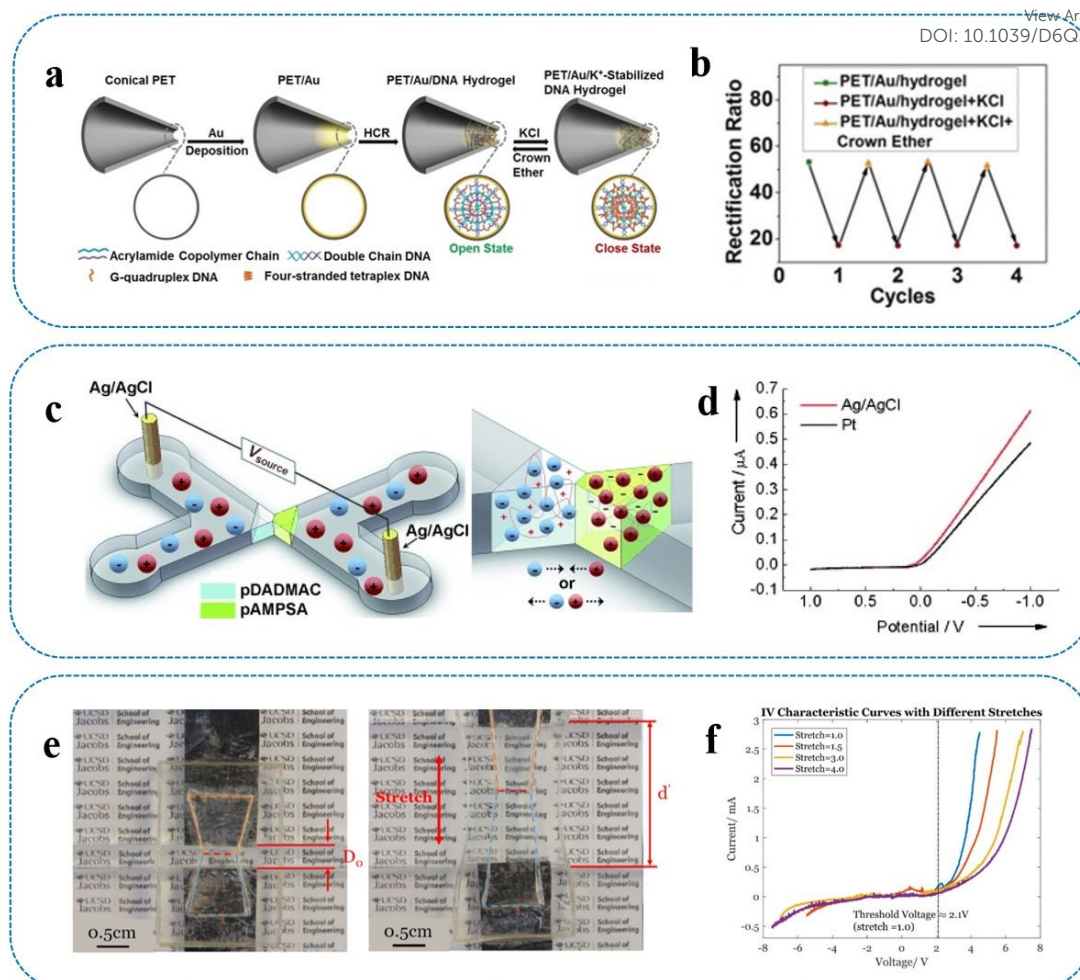


Figure 9. Hydrogel ionic diodes. a) Illustration of the preparation process of the Hydrogel-based conical nanochannel ionic diode. The nanochannel ionic diode can reversibly switch the rectification ratios.¹⁶⁹ b) Schematic illustration of a polyelectrolyte hydrogel ionic diode in a microchip.¹⁶⁹ c) A hydrogel ion diode enabling selective ionic currents.¹⁷⁵ d) Schematic illustration of a stretchable hydrogel ionic diode.¹⁷⁵ e) The rectification ratio of ionic diode decreases slightly as stretching.¹⁷⁰ f) IV characteristic curves with different stretches.¹⁷⁰

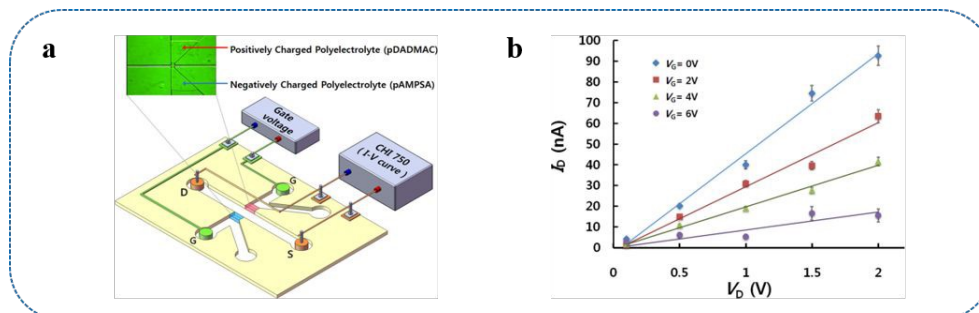
5.2 Ionic transistors

Field effect transistors based on polyelectrolyte hydrogels can also be prepared by using pattern masking photopolymerization.¹⁸⁸ A polycationic hydrogel and a polyanionic hydrogel are formed at two positions on the microchip (**Figure 10a**), allowing the selective passage of anions and cations, respectively. Under the action of the gate voltage, the polyelectrolyte plug extracts ion, resulting in the formation of an ion depletion region and a decrease in source–drain current (**Figure 10b**). The pnp and npn-type ion bipolar junction transistors (IBJT) can be used to construct functional elements similar to CMOS to deliver proper signal amplification and logical gate functions.^{171,172}

However, this ion-based gating mechanism inherently relies on ion migration and diffusion processes, which are significantly slower than electron transport and thus limit the response speed of ionic transistors.¹⁸⁹ In addition, the formation and relaxation of ion depletion/enrichment regions require finite diffusion times, leading to delayed



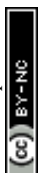
1 switching and hysteresis effects.¹⁹⁰ Furthermore, while ion accumulation at interfaces
 2 can provide high capacitance and efficient gating, it may also induce structural or
 3 compositional changes in the active layer, affecting device stability and
 4 reproducibility.¹⁹¹ Therefore, the design of ionic transistors involves balancing gating
 5 efficiency, response speed, and operational stability.



6
 7 **Figure 10.** Hydrogel ionic field-effect transistor. a) Schematic illustration of a field-effect transistor
 8 in a microchip. By applying the gate voltage, a field-effect transistor induces the main channel
 9 connected to source and drain electrodes.¹⁸⁸ b) Output characteristics of a polyelectrolyte junction
 10 field effect transistor (pJFET).¹⁸⁸

11 5.3 Ionic memristors

12 A memristor is a nonvolatile device that resistive state changes with the current or
 13 voltage history through the device, which can retain memory without power.^{192,193}
 14 Taking advantage of low energy consumption, high data density, and self-adaptation,
 15 memristors may become the core of next-generation storage and computing
 16 devices.^{173,194} A hydrogel-based memristor consists of two liquid metal (EGaIn)
 17 electrodes wrapped and separated by two hydrogels with different pH (**Figure 11a**).¹⁷⁴
 18 The forward bias cause the liquid metal to form an insulating oxide layer (Ga_2O_3) to
 19 inhibit ion current, while the reverse bias can eliminate the metal oxide layer to achieve
 20 conduction of current (**Figure 11b**). It is similar to the accumulation and extrusion of
 21 Ca^{2+} in the synapses. The oxidation and reduction state of the liquid metal depends on
 22 the applied electrical bias and the pH of its local environment. Memristors based on
 23 ion-conductive hydrogels can achieve effective resistance switching and remember the
 24 final state. A memristor is a continuously tunable intelligent device like synapse. It can
 25 adaptively change the performance, suggesting the possibility of developing intelligent
 26 memory and thinking. The favorable biocompatibility of hydrogels makes these devices
 27 promising for bionic systems, such as artificial brain-mimicking or neuromorphic
 28 structures.



