



## Hydrogel soft robotics

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### ABSTRACT

With the rapidly growing attention to human-robot interfaces, soft robotics has attracted a great deal of interest. Soft robots have diverse advantages, including compliancy and safety, which contribute to seamless interactions with humans. To boost progress in the field, there is a need for compliant materials. Hydrogels are promising as compliant materials for soft robots because of their outstanding features, including high stretchability, transparency, ion conductivity, and biocompatibility. Furthermore, hydrogels provide innovative capabilities for soft robotics based on their unique responsiveness to stimuli. In this review, we discuss the unique features of hydrogel-based soft robots, from their fundamental working mechanisms to notable applications. Finally, we suggest perspectives on future directions that addressing potential challenges in the field of hydrogel soft robotics.

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## 1. Introduction

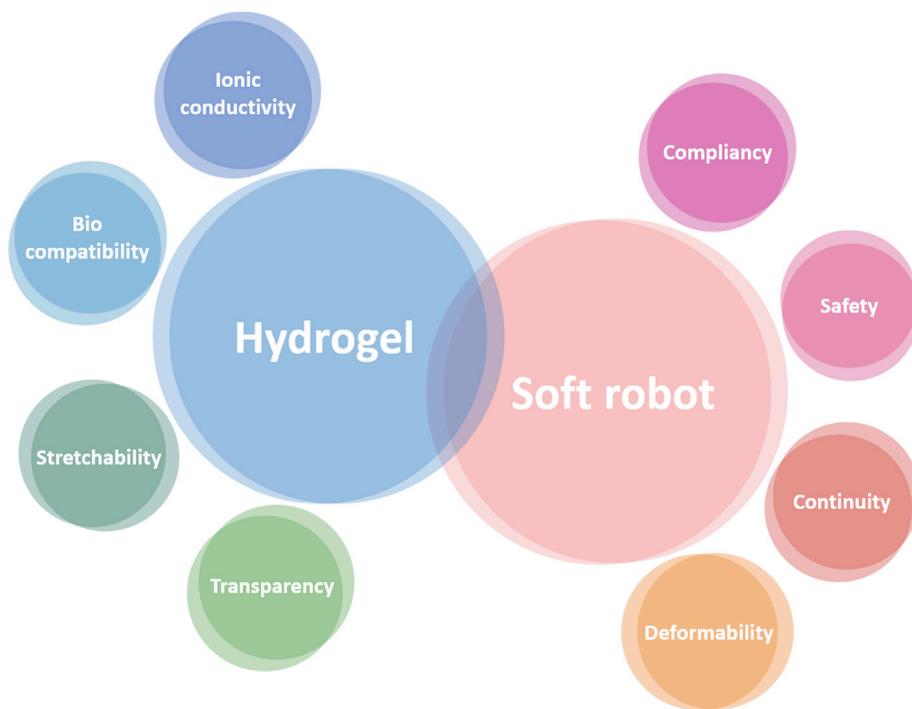
Most robots are built using rigid materials. These robots are designed to efficiently carry out preplanned tasks, and they have contributed to the progression of humanity by significantly improving productivity in industrial fields. Recently, people have begun trying to use robots in daily life. However, conventional rigid-bodied robots struggle to operate in unstructured environments because they are composed of discrete links and joints. Moreover, differences in elastic modulus between robotic components and the tissues of living organisms raise safety concerns. The desire for compliant and safe robots has led to the development of soft-bodied robots [1]. Since the continuously deformable bodies of soft-bodied robots give them a high degree of freedom, they can flexibly handle irregular tasks in a less complicated manner than traditional robots. In addition, they can safely and comfortably interact with humans because they are composed of compliant materials that have an elastic modulus similar to that of the soft tissues found in biological systems. Thus, there has been rapid progress in the development of compliant materials to meet the growing need for soft robots (Fig. 1).

Hydrogels are composed of water and hydrophilic polymer chains arranged in a network structure [2–4], as shown in Fig. 2. Cross-linked polymer chains render hydrogels into a three-dimensional elastic solid. The configuration of the polymer network can be adjusted so that it can endure mechanical strain of up to ~1,000% [5]. The elastic modulus of hydrogels is in the range of 1–100 kPa, which is softer than that of other compliant materials, owing to the large volume fraction of water [2]. A hydrogel is biocompatible if its polymer network consists of non-toxic polymers [4]. Thus, hydrogels can be introduced into the field of soft robotics to broaden the range of biological applications. As with water, the transparency of a hydrogel is up to ~99%, even in the bulk state, because the polymer network of the hydrogel absorbs a negligible amount of visible light [6]. This high degree of transparency allows optical information to be transmitted through a hydrogel. Since hydrogels can respond to diverse external stimuli via interactions between the polymer network and water, they endow soft robots with increased functionality [7,8]. Moreover, mobile ions dissolved in the water enable a hydrogel to act as an ionic conductor with a conductivity of up to ~10 S/m [3]. Although a hydrogel has a higher resistivity than other soft electrical materials, the increases in electrical resistance with strain are orders of magnitude lower, because configuration changes in a polymer network rarely affect the migration of mobile ions [9]. Thus, hydrogels can be used as electrodes when a high degree of stretchability is required, such as in soft robots. In addition, hydrogels can be equipped with a multitude of other functions

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**Fig. 1. Features of hydrogels and soft robots.** A widespread desire for robots in everyday life has led to the development of soft robots. Their continuously deformable bodies allow them to be safe and compliant. Fascinating features, such as biocompatibility, ionic conductivity, stretchability, and transparency make hydrogels a promising material for soft robots.

because they can hold any additives that are larger than the mesh of the polymer network [10,11]. Thus, hydrogels are a promising material for soft robots based on their fascinating features (Fig. 3).

In this review, we discuss hydrogel-based soft robots through the lens of five essential components: actuator, sensor, communicator, power source, and computation circuit. We also cover soft robotic components using organogels and ionogels in which the water is replaced with an organic solvent and an ionic liquid, respectively. We provide an overview of the fundamental working mechanisms of each component and highlight the roles played by the hydrogel. Finally, we suggest future directions for hydrogel-based soft robots.

## 2. Hydrogel-based soft actuators

The actuator of a soft robot is a component that generates mechanical motion. Since hydrogel-based soft actuators can be manipulated by diverse stimuli, various candidates are available to control them. A hydrogel-based actuator is organized into six sections based on input stimuli. We highlight the actuation mechanisms that play pivotal roles in hydrogel-based actuators and summarize their wide variety of applications. At the end of this section, challenges and future work will be discussed.

### 2.1. Thermally responsive actuators

Thermally responsive hydrogel actuators are based on volumetric changes of hydrogels in response to changes in ambient temperature. Such actuators selectively respond to a certain temperature regime to produce large deformations [12–19]. Furthermore, since the critical temperature can be tuned, this attribute has a wide range of applications [19,20]. Thermally responsive hydrogel-based actuators fall into two categories; actuators with lower critical solution temperature (LCST) and upper critical solution temperature (UCST).

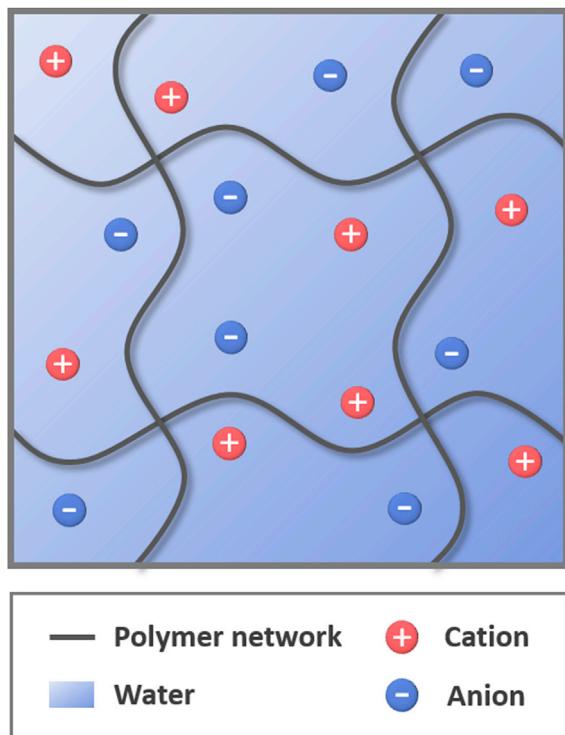
As shown in Fig. 4a, LCST and UCST actuators show opposite responses to temperature [19]. Hydrogels with LCST, such as poly(*N*-isopropylacrylamide) (pNIPAM), experience entropy-driven shrinkage at temperatures above the critical temperature and swell back to their normal state at low temperatures [21–23]. In contrast, hydrogels with UCST, such as poly(acrylic acid-co-acrylamide), experience enthalpy-driven swelling at temperatures above the critical temperature and shrink back to normal at low temperatures [20,21,23]. Based on their unique characteristics, efforts have been made to improve the performance of thermally responsive hydrogels; such efforts include enhancing agility [15,24], minimizing hysteresis during the volume phase transition [16,25,26], and broadening the operating environment (Fig. 4b) [12,18,27].

### 2.2. Chemically responsive actuators

Chemically responsive hydrogel actuators are based on volumetric changes of hydrogels in response to chemical stimuli. The hydrogels can directly convert the chemical potential of the surrounding environment to mechanical motion [12,28–36]. Chemically responsive hydrogel-based actuators fall into several underlying categories, including solvent-responsive actuators, pH-responsive actuators, and biomolecule-responsive actuators.

As shown in Fig. 5a, volumetric change of a solvent-responsive actuator depends on the difference in hydrophobicity between the polymer network and the solvent [32]. A hydrogel can absorb more solvent when the difference in hydrophobicity between the solvent and the polymer network is smaller. Thus, most hydrogels shrink in an organic solvent and return to their initial state in water (Fig. 5b). Based on this fundamental characteristic, intensive studies have been done on ways to control the actuation direction of a hydrogel, e.g. by using geometric design [32,37], modulus gradient [35,38], and application of a passive layer [36,39–41].

As shown in Fig. 5c, volumetric changes of a pH-responsive hydrogel actuator depend on ionization of the polymer network



**Fig. 2. Chemical structure of a hydrogel.** A hydrogel is composed of ample water and a hydrophilic polymer network. Cross-linked polymer chains render hydrogels into a three-dimensional elastic solid. The high volume fraction of water allows dissolution of salt.

under certain pH conditions [28], pH-responsive hydrogels can be classified into polyanionic and polycationic hydrogels based on the charge of ions fixed to the polymer chain. The polymer chains of polyanionic hydrogels are negatively ionized in solutions with a pH greater than their acid dissociation constant ( $pK_a$ ) [42–44]. Conversely, the polymer chains of polycationic hydrogels are positively ionized in solutions with a pH lower than their  $pK_a$ . The ionized functional groups induce electrostatic repulsion between adjacent functional groups and make the polymer network more hydrophilic. Extension of a polymer chain by electrostatic repulsion

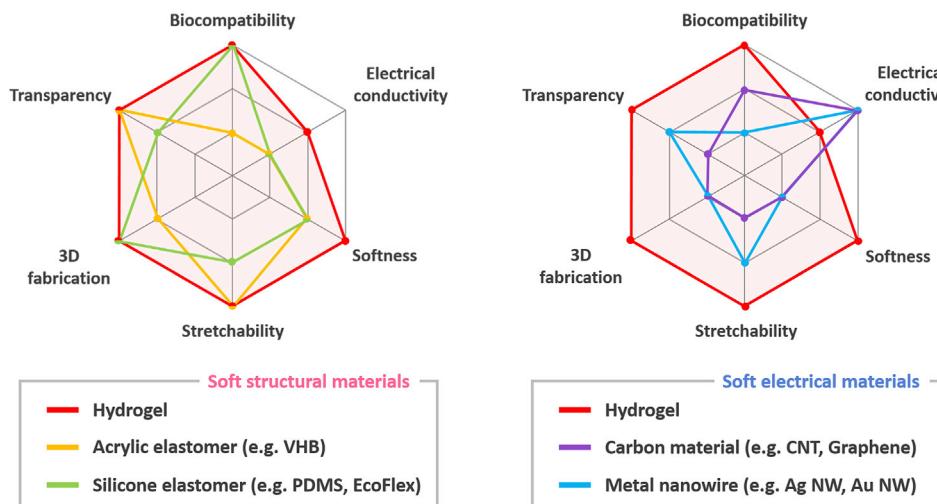
and change in hydrophobicity allows a hydrogel to absorb more water. The volume of the expanded hydrogel reversibly returns to its initial state when the polymer chain is deionized by a pH change. These basic characteristics have led to impressive attempts to enhance the usability of pH-responsive hydrogels, such as by adjusting the operating pH range, improving response time, and controlling the actuating direction [28,31,45–47] (Fig. 5d).

Volumetric changes of biomolecule-responsive actuators are based on reversible association and dissociation of biomolecular complexes [48–52]. Biomolecules such as polynucleotides and antibodies can selectively form molecular complexes with certain complementary biomolecules. Thus, biomolecular complexes fixed to hydrogel polymer networks can act as active hydrogel crosslinks. When input biomolecules diffuse into a biomolecule-responsive hydrogel, biomolecular complexes that act as hydrogel crosslinks associated or dissociated. Changes in crosslinking length or density induce a volumetric phase transition of the hydrogel, as shown in Fig. 5e [49–54]. Since biomolecule-responsive hydrogels selectively respond to specific input biomolecules, multiple domains can be controlled separately (Fig. 5f).

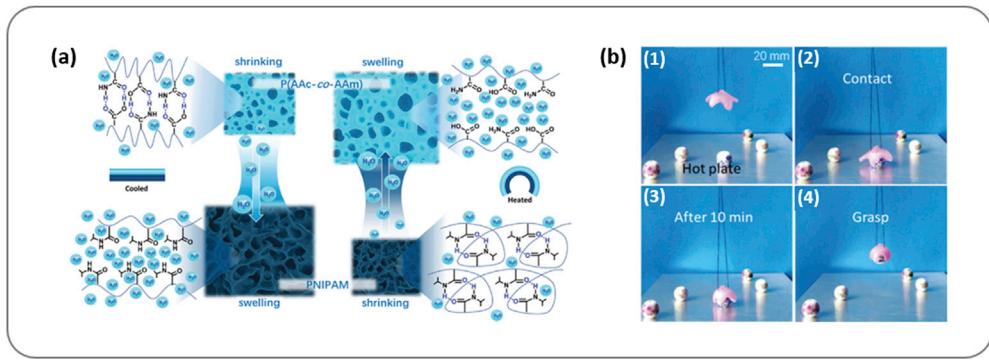
### 2.3. Optically responsive actuators

Optically responsive hydrogel actuators are based on volumetric/shape changes of hydrogels in response to light irradiation. These actuators do not require a physical connection for energy transfer. Furthermore, they can selectively respond to light of certain wavelengths [55–58]. Optically responsive hydrogel-based actuators fall into three categories with distinct underlying working mechanisms.

