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Nanocomposite Hydrogels for Strain Sensing Based on Optical and Electrical Signals: A Review

Youfeng Yue[1,2,*]

[1]Research Institute for Advanced Electronics and Photonics, National Institute of Advanced Industrial Science and Technology (AIST), Tsukuba 305-8565, Japan

[2]Precursory Research for Embryonic Science and Technology (PRESTO), Japan Science and Technology Agency (JST), Kawaguchi 332-0012, Japan

[*]Email: yue-yf@aist.go.jp

Abstract

Unlike metals, ceramics, or plastics, hydrogels are semi-solid, hydrophilic polymer networks with high water content. Embedding nanostructures or nanomaterials into hydrogels can impact special properties to the composites, such as anisotropy, optical or electrical properties. With the development of nanomaterials and advanced synthetic methods, nanocomposite hydrogels have attracted growing research interest in recent years due to their desirable mechanical properties, optical/electrical functions, reversibility, stimulus sensitivity, and biocompatibility. They have opened up a broad range of applications as stretchable strain sensors in mapping of strain distributions, motion detection, health-monitoring, and flexible skin-like devices. This minireview summarizes the recent developments of nanocomposite hydrogels as strain sensors based on optical and electrical signals. The strain sensing performances and their dynamic properties are discussed. Appropriate embedding of nanostructures or nanomaterials in hydrogels and engineering of interactions between nanomaterials and polymer networks can yield significant improvements in the performance of strain sensors.

1. Introduction

Hydrogels are classically soft and wet materials that contain large amounts of water in their three-dimensional (3D) polymer networks. They exhibit many characteristic properties, including stretchability, transparency, viscoelasticity, self-healing, adhesiveness, stimuli-sensitivity and biocompatibility. [1-2] Conventional hydrogels are structurally isotropic; however, embedding periodic nanostructures (e.g., photonic crystal structures) or conductive nanomaterials (e.g., nanoparticles, carbon nanotubes, graphene, and MXene) into hydrogels, can impact the composites with interesting optical or electrical properties. For example, they can exhibit tunable structural color or electrical conductivity in response to external stimuli. Furthermore, they can be designed to transmit external stimuli, such as stress/strain, humidity, pH, and temperature to optical or electrical signals, including wavelength, current, capacitance, or resistance change. Among them, hydrogel strain sensors based on strain-optical and strain-electrical conversions have attracted significant interest owing to their potential applications in flexible displays, skin-like devices, touch screens, movement monitors, and on-demand ability to recognize strain distribution.

In the biological field, hydrogels have also been intensively investigated for applications such as cell culture,[3] tissue engineering,[4] and wound healing,[5] owing to their excellent biocompatibility, high water content (70−90 wt%), stretchability, and adjustable mechanical properties. Thus, strain sensors constructed from nanocomposite hydrogels are more likely to be compatible with biological tissues, providing a foundation for the design of smart skins, implantable devices, and wearable sensors.

Based on their working mechanisms, nanocomposite hydrogels as strain sensors can be mainly divided into strain-optical conversion and strain-electrical conversion. The strain-optical conversion is from hydrogels with photonic crystal structures, and the strain values can be directly visualized by eyes from their structural colors or by monitoring their wavelength shifts. Strain-electrical conversion is based on the change in conductivity (resistance or capacitance) upon the applied strains. They can be further divided into electron-, ion-, and hybrid electron-ion conductive hydrogels according to the conductive mechanisms arising from the transmission of electrons, ions, or both. For strain sensors, sensitivity, linearity, stretchability, and durability are important parameters that need to be considered during material design.

In this minireview, strain sensing performances of hydrogels with different working mechanisms are summarized. Strain dependent optical and electrical responses in hydrogels are comprehensively discussed combined with their chemical-physical properties (**Figure 1**). Hydrogel based on strain-optical (i.e., photonic hydrogels) are classified according to their photonic crystal structures. One is based on ordered layered structures and the other is based on crystalline colloidal arrays. Hydrogels based on strain-electrical (conductive hydrogels) are divided according to their conducting mechanisms and the conductive nanomaterials they contain. A number of classic and interesting nanocomposite hydrogels for strain-sensing have been described. Their fabrication, advantages, limitations, current scientific/technical obstacles and future research perspectives are discussed.

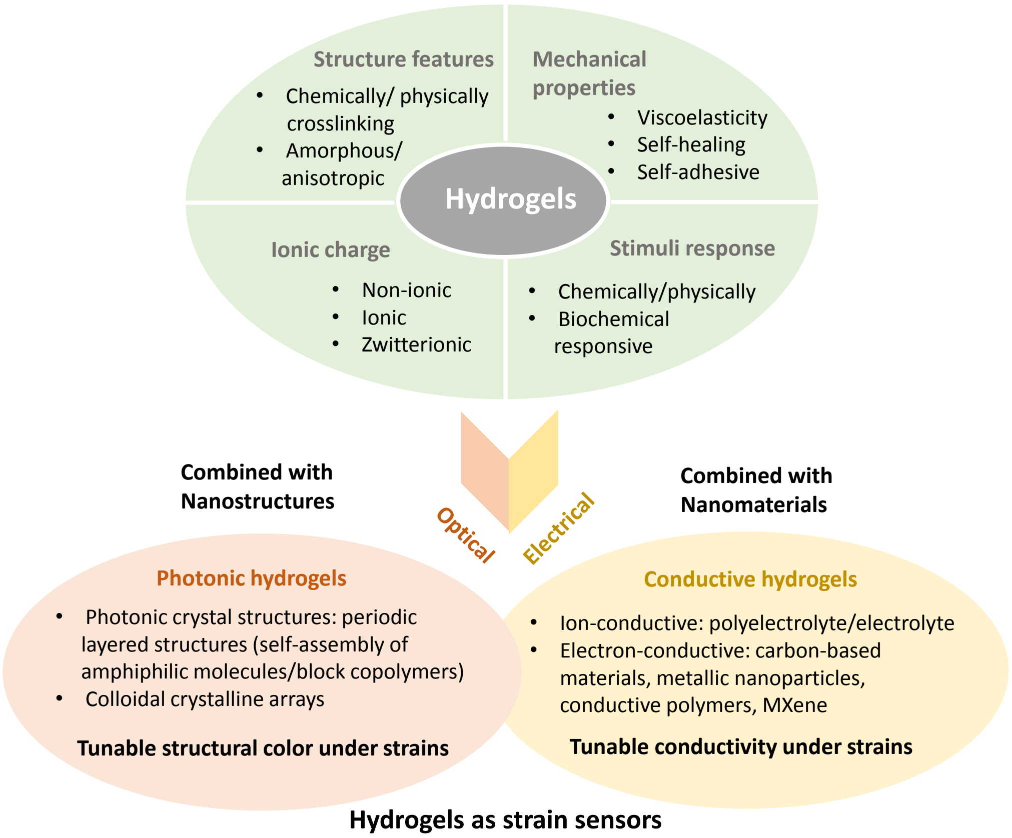

Figure 1 The characteristic of hydrogels and their combination with nanostructures/ nanomaterials to create hydrogel-based strain sensors. They are classified into two types based on the working mechanisms from strain-optical or strain-electrical change.

2. Photonic hydrogels as strain sensors based on strain-optical conversion

In nature, many organisms including insects, birds, fish, and plants, can display iridescent structural colors and achieve specific functions, such as camouflage, reproduction, and signal communication. They are called photonic crystal materials, which have ordered submicroscale structures in which light propagation is affected by periodic variations in their optical dielectric constants.[6, 7] Most photonic crystals produced from hard materials exhibit a limited response to external stimuli. However, embedding photonic crystal structures into hydrogels results in composite materials with tunable structural colors and stimuli-sensitivity.[8,9,10] For example, they exhibit structural color changes upon mechanical stretching or compression. When mechanical stimuli change the lattice distance in photonic crystal structures, the photonic bandgap shifts. Thus, they can also be called mechanochromic materials, which exhibit reversible stress/strain-dependent color changes. The optical effects of these materials make them promising candidates for real-time strain detection and motion monitoring applications. For strain-optical based hydrogel sensors, a broad wavelength shift, fast response, high sensitivity, linearity, and long-term stability are required.