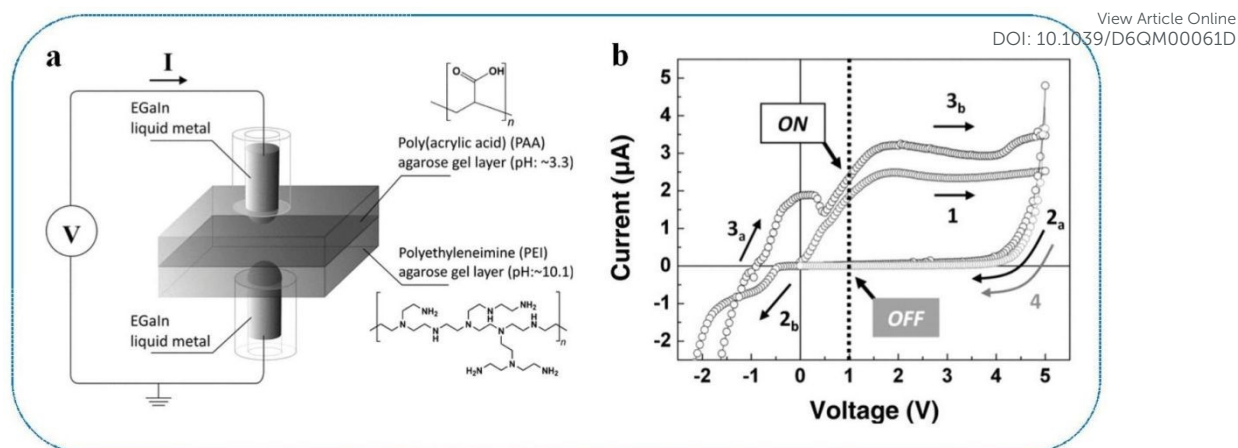


Figure 11. Hydrogel ionic memristors. a) An Ionic memristors is consists of liquid metal electrodes and hydrogel electrolytes with different pH.¹⁷⁴ b) The characteristic curve shows the rectification performance of an ionic memristor.¹⁷⁴

6. Hydrogel ionic effectors

The hydrogel ionic effectors are located at the end of the sensory systems and is an element used to feedback and express the processed information. At present, hydrogel effectors are mainly divided into actuators expressed in a mechanical form and optoelectronic devices expressed based on optical signals.

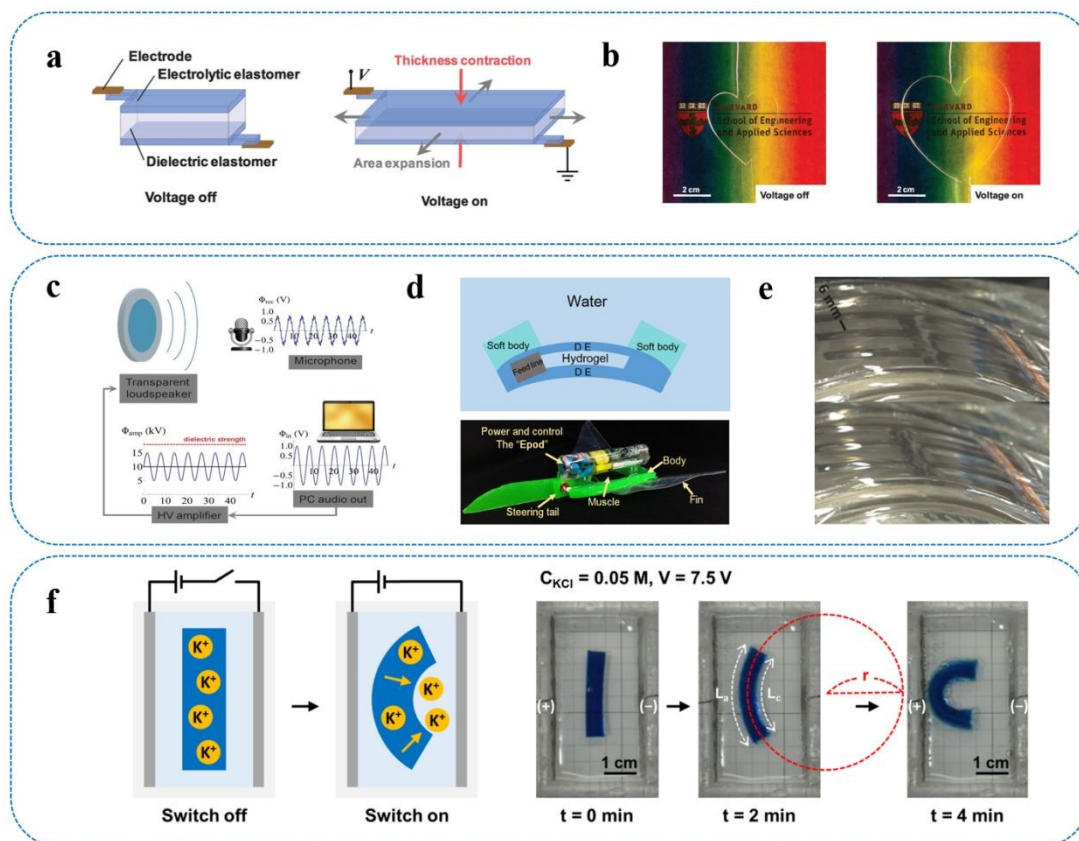
6.1 Ionic actuators

The first type of ionic actuators is based on electrostatic force. The hydrogel actuators can turn the input voltage into mechanical motion. Structure of an actuator is the same as that of a capacitive sensor, including a dielectric elastomer layer sandwiched between two ion-conducting hydrogels (**Figure 12a**). When a high voltage is applied between the hydrogels, the anions and cations in the hydrogels move directionally, forming ionic accumulation layers of opposite charge along each hydrogel/elastomeric interface. The attractive force of opposite charges between the hydrogels would squeeze the elastomer layer (**Figure 12b**).⁴³ The capacitance of the interface between the electric double layer and the dielectric elasticity is very different. In order to meet the charge balance, even if the applied voltage exceeds 1kV, the interface voltage between the hydrogel and the metal remains less than 1V, so that they will not undergo electrochemical reactions. The actuators based on ion-conducting hydrogel have the advantages of highly deformable (fracture stretch 4.6 ± 0.3), fast response ($\tau_{inertia} \approx 1 \times 10^{-3}$ s), lightweight, high transparency (96.95% average transmittance),¹⁹⁵ which can be applied to various forms of feedback expression, such as artificial muscles and fast-moving soft robots that provide feedback through actions (**Figure 12c**),^{43,196,197} surface texture change device for tactile feedback through local deformation (**Figure 12d**),¹³⁵ speakers that express acoustical signals through mechanical vibration (**Figure 12e**).⁴³

The second type of ionic actuators is based on the difference in the motion state of the counterion and polymer chains in the polyelectrolyte hydrogel under the action of an electric field.¹⁹⁸⁻²⁰⁰ When an external electric field is applied to the polyelectrolyte hydrogel, only the counterion moves toward the electrode carry water away from the



1 hydrogel through hydration.¹⁹⁸ This causes anisotropic shrinkage of the hydrogel
 2 accompanied by water extrusion. When the polyelectrolyte hydrogel is placed in an
 3 aqueous solution, the counterions and the ions in the aqueous medium with opposite
 4 charges to the counterions migrate in contrary directions, respectively. The osmotic
 5 pressure brought by the ion gradient caused the asymmetric swelling of the hydrogel
 6 (Figure 12f).¹⁹⁹

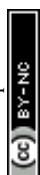


7
 8 **Figure 12.** Hydrogel-based ionic actuators. a) Schematic illustration of the working principle of an
 9 ionic actuator is based on electrostatic force. The application of voltage causes the hydrogel layers
 10 on both sides of the elastomer to produce accumulation layers of opposite charges at the interface,
 11 thereby squeezing the elastomer.⁴³ b) The actuator can produce a large deformation.⁴³ c-e)
 12 Application of actuators: c) Speaker.⁴³ d) Fast-moving soft robots.¹⁹⁶ e) surface texture change
 13 device for tactile feedback.¹³⁵ f) Schematic diagram showing the working mechanism of an ionic
 14 actuators based on the difference in the motion state of the counterion and polymer chains. When
 15 an electric field is applied to the polyelectrolyte hydrogel, the osmotic pressure generated by the
 16 movement of counter ions to the electrode bends the hydrogel.¹⁹⁹

17 6.2 Ionic optoelectronic devices

18 The hydrogel optical effectors can visually express and feedback information
 19 through changes in color and transmittance under electrical stimulation. The ion-
 20 conducting hydrogels with high transparency and high refractive index is the guarantee
 21 for realizing the efficient change of optical information.^{49,201,202} The research on ionic
 22 hydrogel optical communication devices mainly involves three types: electrochromic,
 23 capacitive optoelectronic, and electro-zoom devices.