Photoswitchable moieties such as spiropyran and azobenzene selectively respond to light of certain wavelengths and experience reversible isomerization. Spiropyran dissociates under ultraviolet (UV) light and transforms back into a ring-closed structure under visible light [55,59]. The reversible isomerization results in a volume transition of the hydrogel due to a change in hydrophobicity. A hydrogel-based actuator composed of spiropyran can act as a reversible photo valve for microfluidics [55]. Meanwhile, azobenzene maintains the *trans* form under visible light and transforms into the *cis* form under UV light [56,60–67]. Isomerization of azobenzene induces a change in cross-linking density and length of the polymer chain, as shown in Fig. 6a [60,68–70]. Thus, a hydrogel



**Fig. 3. General characteristics of the materials used in soft robots.** Based on their fascinating features, hydrogels can serve as both structural and electrical materials for soft robots.



**Fig. 4. Thermally responsive hydrogel-based soft actuators.** (a) Schematic diagram showing the working mechanisms of thermally responsive actuators. As temperature increases, a hydrogel with a lower critical solution temperature (LCST) swells and a hydrogel with an upper critical solution temperature (UCST) shrinks [18]. (b) By stacking two different types of temperature-responsive hydrogels, a fabricated soft gripper with a bilayer structure can operate even in open-air environments [18].

with an azobenzene moiety can respond to UV/Vis light irradiation to reversibly swell and shrink (Fig. 6b).

Thermally responsive hydrogels also can be controlled by light irradiation through photothermal conversion (see the section titled “Thermally responsive hydrogel-based actuator” for details on thermally responsive hydrogels). To enhance the photothermal conversion efficiency, additives such as carbon nanotubes [57,71–73] and gold nanoparticles [74–76] are embedded in the hydrogels (Fig. 6c) [75]. Based on the plasmonic effect, the additives selectively absorb light of a certain wavelength [76,77]. This allows the hydrogel to selectively and rapidly respond to light of the desired wavelength (Fig. 6d). Much work has been done to maximize the outstanding features of optically responsive hydrogels, which include enhancing the agility of actuation and maximizing deformation [74,75,78].

Recently, in order to exploit the outstanding performance and efficiency of biological actuators in robots, attempts have been made to combine living cells directly with artificial components [79–81]. Mammalian skeletal muscle cells with blue light-sensitive cation channels can be cultured in a natural hydrogel matrix (Fig. 6e) [81]. As shown in Fig. 6f, the resultant muscle ring can contract under light with a wavelength of 470 nm. Thus, combining an optogenetic muscle ring with an asymmetric hydrogel skeleton results in a bio-hybrid actuator that can convert noninvasive optical stimuli into directional locomotion (Fig. 6g).

#### 2.4. Electrically responsive actuators

Electrically responsive hydrogel actuators are based on volumetric/shape changes of hydrogels in response to electric stimuli. They can be controlled quickly and accurately using computational circuits without the need for conversion of any stimuli [6,82,83]. Electrically responsive hydrogel-based actuators fall into two categories with distinct underlying working mechanisms.

The first type of electrically responsive actuator is based on Maxwell stress [6]. The actuator is called a dielectric elastomer actuator (DEA) and comprises a dielectric elastomer layer sandwiched between two ionically conductive hydrogels. When a high voltage is applied between the hydrogels, ions of opposite charge accumulate along each hydrogel/elastomer interface. This induces Maxwell stress between the hydrogels, resulting in thickness contraction and areal expansion of the elastomer layer (Fig. 7a). The electrically responsive hydrogel-based actuator is an attractive prospect for use in daily life because of its capacity for large deformation, fast response, high transparency, and strong force

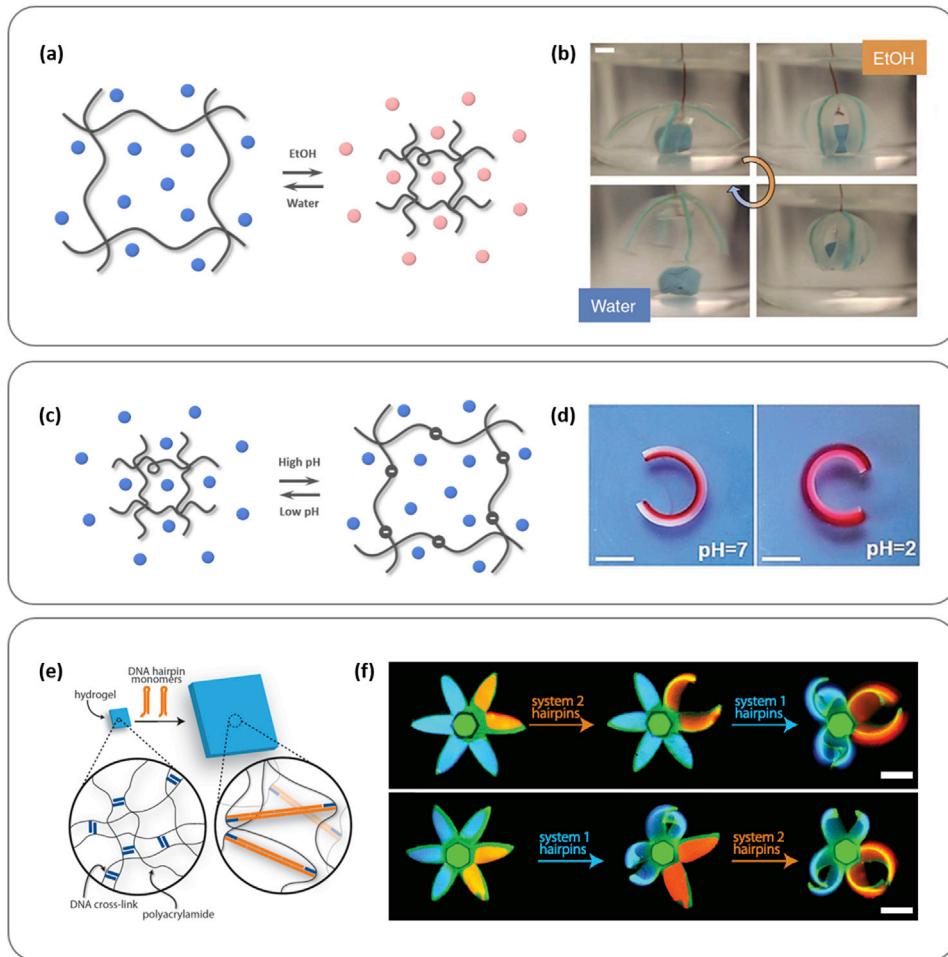
(Fig. 7b). Given the basic characteristics of such actuators, a valuable attempt was made to maximize the vertical amplitude of the DEA by adding an air gap in the middle of the elastomer layer [84]. Moreover, there have been intensive efforts to broaden the scope of applications of such hydrogel-based actuators such as in artificial muscles [6,82,85–90].

The second type of electrically responsive hydrogel-based actuator is based on electrically induced osmotic pressure [83,91,92]. A polyelectrolyte hydrogel is composed of polymer networks with charged functional groups and mobile counter ions; only the counter ions migrate toward the electrode when an electric field is applied to the hydrogel (Fig. 7c) [83]. To satisfy charge neutrality, ions in aqueous medium with the opposite charge as the counterions of the polyelectrolyte hydrogel simultaneously migrate toward the same electrode. Consequently, the formation of ionic gradients creates osmotic pressure that causes the polyelectrolyte hydrogel to swell asymmetrically (Fig. 7d). There have been numerous attempts to apply the capacity for large deformation, fast response time, and strong force associated with such actuators [93–95].

By combining muscle cells with compliant materials, soft actuators with the efficiency and performance of biological systems have been realized (Fig. 7e) [79,96–99]. For example, mammalian skeletal muscle myoblasts were seeded in a natural hydrogel matrix to produce an electrically responsive muscle strip. As shown in Fig. 7f, the contraction of cells in the muscle strip was converted into directional locomotion using three-dimensional printed hydrogel beams with asymmetric structures. Under 1 Hz electrical stimulation, the fabricated bio-bot could move 1.5 body lengths per minute (Fig. 7g).

#### 2.5. Magnetically responsive actuators

Magnetically responsive hydrogel actuators are based on volumetric changes of hydrogels in response to an external magnetic field. They can be controlled wirelessly and have prompt response times [100–104]. As shown in Fig. 8a, such actuators generally comprise hydrogels filled with micro/nano-sized magnetic particles (e.g.  $\gamma$ - $Fe_2O_3$ ,  $Fe_3O_4$ ,  $CoFe_2O_4$ ) [100]. When an external magnetic field is applied, the magnetic particles dispersed in the hydrogel receive the magnetic force and transmit that force to the hydrogel matrix, resulting in a shape change (Fig. 8b). These actuators are promising for applications in artificial muscles and several other biomedical fields [100–102,105,106]. Notably, magnetically responsive hydrogels are suitable for drug delivery because they allow for loading of an abundant quantity of drugs, and they release



**Fig. 5. Chemically responsive hydrogel-based soft actuators.** (a) Schematic diagrams showing the working mechanism of solvent-responsive actuators. A hydrogel can absorb more solvent as the difference in hydrophobicity between the solvent and the polymer network becomes smaller. (b) A fabricated actuator can grip a small object in ethanol (EtOH) and release it in water [32]. (c) Schematic diagram showing the working mechanism of anionic pH-responsive actuators. Ionization of the polymer network under certain pH conditions induces electrostatic repulsion between adjacent functional groups and makes the polymer network more hydrophilic. This allows the hydrogel to absorb more water. (d) By stacking anionic and cationic pH-responsive hydrogels, a fabricated actuator can induce greater deformation [28]. (e) Schematic diagram showing the working mechanism of a biomolecule-responsive actuator. Sequence-specific DNA monomers can insert into crosslinks, inducing 100-fold volumetric expansion of the hydrogel [48]. (f) Multiple domains actuate in response to different DNA sequences [48].

those drugs following stimulation by a non-invasive external magnetic field.

## 2.6. Hydraulically responsive actuators

Hydraulically responsive hydrogel actuators undergo shape changes in response to hydraulic pressure. They can produce higher actuation forces and speeds than other existing hydrogel-based actuators [107]. Furthermore, the high transparency of hydrogels potentially allows for passive camouflage of the actuators in all different kinds of backgrounds (Fig. 9a). Their utility is especially notable in underwater environments due to the very similar refractive indexes of hydrogels and water. In addition, since hydrogels have a similar acoustic impedance as water, they can also be sonically camouflaged in water. Based on these advantages, hydraulically responsive hydrogel-based actuators can be utilized in applications requiring stealthy operation (Fig. 9b).

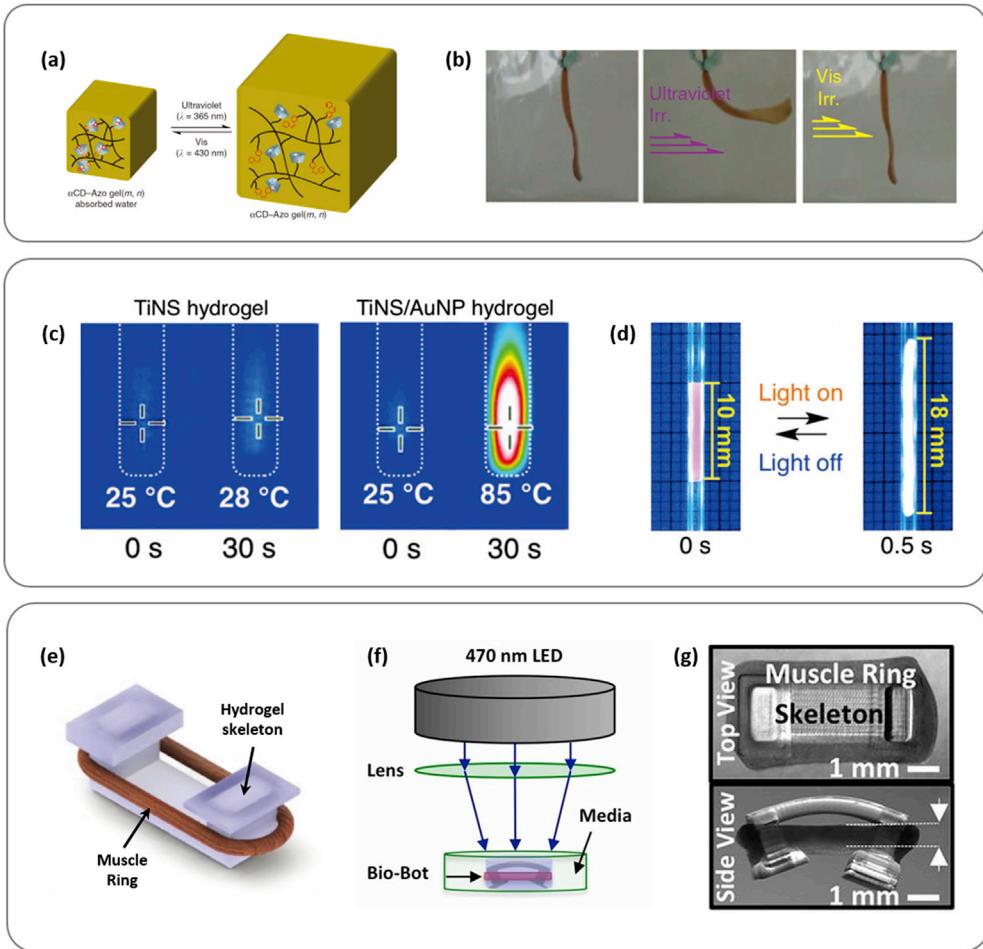
## 2.7. Summary of hydrogel-based soft actuators

This section outlined hydrogel-based soft actuators broken down by type of input stimulus. Although impressive progress has been made in the field, hydrogels have to meet rapid and large

deformation requirements with minimal input stimuli, somewhat limiting their practical usefulness. Furthermore, even though the relatively low modulus of hydrogels is fascinating and allows hydrogel-based actuators to be compliant, they are not suitable for application requiring strong force. It is expected that endowing hydrogels with controllable stiffness, like muscle tissue, will give hydrogel-based actuators both adaptability and strength and help overcome this challenge.