2.1 Layered-structured hydrogels for strain sensing

Nanocomposite hydrogels composed of alternating layers with different reflective index materials and planar interfaces between each layer can exhibit tunable structural colors. The optical properties of a one-dimensional structure can be tuned by changing the number of layers, incident angles, thickness of the layers, uniformity, and interface quality of the layers. The reflection peak wavelength of 1-D structures can be calculated from the Bragg-Snell law: $m\lambda = 2(n_l d_l + n_h d_h)$, where m is the diffraction order ($m = 1$ for the first order), n_l and n_h are the refractive indices of low and high-refractive-index materials, and d_l, d_h are the thickness of the two repeating layers. Either increase in the thickness or an increase in the reflective indices of the repeating layers leads to a shift in the reflection peak wavelength. The variation in the structural colors of the hydrogels is relatively large because the layer distance can be easily decreased in response to stress/strain. The change in the layer distance could result in a more significant structural color tuning than the effects of refractive index change.[8]

In terms of synthesis, self-assemble and consecutive spin-coating/dipping are the main methods used to fabricate layered photonic hydrogels (**Figure 2a-c**). In particular, the self-assembly of amphiphilic molecules,[11] block copolymers (linear and brush),[12] or inorganic 2D nanosheets into layered structures can be used as a basis for the construction of layered-structured hydrogels. Additionally, layered structures can also be achieved by consecutive spin-coating of hydrophilic/hydrophobic polymer-based solutions, nanoparticles/sol-gel precursors.[13, 14]

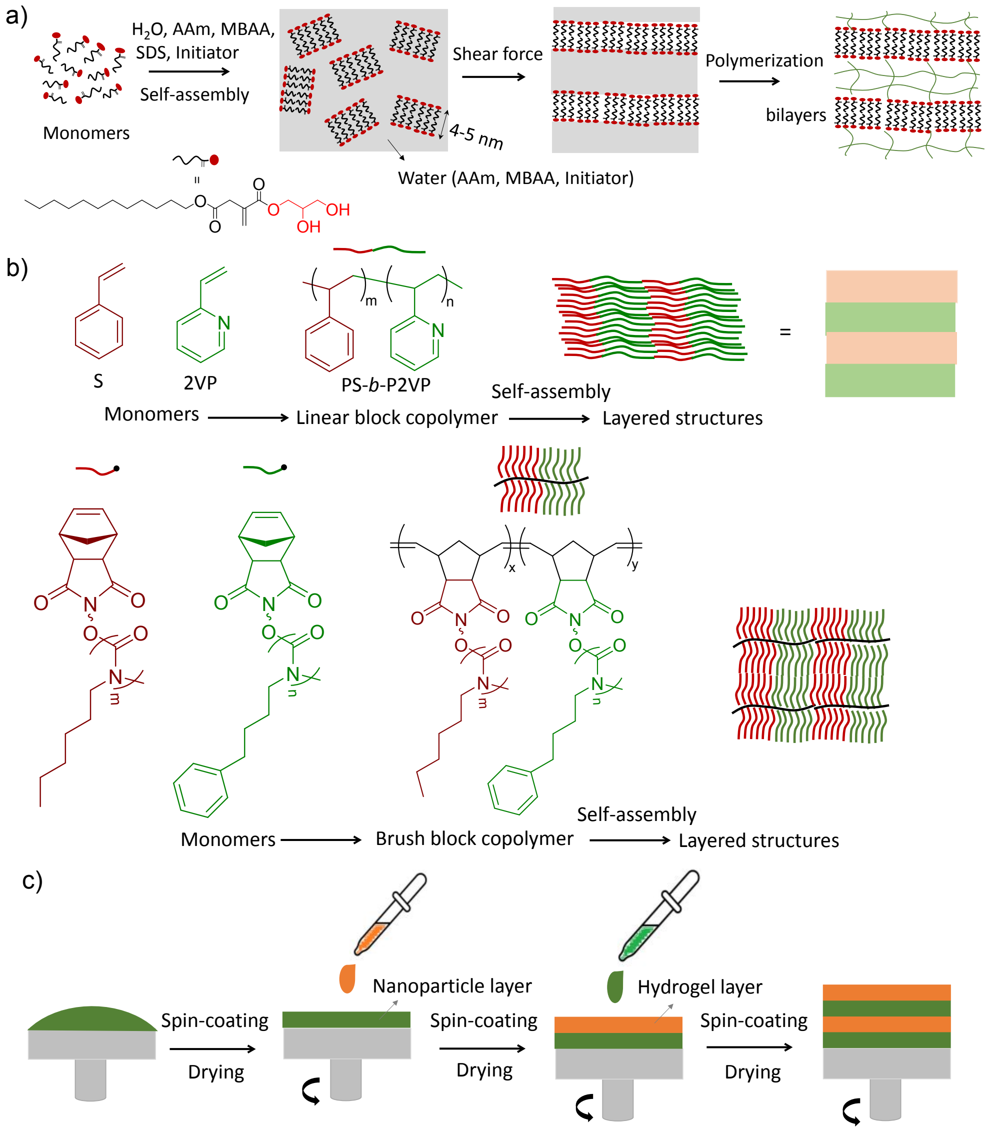

Figure 2 Fabrication of layered-structured hydrogels. (a) Synthesis of layered photonic hydrogels from the self-assembly of amphiphilic molecules. The amphiphilic molecules self-assemble into bilayer structures in water (gray), even when they coexist with acrylamide (AAm) monomers, N, N'-methylene-bis-acrylamide (MBAA) crosslinkers, sodium dodecyl sulfate (SDS) and initiators. After free-radical polymerization, polymer networks (green) are formed (See section 2.1.1. for detail). Panel a was reprinted with permission from Ref. **11**. Copyright 2021 The Chemical Society of Japan. (b) Synthesis of layered photonic hydrogels from self-assembly of block copolymers (See section 2.1.3.). Panel b was reprinted with permission under a Creative Commons CC BY License from Ref. **12**. Copyright 2022 John Wiley and Sons. (c) A general fabrication of layered photonic hydrogels via consecutive spin-coating of nanoparticles and polymer-based aqueous solutions. Panel c was adapted with permission from

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2.1.1 Self-assembly of amphiphilic molecules

Many amphiphilic molecules can self-assemble into bilayer structures in water. This process relies on electrostatic repulsion between the bilayers. To achieve this, a small amount of ionic amphiphilic molecules (e.g., SDS) is co-added to the system to increase electrostatic repulsion. They can form a microscale layered structure with layer spacing of up to several hundred nanometers; however, the self-assembled structure is not highly ordered. Thus, external forces, such as shear, need to be applied to the solutions. The structural color of the solution becomes bright after shearing, indicating an improved orientation of the layered structure.

The formation of structural color in hydrogels with alternating layered structures is challenging due to the difficulties in their synthesis. Tsujii et al. developed photonic hydrogels with self-assembled amphiphilic molecules alternatively embedded in polyacrylamide (PAAm) polymer networks.[15] Without applying shear force to the layered structures, the ordering was limited to the microscale, and the structural color was not uniform. Haque et al. significantly improved the system with shear force during gel preparation and were able to obtain photonic hydrogels that exhibit iridescent structural colors and excellent mechanical properties.[16]

The perfect one-dimensional layer orientation allows the gel to exhibit uniform structural colors, and the color is tuned under mechanical stimuli (**Figure 3a**). This color change was reversible, i.e., the color blue shifted under an applied strain and then returned to its initial color after the removal of the strain. However, because of the existence of many physical bonds, gels are highly viscoelastic and exhibit time-dependent stress relaxation. Therefore, color recovery after stress/strain removal requires a long time (approximately 10 min or more), which limits their application as highly sensitive strain sensors. Yue et al. split the amphiphilic bilayer structure into domains, where the domains play the role of light diffraction without a large energy dissipation.[17] The mechanical properties of the hydrogels changed from viscoelastic to quasi-elastic properties. Then the resulting hydrogels exhibited an ultrafast response (0.1 ms) to mechanical strain, with a large and dynamic structural color change (**Figure 3b, c**). There is a linear relationship between wavelength shift and strains (**Figure 3d**). The hydrogels also exhibit excellent mechanical stability with structural colors that can be reversibly switched more than 10,000 times at high frequency without color degradation (**Figure 3e**). They can even sense small strains with a color change or real-time motion monitoring of an object, such as the movement of human hair under the hydrogels.[17]

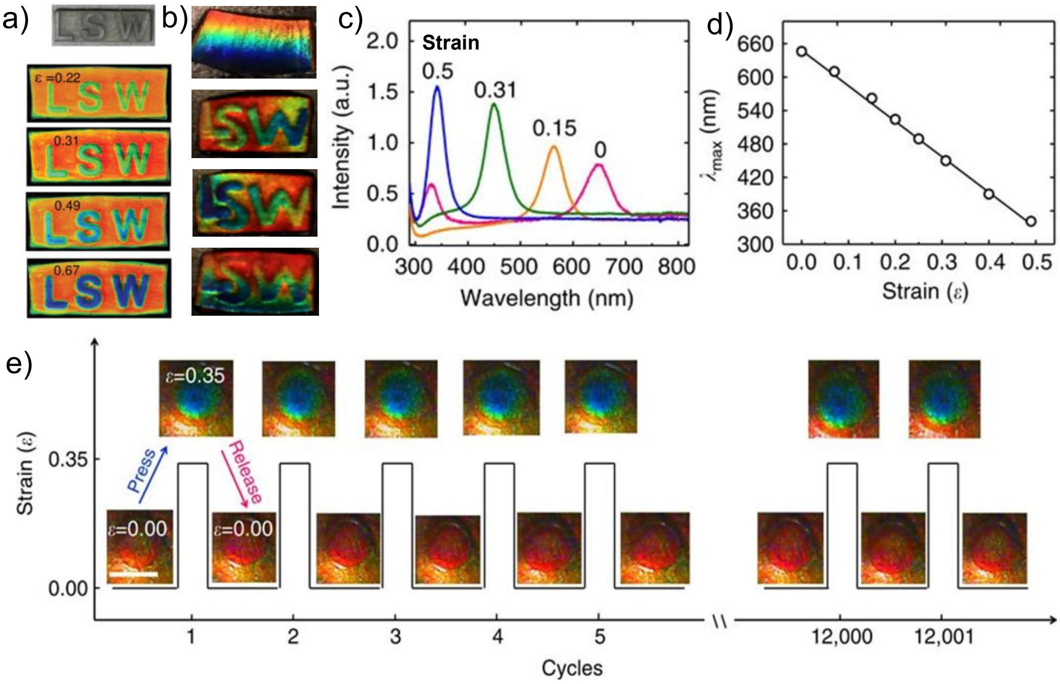


Figure 3 Layered photonic hydrogels as strain sensors based on strain-optical effects. (a) Color change of the hydrogel induced by the strain change. (b) Strain distribution on a highly sensitive photonic hydrogel. (c) Strain-dependent reflection wavelength shift. (d) The peak wavelength decreases linearly with increasing strain. (e) The cycles of strain loading and unloading can be repeated for more than 12,000 times without degradation in the color quality. Panel a was reprinted with permission from Ref. **16**. Copyright 2010 John Wiley and Sons. Panels b-e were reprinted with permission under a Creative Commons CC BY License from Ref. **17**. Copyright 2014 Springer Nature.