24 Electrochromic devices have the advantages of high contrast and low driving

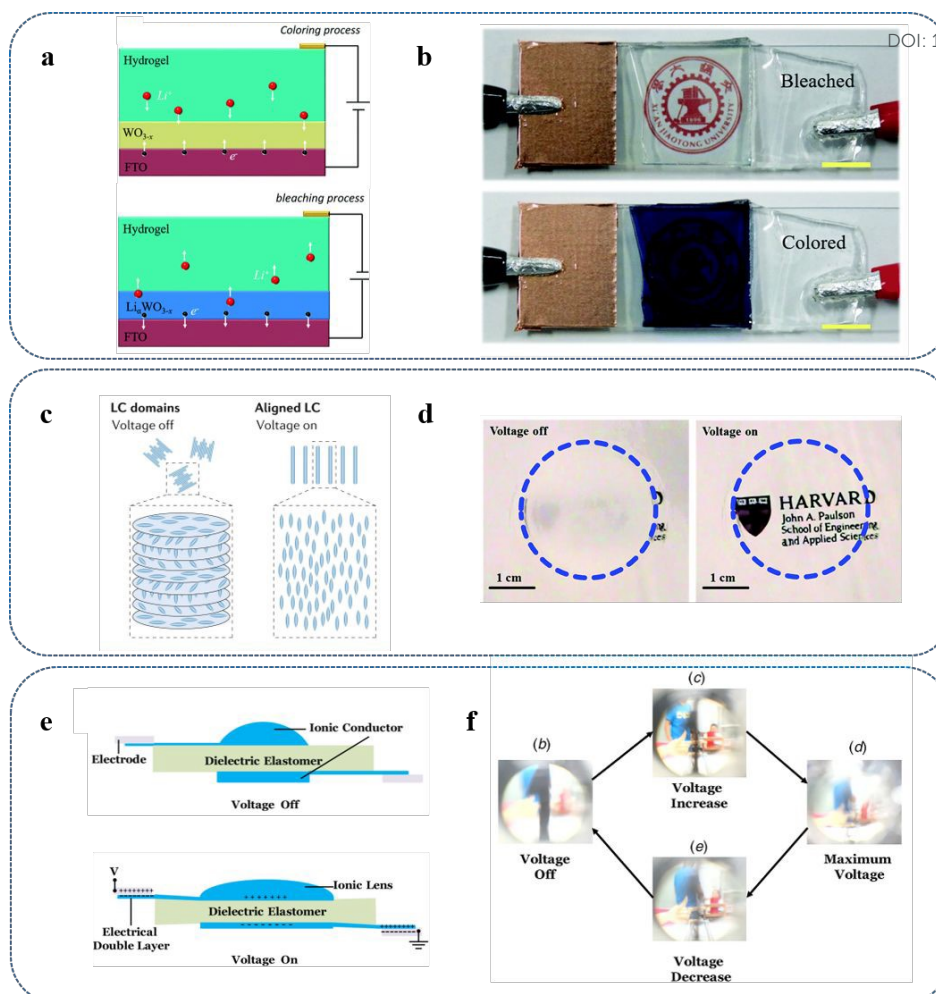


1 voltage.²⁰³ Electrochromic materials including inorganic materials (such as transition
2 metal oxides) and organic materials (such as conjugated polymers), can change their
3 color and transmittance through a reversible oxidation-reduction reaction caused by
4 voltage application.²⁰⁴ With its high transparency and compatibility, a hydrogel can be
5 used simultaneously as a transparent electrode, electrolyte and electrochromic material
6 storage layer, greatly simplifying the device structure to improve performance and
7 reduce manufacturing costs (**Figure 13a**). The hydrogel-based electrochromic device
8 with a three-layer structure can obtain evident transmittance modulation and high
9 coloring efficiency at a lower driving voltage (about 1V) (**Figure 13b**).

10 In a capacitive optoelectronic device, a layer of electroluminescent material (e.g.,
11 liquid crystal²⁰⁵ and phosphor^{46,206}) is sandwiched between two ionic hydrogels and two
12 elastomer layers (**Figure 13c**). When an AC voltage is applied, the hydrogel acts as an
13 ionic conductor to project an electric field to the active layer, causing the device to emit
14 light or change transparency (**Figure 13d**). The similar structure of the capacitive
15 optoelectronic effector and the capacitive sensor suggests its possibility as a dual-
16 function device for sensing and feedback.

17 The electro-zoom device with adjustable focal length is realized by a capacitive
18 actuator.²⁰⁷ Like the human eye, the dielectric elastic body acts like a muscle, and the
19 ion gel acts as the lens and the nervous system for zooming and signal transmission
20 (**Figure 13e**). Benefit from the advantages of stretchable and transparent ionic hydrogel,
21 the optical zoom effector has powerful adjustable optical performance (**Figure 13f**).
22 The ion eye shows fast response performance (3.6 ms) and a wide range of focal length
23 changes (approximately 50%). By cooperating with more ionic sensors, an electro-
24 zoom device is expected to be used in bionic robots and medical treatments based on
25 ion sensor systems as an ionic eye.



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1 **Figure 13.** Hydrogel-based ionic optoelectronic devices. a) Schematic illustration of the working
 2 mechanism of electrochromic device.²⁰⁴ (b) The transmittance and color of the hydrogel can be
 3 effectively adjusted by applying voltage.²⁰⁴ c) Schematic illustration of the working principle of a
 4 capacitive liquid-crystal device. The arrangement direction of the liquid crystal molecules can be
 5 changed by applying voltage, thereby regulating the transparency of the device.²⁰⁵ d) When voltage
 6 is applied, the device changes from opaque to transparent.²⁰⁵ e) Schematic illustration of an electro-
 7 zoom device. When a voltage is applied, the two gel layers squeeze the dielectric elastomer by
 8 electrostatic force to flatten the lens and increase the focal length.²⁰⁷ f) With the help of an
 9 electro-zoom device, the quality of the image can be regulated by adjusting the voltage.²⁰⁷

11 7. Hydrogel ionic power sources

12 Hydrogel ionic sensor systems are inherently flexible, so it has increased the demand
 13 for essential energy supply equipment that maintain high performance and safety under
 14 various deformations, which puts forward higher requirements for the internal
 15 component performance and interface combination of energy storage devices. The
 16 hydrogel ion power sources came into being based on this demand. Our discussion of
 17 hydrogel-based power sources is organized into two sections, energy storage devices
 18 and energy harvesters.

Battery Type	Operating Voltage	Specific Capacity	Cycling Performance	Refs.
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Stretchable battery	Li-ion	1.7 V	28 mAh g ⁻¹	50 cycles with 65% capacity	Chen et al. ²⁰⁸	View Article Online DOI: 10.1039/D6QM00061D
Aqueous battery	Na-ion	1.03 V	160 mAh g ⁻¹	100 cycles with 90% capacity	Zhang et al. ²⁰⁹	
Quasi-solid-state Li-ion battery		3.4 V	160 mAh g ⁻¹	300 cycles with 98.9% capacity	Jiang et al. ²¹⁰	
Wearable battery	Zn-ion	1.5 V	306 mAh g ⁻¹	1000 cycles with 97% capacity	Li et al. ²¹¹	
Ultralong-life based battery	Zn-	1.6-1.8 V	260 mAh g ⁻¹	16000 cycles with 65% capacity	Huang et al. ²¹²	

Table 3. Comparison of the performance of different hydrogel ionic power sources.

7.1 Energy storage devices

Currently, the most common flexible energy storage devices are batteries and supercapacitors. Electrolyte is an important element that determines the level of the electrochemical window and ion transmission efficiency of a battery or supercapacitor. Conventional flexible batteries or supercapacitors rely on membranes permeated with liquid electrolytes to act as solid electrolytes. However, there are clear drawbacks to this type of electrolyte: low ionic conductivity, poor mechanical strength, and high risk of short circuit.²¹³ At the same time, most electrolyte solvents are toxic and flammable organic solvents. Compared with organic solvent electrolytes, aqueous electrolytes have outstanding advantages in terms of safety and environmental performance. The ion-conducting hydrogel containing massive aqueous electrolyte offers an ideal electrolyte with flexibility, high ionic conductivity, and high safety for flexible energy storage devices. As shown in **Figure 14a**, a hydrogel acts as both an electrolyte and a diaphragm to prevent short circuit caused by the contact between the cathode and the anode.

A battery is a device that stores energy in the form of chemical energy. They have a sufficiently high energy density and are suitable for long-term energy storage. At present, common batteries based on hydrogel electrolytes include lithium ion batteries, sodium ion batteries and zinc ion batteries.

Lithium-ion batteries (LIB) are widely used due to their high energy density (200 W h kg⁻¹), high power density (10 kW kg⁻¹) and long-term stability (delivered 200 mA h g⁻¹ for 100 cycles).²¹⁴ Sodium-ion batteries (SIB) have attracted extensive attention as an alternative method of lithium ion energy storage technology due to its rich composition and low cost.²¹⁵ With the rapid development of flexible and wearable electronic devices, in addition to achieving higher energy density and longer service life, the development of flexible lithium or sodium ion energy storage technology has become an emerging research field.²¹⁶ However, the high cost and safety issues brought about by the organic solutions used in lithium-ion and sodium-ion batteries are required to develop alternative energy storage solutions²¹⁷. The use of safe and relatively inexpensive hydrogel electrolytes instead of flammable and expensive organic electrolytes is a competitive candidate.^{210,213,218,219} The main bottleneck of the application of hydrogels to lithium-ion batteries and sodium-ion batteries is that the electrochemical window of the aqueous electrolyte is very narrow (1.23 V), which



1 limits the performance of batteries. To solve the dilemma, revolutionary “water-in-salt”
2 electrolyte (WiSE) is introduced into hydrogels to prepare high-performance lithium-
3 ion batteries. As an example, a polyacrylamide hydrogel containing WiSE can provide
4 a working window of 3.28 V (1.95 to 4.93 V).²⁰⁸ On the other hand, the electrochemical
5 window is significantly expanded as the polymer content increases. Because the
6 polymer network appropriately restrict the movement of water molecules, thereby
7 inhibiting water-induced side reactions, including hydrogen release and passivation.⁵⁶
8 The electrochemical window of the hydrogel electrolyte with a polymer content
9 exceeding 55% can be extended to 5 V.²⁰⁹