## 3. Hydrogel-based soft sensors

The sensor of a soft robot is a component that measures changes in the surrounding environment. It converts an external stimulus into a form that can be quantitatively analyzed. Since a hydrogel-based soft sensor can convert external stimuli into diverse forms, their use has broadened the applications of soft robots. Hydrogel-based sensors can be divided into five groups based on their target stimuli. We highlight sensing mechanisms that play pivotal roles in hydrogel-based sensors and summarize their applications. At the end of this section, challenges and future work will be discussed.



**Fig. 6. Optically responsive hydrogel-based soft actuators.** (a) Schematic diagram showing the working mechanism of an optically responsive actuator, which is based on isomerization of photoswitchable moieties. Photo-isomerization of an azobenzene moiety induces volume transition of the hydrogel by changing the density of molecular complexes that act as crosslinks [60]. (b) Hydrogel deformation by ultraviolet irradiation and restoration to its initial state by visible (Vis) light irradiation [60]. (c) Photographs showing the working mechanism of a photo-responsive actuator based on a thermally responsive hydrogel. When a laser with a wavelength of 445 nm was projected onto the actuators, gold nanoparticles (AuNPs) effectively converted light into heat by plasmon absorption, and the thermally responsive hydrogel expanded [75]. (d) A cylindrical hydrogel expanded 1.8 times in length within 0.5 s when the laser was turned on [75]. (e) Schematic illustration of a bio-bot based on optically responsive bio-hybrid actuators. A natural hydrogel was used as a matrix for mammalian skeletal muscle cells with blue light-sensitive cation channels [80]. (f) Contraction occurs when light with a wavelength of 470 nm is focused on the bio-bot [81]. (g) For directional movement, the hydrogel skeleton was 3D-printed in an asymmetric structure [81].

### 3.1. Chemical sensors

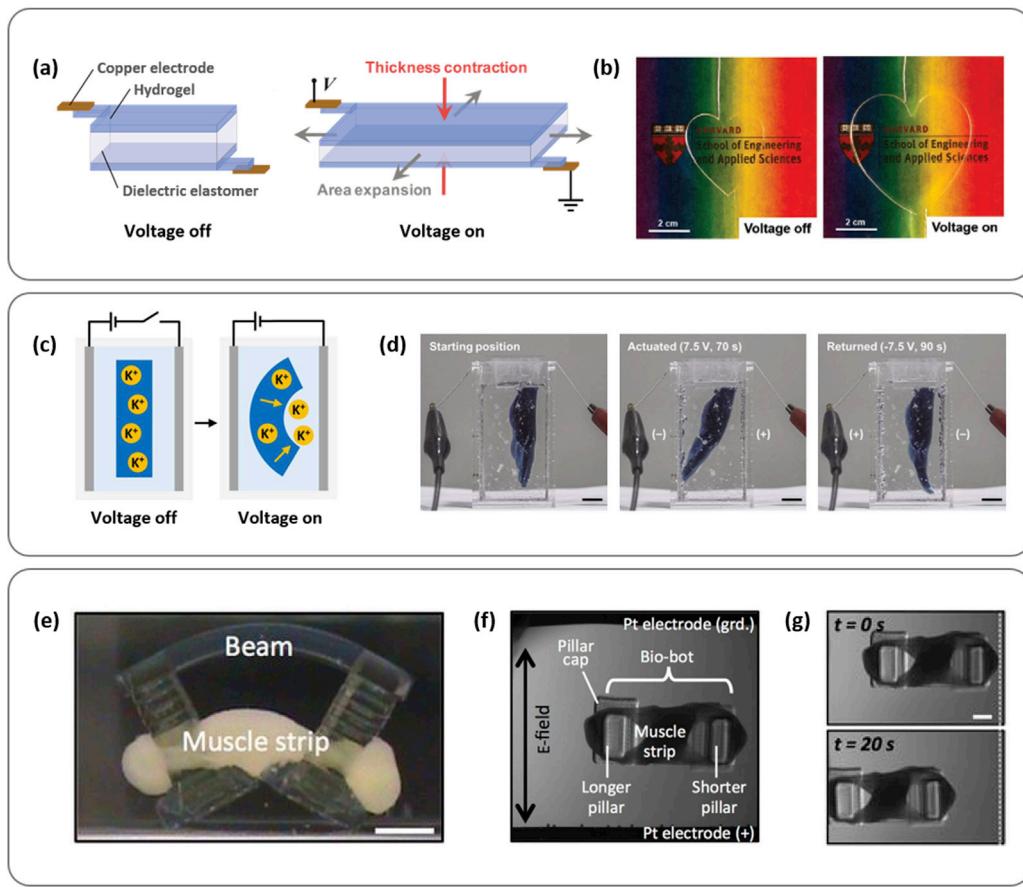
Hydrogel-based chemical sensors convert external chemical stimuli into electrical and optical form. They are particularly adept at visualization of chemical stimuli for quick and simple analysis [10,108–110]. Hydrogel-based chemical sensors fall into three categories, sensors based on polymer-solvent interactions, those that use ionic conductivity, and sensors that take advantage of additional functionalities of additives.

Chemical stimuli can be quantitatively measured using volumetric changes of chemically responsive hydrogels that occur due to interactions between the polymer network and the solvent (Fig. 10a) (see the section titled “**Chemically responsive actuator**” for details on chemically responsive hydrogels). If the thickness of the hydrogel layer [108] or the spacing of periodic structures embedded in the hydrogel [111–117] is adjusted on the nano scale, a hydrogel can diffract visible light in a manner that satisfies Bragg's law. As shown in Fig. 10b, the swelling and shrinking of a chemically responsive hydrogel changes the color given off by light diffraction,

thus enabling visualization of chemical stimuli. In addition, the integration of a chemical-responsive hydrogel with a pressure sensor allows chemical stimuli to be measured by pressure changes [118].

As shown in Fig. 10c, a parallel electrode array encapsulated by a dielectric can identify different liquid molecules using capacitance responses [119]. When the electrodes are connected to an AC power source, the capacitance between electrodes depends on the physical properties of the liquid, such as polarity, volatility, and wettability. Thus, by using an ionogel as a compliant electrode, a soft chemical sensor capable of recognizing different polar molecules can be realized (Fig. 10d).

Hydrogels can contain functional particles [109,120]. Moreover, cells can be cultured three-dimensionally in a hydrogel because the structure of a hydrogel is similar to the extracellular matrix of biological tissue (Fig. 10e) [10,110,121]. Thus, if chemically responsive additives are used, a hydrogel can be employed as a soft chemical sensor. When a target chemical comes into contact with the hydrogel, it diffuses to the additives in the hydrogel. In response

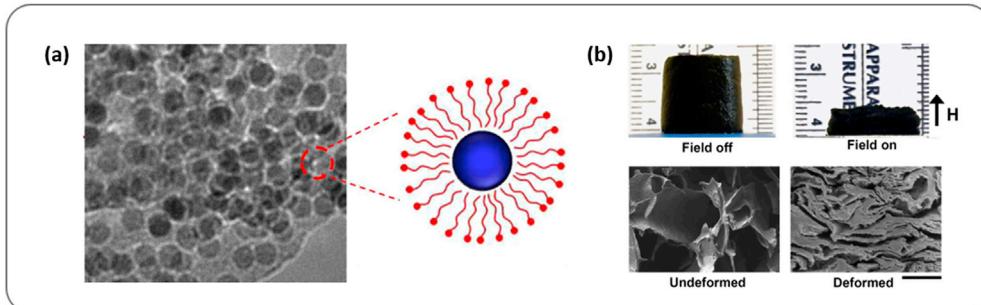


**Fig. 7. Electrically responsive hydrogel-based soft actuators.** (a) Schematic diagram showing the working mechanism of a dielectric elastomer actuators (DEA). Applying voltage to the hydrogel layer induces an electric field through the dielectric elastomer. Electromechanical transduction is achieved by the resulting Maxwell stress, which pressurizes the dielectric elastomer between the hydrogel layers [6]. (b) The transparent actuator is capable of high-speed and large-strain deformation [6]. (c) Schematic diagram showing the working mechanism of electric-responsive actuators based on electrically induced osmotic pressure. When an electric field is applied to the polyelectrolyte hydrogel, only mobile ions migrate toward the electrode and the hydrogel bends by electrically induced osmotic pressure [83]. (d) A 3D-printed hydrogel hand can reversibly actuate in response to applied voltage [83]. (e) Photograph of bio-bot based on electrically responsive bio-hybrid actuators. Mammalian skeletal muscle cells cultured in a natural hydrogel matrix were fixed to the hydrogel beam [96]. (f) The muscle cells contract in response to electrical stimulation, and the asymmetric structure of the hydrogel beam converts the contraction into directional movement [96]. (g) The fabricated bio-bot can move 1.5 body lengths per minute [96].

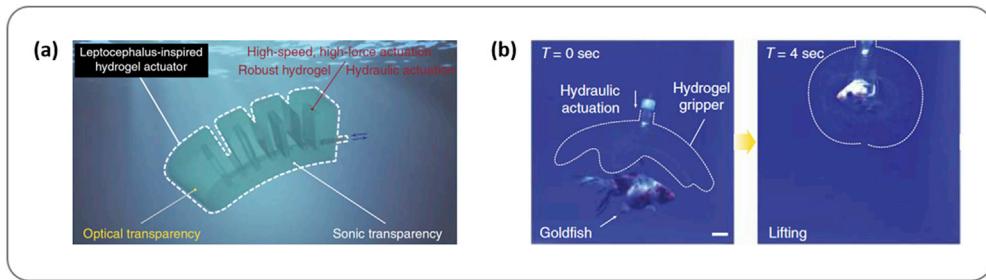
to the chemical stimulus, the optical properties of the additives change and the generated optical signals are transmitted through the transparent hydrogel. Plasmonic metal nanoparticles [120], pH-sensitive organic dyes [109], and genetically engineered bacterial cells [10,110,121] have been used as chemically responsive additives for applications such as long-term wound monitoring, in vivo cellular cytotoxicity sensors, and wearable chemical sensing patches (Fig. 10f).

### 3.2. Temperature sensors

Hydrogel-based temperature sensors convert temperature stimuli into volumetric and electric responses. They are essential component for estimating the thermodynamic state of a system. Hydrogel-based temperature sensors can be divided into two underlying categories, sensors using polymer-solvent interactions and sensors using ionic conductivity.



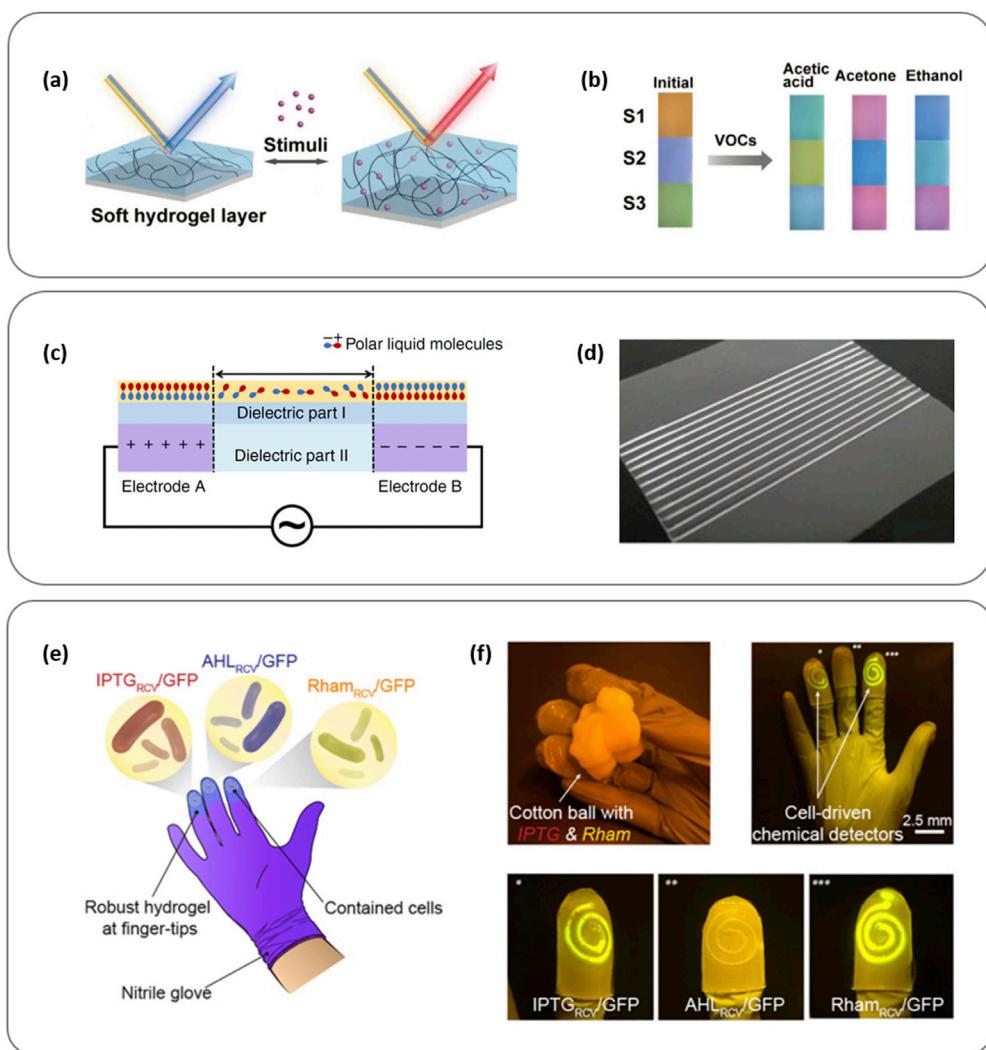
**Fig. 8. Magnetically responsive hydrogel-based soft actuators.** (a) Transmission electron microscope images of iron oxide nanoparticles coated with Pluronic F127. Under an external magnetic field, embedded iron oxide particles induced deformation of the hydrogel scaffold [100]. (b) The volume of the macroporous hydrogel decreased by 70% under a magnetic field with a gradient of  $38 \text{ A/cm}^2$  [100].