2.1.2 Self-assembly of inorganic nanoplatelets

Self-assembly behavior exists not only in amphiphilic molecules, but also in inorganic material systems. For example, dispersions of clay or titanate nanoplatelets in water can produce structural color under appropriate electrostatic repulsion and shear forces.[18,19] Yang et al. synthesized perovskite nanosheet-based poly(N-isopropylacrylamide) (PNIPAAm) hydrogels, in which the perovskite nanosheets are periodically aligned to the gel surface with a layer distance of 300 nm. The structural color gel showed linear and reversible mechanochromic response that detects weak stress of 1 kPa with the quick response time less than 1 ms (**Figure**

4a-c).[20]

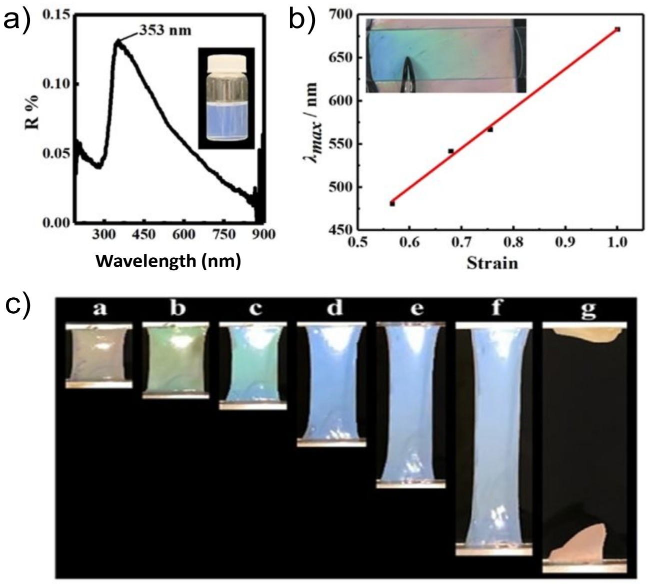


Figure 4 Self-assembly of 2D perovskite nanosheets in synthetic layered hydrogels for strain sensing. (a) Reflectance spectra and photographs of the colloids with the perovskite nanosheet concentration of 0.1 wt%. (b) Linear relationship between the compressive strain and peak reflection wavelength of the 2D nanosheet based gel. (c) Photographs of the gel with 3 wt% of perovskite nanosheet during the stretching. Panels a-c were reprinted with permission from Ref. **20**. Copyright 2021 John Wiley and Sons.

2.1.3 Self-assembly of block copolymers

The self-assembly of block copolymers provides another platform for constructing layered photonic hydrogels with tunable structural colors. Chemical incompatibility between the polymer blocks causes spontaneous phase separation to achieve the lowest energy dissipation. The self-assembly of block copolymers can generate various well-ordered periodic nanostructures such as spheres, cylinders, gyroids, and lamellae. Among them, the lamellar phase, with a large layer distance of up to hundreds of nanometers, can produce layered photonic hydrogels. The layer distance is mainly affected by the molecular weight, additives, and solvents. Different reflective indices between the blocks are important for the generation of structural colors. Similar to the aforementioned layered hydrogels, block copolymer based photonic hydrogels can also deform under an applied strain, which causes a decrease in the layer distance and thus the color tuning ability.

One of the most widely investigated block copolymers is polystryrene-*block*-poly(2-vinyl pyridine) (PS-*b*-P2VP). However, it is still challenging to produce a photonic hydrogel directly from PS-*b*-P2VP due to the similar refractive indices between the two blocks and limited chain lengths. Thus, it is often necessary to dope the layered structure by swelling the P2VP domains with aqueous salt solution.[21]

Block copolymer-based photonic hydrogels are usually fabricated by spin-coating block copolymers on a substrate, followed by solvent-assisted self-assembly. For example, Park et al. reported a self-assembled block copolymer photonic hydrogel that exhibited a fast strain response with a multi-cycle structural color change over 1000 cycles, allowing for convenient visible recognition of finger bending, localized pressure, and the shape of a strained object. [22] This block-copolymer based layered hydrogels exhibited a linear color change with strains and could be utilized for sensing a large strain of up to 100% (**Figure 5a-c**).

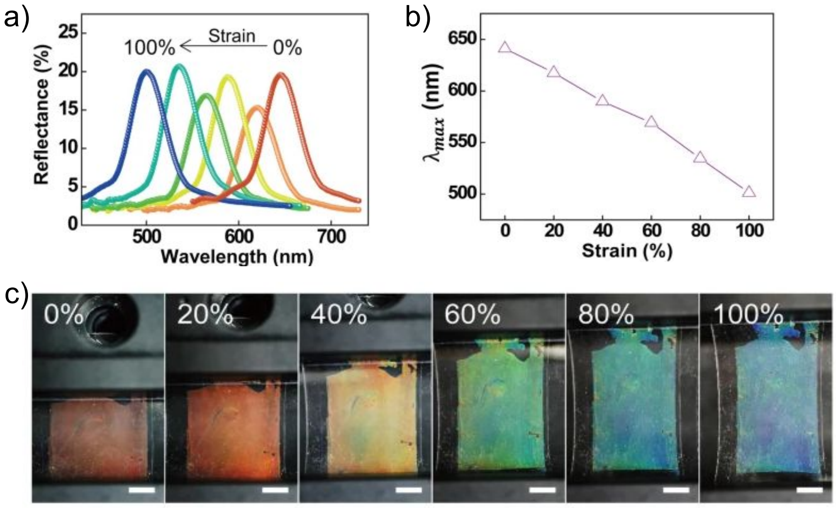


Figure 5 Self-assembly of block-copolymers in synthetic layered hydrogels for strain sensing (a) Block copolymer based photonic hydrogel exhibit blue-shifted structural color with strains. (b) Linear relationship of the strains and peak wavelength of block-copolymer based photonic gels. (c) Photographs of the corresponding structural color as a function of strains. Panels a-c were reprinted with permission under a Creative Commons CC BY License from Ref. **22**. Copyright 2018 Springer Nature.

2.2 Crystalline colloidal array-based hydrogels for strain sensing

The self-assembly of colloidal nanoparticles into photonic crystals has been widely observed in natural materials. Different with the above 1D photonic crystals structures, they are 3D photonic crystals. For example, opals mined on Earth are 3D photonic crystals formed from the close-packed self-assembly of silica nanoparticles.[23] However, these natural photonic crystal materials lack softness and flexibility in response to external strains. Inspired by natural principles, the classic fabrication strategy involves the formation of face-centered cubic (fcc) lattices of silica nanoparticles or polystyrene as close-packed colloidal crystalline arrays within hydrogels. Sometimes in order to further improve the flexibility, the nanoparticles are removed by etching the colloidal crystals, forming an inverse fcc lattice of air holes (**Figure 6a**). Beside close-packed self-assembly of silica nanoparticle, non-close-packed fcc arrays of nanoparticles in a soft and stretchable material is also used to fabricate mechanochromic materials (**Figure 6b**).[24]

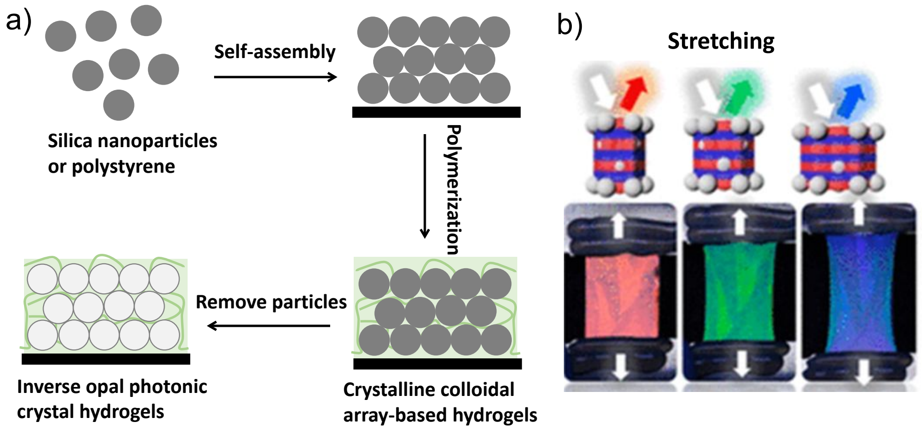


Figure 6 Crystalline colloidal array-based hydrogels for strain sensing. (a) Fabrication of close-packed crystalline colloidal array-based hydrogels and inverse opal photonic crystal hydrogels. (b) The mechanochromic elastomer is fabricated by non-close-packed fcc array of silica particles embedded in a stretchable matrix. Panel b was reprinted with permission from Ref. **24**. Copyright 2017 American Chemical Society.