10 Zinc-ion batteries (ZIB) are inherently safe and environmentally friendly, because
11 they are not subject to environmental restrictions of anhydrous and/or anaerobic
12 assembly and are suitable for aqueous electrolyte environments. The high natural
13 abundance of zinc is conducive to large-scale and low-cost production. Combined with
14 its low electrochemical potential (-SHE of -0.76 V), high capacity (5850 mAh cm⁻³),
15 zinc-ion batteries are considered to be the most promising alternatives to commercial
16 lithium-ion batteries and holds the greatest promise for future energy applications.^{220,221}
17 For instance, a polyacrylonitrile electrospun fiber membrane filled with gelatin
18 modified by polyacrylamide play the role of hierarchical structured hydrogel electrolyte,
19 which was further assembled into a flexible Zn-MnO₂ rechargeable battery with
20 outstanding safety.²¹¹ Flexible solid-state ZIB can provide high power density of 148.2
21 mW cm⁻². The thickness of the entire device is close to A4 paper. More importantly,
22 the solid ZIB exhibits extremely high safety performance when it works in various
23 harsh environments, such as being bent, hammered, punctured, cut, sewed, washed in
24 water and put on fire (**Figure 14b**). As another example, a sodium polyacrylate
25 hydrogel electrolyte was applied to solid Zn // NiCo and Zn-air batteries with ultralong
26 lifetime and high capacity.²¹² Thanks to the inherent high ionic conductivity of 0.17 S
27 cm⁻¹ and strong water-retaining capacity of the sodium polyacrylate hydrogel
28 electrolyte, the prepared battery exhibits high electrical capacity (Zn // NiCo battery is
29 ≈260 mAh g⁻¹, Zn-air battery is ≈800 mAh g⁻¹), ultra-long cycle stability (Zn // NiCo
30 battery has a capacity of 16,000 cycles of 65% and Zn-air battery withstands 800 cycles
31 of 160 h).

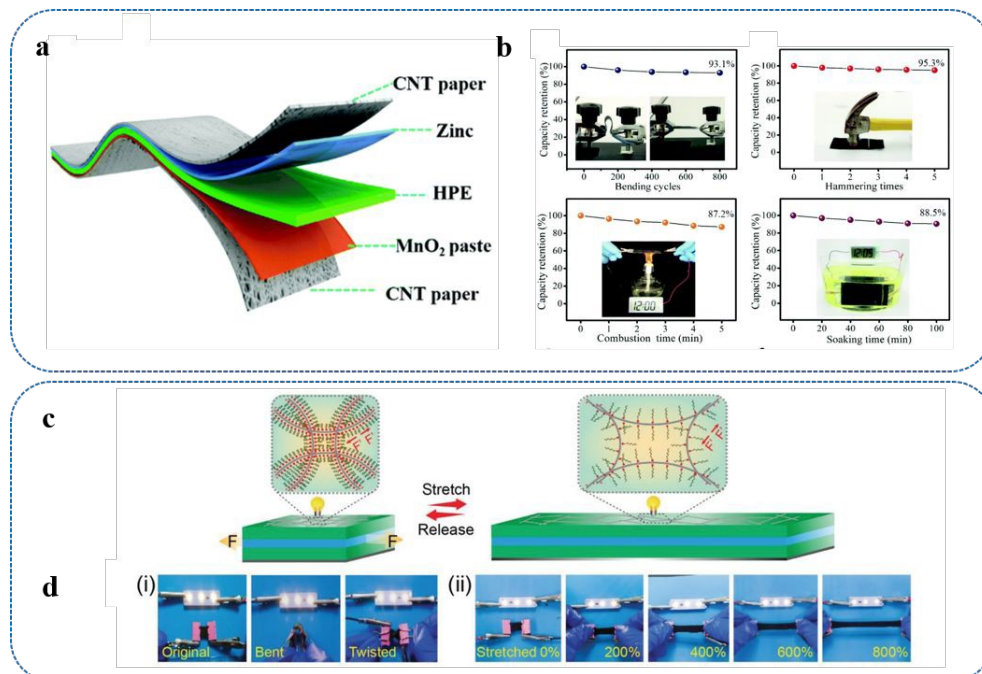
32 Supercapacitors have high power density and the ability to charge and discharge
33 rapidly. There are two ways to store energy in supercapacitors. The first is through the
34 surface adsorption of electrolyte ions (electric double layer capacitors).^{222,223} The
35 electrodes of this type of capacitor are generally carbon materials. Its performance is
36 related to effective surface area of the electrode. The second type of supercapacitors
37 store electrical energy through fast surface redox reactions (pseudocapacitor),^{213,224}
38 whose electrodes mainly include transition metal oxides and conducting polymers, and
39 their performance is closely associated with the electrode materials. As shown in
40 **Figure 14c-d**, by combining flexible electrodes (such as carbon cloth loaded with active
41 electrode materials), solid-state supercapacitors based on ionic hydrogel electrolytes
42 can maintain stable electrical performance under deformation.^{64,225}

43 Through the design of devices and hydrogel materials, adding additional functions
44 to energy storage devices can make them versatile for complicated working



1 environment. For example, by combining intrinsic or structural stretchable electrodes,
 2 stretchable batteries or capacitors can be prepared.²²⁶⁻²²⁹ Purposeful design and
 3 preparation of hydrogels can endow energy storage devices the functions of self-
 4 heal,²³⁰⁻²³² antifreeze,²³³⁻²³⁵ shape memory,^{236,237} overheat protection,²³⁸⁻²⁴⁰ and self-
 5 discharge suppression.²⁴¹

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6
 7 **Figure 14.** Hydrogel energy storage devices. a) Schematic illustration of the structure of the solid-
 8 state ZIB. The hydrogel acts as both an electrolyte and a diaphragm to prevent short circuit caused
 9 by the contact between the cathode and the anode.²¹³ b) The hydrogel-based flexible ZIB can work
 10 normally under various severe tests.²¹³ c) Schematic diagram of a stretchable supercapacitor based
 11 on hydrogel. The hydrogel electrolyte and electrodes maintain superior stability during process of
 12 stretch–release.²³⁰ d) A supercapacitor powers LEDs under various deformation and stretching.²³⁰

13 7.2 Energy harvesters

14 Ionic hydrogel-based dielectric elastomer generate Hydrogel ionic sensor systems
 15 need portable functional methods. Energy storage devices have a limited lifetime and
 16 presents potential hazards to health and the environment. The tremendous development
 17 of sensing equipment urgently requires the development of new technologies that can
 18 obtain sustainable energy from the environment. Hydrogel-based energy harvesters are
 19 used to collect energy wasted in the environment and convert it into electrical energy.
 20 Currently, the harvesters mainly collect three forms of energy: chemical energy,
 21 mechanical energy and salinity gradient energy.

22 Atmospheric moisture is a ubiquitous and sustainable energy source that has
 23 recently attracted increasing interest for decentralized energy harvesting.²⁴² Ionic
 24 hydrogel-based moisture-electric generators (MEGs) convert chemical energy directly
 25 to electricity via functional material–humidity interactions, offering a different energy
 26 conversion pathway from conventional photovoltaic or thermoelectric systems. For
 27 example, by introducing ionic liquids into the double-network hydrogel system formed
 28 by poly(2-acrylamido-2-methylpropanesulfonic acid) (PAMPS) and polyvinyl alcohol



1 (PVA) to induce microphase separation, PAB ionogels can be obtained, thereby
2 constituting the PAB - MEG system (**Figure 15a**).²⁴³ The PAB-MEG system can
3 achieve stable power transmission without the need for external energy storage or
4 rectifier circuits. In practical tests, a 1×4 series-integrated PAB-MEG array can charge
5 a smartwatch based on motion, providing a 100 - hour battery life (**Figure 15b**).