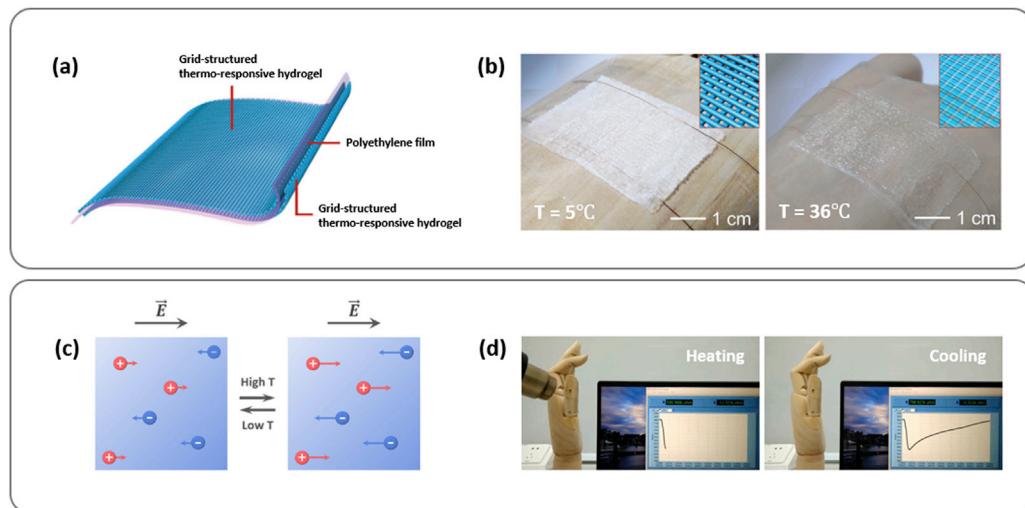
**Fig. 9. Hydraulically responsive hydrogel-based soft actuators.** (a) Schematic illustration of hydraulic responsive actuators. Such actuators can be optically and sonically camouflaged in water [107]. (b) A transparent hydrogel gripper catches a live goldfish in water [107].

Based on changes in the interactions between a polymer network and a solvent, a thermally responsive hydrogel undergoes a volumetric transition in response to an ambient temperature change (see the section titled “Thermally responsive actuator” for details on thermally responsive hydrogels). As shown in Fig. 11a, when a dielectric layer is sandwiched between two

thermally responsive hydrogel electrodes laid out in a grid structure, the contact area between the electrodes and the dielectric depends on the diameter of the hydrogel fibers [122]. Thus, swelling and shrinking of the hydrogel fibers can be converted into changes in capacitance between the electrodes (Fig. 11b). Moreover, by converting volumetric changes of a thermally



**Fig. 10. Hydrogel-based soft chemical sensors.** (a) Schematic diagram showing the working mechanism of a chemical sensor based on chemically responsive hydrogels. The volume of the hydrogel layer changes in response to chemical stimuli and diffracts light of different colors [108]. (b) Via integration of hydrogel layers with different thicknesses, volatile organic compounds (VOCs) can be recognized based on their distinct color patterns [108]. (c) Schematic illustration of a capacitive chemical sensor. When two electrodes are connected to an AC power source, the sensor can identify different liquid molecules based on their capacitive responses [119]. (d) A sensor array fabricated by a 3D printer [119]. (e) Schematic illustration of a bacterial cell-based chemical sensor. Each genetically engineered bacterial cell, contained in a hydrogel matrix, produces green fluorescent protein in response to different chemicals [110]. (f) Chemical sensing patches attached to the fingers [110].



**Fig. 11. Hydrogel-based soft temperature sensors.** (a) Schematic illustration of a capacitive temperature sensor with a grid-structured thermally responsive hydrogel electrode [122]. (b) As the temperature increases, the thermally responsive hydrogel fiber expands, and the capacitance between the two electrodes increases [122]. (c) Schematic diagram showing the working mechanism of a resistive temperature sensor. As temperature rises, the migration rate of ions under the same electric field increases and electrical resistance decreases. (d) Electrical resistance decreases when the sensor is heated and is restored to its initial state when the heating source is removed [119].

responsive hydrogel into optical signals using Bragg diffraction [123–126] or the plasmon effect [127], changes in ambient temperature can be visualized.

The electrical resistance of a hydrogel depends on the migration rate of mobile ions in the hydrogel under an electric field. Thus, temperature can be measured using electrical resistance since the migration rate of mobile ions is proportional to temperature (Fig. 11c) [119,128]. By simply connecting a piece of the hydrogel to an LCR meter, ambient temperature can be measured in real time (Fig. 11d).

### 3.3. Stress/strain sensors

Hydrogel-based stress/strain sensors convert mechanical stimuli into optical and electrical form. They can prevent mechanical failures in a soft robot by monitoring the applied stresses and strains. Hydrogel-based stress/strain sensors can be organized into four categories based on their working mechanism.

A highly stretchable and transparent capacitive stress/strain sensor can be created by sandwiching a dielectric elastomer between two hydrogels [9,82,119,122,128–131]. When the sensor is deformed by a mechanical stimulus, the contact area between the hydrogel and the dielectric increases, and the distance between the two hydrogels decreases (Fig. 12a). Thus, if the sensor is connected to an LCR meter, stress and strain can be measured using the change in capacitance between the hydrogels (Fig. 12b). Furthermore, by using an electrical double layer (EDL) form at the interface of the ionic and electronic conductors, a supercapacitive stress sensor can be made without any dielectrics [132–137]. When the voltage across the EDL is within an electrochemical window, no electrochemical reaction occurs, and the EDL behaves like a capacitor with large areal capacitance [6]. Thus, using the ion-squeezing effect [132,133] or structural design [134–137], which can convert mechanical stress into changes in EDL capacitance, a supercapacitive sensor can measure applied stress.

Since the migration of mobile ions is negligibly impacted by changes in configuration of a polymer network, increases in electrical resistivity with strain can be ignored in a hydrogel [2]. However, when a hydrogel is stretched to  $\lambda$  times its original length,

the cross-sectional area is reduced by a factor of  $\lambda$  because a hydrogel is incompressible. Thus, the electrical resistance of the stretched hydrogel increases by a factor of  $\lambda^2$  [2,6,138]. A hydrogel-based resistive strain sensor uses the relationship between electrical resistance and mechanical strain to measure the latter (Fig. 12c) [119,128,138–146]. Because of the high stretchability of hydrogels, such a strain sensor is capable of measuring large strains (Fig. 12d). Furthermore, by converting mechanical stress into changes in the contact area between two electrodes, resistive stress sensors can be created [147]. When external stress is applied to a sensor with a unique structure, the resistance between the two electrodes decreases as the contact area at the interface increases. Thus, a stress sensor can measure applied stress based on the decrease in electrical resistance.

Because of their high transparency and stretchability, hydrogels can be used as stretchable optical waveguides. If a hydrogel contains an absorber, optical loss increases exponentially as the hydrogel optical waveguide is stretched (Fig. 12e) [148]. Thus, strain can be sensed by measuring the magnitude of absorption in a dye-loaded hydrogel (Fig. 12f). In addition, if a hydrogel has multiple regions doped with different dyes, local strain can be sensed by measuring the magnitude of absorption at the peak wavelength of each dye.

If a periodic structure forms in a hydrogel, it will diffract light in a manner that satisfies Bragg's law. When a hydrogel is stretched, the spacing between the elements of the periodic structure decreases and the hydrogel diffracts light of shorter wavelengths (Fig. 12g). Thus, if the spacing between the elements is in the nanometer range, the hydrogel will diffract visible light and mechanical strain can be visualized in colorimetric form (Fig. 12h) [149–151].

### 3.4. Acoustic sensors

Hydrogel-based acoustic sensors are highly sensitive supercapacitive stress sensors that can measure sound pressure in an electrical manner (Fig. 13a) [152]. Their ability to distinguish sound in the range of the human voice is important as it allows soft robots to interact smoothly with humans. When a dendritic network of metal nanoparticles is introduced into a hydrogel and deformed by

mechanical stress, the capacitance of the EDL formed on the surface of the metal nanoparticles changes (Fig. 13b). Because the network of metal nanoparticles can be deformed by minute mechanical stimuli, a sensor can convert sound pressure to changes in EDL capacitance. Moreover, it can be used in underwater environments since the acoustic impedance of a hydrogel is almost perfectly matched with that of water.

### 3.5. Touch sensors

Hydrogel-based touch sensors can detect physical contact with a target in an electrical manner. They allow humans to easily and intuitively interact with robots. Hydrogel-based touch sensors can be organized into two categories based on their underlying mechanisms.

In a surface-capacitive touch sensor, electrodes are connected to all corners of a hydrogel and the same voltage is applied to each electrode [153]. When a grounded conductor such as a human finger touches the surface of the hydrogel, a potential difference is generated between the touch point and each electrode (Fig. 14a). The magnitude of current flowing through the electrode is inversely proportional to the distance between the touch point and the electrode. Thus, by comparing the magnitude of the current at each electrode, the position of the touch point can be estimated. Because of the high stretchability of the hydrogel, a surface-capacitive touch sensor can maintain its functionality even when 1,000% areal strain is applied. The high transparency of the hydrogel allows the sensor to transmit optical information through the sensor. Moreover, the sensor can be comfortably attached to human skin because hydrogel has a low elastic modulus (Fig. 14b).

A hydrogel encapsulated by a dielectric elastomer acts as a triboelectric touch sensor that operates based on the combined effect of contact electrification and electrostatic induction [41,154,155]. When the target touches the sensor, contact electrification occurs at the interface due to a difference in electron affinity, resulting in the accumulation of static charges on each surface. Because of the accumulated static charge, electrostatic induction occurs when the target attaches and detaches from the sensor, and a voltage signal is induced (Fig. 14c). Since a triboelectric touch sensor converts mechanical energy to an electrical signal, it can operate without a power source. Based on high stretchability and softness of hydrogels, an array of sensors can be used as a self-powered wearable keyboard, as shown in Fig. 14d.

### 3.6. Proximity sensors

Hydrogel-based proximity sensors can detect the presence of an object by measuring changes in an electric field. Because they can locate a target even if they are not in physical contact with that target, they expand the spatial detection range of soft robots. Hydrogel-based proximity sensors can be organized into two categories based on their underlying mechanisms. A capacitive proximity sensor is composed of a hydrogel electrode array and a dielectric elastomer (Fig. 15a) [156]. Disc-shaped row electrodes and loop-shaped column electrodes are separated by elastomers and project vertical electric fields when they are connected to a power source. As shown in Fig. 15b, a nearby object capacitively couples with the projected field, reducing the coupling between electrodes. Thus, by measuring the capacitance between the column and row electrodes, the lateral location of the target can be sensed. Because of the high stretchability of hydrogel, such a

proximity sensor can sense a target even when it is bent or stretched (Fig. 15c).

When a target comes into contact with its surroundings, it becomes charged through contact electrification (Fig. 15d). As shown in Fig. 15e, when a charged target approaches a proximity sensor, a voltage is induced in the sensor by electrostatic induction. By measuring the potential difference across an external load connected to the sensor, the relative distance between the sensor and the target can be estimated (Fig. 15f) [84]. This sensing mechanism means that the sensor is self-powered, and does not require any external power source. Moreover, because this type of sensor only requires an electrode to detect a target, it is potentially applicable to other existing robots that include a conductor. It is expected to endow such robots with proximity sensing capabilities and broaden their scope of applications without necessitating major structural modifications.

### 3.7. Summary of hydrogel-based soft sensors

This section outlined hydrogel-based soft sensors broken down by type of target stimulus. Despite explosive progress in this field, the durability of such components remains an issue. Many fascinating properties of hydrogels stem from their high volume fraction of water, but dehydration deteriorates their softness, transparency, and conductivity. Hydrogel-based soft sensors are not free from the dehydration issue because most of them operate in open-air environments. Addressing the durability concerns without sacrificing the outstanding features of hydrogels will allow for further enhancements in soft robotics.

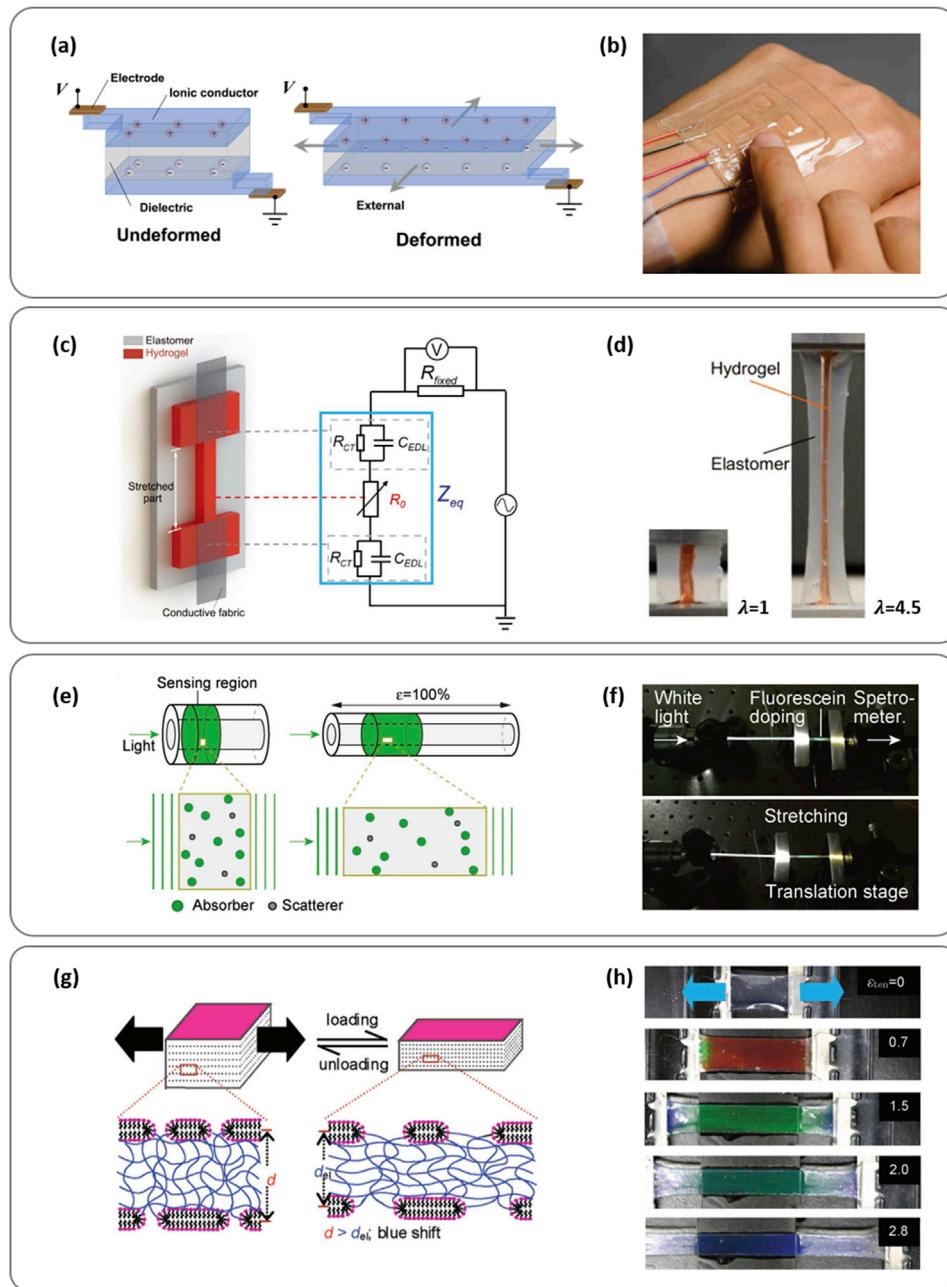
## 4. Hydrogel-based soft communicators

A communicator of a soft robot is a component used to convey information to humans in a perceptible form. For purposes of rapid expression, most communicators are electrically controlled. The ionic conductivity and transparency of hydrogels makes them a fascinating option for the transparent electrodes in a communicator. Hydrogel-based communicators can be divided into three subtypes based on their sensory organ targets. We highlight the working mechanisms that play pivotal roles in hydrogel-based communicators and summarize their many applications. At the end of this section, challenges and future work will be discussed.