Self-assembled colloidal crystalline arrays with high ordering also rely on electrostatic repulsion between charged colloidal particles, where their surfaces are modified with ionic groups. The electrostatic interactions were large and occurred over long distances (1 μm) which enabled the formation of macroscopic photonic hydrogels. Additionally, PAAm is optically transparent, can withstand considerable mechanical stretching, and is commonly used to fabricate hydrogel strain sensors. Hong and coworkers reported an approach for synthesizing a highly stretchable photonic hydrogel that was physically cross-linked via a crystalline colloidal array without any chemical cross-linker.[25] The colloidal crystalline arrays cross linked the PAAm hydrogel and reinforced the polymer networks. The gel has a full-color tunable wavelength based on colloidal crystalline arrays, exhibits reversible 460 nm photonic band gap shifts actuated by a small compressive stress (12.7 kPa), and can be ink-printed to form free-standing structures.[25]

As strain sensors, photonic hydrogels are suggested to be used to detect strains generated by body movements or even the motion of biological cells owing to their high water content and biocompatibility.[17, 26] For example, cardiomyocytes can rapidly adhere, spread, and grow on hydrogel surfaces. When cardiomyocytes are seeded on inverse photonic hydrogels, they can record the spontaneous beating frequency **(Figure 7a-c)**.[26] Thus, highly sensitive photonic hydrogels can visualize the time evolution of complicated strain distribution, for example, generated during the motion of biological cells (cell elongation and contraction) with tunable structural colors.

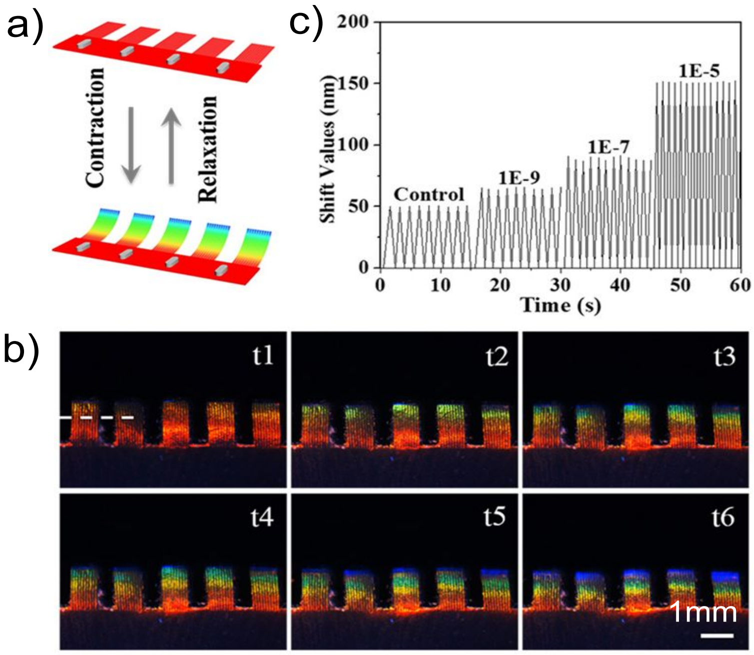


Figure 7 Inverse opal photonic crystal hydrogels as strain sensors. (a) Schematic of the bent-up process of the photonic hydrogels in heart-on-a-chip. (b) Relationship between the reflection peak wavelength and the beating velocity of the photonic hydrogels. (c) Dynamic

optical microscopy images of photonic hydrogels in a heart-on-a-chip system for one myocardial cycle. Panel a-c were reprinted with permission from Ref. **26**. Copyright 2018 The American Association for the Advancement of Science.

Table 1. Performance comparison of some typical photonic hydrogels as strain sensors based on various structures

Structures	Fabrication	Wavelength shift	Specific features	Refs.
Layered photonic hydrogels	Self-assembly, shear force	415-600 nm, 185 nm	Anisotropic modulus, tough, close to linear	Ref. 16
Layered photonic hydrogels	Self-assembly, shear force	340-640 nm, 300 nm	Fast response (0.1 ms), >10000 cyclic test, linear	Ref. 17
2D (perovskite) nanosheet based-hydrogels	Nanosheet assembly	481-683 nm, 200 nm	Fast response time (1 ms), linear	Ref. 20
Block copolymer -based hydrogels	Spin-coating, self-assembly	500-640 nm, 130 nm	>1000 cyclic test, linear	Ref. 22
Crystalline colloidal array-based hydrogels	Particle assemble, interparticle repulsion	490-620 nm, 130 nm	>20 cyclic test, linear	Ref. 24
Crystalline colloidal array-based hydrogels	Particle assemble	495-955 nm, 460 nm	Ink-written to form patterns, nonlinear	Ref. 25
Inverse opal–structure hydrogel	Particle assemble, etching	425-600nm, 175 nm	Beating processes of cells, close to linear	Ref. 26

To compare the performance of photonic hydrogels based on different structures, fabrication, color shifts, and specific features, some exampled photonic hydrogels are summarized in **Table 1**. Photonic hydrogels used as strain sensors are generally required to respond quickly and reversibly to applied strains, with high color quality, fast response and broad wavelength shifts. The linearity between the applied strain and the peak wavelength is an important parameter for strain sensors. Due to energy dissipation, hydrogels with high toughness often exhibit hysteresis during stretching/releasing, which can lead to the nonlinearity in hydrogel-based strain sensors. Additionally, durability is another parameter that indicates the ability to withstand long-term stretching/releasing cycles with stable optical functionality and mechanical integrity.

3. Conductive hydrogels as strain sensors based on strain-electrical conversion

In addition to the aforementioned strain-optical conversion hydrogels, strain-electrical conversion is widely used to construct hydrogel strain sensors. Embedding conductive nanomaterials, such as electrolytes, carbon-based materials, metallic nanoparticles, MXenes, or conductive polymers, into hydrogels will endow the materials with tunable electrical signals (resistance or capacitance change) in response to mechanical stimuli. They can change their electrical properties in response to external mechanical stimuli with a large stretchability of up to 10000%, whereas the current commercially available strain sensors based on elastomers have limited stretchability of up to 200%.[27,28] Thus, the electrical signal change from stretchable hydrogels is a promising candidate for strain sensor applications. In particular, when hydrogel-based strain sensors are attached to the skin, they can detect body movements (e.g., breathing pulse, heart rate, and joint twist) that convert applied stress/strain into electrical signals.

Based on their conductive mechanisms, conductive hydrogels can be divided into three main types: electron-, ion-, and hybrid electron-ion conductive hydrogels. The type of conductive component and incorporation method play an important role in the conductivity of hydrogels. The conductivity of ion-conductive hydrogels depends on the movement of ions along with water. Therefore, they can generate ionic gradients within hydrogels, and the conductivity is significantly influenced by water content. The conductivity of electron-conductive hydrogels mainly originates from the moving electrons via the contact effect. They usually have excellent electrical conductivity, and the entrapped water molecules do not significantly influence conductivity.

Compared to the fabrication of photonic hydrogels with tunable strain-optical effects, the synthesis of conductive hydrogels is relatively easier and can be prepared by in situ formation of a conductive network within the pre-formed hydrogels or polymerization of a precursor solution containing conductive materials.

For conductive hydrogels as strain sensors, the gauge factor (GF) is used to define the strain sensitivity, which is evaluated by the relative change in the resistance or capacitive value ($\Delta R/R_0$, $\Delta C/C_0$) versus the applied strain. For example, when the hydrogel was stretched to 100%, the resistance increased by approximately 250%. Then the hydrogel has a strain sensitivity of GF = 1.5.

3.1 Ion-conductive hydrogel strain sensors

In ion-conductive hydrogels, electrical currents are transported by ions owing to the motion of the ionic charge. Inorganic materials, such as metals, conduct current through electrons, whereas electrical conduction in biological tissues (0.1 to 1 S m^{-1}) is often due to the migration of ions.[29] From this perspective, ion-conductive hydrogels are more likely to be biocompatible.

Compared with electron-conductive hydrogels, ion-conductive hydrogels can be prepared with high optical transparency owing to the lack of dark-colored conductive additives, facilitating their application in photoelectronics or bioelectronics. The main parameters that affect ionic conductivity are molecular mobility and ion concentration. A high water content increases the molecular mobility of ions; thus, the ionic conductivity is easily affected by the water in the hydrogels.