6 The triboelectric nanogenerators (TEGs) is mainly based on contact
7 electrification and electrostatic induction to realize the conversion of mechanical
8 energy into electrical energy. During the periodical contact and separation of the object
9 with the TENG, ions flow repeatedly in the hydrogel and induce the external load to
10 generate an alternating current through capacitive coupling (**Figure 15c**). The ion-
11 conducting hydrogel TENG can output electricity with an instantaneous peak power
12 density of 35 mW m^{-2} , which is close to TENGs based on electronic conductors, and
13 converts energy through human motion to drive wearable electronics (**Figure**
14 **15d**).^{244,245} Ionic hydrogel-based TENGs have the advantages of large output power,
15 light weight and simple manufacturing, while also exhibiting high stretchability and
16 transparency.²⁴⁵⁻²⁴⁹

17 DEGs are also used for the collection of mechanical energy, which are
18 composed of dielectric elastomer film sandwiched between two ion-conducting
19 hydrogels (**Figure 15e**).^{250,251} The dielectric elastomer generator will absorb the charge
20 from the external power source to increase the capacitance during the stretching state.
21 When the power is disconnected and the DEGs is released, the capacitance of the DEGs
22 decrease with the elastic recovery of the shape, resulting in an increase in the voltage
23 between the electrodes. Many studies have shown that the efficiency of energy
24 conversion of DEGs can be effectively improved by equi-biaxial stretching (**Figure**
25 **15f**). The capacitance C scales with the fourth power of stretch ratio λ : $C \propto \lambda^4$.²⁵²⁻²⁵⁴

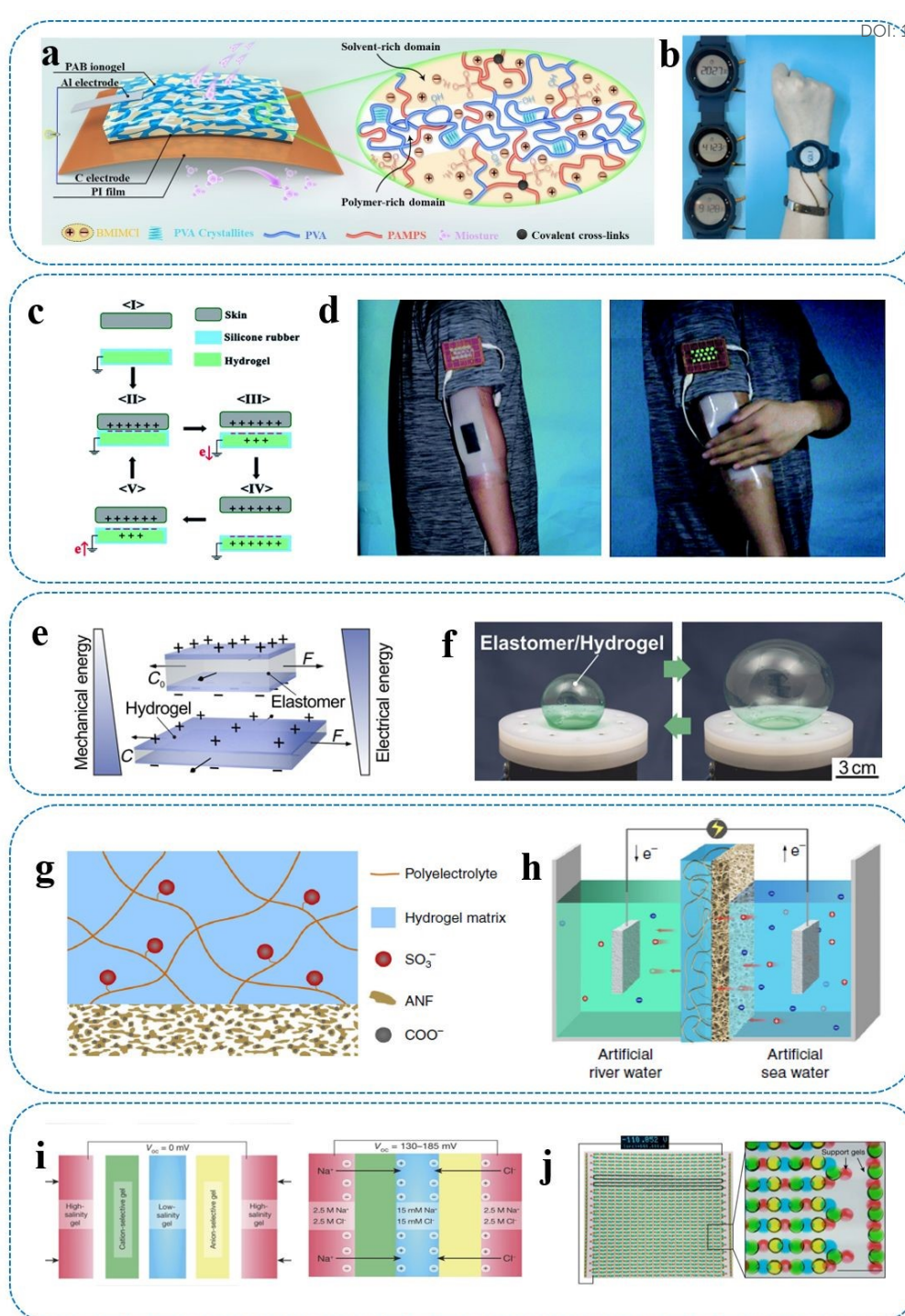
26 The salinity gradient between river water and sea water is a potential renewable
27 energy source.^{255,256} The desire to collect clean energy from water to replace fossil fuels
28 has promoted the development of salinity gradient energy harvesters. The electric eel
29 inspired a new type of hydrogel-based energy harvester that uses ion gradients to collect
30 energy.²⁵⁷ The power source is created by sequentially stacking a high salinity hydrogel,
31 a cation selective hydrogel, a low salinity hydrogel, an anion selective hydrogel, and a
32 second-high salinity hydrogel (**Figure 15g**). The selective diffusion of ions creates an
33 ion gradient to generate energy, which can generate a voltage of up to 110 V reach up
34 by stacking the units in series (**Figure 15h**). By increasing the asymmetry between the
35 membrane layers, the unidirectional ion diffusion can be effectively promoted, resulting
36 in an increase in the ability to use the difference in ion concentration to generate
37 electricity. A heterogeneous membrane made of polyanion electrolyte hydrogel and
38 porous aramid nanofiber (ANF) is used to generate electricity via salinity gradients
39 (**Figure 15i**).²⁵⁸ Inherent synergistic effects of static electricity, chemistry, and
40 structural asymmetry make the system have a stable ion diode effect, which greatly
41 promotes the unidirectional transmission efficiency of cations to enhance the power
42 generation (**Figure 15j**). The power output of heterogeneous membranes has been
43 significantly increased to $\sim 5.06 \text{ W m}^{-2}$.

44 Despite the rapid development of hydrogel-based energy harvesters, power output

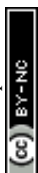


1 and energy conversion efficiency remain key limiting factors for practical applications.
2 In most cases, the generated power density is still relatively low and highly dependent
3 on external stimuli such as mechanical motion, thermal gradients, or environmental
4 humidity, resulting in intermittent and unstable energy supply. This limitation
5 originates from the fundamental working mechanism of ionic power generation, which
6 relies on ion diffusion, charge separation, and concentration gradients rather than fast
7 electron transport.²⁵⁹ Therefore, the energy conversion process is inherently diffusion-
8 limited and exhibits relatively slow response dynamics, which is consistent with ionic
9 energy harvesting systems, where device performance and power output are
10 fundamentally governed by ion transport kinetics.²⁶⁰ Such characteristics restrict the
11 ability of these systems to continuously power integrated electronic components. These
12 constraints have been widely recognized in ionic energy systems, where power
13 generation is often governed by ion transport kinetics and environmental conditions.¹⁶⁶





1
 2 **Figure 15.** Hydrogel energy harvesters. a) Schematic diagram of one-single PAB-MEG device with
 3 asymmetric moisture penetration layers.²⁴³ b) Integrated PAB-MEG harvest moisture energy to
 4 power a watch.²⁴³ c) Schematic diagram showing the working principle of Hydrogel-based TENG
 5 at single-electrode mode. TENG converts mechanical energy into electrical energy through contact
 6 charging and electrostatic induction.²⁴⁵ d) A TENG harvested energy of percussion to power the
 7 LEDs.²⁴⁵ e) Schematic illustration of working mechanism of a dielectric elastomer generator. A
 8 dielectric elastomer generator can convert mechanical energy into electrical energy through the
 9 capacitance change caused by stretching.²⁵⁰ f) An efficient energy conversion rate of 11% can be
 10 effectively achieved by inflating the balloon.²⁵⁰ g) Schematic illustration of hybrid membrane with



1 asymmetric structure composed of polyelectrolyte hydrogel and aramid nanofiber membrane.²⁵⁸ **1a)** View Article Online
2 Schematic of the osmotic energy conversion process.²⁵⁸ **i)** Schematic illustration of working
3 mechanism of all-gel energy harvester of salinity gradient. The selective diffusion induced by
4 hydrogels with different concentrations and ion types would convert the salinity gradient into
5 electrical potential differences.²⁵⁷ **j)** The voltage of the energy harvester in series can be up to 110
6 V.²⁵⁷ DOI: 10.1039/D6QM00061D

7 **8. Aging mechanisms and stability enhancement strategies of hydrogel** 8 **ionic systems**

9 Hydrogel-based ionic systems exhibit excellent mechanical compliance, ionic
10 conductivity, and biocompatibility. However, their long-term stability remains a critical
11 challenge that limits practical deployment, particularly under real-world environmental
12 and operational conditions. The aging of hydrogel ionic systems generally originates
13 from three primary mechanisms: dehydration, ion leaching, and mechanical fatigue.
14 Understanding these degradation pathways is essential for the rational design of durable
15 ionic devices.