### 4.1. Optical communicators

Hydrogel-based optical communicators express optical information via a change in color, light intensity, transmittance, etc [157–160]. Owing to their high transparency, hydrogels are used as transparent electrodes that allow for the flow of optical information. Hydrogel-based optical communicators fall into three categories: mechanochromic, electrochromic, and capacitive optoelectronic devices.

The high transparency of hydrogels makes them attractive for use as transparent matrices in mechanochromic communicators [149,150,161,162]. As shown in Fig. 16a, periodic structures that form in a hydrogel can diffract visible light (see the ‘Chemical sensors’ section for details of Bragg diffraction) [11]. When mechanical strain is applied to a hydrogel with periodic structures, the color of diffracted light changes as the spacing between periodic structural elements changes. Thus, a mechanochromic optical communicator can be constructed by combining an actuator with a



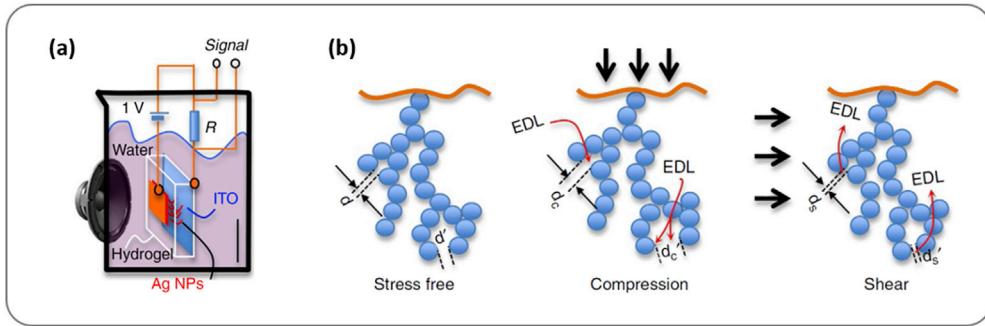
**Fig. 12. Hydrogel-based soft stress/strain sensors.** (a) Schematic diagram showing the working mechanism of capacitive stress/strain sensors. The capacitance between two hydrogel electrodes increases when the sensor is deformed by external forces [130]. (b) The transparent and stretchable sensor array was operated on the back of a hand [130]. (c) Schematic illustration of resistive strain sensors. The resistance of a hydrogel increases when the sensor is stretched by external forces [138]. (d) The sensor can measure a strain of up to 450% [138]. (e) Schematic diagram showing the working mechanism of a strain sensor based on optical loss. Since the sensing region contains an absorber and a scatterer, the optical loss increases when the hydrogel optical waveguide is stretched [148]. (f) A white light source and a spectrometer were used to measure the strain-dependent optical loss of hydrogel fibers [148]. (g) Schematic diagrams showing the working mechanism of a mechanochromic strain sensor. During tensile loading, the periodic distance of the bilayer decreases and the hydrogel diffracts shorter wavelengths of light [150]. (h) The sensor was initially transparent and rapidly changed in color from red to blue under tensile strain [150].

mechanochromic transductive gel (Fig. 16b) [11,163]. This is a fascinating option for outdoor use due to its high visibility under bright ambient light and low power consumption.

Electrochromic materials including some inorganic metal oxides [164,165], organic molecules [157,158,166–168], and conducting polymers [169], can change their color and transmittance through reversible redox reactions induced by applied voltage. Given their high transparency and ion conductivity, hydrogels have been used as the electrolytes in electrochromic optical communicators. Since hydrogels are in the solid state, they address concerns about

leakage without necessitating a complicated encapsulation process and enable formation of stretchable communicators (Fig. 16c and d). Such optical communicators have the advantage of requiring low operating voltage slightly above the electrochemical window.

In capacitive optoelectronic communicators, the hydrogel acts as a transparent and soft ionic conductor. As shown in Fig. 16e, an active layer, which expresses optical information, is sandwiched between hydrogels [9,159,160]. When high AC voltage is applied, the hydrogel projects a strong alternating electric field to the active materials change



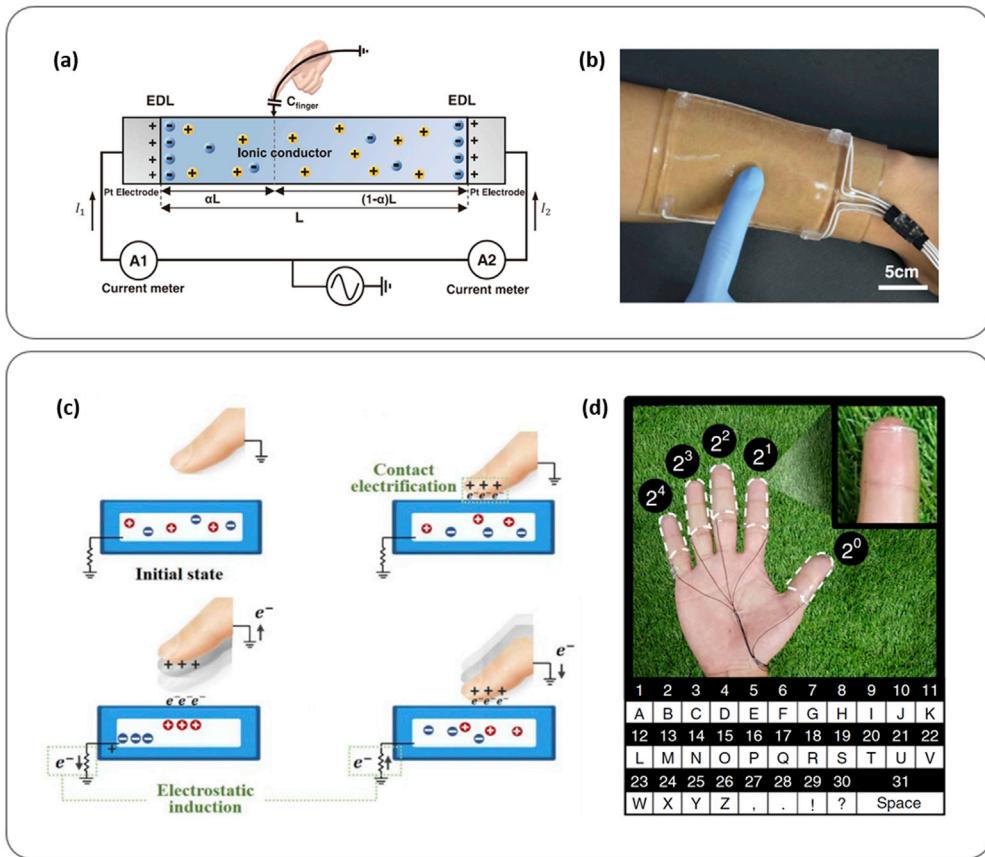
**Fig. 13. Hydrogel-based soft acoustic sensors.** (a) Schematic illustration of a capacitive acoustic sensor with a hydrogel containing a silver nanoparticle (Ag NP) network [152]. (b) Pressure induced by a sound wave deforms the Ag NP network and changes the capacitance of the electrical double layer (EDL) that formed along the surface of the Ag NP network [152].

their optical properties. Since the capacitance of the EDL formed at the interface of the electronic conductor and the hydrogel is an order of magnitude higher than that of the active layer, the voltage drop at the EDL is negligible. Thus, a hydrogel is capable of efficiently delivering electrical stimuli to the active layer. Although the communicators require high voltage to operate, they have the advantage of low power consumption because current flow through the active layer is negligible. Furthermore, the high stretchability of the hydrogel allows optoelectronic communicators to maintain functionality even under large deformation (Fig. 16f). Based on these useful characteristics, impressive efforts are under

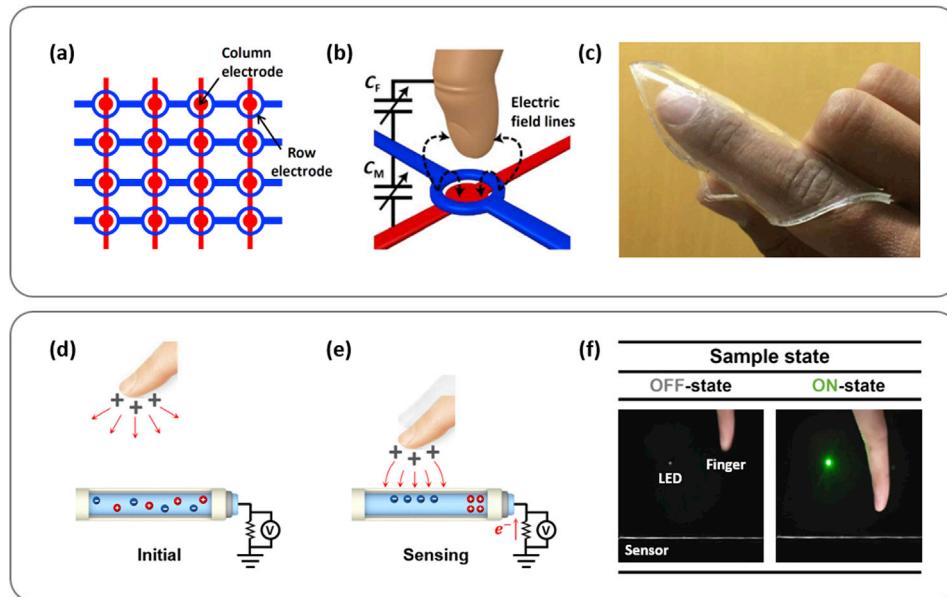
way to broaden the scope of their applications by, for example, using liquid-crystals [160] and phosphors as active materials [159].

#### 4.2. Acoustic communicators

Hydrogel-based acoustic communicators express acoustic information based on mechanical vibration generated by electrically responsive actuators. Expressing acoustic information is an effective means of simultaneously communicating with numerous targets. Based on Maxwell stress, a DEA converts an electrical signal into a corresponding sound (Fig. 17a) (see the “Electrically



**Fig. 14. Hydrogel-based soft touch sensors.** (a) Schematic illustration of a surface-capacitive touch sensor. When a grounded conductor such as a human finger touches the surface of the hydrogel, a potential difference is generated between the touch point and each electrode. The magnitude of current flowing through the electrode is inversely proportional to the distance between the touch point and the electrode [153]. (b) The transparent and stretchable touch sensor was operated on an arm [153]. (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 [155]. (d) The transparent and stretchable sensor array was attached to the fingers to express the alphabet [155].



**Fig. 15. Hydrogel-based soft proximity sensors.** (a) Schematic illustration of a capacitive proximity sensor. The disc-shaped row electrodes and loop-shaped column electrodes project a vertical electric field [156]. (b) A nearby object capacitively couples itself with the projected field and reduces coupling between electrodes [156]. (c) The transparent and stretchable sensor array was wrapped around a finger [156]. (d) Schematic diagram showing the working mechanism of a proximity sensor based on electrostatic induction. A charged target creates an electric field [84]. (e) Approach of a charged target induces a voltage in the sensor by electrostatic induction. By measuring the voltage across an external load, the sensor can measure the relative distance to the target [84]. (f) A light emitting diode is turned on only when the target approaches the proximity sensor [84].

“responsive actuator” section for details of a DEA) [6]. The outstanding capability of a DEA to generate large-strain and high-speed deformation has led to it being considered a good candidate for sound expression throughout the entire audible frequency range, from 20 Hz to 20 kHz (Fig. 17b). Furthermore, if the hydrogel-sandwiched elastomer layer is transparent, the entire communicator can also be transparent, thus broadening the range of potential applications (Fig. 17c). Impressive efforts have been made to broaden the scope of potential applications by integrating optical communicators. Such communicators express not only sound but also color via a single input control [11].

#### 4.3. Tactile communicators

Hydrogel tactile communicators are based on the transmission of mechanical stimuli to human skin. They expand the options for robot-to-human communication beyond color and sound information. To date, the functional mechanisms of hydrogel-based tactile communicators fall into two categories.

The first category is based on optically responsive hydrogels (see the “Optically responsive actuator” section for details of optically responsive hydrogels) [170,171]. Thermally responsive hydrogels arranged in an array can be controlled simultaneously and individually with light irradiation through photothermal conversion (Fig. 18a). Light-irradiated elements shrink and become stiff, while non-irradiated elements stay swollen and soft. As shown in Fig. 18b, this leads to a topological transformation on the surface of the communicator. Consequently, the system feels different to the touch. Furthermore, since shrinkage causes the hydrogel to become opaque, it is expected that such communicators will be useful in applications that express not only tactile but also optical information.