3.1.1 Polyelectrolyte-conductive hydrogels

Polyelectrolyte-based hydrogels are among the main ion-conductive hydrogels. Depending on the type of electrolyte group, they can be classified into three groups: polycations, polyanions, and polyampholytes (zwitterionic polyelectrolytes). Among them, zwitterionic-based hydrogels have been widely used as strain sensors, based on the commonly used method of copolymerization of their cationic and anionic monomers. First, the presence of abundant zwitterionic groups promotes ionic conductivity because ions are able to migrate along with a highly depolarized zwitterionic skeleton, providing hydrogels with good conductive properties. Second, the association of zwitterions can work as physical cross-linkers to dissipate energy and enhance the mechanical properties (e.g., toughness, self-assembly, and viscoelasticity) of the hydrogels.[30, 31] Third, zwitterionic-based hydrogels have high dipole moments because the polymer chains have both positive and negative charges, and therefore the hydrogels have significant self-adhesive properties, allowing them to stick to skins for wearable strain sensors.[32]

Zwitterionic-based hydrogels exhibited higher ionic conductivity in high concentration of salt solutions, compared to cationic and anionic polyelectrolyte hydrogels.[33] Although all polyelectrolyte hydrogels obtained better ionic conductivity with increasing salt concentrations, zwitterionic hydrogels exhibited a higher rate of increase, which surpasses that of cationic/anionic hydrogels in 1 M $MgCl_2$ electrolyte solutions.[34] It is still not clear how the functional groups in zwitterionic hydrogels affect the ion diffusion. It is postulated that the zwitterion would provide a polar medium or migration channels for movement of other ions through the polyelectrolyte, which significantly facilitate ion transport. [35, 36]

Lei et al. developed a type of zwitterionic hydrogel from the perspective of molecular interactions, which successfully combined ultrastretchability (>10000%), self-healability, 3D printability, strain-responsibility, biocompatibility, and antibacterial activity.[37] Wang and coauthors prepared highly conductive zwitterionic hydrogels that showed a high fracture elongation of up to 2000%, a fracture strength of up to 0.27 MPa, and a fracture toughness of up to 2.45 MJ/m^3.[31] The combination of these properties enables hydrogels to act as strain sensors with high sensitivity (gauge factor = 1.8).

Xu et al. prepared zwitterionic hydrogels that generate ions under an applied force in a mobile-ion-free system.[38] Under a tiny pressure, deformation of the polymer network in the hydrogel increased the Coulomb interactions between zwitterions, causing water dissociation and subsequent OH^- generation (**Figure 8a**) . The instantaneous increase in ionic concentration generated ionic currents that improved the sensing sensitivity of the hydrogel. The signal response time is about 38 ms, which is comparable to that of natural skin. The skin hydrogel sensor is integrated into a universal throat-worn silent speech recognition system that converts minute signals from mechanical strain vibrations of the throat into silent speech (**Figure 8b-e**).[38]

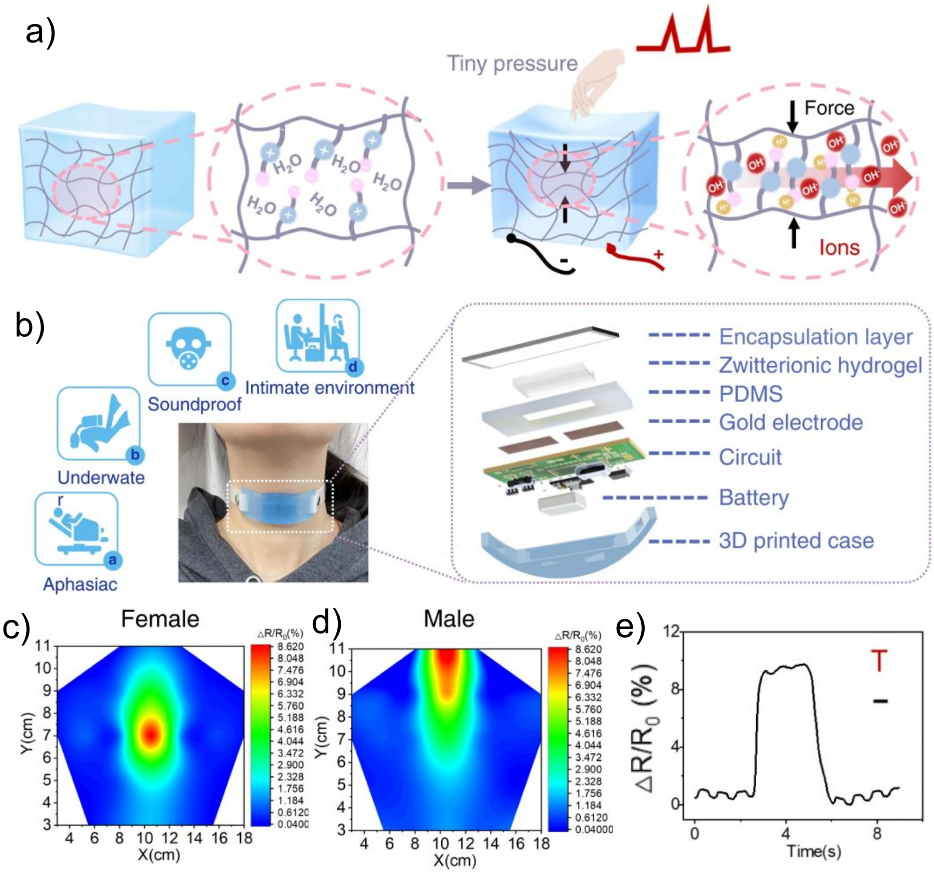


Figure 8 Zwitterionic hydrogel as strain sensors. (a) The deformation decreases the distance

between the zwitterions on the polymer backbones, dissociating water into mobile OH^-. (b) Specific situations serviced by silent-speech interfaces and a photograph of the throat-worn sensor. (c, d) Resistance signal mappings of the throat vibrations of female and male volunteer. (e) Recognition representative silent-speech signal: T. Panel a-e were reprinted with permission under a Creative Commons CC BY License from Ref. **38**. Copyright 2023 Springer Nature.

3.1.2 Electrolyte-based hydrogels

In addition to covalently bonded ionic groups, free ions in hydrogels also facilitate good electrical conductivity. Thus, electrolyte-based hydrogels are another type of ion-conductive hydrogels. The simple synthesis method for these hydrogels involves the direct dissolution of the electrolytes (free ions) in the hydrogel precursor solutions or post-modification to include electrolytes in the hydrogels. There are various types of electrolytes, such as NaCl, KCl, LiCl, $FeCl_3$, Na_2HPO_4, and $CaCl_2$, which can be doped hydrogels to improve the ionic conductivity to higher than 10 S m^{-1}. Wu and coauthors developed antifreezing and antidrying PAAm organohydrogels for high-performance strain sensing by solvent replacement and doping with KCl electrolyte. The movement of K^+ and Cl^- ions in the solvents contributes to the conductivity of the organohydrogels, which can monitor various human motions with high sensitivity (GF = 6), wide strain range (0.5–400%), wide temperature range, and excellent stability. [39]

3.2 Electron-conductive hydrogel strain sensors

In addition to the aforementioned ion-conductive hydrogels, electron-conductive hydrogels are another type of hydrogel used as strain sensors. Strain sensors based on electron-conductive hydrogels can quickly monitor human movements (e.g., elbow rotation, knee bending, and swallowing) through stable and repeatable changes in electrical signals, indicating their potential applications in personal health monitoring, human motion detection, and soft robotics. Doping with conductive materials, such as carbon nanotubes, metallic nanoparticles, or conductive polymers via physical or chemical interactions, can not only impact materials with high electron conductivity, but also improve the mechanical properties of the hydrogels. However, disconnected nanoaggregates of the doped materials or poor miscibility of the included components may also restrict their electrical or mechanical properties.

3.2.1 Carbon-based hydrogels

Carbon-based nanomaterials have diverse structures and properties because of the various hybridization modes (sp, sp2, and sp3) between carbon atoms.[40] Carbon-based nanomaterials are mainly classified as carbon dots, carbon nanotubes, graphene, and their derivatives. Among them, carbon nanotubes with high electrical conductivity, flexibility, and mechanical strength are usually combined into hydrogels to endow composites with good conductivity and excellent mechanical properties. However, one of the main disadvantages of using carbon nanotubes is that they are immiscible with most media (poorly dispersed in solution) because of their strong tendency to establish van der Waals and π-π interactions, causing them to form bundles.[41, 42] Thus, the uniform dispersion of hydrophobic carbon-based materials in hydrophilic polymer networks is crucial for the fabrication of these types of hydrogels. Physical interactions, such as hydrogen bonding, facilitate the dispersion of carbon nanotubes in polymer networks. Additionally, functionalized carbon nanotubes with substances containing functional groups, such as carboxylic acid groups, help them to hybridize in the hydrophilic polymer network from hydrogels. Sun et al reported nanocomposite hydrogels composed of oxidized multi-walled nanotubes and PAAm, where the nanotubes are uniformly dispersed in the presence of the gelatin via hydrogen bonding.[43] The resulted nanocomposite hydrogel exhibited excellent strain sensitivity (GF= 3.39 at 250–700% strain), fast response (300 ms) and high durability (over 300 cycles) due to the conductive pathways formed by carbon nanotubes. These carbon nanotube-based hydrogels could detect the wrist bending and identify complex skin stretching over the human throat associated with speaking “goodbye” repeatedly (**Figure 9a-d**).