16 **8.1 Dehydration-induced performance degradation**

17 Dehydration is one of the most fundamental aging mechanisms in hydrogels due
18 to their intrinsically high water content. When exposed to ambient or elevated
19 temperatures, water evaporation leads to significant shrinkage of the polymer network,
20 resulting in increased stiffness, reduced elasticity, and diminished ionic conductivity.²⁶¹

21 More importantly, dehydration alters the microstructure of hydrogels by
22 collapsing ion transport pathways and increasing ion-polymer interactions, thereby
23 severely suppressing ion mobility.²⁶² In extreme cases, dehydration-induced stress can
24 even lead to crack formation or structural failure. Additionally, under freezing
25 conditions, localized dehydration driven by ice formation (cryosuction) can further
26 induce fracture and irreversible damage, highlighting the coupled effects of dehydration
27 and environmental stress.²⁶³

28 To address dehydration, several approaches have been developed. For instance,
29 coating hydrogels with elastomeric or polymeric barrier layers effectively reduces
30 water evaporation and enhances environmental stability.²⁶² As another example,
31 replacing part of water with hygroscopic solvents can significantly suppress
32 evaporation and maintain conductivity.²⁶⁴

33 **8.2 Ion leaching and compositional instability**

34 Another critical aging pathway is ion leaching, particularly in open or bio-
35 interfaced systems. Due to concentration gradients between the hydrogel and
36 surrounding media, mobile ions tend to diffuse outward, leading to gradual depletion
37 of charge carriers³¹. This process results in reduced ionic conductivity, signal drift, and
38 degraded device performance over time. Ion leaching is especially problematic in
39 bioelectronic interfaces and aqueous environments, where continuous ion exchange can
40 disrupt both device functionality and biological compatibility. Moreover, leaching may
41 alter the internal ionic balance, affecting electrochemical stability and device
42 reproducibility.²⁶⁵



1 To mitigate ion leaching, substantial efforts have been devoted to regulating ion
2 transport and confinement within hydrogel networks. One effective strategy involves
3 the incorporation of fixed charges through polyelectrolyte or zwitterionic architectures,
4 which can immobilize counterions and suppress their outward diffusion by electrostatic
5 interactions.²⁶⁶ In addition, spatial confinement approaches such as aqueous two-phase
6 systems (ATPS) enable the localization of conductive ionic species within distinct
7 domains, thereby maintaining high ionic conductivity while minimizing ion exchange
8 with the surrounding environment.¹⁵⁸ Furthermore, physical encapsulation using
9 elastomeric or polymeric barrier layers has been widely adopted to isolate hydrogels
10 from external media, effectively reducing electrolyte leakage and preventing instability
11 or even short-circuit risks in aqueous systems.²⁶⁷ Collectively, these strategies highlight
12 the importance of controlling ion mobility and interfacial exchange to ensure long-term
13 compositional stability of hydrogel ionic systems.

14 **8.3 Integrated strategies for long-term stability**

15 In practical applications, these aging mechanisms often occur simultaneously and
16 are strongly coupled. For example, dehydration can significantly alter the
17 microstructure of hydrogel networks by reducing water content and increasing polymer
18 chain density, which in turn affects mechanical properties and may accelerate fatigue-
19 induced damage.²⁶⁸ Meanwhile, dehydration driven by osmotic imbalance or
20 environmental exposure can lead to structural shrinkage and failure, further
21 exacerbating mechanical instability.²⁶² In parallel, ion transport and retention in
22 hydrogels are intrinsically governed by diffusion and thermodynamic equilibrium,
23 implying that ion leaching can disrupt internal ionic balance and indirectly influence
24 mechanical integrity and electrochemical stability.²⁶⁹ These coupled effects highlight
25 that individual degradation pathways cannot be considered in isolation.

26 Therefore, future designs should adopt multifunctional and synergistic strategies,
27 integrating multiple stabilization mechanisms within a single system. For instance,
28 combining encapsulation layers to suppress water evaporation and ion exchange,
29 antifreezing or antidehydration solvent systems to maintain hydration under extreme
30 conditions, and dynamic or self-healing polymer networks to resist mechanical fatigue,
31 represents a promising direction for achieving durable performance. Such integrated
32 designs enable simultaneous regulation of water retention, ion transport, and network
33 integrity, thereby addressing the interdependent nature of hydrogel aging processes.

34 **9. Future development directions for hydrogels**

35 Based on the unique properties of hydrogels, various innovative hydrogel ionic sensing
36 devices have been constructed. However, in the face of broad development prospects,
37 the properties of hydrogels should be continuously improved to meet more
38 comprehensive needs and development new functional to open untapped opportunities.
39 We will discuss future research directions for hydrogels on three major aspects (**Figure**
40 **16**): 1. development of hydrogels with extraordinary properties 2. integration of
41 hydrogels 3. weather resistance improvement of hydrogels.

42 **9.1 Hydrogels with extraordinary properties**

43 *Hydrogels with high strength and high toughness*



1 Demonstrating strength and toughness in the hydrogels at the same time, which
2 are usually contradictory mechanical properties, is beneficial to prolong service periods,
3 endure high loads and large deformation, and improve impact resistance.

4 By introducing the energy dissipation mechanism during the loading process into
5 hydrogels, for example, including dual-network hydrogels,^{45,270,271} hydrogels with
6 chemical and physical hybrid crosslinking agents,^{272,273} creating nanocrystalline
7 domains^{274,275} or improving the homogeneity of the hydrogels, examples include
8 topological hydrogels with sliding crosslinkers,^{276,277} and tetra-arm polymer
9 hydrogels²⁷⁸, many hydrogels have shown tremendous enhancements of strengthen and
10 toughen over their conventional counterparts.

11 A breakthrough strategy to produce ultrastrong nanocomposites gel with highly
12 ordered layered structures has been proposed (**Figure 16a**).²⁷⁹ At the immiscible
13 hydrogel/oil interface, two-dimensional nanosheets are arranged in an orderly manner
14 under the induction of shearing force in the hydrogel prepolymerization solution and
15 polymerized. A proper weight percentage of the nanosheets would cause the polymer
16 chains to harden effectively under the strong constraints of the nanosheets. The gel
17 reinforced by two-dimensional nanosheets exhibits high strength of up to $1,215 \pm 80$
18 MPa and a toughness can reach 36.7 ± 3.0 MJ /m³. This work provides a generic and
19 facile approach for high-strength and high-toughness composite materials.

20 High strength nanocomposites polymer can also be fabricated via an interfacial
21 co-crystallization strategy. For instance, Liu et al. constructed a sub-molecularly flat
22 interface between ferromagnetic vanadium diselenide (VSe₂) and ferroelectric
23 poly(vinylidene fluoride) (PVDF) nanocrystals, resulting in a high-performance
24 magnetoelectric polymer composite (**Figure 16b**).²⁸⁰ The obtained material exhibits a
25 tensile strength exceeding 100 MPa and a magnetocapacitive coefficient of 23.6%,
26 demonstrating outstanding mechanical robustness and magnetoelectric coupling.
27 Owing to these advantages, this composite shows great potential for applications in
28 wearable magnetoelectric sensors.

29 *Programmable hydrogels*

30 At present, hydrogel materials lack the ability to control and adjust the mechanical
31 properties stably, which limits their engineering applications in complex
32 environments.²⁸¹ In nature, many biological soft tissues with synergistic
33 heterostructures, such as sea cucumbers, skeletal muscle and cartilage, can adapt to
34 complex environments by adaptive mechanical properties.²⁸²⁻²⁸⁴ Inspired by them, the
35 researchers designed and prepared a heterogeneous gel network with excellent
36 mechanical adaptability.²⁸⁵⁻²⁸⁸ Incorporating oil-phase monomers with phase change
37 behavior triggered by temperature into hydrogels, renders hydrogels excellent
38 switchable mechanical and shape memory properties.²⁸⁵ By using this strategy on a
39 metal-supramolecular hydrogel framework, a dual-programmable shape-deformable
40 organic hydrogel was further prepared.²⁸⁶ In order to break through the simple bistable
41 mechanical state of rigid and soft, a variety of noneutectic phase transition components
42 are incorporated into continuous elastic hydrogel networks to construct a programmable
43 organic hydrogel with multi-stable mechanical states. A multi-programmable hydrogel
44 can stably switch mechanical modulus stepwisely under heat induction (i.e., triple,



quadruple, and quintuple switching).²⁸⁷ Smart hydrogels with precisely controllable mechanical properties provide valuable breakthroughs practical applications, including wearable devices, biomedical engineering, and soft robots.²⁸⁹⁻²⁹¹