The other subset of this type of communicator is based on the DEA (Fig. 18c) (see the “Electrically responsive actuator” section for details of the DEA) [172–174]. An elastomer is partially sandwiched between hydrogels, which act as conductors. The region covered with hydrogels experiences thickness contraction and areal

expansion when a voltage is applied. The surrounding region, which is not covered with hydrogels, bulges due to squeezing by shear stress from the region covered with hydrogels (Fig. 18d). Consequently, the skin can perceive surface textural changes of the communicator.

#### 4.4. Summary of hydrogel-based soft communicators

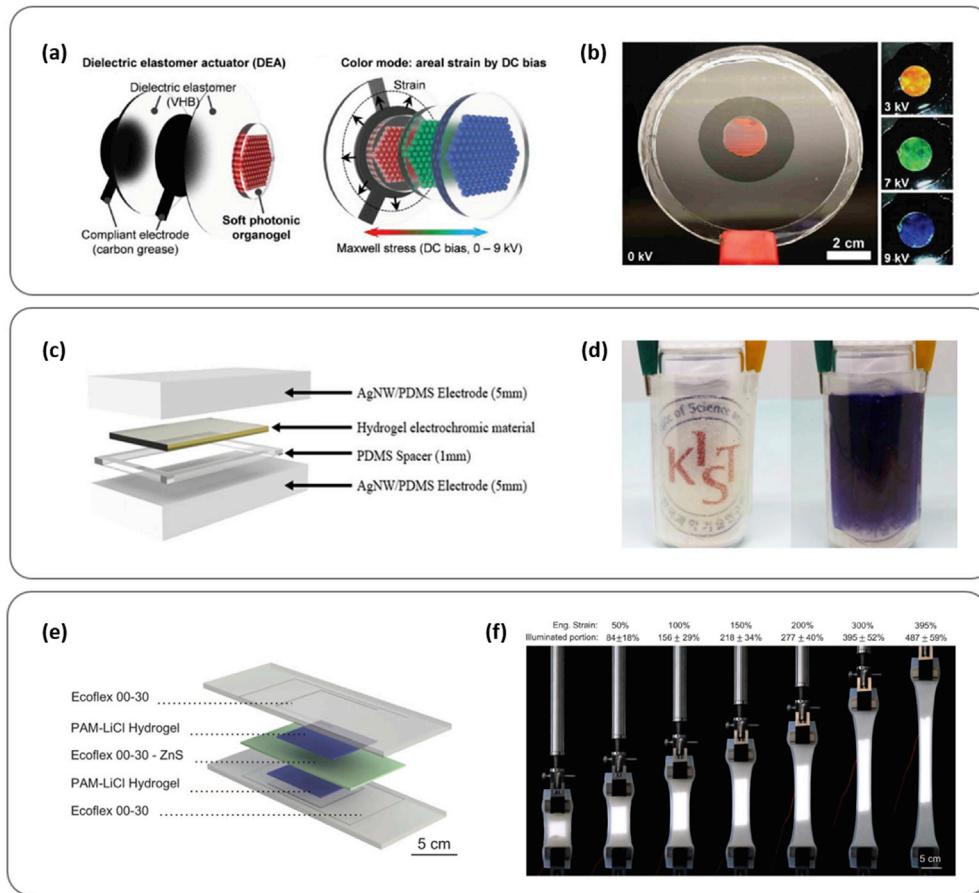
This section outlined hydrogel-based soft communicators broken down by sensory organ target. Although explosive progress has been made in this field, it remains difficult to express complex information for practical use. An approach to the development of array-type communicators that downsizes each unit is expected to help solve this challenge. Exquisite fabrication methods such as 3D printing and photo lithography will be required to realize next-generation soft communicators.

### 5. Hydrogel based soft power sources

The power source of a soft robot is a component that supplies energy to other components. Gel electrolytes allow a power source to have both high compliancy and excellent performance, because they can maintain high ionic conductivity even in the solid state [6]. Our discussion of hydrogel-based power sources is organized into two sections, energy storage devices and energy generators. We highlight the working mechanisms of hydrogel-based power sources and summarize their various applications. At the end of this section, challenges and future work will be discussed.

#### 5.1. Energy storage devices

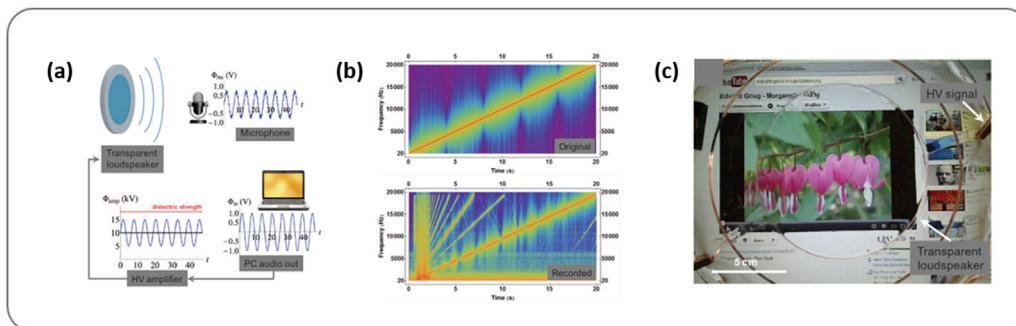
Hydrogel-based energy storage devices can store energy in electrical and chemical form. Since energy storage devices can supply energy to a desired location at a desired time, it allows untethered operation of soft robots. Hydrogel-based energy storage devices fall into three categories based on their storage mechanism.



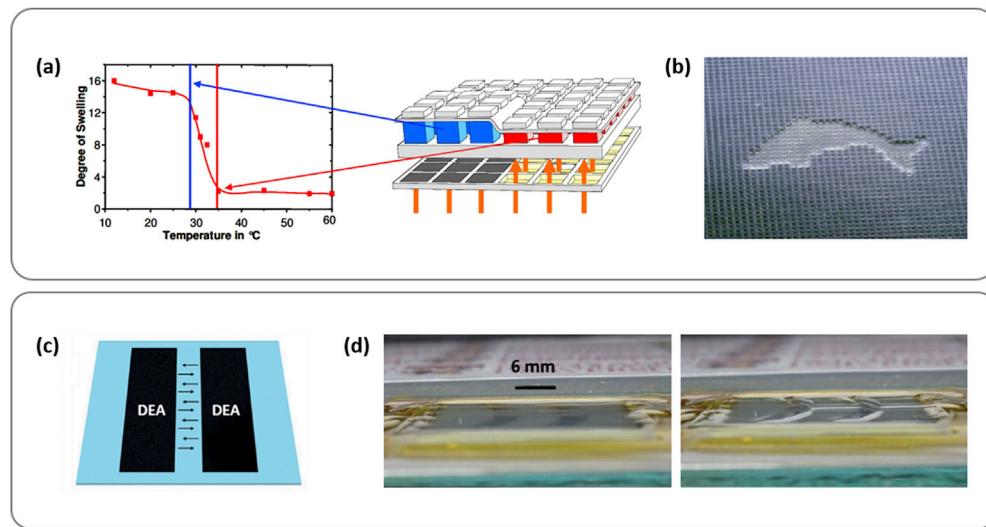
**Fig. 16. Hydrogel-based soft optical communicators.** (a) Schematic diagram showing the working mechanism of a mechanochromic optical communicator. As the interparticle distance of polystyrene beads changes due to areal strain derived from DEA, the color of the light diffracted by the organogel changes [11]. (b) As the DC bias increases from 0 to 9 kV, the color of the gel changes from red to blue [11]. (c) Schematic illustration of electrochromic optical communicators. Absorbance, reflectance, and transmittance of the hydrogel layer can be controlled through an applied voltage [168]. (d) The device is transparent when voltage is off and purple when voltage is on [168]. (e) Schematic illustration of electroluminescent optical communicators. As a high AC voltage is applied across the transparent hydrogel electrodes, excited zinc sulfide phosphors emit light [9]. (f) The fabricated device can stretch up to a strain of 500% [9].

The most commonly used energy storage device that can store energy in chemical form is the battery. A battery converts stored chemical energy into electrical energy using electrochemical reactions and delivers it to external circuits. Although batteries have relatively low power density compared to capacitors, they have sufficiently high energy density and are suitable for long-term energy storage. Gels, including hydrogels [175] and ionogels [176], have been used as solid-state electrolytes in flexible batteries.

Since they can maintain high ionic conductivity even in the solid state, gel electrolytes ensure that batteries are compliant and safe, without necessitating a complicated encapsulation process. Moreover, gel electrolytes can also act as separators because they can prevent electrical short circuiting by maintaining a divide between a cathode and an anode (Fig. 19a). Batteries fabricated using gel electrolytes with zinc and manganese dioxide paste can stably



**Fig. 17. Hydrogel-based soft acoustical communicators.** (a) A transparent acoustical communicator based on DEA transformed an electrical signal from the laptop into sound, and the microphone was used to record it [6]. (b) The communicator can produce sound with frequencies from 20 to 20,000 Hz, corresponding to the entire audible frequency range [6]. (c) Optical information from a laptop is transmitted through the transparent communicator [6].



**Fig. 18. Hydrogel-based soft tactile communicators.** (a) Schematic diagram showing the working mechanism of tactile communicators based on thermally responsive hydrogels. When a digital micro-mirror project light, the thermally responsive hydrogel contracts as the substrate converts the light into heat [170]. (b) The fabricated communicator has 297 actuators per  $\text{cm}^2$  [170]. (c) Schematic illustration of tactile communicators based on DEA. As a series of DEAs actuate, the elastomer in the inactive region is deformed by shear stress [172]. (d) The fabricated communicator can provide localized tactile feedback [172].

supply energy to external circuits even under deformation (Fig. 19b).

Electrical double layer supercapacitors (EDLCs) are energy storage devices with high power density that store energy in an electric field. They are suitable for applications requiring rapid charge/discharge cycles rather than long-term energy storage. Since EDLCs store energy using electrical double layer forms at the interface between an electrode and an electrolyte, the effective surface area of the electrode is directly related to performance. Hydrogels based on soft conductive materials such as graphene [177,178] and conducting polymers [179–183] can be used as compliant electrodes with high specific surface area; their highly porous networks allow free diffusion of electrolytes (Fig. 19c and d). Furthermore, gels are used as solid-state electrolytes in EDLCs to prevent leakage [184–191]. Based on the high specific surface area of gel electrodes and compliancy of gel electrolytes, fabricated EDLCs show high performance even under deformation (Fig. 19e).

Recently a new type of hydrogel-based energy storage device that stores energy using an electrolyte salinity gradient was developed [192]. By sandwiching polyelectrolyte hydrogels between high- and low-salinity hydrogels, and selective diffusion of ions converts the chemical potential difference between the electrolytes into an electrical potential difference (Fig. 19f). An energy storage device made of three hydrogel layers can generate a voltage of 80 mV, as shown in Fig. 19g. Since the device is only composed of hydrogels, it is flexible, transparent, and biocompatible. Furthermore, by stacking the units in series, the fabricated cell can generate a voltage of up to 110 V in an open circuit.

## 5.2. Energy generators

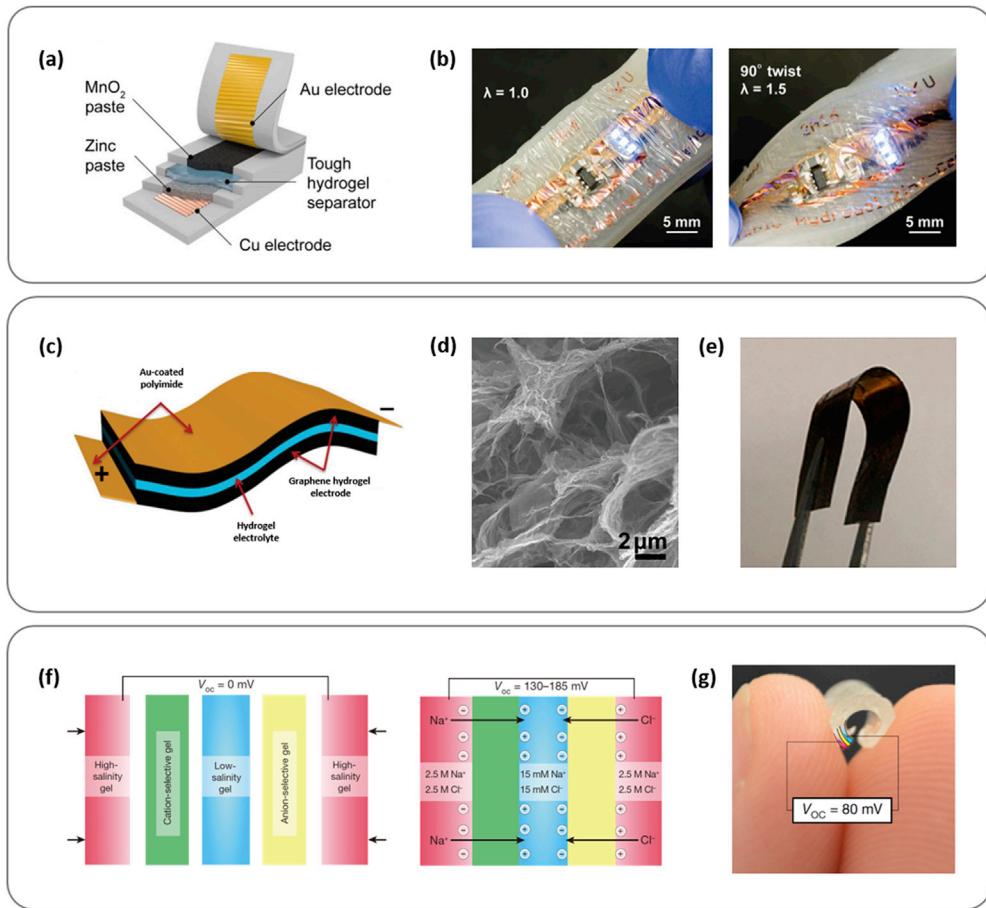
Hydrogel-based energy generators are based on harvesting wasted mechanical energy and turning it into electrical energy. They play an important role in meeting the growing need for wireless, lightweight power supplies for soft robots [155,175,193]. Hydrogels are attractive options for electrodes in the generators due to their high ionic conductivity and stretchability. Hydrogel-based soft power generators fall into two categories, dielectric elastomer generators and triboelectric nanogenerators.