Graphene, a 2D carbon-based materials, exhibits high conductivity, a large surface area, excellent electrical conductivity, and mechanical strength (Young’s modulus of 1.0 TPa). Graphene and its derivatives, such as graphene oxide (GO), and reduced graphene oxide (rGO) have also been widely used to produce conductive hydrogels. Jing and coworkers prepared a nanocomposite hydrogel composed of polyacrylic acid (PAAc) and reduced graphene oxide, which exhibited high stretchability (600%), high mechanical strength (400 kPa), and excellent self-healing properties (75%).[44] Chen et al. fabricated conductive hydrogels by adding Ag nanoparticle-coated graphene oxide (Ag/TA@GO)-based nanocomplexes to a PAAm hydrogel. The nanocomposite hydrogel showed a high stretchability of 1250%, excellent conductivity (0.15 $S \cdot m^{-1}$), and strain sensitivity of GF = 3.1. [45]

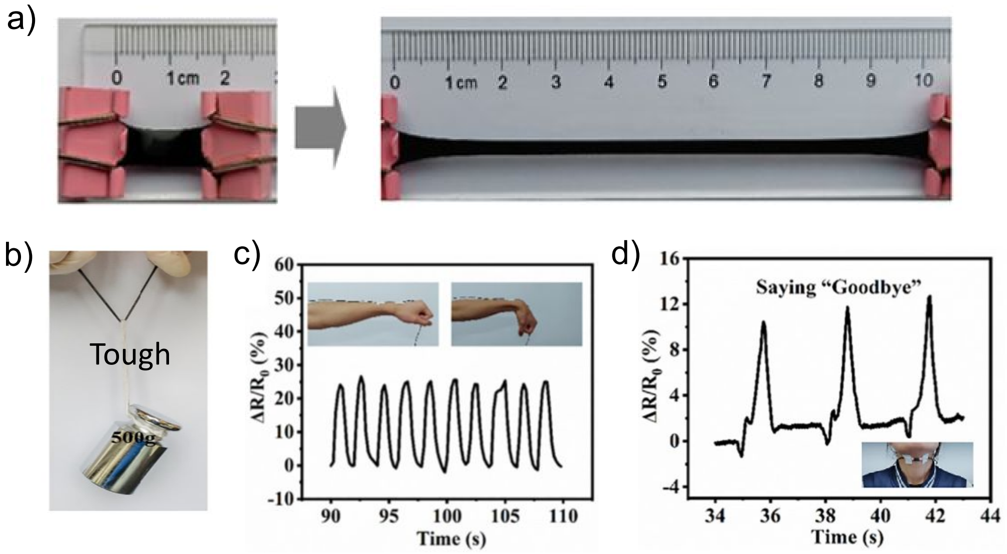


Figure 9 Carbon nanotube-based hydrogels as strain sensors. (a-b) Images of carbon nanotube-based hydrogels under stretching and holding a weight of 500g. (c-d) Carbon nanotube-based hydrogels detect the wrist bending and identify complex skin stretching over the human throat associated with speaking "goodbye" repeatedly. Panel a-d were reprinted with permission from Ref. **43**. Copyright 2020 Elsevier.

3.2.2 Metallic nanoparticle-based hydrogels

Metallic nanoparticles have been widely used to fabricate conductive hydrogels for strain sensor applications. For example, Ag, Au, and Cu nanoparticles have been incorporated into polymer networks to impact hydrogels with appropriate electrical conductivity. In particular, metallic nanoparticles should be homogeneously dispersed and stabilized within polymer networks to improve their conductivity. However, homogeneous dispersion of metallic nanoparticles in hydrogel matrices is not easy due to the tendency of nanoparticles to agglomerate and high viscosity of hydrogels.[46] Thus, physical or chemical bonds need to be incorporated into the hybrid systems. As a typical example, Au nanoparticles are usually incorporated into hydrogel matrices (e.g., polyacrylamide) that contain -SH, -CN, and -NH_2 groups by forming covalent bonds.[47] To avoid the agglomeration of metallic nanoparticles, the metal ions (e.g., Ag^+ or Cu^{2+}) are anchored in the polymer networks by the carboxylic groups of the hydrogels before in situ reduction. This facilitates the uniform incorporation of metallic nanoparticles into hydrophilic polymer networks after reduction.[46]

Therefore, the corresponding doping methods can be divided into two types. One directly incorporates the nanoparticle precursor (metal ions) into the hydrogel precursor solution, and then the reduction of metal ions into nanoparticles occurs together with hydrogel synthesis. The other is the incorporation of nanoparticles into gels after hydrogel formation. However, there are several challenges in terms of material stability, mass production, and effectiveness.[48]

Lin et al. prepared conductive hydrogels by incorporating Ag nanoparticle-coated cellulose nanocrystals into the polyvinyl alcohol (PVA) matrix, allowing enhancement of the conductivities (4.61 S m^{-1}), stretchability (>4000%), antibacterial properties, and repeatable self-adhesiveness of composite hydrogels.[49] There is a trade-off between conductivity and stretchability. Yue et al. embedded Au nanosheets in a small amount of organic matrix, which exhibited strain-dependent changes in conductivity because the sliding of Au nanosheets under compression leads to significant changes in the length of conductive paths.[50] However, without sufficient polymer networks, the composites exhibited high electrical conductivity but poor reversibility. In addition, direct doping of metal nanomaterials into hydrogel networks has less effect on enhancing their mechanical properties compared with 1D or 2D carbon-based doped materials.

3.2.3 Conductive-polymer-based hydrogels

Conductive polymers, such as poly (3, 4-ethylenedioxythiophene): polystyrene sulfonate (PEDOT: PSS), polyaniline, polypyrrole, and polythiophene, exhibit good conductivity due to the highly delocalized, polarized, and electron-dense π bonds on the conjugated carbon chains. The conductivity of undoped polymers is 10^{-6}–10^{-10} S cm^{-1}, which is among the range between semiconductor and insulator. The conductivity can increase up to more than 10^4 S cm^{-1} after doping, which is comparable to that of metallic conductor.[51]

Conventional hydrogels typically exhibit an intrinsic ionic conductivity from 10^{-5} to 10^{-1} S cm^{-1} in physiological conditions, while the conductivity of conductive-polymer-based hydrogels can change from 0.003 to 0.27 S cm^{-1} depending on the type of conductive polymer doped.[52] Although the pure conductive polymers have a high electrical conductivity and flexibility, they face problems in their stretchability. The introduction of conductive polymers into hydrogels can significantly improve their stretchability.

For fabrication, conductive polymers can be directly introduced into the hydrogels or via post-treatment of the prepared hydrogels. PEDOT:PSS is considered to be one of the most promising additives for the synthesis of polymer-based hydrogels owing to its high water dispersibility. Shen et al. prepared a highly stretchable and hysteresis-free strain sensor based on conducting polymer hydrogels for use in wearable skin and soft robots.[53] The hydrogels were composed of PEDOT:PSS nanofibers and PVA by combining 3D printing and successive freeze-thawing. The GF of the strain sensor was calculated to be 4.07, indicating high sensitivity. By recording the resistance changes of all the five channels, the wearable PEDOT:PSS based hydrogels could be used to precisely recognize hand gestures (**Figure 10**).

Post-treatments of the prepared hydrogels, such as second polymerization to form interpenetrating polymer networks or double polymer networks, are generally used to form stable conductive hydrogels. For example, Chen et al. developed conductive hydrogels that combined high conductivity with stretchability, injectability, and rapid self-healing ability in one platform by introducing 2-ureido-4[1H]-pyrimidinone groups into fragile conducting polyaniline/poly(4-styrenesulfonate) polymers based on hydrogen-bonding and electrostatic interactions.[54] The interpenetrating polymer networks not only provides mechanical enhancement but also impacts ionic transport-assisted electronic conduction, where the hybrid hydrogel shows an excellent conductivity of 0.13 S cm^{-1} with a linear response (GF = 3.4) to external strain (300%). Additionally, the thermos-responsive association/dissociation of the H-bonds offered the hydrogel injectability, which can be molded into different shapes.