Self-healing hydrogels

Biological organisms have built-in repair mechanisms to ensure stable physiological functions.²⁹² For example, Human skin produces collagen through the stimulation of inflammation to regenerate epithelial cells and tissues.²⁹³ Plants can induce repair through oligopeptides, oligosaccharides or other molecules.²⁹⁴ Development of self-healing materials can not only extend the service life of the material, but also improve the reliability of the material by avoiding failures caused by the accumulation of cracks.²⁹⁵ Abundant functional groups endow the hydrogel materials with great designability and allow them to have fast self-healing properties after purposefully designed. There are two main repair mechanisms of self-healing hydrogels: dynamic covalent reaction (chemical cross-linking) and non-covalent reaction (physical cross-linking).²⁹⁶ Common cross-hydrogels are cross-linked by irreversible chemical bonds, so it is difficult for hydrogels to self-healing. The dynamic covalent bonds are not permanent cross-linking, which can reversibly recombine after breaking. Reactions that can form dynamic covalent bonds include phenylboronic acid complexation reaction,^{297,298} disulfide bond,^{299,300} imine bond,^{301,302} acylhydrazone bond,^{303,304} and Diels-Alder reaction^{305,306} and others. The process of repairing hydrogels based on dynamic covalent bonds often requires the application of external stimuli, such as UV light,²³⁰ alternating current³⁰⁷ and pH³⁰⁸. In the case of physically self-healing hydrogels, noncovalent interactions are generally utilized, such as hydrogen bonds,^{308,309} hydrophobic interactions,³¹⁰ host-guest interactions,³¹¹ and ionic interactions,³¹² nanocomposite effect,³¹³ crystallization effect^{314,315} and π - π stacking effect.³¹⁶ The durability and stability of various flexible devices based on self-healing hydrogels are greatly improved, which also means less energy consumption and less waste generation.

Without introducing new functional groups and special designs, the method of preparing self-healing hydrogels by increasing the preparation temperature provides a valuable insight.³¹⁷ A hydrogel prepared at high temperature contains a loose network of highly mobile short polymer chains, which can promote the exchange of hydrogen bonds to facilitate self-healing. It exhibits rapid self-healing properties at room temperature (below its T_g).

9.2 Integration of hydrogels

Functional integration of hydrogels

In order to meet more complex and comprehensive application requirements, the multi-functionalization of hydrogel-based equipment is the future development trend. A large number of integrated applications have been explored.³¹⁸ For instance, various stimulus-sensitive sensors are integrated to mimic the mechanical, temperature, humidity receptors of human skin, respectively.³¹⁹ Integrating a flexible powers or the self-charging devices in the system can realize get rid of the limitation of the fixed power.^{227,320} The introduction of wireless communication devices such as WiFi transmitters into the flexible strain sensor is able to implement effectively detect human



1 movement remotely, which improves portability and comfort.³²¹ Hydrogels with
2 multiple functions are coupled into an integrated system of soft robots that can perform
3 complex actions.^{322,323} For example, magnetic particles are introduced into a
4 temperature-sensitive hydrogel head as a magnetic drive unit, and a non-magnetic
5 hydrogel tail as a functional unit to obtain a segmented hydrogel robot that can carry
6 out multi-modal motion.³²² These sophisticated hydrogel soft robots are envisioned to
7 be used in medical operations.

8 *Materials integration of hydrogels*

9 A complete hydrogel-based device is usually consisted of multiple materials with
10 corresponding functional merits. Therefore, the adhesion of hydrogels to different
11 materials (e.g., elastomers, metals, and glass) is a key factor that determines the stability
12 of devices.³¹ Among them, the adhesion between hydrogels and elastomers is the most
13 notable.¹ Elastomers with low water and solute permeability are equivalent to coats for
14 hydrogels, which can effectively prevent the dehydration of hydrogels or the exchange
15 of substances with the external environment when the device is immersed in solution,
16 thereby improving the mechanical reliability of hydrogels under harsh temperature and
17 chemical conditions. Conversely, hydrogel as a coating for elastomers such as medical
18 catheters can provide a biocompatible interface while reducing mechanical mismatch
19 and foreign body reactions.^{324,325} Currently, the interfacial adhesion of hydrogels is
20 mainly achieved through physical interaction,^{326,327} covalent anchoring,^{328,329} and
21 interfacial interpenetration.^{50,324,330}

22 Compared with physical interaction based on weak non-covalent bonds such as
23 electrostatic interactions, hydrogen bonds, the interface formed by covalent anchorage
24 is more stable. As an example, the unreacted double bond can be introduced into the
25 hydrogel by reacting the hydroxyl group with acryloyl chloride.³²⁸ The organogel-
26 hydrogel hybrid is obtained by polymerizing organogel monomers on the surface of the
27 modified hydrogel. However, the low toughness and brittle interfaces formed by
28 covalent anchoring will still crack under high mechanical load.

29 The way of interfacial interpenetration makes the two polymer networks produce
30 molecular entanglement to obtain a firm interface, which is strong enough to elicit
31 Lake-Thomas mechanism and significant hysteresis in the bulk. The interpenetration of
32 stitching polymers (such as silane coupling agents) can bridge hydrogels and
33 elastomers to improve adhesion.³³⁰ The silane coupling agent is added to the precursor
34 of both the hydrogel and elastomer. During the manufacturing process of the polymer,
35 the reactive silane coupling agent produce a strong bond on the interface. Alternatively,
36 adhesion can be improved by using benzophenone to modify the permeability of the
37 elastomer surface to hydrophilic polymers and monomers, which allowing the hydrogel
38 network to interpenetrate into the elastomer surface to form a firm bond.³²⁵

39 In addition, the integration of elastic networks with gels can also be achieved by
40 infiltrating high energy-dissipative polymeric fluids into an elastic polymer matrix
41 **(Figure 16c,d)**.^{331,332} For example, Liu et al. prepared polymer-fluid-gels (PFGs) by
42 photoinitiated free-radical polymerization of a prepolymer solution containing n-butyl
43 acrylate monomer, poly (n-butyl acrylate) (PBA) fluid, ethylene glycol dimethacrylate,
44 and 2,2-diethoxyacetophenone.³³² The resulting PFGs exhibit pronounced energy



1 dissipation over a broad frequency range (10^{-2} - 10^8 Hz), with loss factors exceeding 0.5, demonstrating their potential as high-performance impact-resistant materials. Owing to
2 this combination of high dissipative capacity and elastic integrity, PFGs hold promise
3 for applications in actuators, wearable devices, flexible electronics, and soft robotics.

5 **9.3 Weather resistance improvement of hydrogels**

6 *Improvement in anti-swelling property*

7 Considering that hydrogels will be used in vivo, high humidity environment and
8 even underwater in the future, its anti-swelling performance should be effectively
9 improved. The difference in osmotic pressure causes the swelling of hydrogels.^{333,334}
10 Swelling significantly weakens the mechanical toughness and limits applicability of
11 hydrogels.^{90,333} The development of anti-swelling hydrogels becomes an important but
12 challenging proposition. The degree of swelling is mainly affected by the hydrophilicity
13 of the polymer and the crosslinking degree.⁹⁰ Bearing in mind the impact of hydrophilic
14 polymers, a heteronetwork organohydrogels is obtained by combining the oleophobic
15 polymer network and hydrophobic polymer network to limit the swelling.^{335,336}
16 Because of the mutual restriction between the two networks, the organohydrogel can
17 maintain its original volume in various immersion environments with different
18 hydrophilic and hydrophobic properties. At the same time, the integration of
19 heteronetwork endows organohydrogels adjustable hydrophobic and oleophobic
20 properties according to soaking environment and stable elasticity over a wide
21 temperature range. For example, the poly(vinyl alcohol)/poly(butyl methacrylate)-co-
22 poly(lauryl methacrylate) multiple network organohydrogels (MN-OHGs) prepared by
23 Liu et al.³³⁷ When the ratio of the oleophilic polymer network (OPN) to the hydrophilic
24 polymer network (HPN) is 2/1, MN-OHGs can maintain the same swelling ratio in
25 water and n-dodecane. Moreover, the surface of MN-OHGs can dynamically transition
26 between hydrophobic (in oil) and oleophobic (in water) states (**Figure 16e**). The way
27 to control swelling by introducing thermoresponsive segments is also worth learning.
28 When the temperature rises to the critical temperature (T_c), the hydrophobic effect
29 caused by the thermoresponsive segments will dominate, causing a sharp shrinkage of
30 the hydrogel volume.³³³ On the other hand, enhancing the degree of crosslinking of
31 hydrogels is also an effective method for inhibiting swelling of hydrogels. For example,
32 coordination bonds or hydrogen bonds have been introduced into the network of a
33 crosslinked hydrogels to form a dual-crosslinked hydrogel, which has a significantly
34 reduced swelling rate while exhibiting excellent mechanical strength in a balanced
35 swelling state.³³⁸⁻³⁴⁰ In addition, collaborative strategy of combining organohydrogels
36 and dual-crosslinked can also effectively achieve the effect of inhibiting swelling.³³⁴

37 *Improvement in temperature adaptability*

38 Conventional hydrogels contain a large amount of water, which inevitably freezes
39 to lose flexibility and limit the transport of ions at sub-zero temperature environment
40 and dehydration at high temperature, which seriously affect the performance of
41 hydrogel-based devices. The freezing of water at low temperatures is a crystallization
42 phenomenon caused by strong hydrogen bonds between water molecules.³⁴¹