As shown in Fig. 20a, dielectric elastomer generators (DEGs) are composed of a dielectric elastomer film sandwiched between two compliant electrodes [175,194–196]. The generator converts mechanical energy into electrical energy using the capacitance variation induced by cyclic deformation. When the DEG is stretched by mechanical stress, an external power source connected to the electrodes of the DEG supplies a charge. After charging, the power source is removed. Then, the mechanical stress is released and the capacitance of the DEG decreases as it returns to its undeformed state via elastic force. Since the amount of charge accumulated at the electrodes is constant, the decrease in capacitance induces an increase in voltage between the electrodes. Thus, electromechanical coupling allows high-energy-density energy transduction. Based on their softness and stretchability, hydrogels are attractive options for compliant electrodes for DEGs (Fig. 20b) [175].

Triboelectric touch sensors can also be utilized as electromechanical transducers, so-called triboelectric nanogenerators (TENGs) (see the “Touch sensor” section for details of triboelectric touch sensors) [41,155,197]. During cyclic attaching and detaching of the generator, charges are electrostatically induced and released repeatedly in the hydrogel electrode [155]. Consequently, an alternating electric current is generated across the external load. TENGs have earned a great deal of attention as wearable power sources because of their many advantages, which include a high capacity for voltage generation, simplicity of fabrication and structure, low weight, flexibility, etc. [154,198–202]. Recently, the introduction of hydrogels to the generators has provided a promising means through which to equip them with both high transparency and stretchability (Fig. 20c) [155]. Such a generator allows the gentle touch of a finger to light up a series of light-emitting diodes, as shown in Fig. 20d.

## 5.3. Summary of hydrogel-based power sources

This section outlined hydrogel-based power sources. Although impressive progress has been made, the field is still struggling due to the low compatibility between these power sources and other soft robotic components. The hydrogel-based power sources that have been developed to date can provide only electrical energy, but other existing soft robotic components require various forms of



**Fig. 19. Hydrogel-based soft energy storage devices.** (a) Schematic illustration of a stretchable battery that uses a hydrogel as a separator. The hydrogel separator prevents an electrical short circuit by keeping the cathode and anode apart [175]. (b) The stretchable battery can supply energy to an external circuit even under deformation [175]. (c) Schematic illustration of flexible solid-state electrical double layer supercapacitors based on graphene hydrogel electrodes. The three-dimensional graphene hydrogel enables an unprecedented areal specific capacitance of 372 mF/cm<sup>2</sup> by preventing parallel restacking of the graphene sheets [178]. (d) SEM image of a graphene hydrogel electrode [177]. (e) The fabricated supercapacitor showed a negligible change in its performance even under a bending angle of 150° [177]. (f) Schematic diagrams showing the working mechanism of a soft energy storage device that stores energy using a salinity gradient of electrolytes. The energy storage device is composed only of hydrogels, including high/low salinity gels and cation/anion-selective hydrogels. Selective diffusion of ions converts the chemical potential difference of the electrolytes into electrical potential differences [192]. (g) A transparent and flexible energy storage device, made of three layers of hydrogel, can generate a voltage of 80 mV [192].

energy, such as thermal, chemical, radiant, electromagnetic, and so on. This mismatch raises the need for soft power sources capable of supplying various types of energy. An approach that uses hydrogel as a liquid fuel storage material may help address this concern. A hydrogel can maintain its solid state even with a high volume fraction of solvent. Thus, it will allow for safe and compliant storage of liquid fuel for soft power sources which can supply thermal, chemical, and hydraulic stimuli.

## 6. Hydrogel-based soft computation circuits

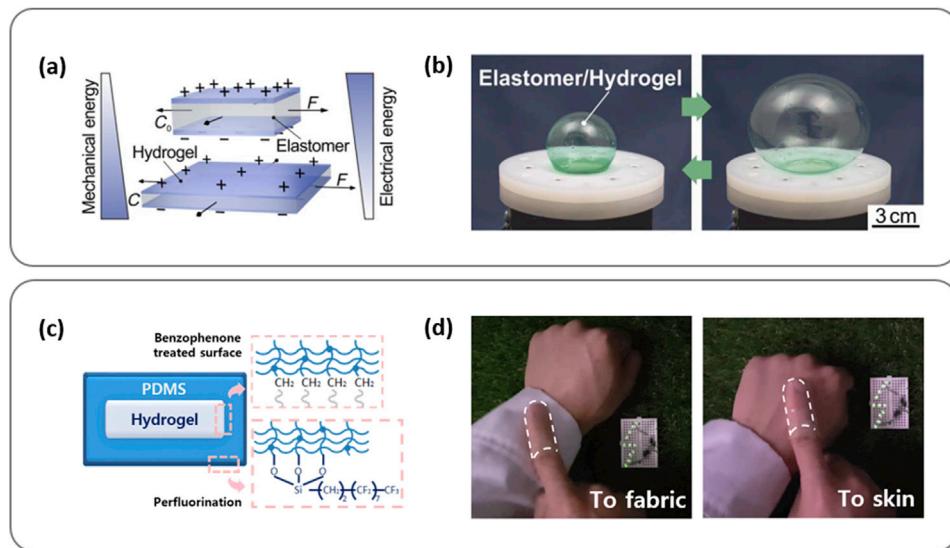
The computational circuit of a soft robot is a component that processes information and controls other components. Since hydrogels can serve as stretchable ionic conductors, electronic conductors, and even ionic semi-conductors, they may be a fascinating material for use in intrinsically stretchable computational circuits. Hydrogel-based computational circuits are organized into four subtypes based on their functions. We highlight the working mechanisms that play pivotal roles in hydrogel-based

computational circuits and summarize the applications of such devices. At the end of this section, challenges and future work will be discussed.

### 6.1. Conductors

A conductive hydrogel serves as a medium to transmit charge carriers. The innately high stretchability of hydrogels may meet the growing need for stretchable electronics. Hydrogel-based conductors fall into two categories based on the charge carriers used: ions and electrons.

A hydrogel is an ionic conductor owing to its high volume fraction of water. It allows dissolution of a large amount of salt, and dissolved ions act as charge carriers under an electric field. Since ions are massive carriers, hydrogels exhibit a low conductivity of ~10 S/m at room temperature [2,6,119,145,146,175,202–204]. However, this is still an attractive option because hydrogels promise lower sheet resistance compared to existing soft conductors under high applied strain (see the “Introduction” section for details). Meanwhile, there is a significant challenge: it would be



**Fig. 20. Hydrogel-based soft energy generators.** (a) Schematic diagram showing the working mechanism of dielectric elastomer generators. Using electromechanical coupling, a dielectric elastomer generator can convert mechanical energy to electrical energy [175]. (b) The balloon-shaped generator can transform 500 mJ of mechanical energy into 54 mJ of electrical energy per cycle [175]. (c) Schematic illustration of a triboelectric nanogenerator with a hydrogel electrode. Chemical anchoring between the hydrogel and polydimethylsiloxane (PDMS) allows for mechanical stability under deformations, and perfluorination on the surface of PDMS enhances output performance [155]. (d) The transparent and stretchable generator can turn on a series of light emitting diodes (LEDs) with a single touch with various materials [155].

difficult to use a hydrogel in an application requiring DC current without an electrochemical reaction occurring at the interface between the hydrogel and the electronic device, causing diverse side effects. Despite the challenges, ionic conductive hydrogels are promising in applications requiring AC current because they are highly transparent and stretchable [6]. Based on these fascinating features, hydrogels have been spotlighted in the field of stretchable electronics (Fig. 21a and b) [204].

Recently, valuable attempts have been made to endowing hydrogels with electronic conductivity by introducing a conducting polymer, such as polypyrrole, poly(3,4-ethylenedioxythiophene);poly(styrene sulfonate), polyaniline, etc. (Fig. 21c and d) [144,205,206]. This is considered a promising approach to address the conductivity concern. However, the approach remains challenging; for example, it is still not suitable for DC current conduction and it sacrifices the transparency of the hydrogel. The removal of ions from the gel is one direction to explore in the effort to conduct DC current without unwanted electrochemical reactions [205]. Meanwhile, securing high transparency remains an open challenge in the field.

## 6.2. Semiconductors

Hydrogel-based semiconductors make use of Donnan exclusion at the polyelectrolyte junction. Since hydrogel-based semi-conductors can directly process ionic signals from biological systems, they may be very useful at human-robot interfaces. Furthermore, they are potentially applicable to wearable electronics because they can maintain their functionality even under mechanical strain. Hydrogel-based semi-conductors fall into two underlying categories, diodes for rectifying ionic signals and transistors for regulating and amplifying ionic signals.

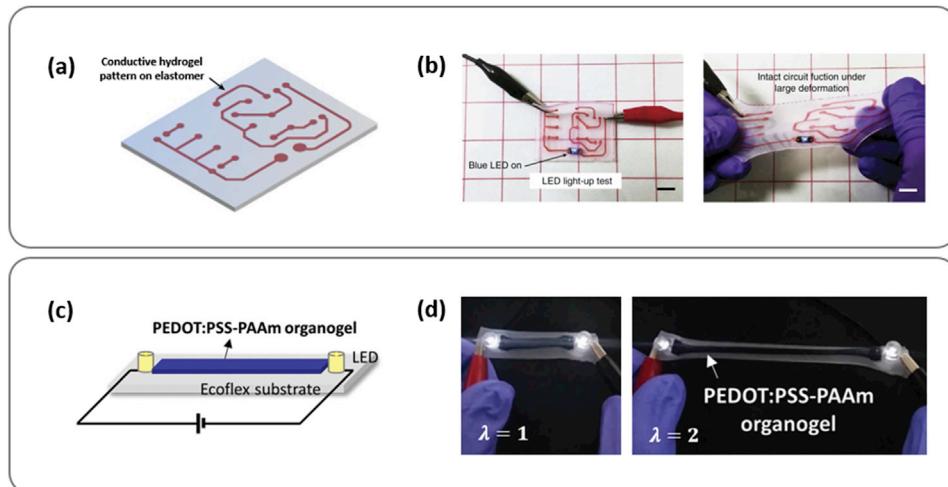
The polymer network of a polyelectrolyte hydrogel has charged functional groups, and mobile counterions with the opposite charge [3]. Depending on the charge of the mobile counterions,

polyelectrolyte hydrogels are classified into p-type (positively charged counterions) and n-type (negatively charged counterions). When two different types of polyelectrolyte form a junction, it acts in ways that are similar to a silicon-based p-n junction [207,208]. Under forward bias, electrolyte ions can migrate through junctions because the polyelectrolyte hydrogel is in an accumulated state (Fig. 22a). However, under reverse bias, counterions in the polyelectrolyte hydrogel migrate toward the electrolyte, and an ion depletion region is formed at the junction. As a result, ion of electrolyte cannot pass through the polyelectrolyte hydrogel due to the strength of Donnan exclusion (Fig. 22b). Thus, the polyelectrolyte junction exhibits different resistance behaviors depending on the direction of bias.

Since a polyelectrolyte hydrogel junction only allows ionic current to flow in one direction, it can be used as a diode (Fig. 23a) [207–212]. Non-polarizable Ag/AgCl electrodes have been employed to prevent a drop in potential caused by EDL formation at the interface between the electrode and electrolyte. By using a neutral hydrogel as a compliant solid-state electrolyte, a polyelectrolyte hydrogel diode can maintain its functionality even under 300% strain and can be used as a wearable ionic circuit (Fig. 23b).

Furthermore, a hydrogel-based ionic diode with an open junction can directly amplify minute ionic signals (Fig. 23c) [213]. In the initial state, reverse bias is applied to the diode, and the mobile ions in the electrolyte are piled up at the electrolyte/gel interface by Donnan exclusion. When an ionic signal is injected into the open junction of the diode, a depletion region is temporarily neutralized, and ions piled up at the interface migrate through the junction. Ionic signals are amplified up to 18.9 times by the induced breakdown current, as shown in Fig. 23d.

A polyelectrolyte hydrogel can also be used to regulate ionic signals. When a p-type and an n-type gel are arranged facing each other at the wall of an ion channel, they act as a field effect



**Fig. 21. Hydrogel-based soft conductors.** (a) Schematic illustration of an ionically conductive hydrogel circuit patterned on an elastomer. Mobile ions dissolved in the solvent allow the hydrogel to be used as an ionic conductor [204]. (b) The stretchable hydrogel circuit can maintain its electrical conductivity even under large deformation and succeed in lighting up the LED [204]. (c) Schematic illustration of a gel electronic conductor based on conducting polymers. By combining conducting polymers with hydrated matrix polymers, an electrical path is formed inside the gel that allows it to be used as an electronic conductor [205]. (d) Through removal of ions by a dialysis process, the stretchable conducting polymer-based gel can light up the LEDs without an electrochemical reaction even when it is connected to a DC power source [205].

transistor (Fig. 23e) [214]. In the initial state, no gate voltage is applied, and ionic signals can freely be transferred from source to drain through the ion channel. However, with application of a gate voltage, mobile cations and anions in the ion channel are selectively extracted by the p-type and n-type hydrogels, respectively. This extraction results in formation of an ion depletion region in the ion channel and decreases ionic conductivity. Thus, by adjusting the gate voltage, ion signals can be regulated quite easily as shown in Fig. 23f.

### 6.3. Circuit boards

The biocompatibility and softness of hydrogels makes them promising materials for a substrate of wearable electronics [2]. The elastic modulus of hydrogels closely resembles those of biological tissues, ensuring a comfortable human-machine interface. Recent research regarding tough bonding hydrogels to diverse materials including metals [215,216] and polymers [175,204,217,218] has shown that hydrogels can stably contain diverse elements of a circuit, even under large deformation (Fig. 24a). When a strain is applied to a hydrogel with rigid circuit elements, the strain is concentrated in the gel due to the large difference in elastic modulus between the hydrogel and the rigid circuit elements (Fig. 24b). Consequently, isolation of rigid elements from strain allows the entire circuit to be stretchable.