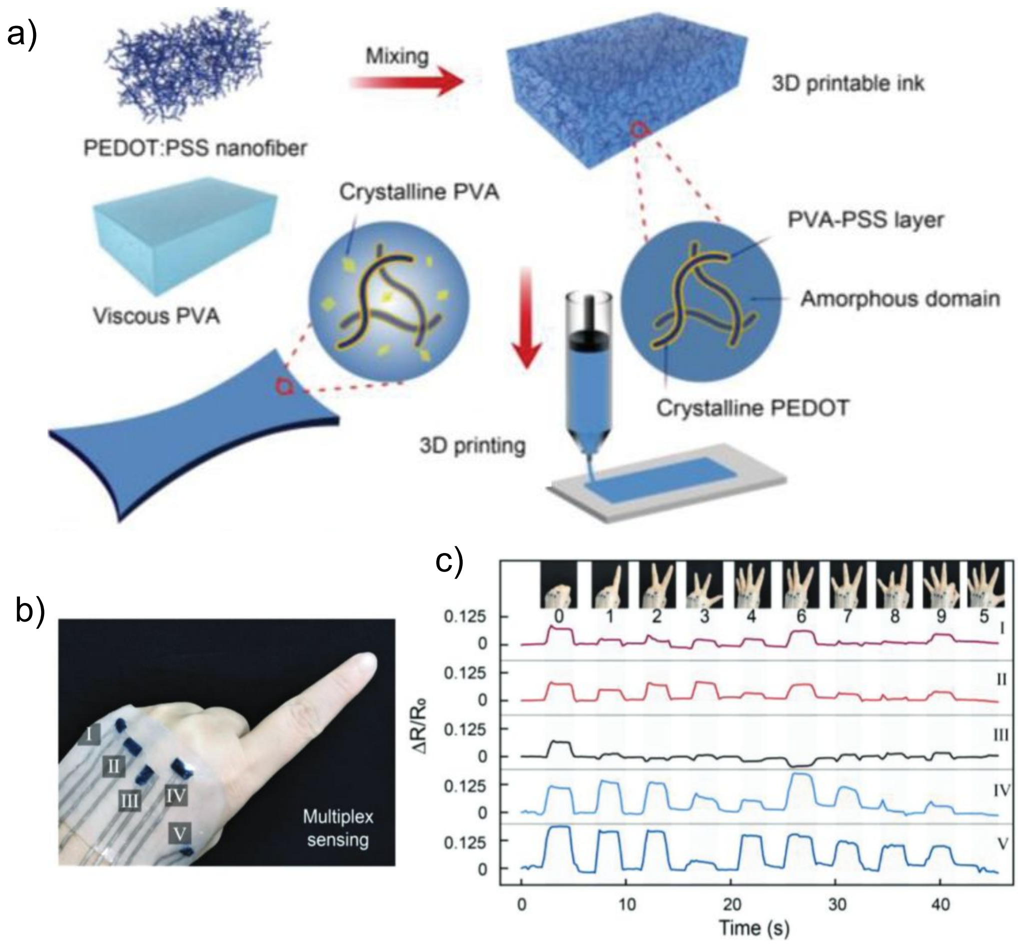


Figure 10 Conductive polymer-based hydrogels as strain sensors. (a) Simplified fabrication process of the PEDOT:PSS-PVA conducting polymer hydrogel. (b) PEDOT:PSS conductive polymer-based hydrogels used as wearable electronic skin mounted on the back of a hand which provided multiplex sensing capability for gesture recognition. (c) Their relative changes in resistance of five channels in the electronic skin when the subject performs gestures from 0 to 9. Panels a-c were reprinted with permission from Ref. **53**. Copyright 2022 John Wiley and Sons.

3.2.4 MXene-based hydrogels

MXenes ($Ti_3C_2T_x$), a new family of transition metal carbides/carbonitrides, possess hydrophilic surfaces, two-dimensional layered structures, and high electrical conductivity.[55] MXene nanosheets with abundant surface functional groups (e.g., −OH, −F, and −O) can be incorporated into hydrophilic polymer networks to endow the composites with good mechanical properties and high stretchability. To date, MXenes have been combined with a wide range of polymers to prepare hydrogels for strain sensor development. They show promise for piezoresistive sensing materials, as the sliding and stacking of MXene nanosheets under tension or compression lead to significant changes in the number and length of conductive paths, further leading to dramatic changes in resistance.[56, 57]

For instance, Zhang and coauthors incorporated MXene into low-cost hydrogels, “crystal clay”, to monitor the direction of object movement. The nanocomposite hydrogel exhibited a remarkable stretchability of more than 3400%, instantaneous self-healing ability, high strain sensitivity (GF = 25 under tensile strain; GF = 80 under compressive strain), excellent conformability, and adhesiveness to various surfaces.[58] The compressive force reduced the gap between the MXene nanosheets and establish more surface contact between the MXene nanosheets. Thus, the resistance changed repeatability when the finger was bent, and the compressive force from handwriting also induce characteristic resistance change, which can be used for signature recognition (**Figure 11**). Li *et al.* developed a biocompatible nanocomposite hydrogel, fabricated from the conformal coating of MXene networks using PAAc polymer networks.[59] The hydrogels sensitively detect human motion with a fast response time (20 ms) and serve as electronic skins for wirelessly monitoring electrophysiological signals. The GF values were 1.51 in the strain range of 0.3−30% and 10.79 in the strain range of 30−450%. Cai and coauthors developed a modified PAAm hydrogel combined with MXene nanosheets and polypyrrole nanowires in a layer-by-layer manner. The multidimensional configurations endowed the hydrogel composites with an extraordinary working range (2800%), ultrafast responsiveness (90 ms), good linearity (0–800% strain), reliable strain sensitivity (GF = 16.9), and excellent reproducibility (>5000 cycles). [60]

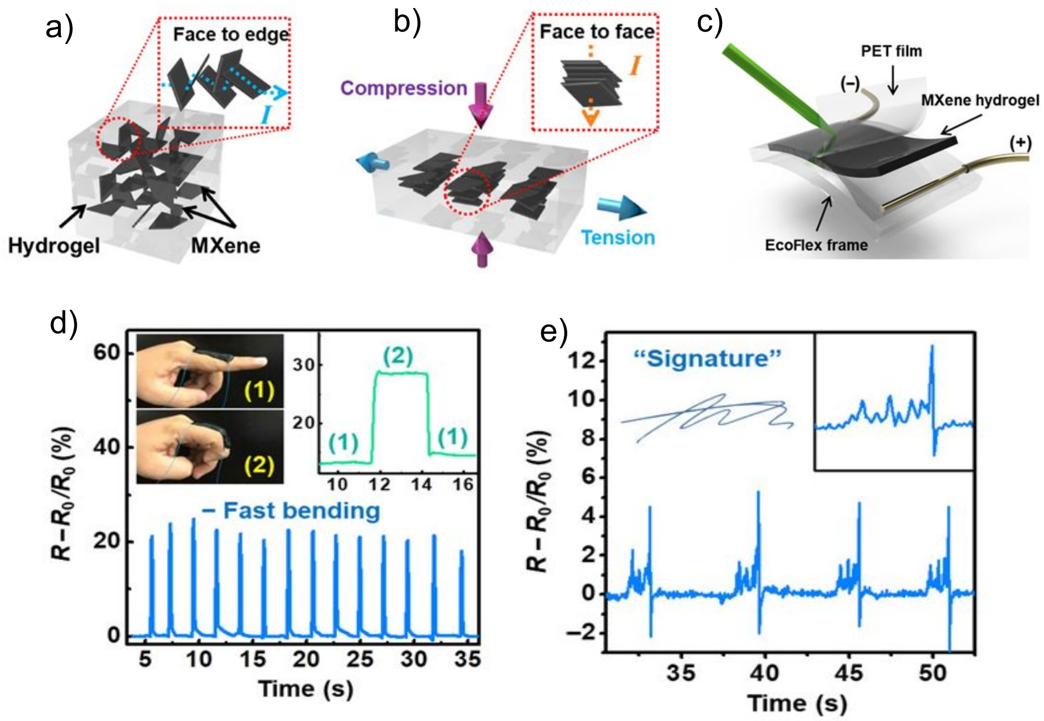


Figure 11 MXene-based conductive hydrogels as strain sensors. (a-b) Schematic illustration of the electromechanical responses of MXene based hydrogel. (c) Schematic for signature sensing. (d) Resistance changes during finger bending. (e) Resistance changes to signature. Panels a-e were reprinted with permission under a Creative Commons CC BY License from Ref. **58**. Copyright 2018 The American Association for the Advancement of Science.

A summary of conductive hydrogels based on different structures is listed in **Table 2**. There are several main types of conductive hydrogels which are generally used as stretchable soft materials to detect the strain distribution or real-time monitor the movements of subjects. Among them, hydrogels from ion-conductive mechanisms are usually transparent or light colored, while the hydrogels based on carbon-, conductive polymers, or MXene materials are dark-colored. When an efficient dopant is applied, hydrogels based on conductive polymers exhibit higher conductivity than other hydrogels due to the formation of continuous conductive pathways along the polymer skeleton. Electron-conductive hydrogels are often prepared by dispersing or mixing hydrophobic conductive materials with other monomers followed by a polymerization or chemical reaction to form solid-state materials. It is sometimes not easy because the conductive materials are usually hydrophobic, which is difficult to uniformly mix within hydrophilic polymer networks. The non-uniformity problem will consequently affect the functional properties of hydrogels, including conductivity, sensitivity, linearity, and mechanical properties. For example, once the non-uniform microstructures become microcracks during stretching, strain sensors respond to the applied strain in a nonlinear manner, which is one of the major drawbacks of conductive hydrogels as strain sensors. Thus, the surface modification or introducing of strong physical interactions between the nanomaterials and polymer networks are applied to produce uniform hydrogels. MXene-based hydrogels are reported to exhibit a high sensitivity with high GF values, when they are combined with other conductive polymers or nanowires within polymer networks. They also show promise for piezoresistive sensing materials, as the sliding of MXene nanosheets under stretching or compression further leads to significant changes in the length of conductive paths, resulting in changes in resistances.