43 Methods to inhibit the freezing of hydrogels can be divided into two categories:
44 The first type of mechanism is to introduce additives to combine with water molecules

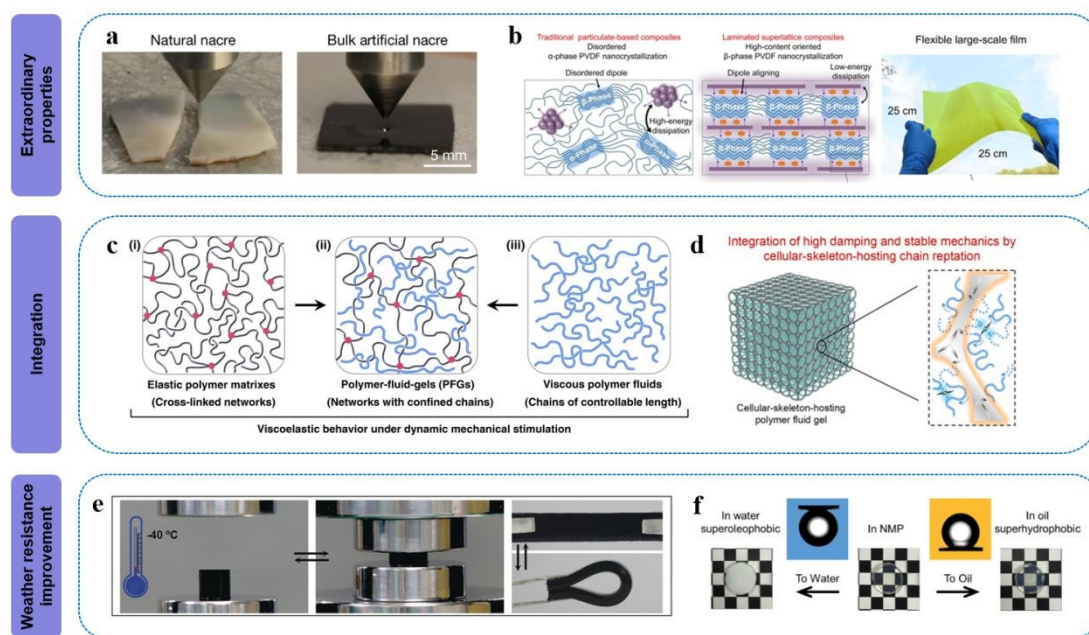


1 to weaken the interaction between water molecules. This method can reduce the
2 freezing point of the aqueous solution. Specific solutions include adding ionic salts³⁴²⁻
3³⁴⁵ or organic solvents.^{119,233,234,346} An antifreeze hydrogel that maintains flexibility at -
4 57 °C by adding CaCl₂(when CaCl₂ is 30 wt %).³⁴² Liu et al. added ethylene glycol to
5 the PVA hydrogel to prepare a conductive hydrogel that has stable flexibility and a high
6 ion conductivity of 2.38 mS cm⁻¹ at -40 °C (**Figure 16f**).¹¹⁹

7 The second option is to introduce nano-constraints of water molecules to inhibit
8 the ice recrystallization and growth. Studies have shown that hard confinement of water
9 clusters smaller than 10 nm can effectively prevent water crystallization.^{347,348}
10 Therefore, the introduction of a hard polymer network structure into the hydrogel s also
11 conducive to the antifreeze of hydrogels .

12 By introducing a lipophilic network that reducing the size of ice crystals into the
13 hydrogel network to suppresses water crystallization, the antifreeze hydrogel maintain
14 a stable modulus over a wide range of temperature, from -78 °C to 80 °C.³³⁵

15 Hydrogels are susceptible to dehydration in air , and the increase in temperature
16 will accelerate dehydration process, which is also a bottleneck in its application. Owing
17 to the strong interaction of the above-mentioned ionic salt and organic solvent for anti-
18 freeze, they can effectively stabilize water and prevent the volatilization of water.^{343,346}
19 Encapsulating hydrogel with an elastomer film can also effectively weaken the exchange
20 of substances between the hydrogel and the outside world, thereby reducing water
21 loss.^{324,349}



22 **Figure 16.** Future development directions for hydrogels. a-b) Hydrogels with extraordinary
23 properties: a) high strength and toughness nanocomposite gel.²⁷⁹ b) high strength and
24 magnetolectric performance nanocomposite polymer.²⁸⁰ c-d) Integration of hydrogels: c)
25 Construction of polymer-fluid-gels.³³² d) Cellular-skeleton-hosting polymer fluid gel.³³¹ e-f)
26 Weather resistance improvement of hydrogels: e) Enhanced temperature adaptability of
27 hydrogels.¹¹⁹ f) Enhanced swelling resistance of hydrogels.³³⁷

29 10. Conclusions and outlook



1 We have focused on the unique performance and working mechanisms of hydrogel-based sensing systems, as well as their potential applications. Although various functions of ion transport have been achieved based on hydrogels, the field is still in the toddler stage and have ample room for future development. Here, we will discuss the challenges faced by the hydrogel ion transport systems from four aspects: improving selectivity, synergy and systematization, bionic intelligence, enhancing performance.

7 **10.1 Improving selectivity**

8 The protein channel can precisely control the opening and closing of the channel under the synergistic effect, performing the selective input and output of ion Na^+/K^+ in the cell to further perform the generation and conduction of nerve impulses.

11 However, the ion selectivity and rectification of the hydrogel ion sensor system can only be limited to the choice of anions and cations. The separation of lithium in magnesium/lithium brine using nano-heterogeneous membranes has given us enlightenment, but there are still limitations.³⁵⁰ Solving the above insurmountable challenges will help to realize more advanced ion sensing functions.

16 Numerous receptors in the human body can clearly distinguish a variety of stimuli. In order to realize this multi-stimulus perception, integrated sensors have been developed. However, the current sensor systems lack a differentiated monitoring mechanism for different stimuli, which leads to the generation of stimulus interference. How to accurately distinguish multiple stimuli simultaneously to meet reliable sensing requirements is a challenging topic.

22 **10.2 Inter-module coupling challenges**

23 Hydrogel-based ion sensing modules (sensors, transporters, processors, effectors, and power sources) have shown unique effects in many aspects. However, we cannot ignore that its systematic research progress remains in its infancy. The practical implementation of hydrogel ionic sensory systems critically depends on the efficient coupling between these modules. Nevertheless, fundamental differences in charge carriers, transport mechanisms, and interface properties introduce several key challenges, including impedance mismatch, signal attenuation, and cross-interference. To address these challenges, several strategies have been explored. Structurally, designing graded or hybrid interfaces can reduce impedance mismatch and enhance charge transfer efficiency.^{1,351} Functionally, ion-selective channels, nano-confined pathways, and heterostructured hydrogels can regulate ion transport and suppress signal dispersion.³⁵² Additionally, encapsulation strategies and multi-phase systems can minimize ion leakage and environmental interference, thereby improving signal stability.³⁵³ Finally, integrating signal amplification and rectification units within the system can compensate for signal attenuation and enhance overall robustness.³⁵⁴

38 Overall, achieving seamless coupling between modules remains a central challenge for hydrogel ionic sensory systems. Future progress will rely on the co-design of materials, interfaces, and device architectures to enable efficient, stable, and low-noise signal transmission across multi-component ionic circuits

42 **10.3 Bionic intelligence**

43 Imitating creatures to make autonomous decisions is the future development direction of intelligent systems. On the one hand, studies have confirmed that animals



1 can respond to stimuli through an autonomous, non-centrally controlled system. Thus, the dynamic structure of the ionic decision maker is expected to achieve adaptive decision-making of the system, which will significantly reduce computing requirements and simplify systems. On the other hand, the combination of machine learning can help the hydrogel ion sensing systems make decisions with minimal human interference by learning from data. A more autonomous and self-improving sensor system will perform tasks in a smarter way.

10.4 Enhancing performance

There is ample room for improvement in the performance of hydrogels in numerous applications. For example, the ion conductivity (10^{-3} - 10^3 S m^{-1}) and thermal conductivity of hydrogels (~ 0.6 W m^{-1} K $^{-1}$) are much lower than that of metals; the electrochemical window of hydrogels is narrow, which makes it difficult to meet high-voltage direct current applications; compared to inorganic nanomaterials, the sensitivity of hydrogel-based strain sensors are comparatively low; the power density of the hydrogel actuator is only in the range of 0.1 to 10^3 W m^{-3} , which is far from the actuation power density of mammalian skeletal muscle; Breaking through the aforementioned challenges in the performance of hydrogels will effectively extend the range of applications of hydrogels sensing systems

With the development of hydrogel-based ion sensing technology, more "human-like" devices will appear. By integrating these devices, a mature hydrogel ion sensory system will be established, which can operate seamlessly between artificial machines and biological systems.

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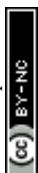
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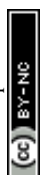
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Data Availability Statement

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This study is based on previously published data, which are cited in the article. No new data were generated or analyzed.