### 6.4. Bio-computational circuits

Unlike commercial computational methods, which use semiconductors to process electric signals, genetically programmed cells cultured in a hydrogel matrix can process chemical signals (Fig. 25a) [10,110]. In a bio-computational circuit, each cell can only perform simple operations. However, by integrating them into hydrogels with three-dimensional structures, complex logic operations can be achieved (Fig. 25b). In the hydrogel matrix, each cell can communicate via the diffusion of chemicals, and a well-

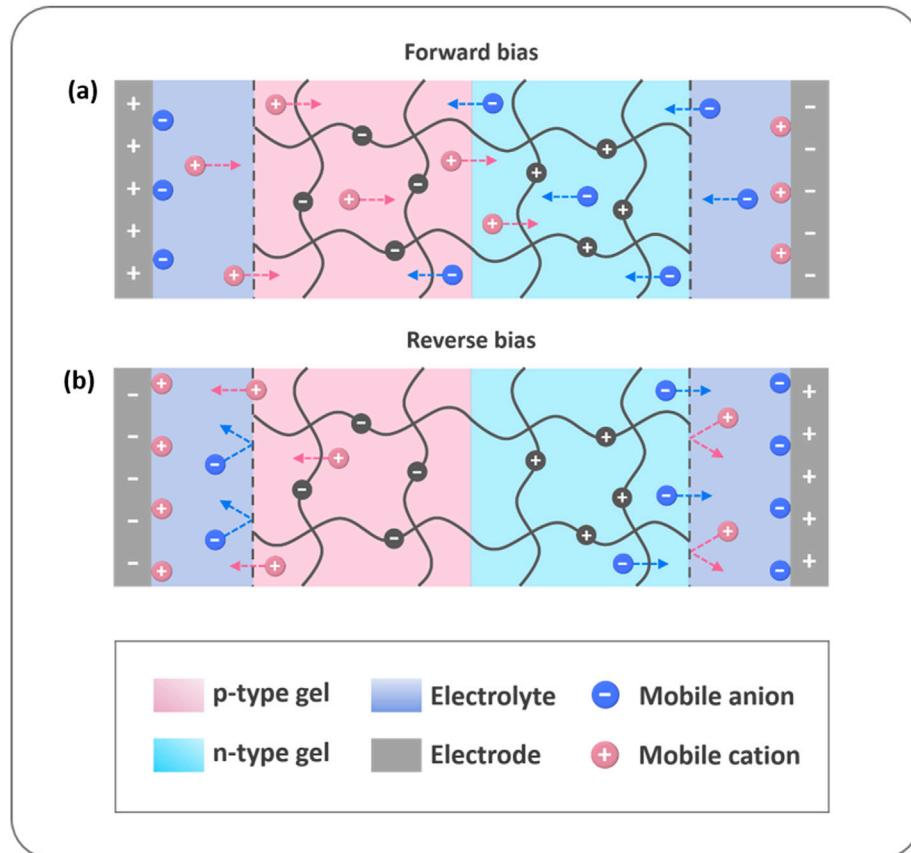
organized cellular network can operate as a set of logic gates, such as Buffer, NOT, AND, OR, and NAND. Integrating such a network with other chemically responsive soft robotic components is a promising direction for computational circuits in the field of soft robotics.

### 6.5. Summary of hydrogel-based soft computational circuits

This section outlined hydrogel-based soft computational circuits based on their functions. Although attempts have been made to create soft circuits with hydrogels, the field is still at the proof-of-concept level. There are a number of issues that need to be addressed to make hydrogel-based circuits practical, such as their sub-optimal performance and reliability. However, if more studies are done, hydrogel-based computational circuits may play an active role in applications that require stretchability and biocompatibility.

## 7. Conclusions and outlook

Here, we have highlighted the unique features of hydrogel-based soft robots and described their potential applications as well as their fundamental working mechanisms. The past decade has brought an explosion of hydrogel-based applications to the field of soft robotics. The unique properties of hydrogels have allowed them to play outstanding roles and endow soft robots with innovative functionalities. Even though recent efforts have proved the feasibility of hydrogels as a material for soft robots, the field is still in its early stages and such robots are far from use in daily life. Approaches beyond the proof-of-concept level pose several issues that need to be explored. Here, we suggest future directions for hydrogel-based soft robotics in three realms: endowing stimuli-selectivity, improving durability, and systemization.



**Fig. 22. Polyelectrolyte hydrogel junction.** A schematic illustration of a junction formed between p-type and n-type gels. (a) Under forward bias, ions of electrolytes can migrate through the junction because the polyelectrolyte hydrogels are in accumulated state. (b) Under reverse bias, counterions of polyelectrolyte hydrogels migrate toward the electrolytes, and a depletion layer is formed. As a result, ions of electrolytes cannot pass through the hydrogel junction due to Donnan exclusion.

### 7.1. Endowing stimuli-selectivity

With the recent increased interest in versatile soft robots, a tremendous amount of effort has been focused on hydrogels because most of them can respond to diverse stimuli, such as solvents, temperature, humidity, and so on [14,18,32,34]. Multi stimuli-responsiveness is an attractive feature that extends the range of applications of hydrogels in the field of soft robotics [13,30,219–222]. However, it raises a new issue originating from stimulus interference. To secure reliability, a hydrogel must be capable of distinguishing between a desirable stimulus and undesirable stimuli in accordance with a given situation. This remains an unaddressed challenge and open area of exploration. The introduction of stimuli-selectivity will spur significant developments in hydrogel-based soft robotics by securing reliable controllability.

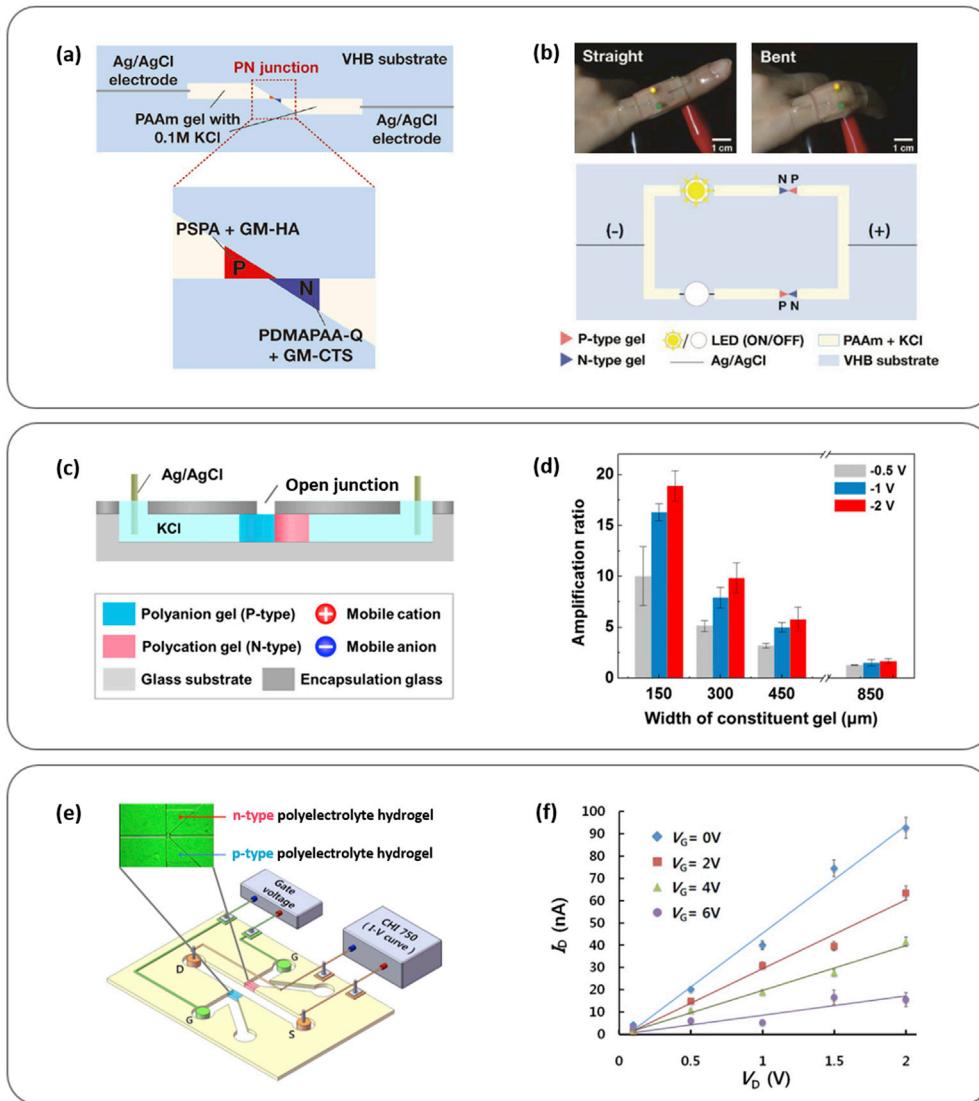
### 7.2. Improving durability

Although there have been explosive efforts that have enhanced the mechanical [5,223] and electrical [6,205] properties of hydrogels, the challenge of durability has rarely been met. First, dehydration of the hydrogel is a critical issue in open-air environments because evaporation deteriorates the properties of a hydrogel, including softness, transparency, and ionic conductivity. Recently, several attempts have been made to address this challenge. For instance, introducing humectants such as hygroscopic salts is a simple strategy

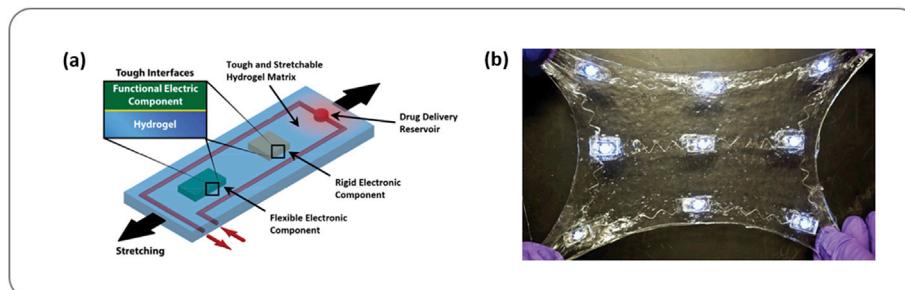
to reduce the vapor pressure of water [153]. However, it is not ideal because hydrogels containing humectants can undesirably swell in humid conditions [224]. Using an elastomeric coating as a diffusion barrier against water is another noteworthy strategy. However, this approach has trade-offs because it deteriorates some fascinating features of the material, such as biocompatibility, stretchability, transparency, etc. [155,204,224]. Second, electrochemical stability is a crucial issue because electrochemical reactions inevitably occur at the interface between a hydrogel and an electronic conductor when the applied voltage exceeds the electrochemical window. This causes undesirable side reactions and reduces the concentration of the ionic charge carrier [6]. While there have been valuable attempts to use hydrogels under high-voltage conditions, their utility remains limited to capacitive operating systems or AC voltage applications. Addressing the elusive challenges mentioned above without sacrificing the fascinating properties of hydrogels will help to provide further enhancements.

### 7.3. Systemization

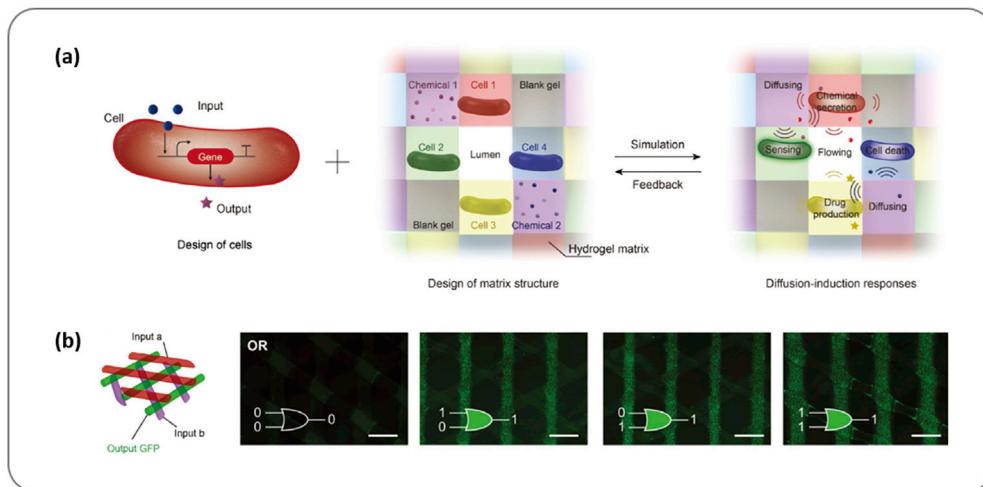
With the advent of hydrogel-based soft robotics, researchers have proven the potential of hydrogel-based robotic components, including actuators [6,18,83], sensors [130,153,155], communicators [6,11,172], power sources [155,175,192], and computational circuits [205,209,213]. While the individual components have been tremendously well-researched, the effort to combine individual



**Fig. 23. Hydrogel-based soft semi-conductors.** (a) Schematic illustration of a stretchable ionic diode. Since a polyelectrolyte hydrogel junction only allows ionic current to flow in one direction, it can be used as a diode [209]. (b) The stretchable and transparent diode can rectify ionic current even under 300% strain [209]. (c) Schematic illustration of an ionic transistor. A minute ionic signal injected into the open junction is amplified by the induced breakdown currents [213]. (d) By using an open-junction ionic diode, ionic signals can be amplified up to 18.9 times [213]. (e) Schematic illustration of an ionic field effect transistor. By applying a gate voltage to polyelectrolyte hydrogels, an ion depletion region is formed in an ion channel and ionic conductivity is decreased [214]. (f) Ionic signals can be regulated by adjusting the gate voltage [214].



**Fig. 24. Hydrogel-based soft circuit boards.** (a) Schematic illustration of a stretchable hydrogel circuit boards. Since the hydrogel has a low elastic modulus and high stretchability, it can be used as a compliant circuit board [216]. (b) A stretchable and transparent hydrogel circuit board containing an array of LEDs and metal wires can be stretched without failure [216].



**Fig. 25. Hydrogel-based soft bio-computational circuits.** (a) Schematic illustration of bio-computational methods based on living cells containing hydrogels. Since chemicals can diffuse in the hydrogel matrix, genetically programmed bacterial cells, cultured in the hydrogel matrix, can process chemical signals [10]. (b) A hydrogel with a three-dimensional structure allows bacterial cells to perform complex logic operations such as double-input and single-output Boolean logic gates [10].

components into a single system remains in its infancy. Systemization of the well-established individual components is an essential prerequisite for real-world application of hydrogel-based soft robotics. Beyond simple physical combining, complementary interactions between all components promise to fulfill the maximum potential of soft robotics – the ability to operate without further intervention. At the same time, the systemization process should not deteriorate the compliance of the soft robots; care must be taken to preserve their unique features. Fundamental insights into the underlying working principle of each individual component gleaned from a multidisciplinary approach may help to address daunting challenges and open new avenues for further innovation in the field of soft robotics.

#### Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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