Table 2. Performance comparison of some typical conductive hydrogels as strain sensors based on various structures

Types	**Synthesis & Polymer networks**	**Conductive materials**	**Sensitivity**	**Specific features**	**Refs.**
Polyelectrolyte -based hydrogels (Ion-conductive)	Free radical polymerization copolymer, zwitterionic	Free ions, 0.099 S m^{-1}	GF = 1.8	Transparent, self-healing	Ref. 31
	Radical copolymerization, zwitterionic	Free ions	9 to 0.9 kPa^{-1} %	Biocompatible, Antimicrobial, Adhesion	Ref. 37
	Radical polymerization, zwitterionic	Free ions, H^+/OH^-	5.92 ± 0.45 kPa^{-1}	Generate ions under force, response time (~38 ms)	Ref. 38
Electrolyte-based hydrogels (Ion-conductive)	One-pot polymerization Solvent-replacement	Free ions, K^+ and Cl^-	GF = 6	Antifreezing, self-healing	Ref. 39
Carbon-based hydrogels (Electron-conductive)	Dispersion and polymerization CNTs, PAAm	CNTs, 0.067 $S \cdot m^{-1}$	GF = 3.39	Fast response (300 ms), durability (>300 cycles)	Ref. 43
	Dispersion and polymerization, rGO, PAAc	rGO	-	Self-healing, biocompatible	Ref. 44
	Dispersion and polymerization Ag/TA@GO, PAAm	Ag/TA@GO, 0.15 $S \cdot m^{-1}$	GF = 3.1	Detect large and delicate motions	Ref. 45
Metallic nanoparticle -based hydrogels (Electron-conductive)	Ag nanoparticle-coated cellulose PVA	Ag nanoparticles	-	Self-healing, adhesive, antibacterial	Ref. 49
Conductive-polymer -based hydrogels (Electron-conductive)	Dispersion, physical crosslinking, PVA	PEDOT:PSS nanofibers	GF = 4.07	Hysteresis-free, 3D printable, 2000 cyclic test	Ref. 53
	Dispersion and polymerization polyaniline, poly(4-styrenesulfonate)	Polyaniline, 13 $S \cdot m^{-1}$	GF = 3.4	Injectable, self-healing, thermoplasticity	Ref. 54
MXene-based hydrogels (Electron-conductive)	Mixing MXene sediment with PVA	MXene ($Ti_3C_2T_x$)	GF = 25 GF = 80 (compressive)	Self-healing, adhesive to various surfaces	Ref. 58
	Mixing and reaction, MXene, PAAc, calcium carbonate	Mxene ($Ti_3C_2T_x$)	GF = 1.51 (0.3-30% strain) GF= 10.79 (30-450% strain)	Self-healing, degradable	Ref. 59
	Mxene, silica nanopartic PAAm, polypyrrole nanowires	Mxene ($Ti_3C_2T_x$), polypyrrole	GF =16.5	Capacitive effect fast response, >5000 cycles	Ref. 60

4. Hydrogel strain sensors based on both optical and electrical change

In addition to the aforementioned photonic hydrogels and conductive hydrogels used as strain sensors, some hydrogels have been designed with both optical and electrical tuning under mechanical stimuli. To fabricate these hydrogels, not only photonic crystal structures but also conductive nanomaterials need to be included in the hydrogels.

Compared with the commonly reported single-signal strain sensor, these hydrogels can be used as multi-signal response films to achieve real-time color sensing and electrical response during human motions, exhibiting dual-signal response with visualization and self-reporting. For example, Liu et al. developed an inverse opal photonic crystal hydrogel containing acrylic acid, acrylamide, chitosan, and carbon nanotubes, which could output an electronic signal together with structural color changes.[61] This photonic crystal hydrogel provides quantitative feedback to external stimuli, such as stretching, bending, pressing, and thermal stimuli, with brilliant color change and electronic signal outputs simultaneously. Zhang et al. developed a composite hydrogels using the mixture of PAAm, silk cellulose, PEDOT:PSS and graphene oxide to replicate colloidal crystal templates and constructed inverse photonic crystals. Hydrogels have vivid structural color and high conductivity, and exhibit color and resistance changes during stretching or bending (**Figure 12**). When the film was stretched from 0% to 150%, the structural color blue shift from 653 nm to 507 nm and the resistance of the hydrogels changed significantly (120%).[62] The dual-signal response performance in real-time color sensing and electrical signal monitoring suggests that the film can be explored as a potential candidate for monitoring motion.

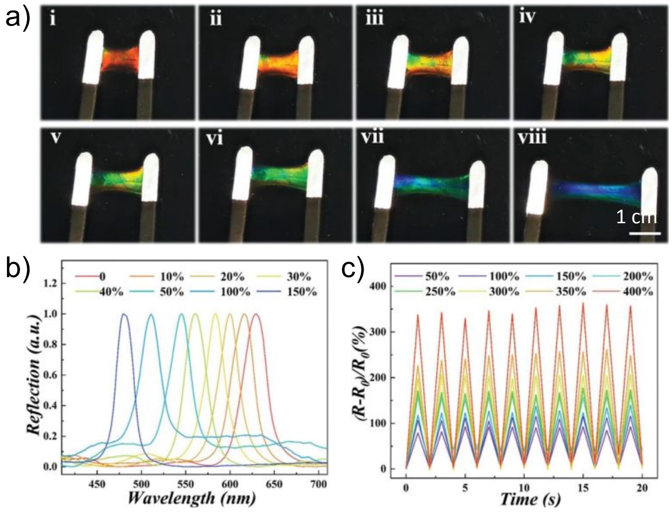


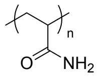
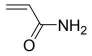
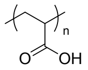
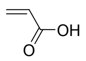
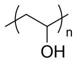
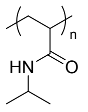
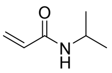
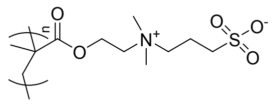
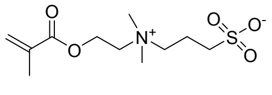
Figure 12 Photonic hydrogels combined with conductive polymers/graphene oxide can output

an electronic signal together with structural color changes. (a) Optical images of the composite hydrogel film under different strains. (b-c) The reflection wavelength and relative resistance changes of composite hydrogel film under stretching. Panel a-c were reprinted with permission under a Creative Commons CC BY License from Ref. **62**. Copyright 2021 John Wiley and Sons.

5. Polymer structures and their interactions with nanostructures/nanomaterials

The selection of appropriate chemical structures of polymers should be important for their water retention, mechanical properties, and miscibility of the nanostructures/nanomaterials in the hydrogels towards strain sensing applications. The mostly reported polymer matrix used in hydrogel-based strain sensors are PAAm, PAAc, PVA, PNIPAAm, and some representative zwitterionic polymers (**Table 3**).

Table 3. Some typical polymer matrix and their monomers that frequently used in hydrogel-based strain sensors

Types	Chemical structures	Monomers	Specific features for the hydrogels	Refs.
PAAm			Amide groups, hydrophilic, transparent, H-bonds	Ref. 11, 25
PAAc			Carboxylic groups, hydrophilic, transparent, self-healing H-bonds, coordination interactions with metal ions	Ref. 44, 68
PVA		Hydrolysis of poly(viny acetate)	Hydroxyl groups, hydrophilic, self-healing	Ref. 53, 76
PNIPAAm			Switching between hydrophilic/hydrophobic with temperature, volume phase transition	Ref. 20, 77
PSBMA			High dipole moment, zwitterionic, self-adhesive	Ref. 38, 78

5.1 PAAm-based polymer matrix

PAAm is a stable hydrogel material that is highly transparent, stretchable, nontoxic, nonimmunogenic, and biocompatible. They are commonly used in nanocomposite hydrogels towards strain sensing applications based on strain-optical or strain-electrical conversions. For example, layered photonic hydrogels use PAAm polymers because of their high optical transparency and hydrophilicity.[16] Especially, the interfacial H-bonds between hydrophilic soft PAAm and hydrophobic hard bilayers significantly improve the mechanical stretchability of the hydrogels (up to 1400%).[63] Although the mechanical properties of single PAAm polymers are weak, their structures can be modified using a variety of strategies to improve mechanical properties. First, PAAm is often used in double network hydrogel synthesis, where poly (2-acrylamido-2-methylpropanesulfonic acid) (PAMPS) polyelectrolyte is used as the first network and PAAm neutral polymer as the second network. Such double-network principle is incorporated in the synthesis of hydrogel-based strain sensors with high toughness.[64] Second, PAAm polymer chains have many amide groups (-$CONH_2$), which are prone to form H-bonds with other polymers or nanomaterials containing carboxylic groups (-COOH) or hydroxyl groups (-OH). It is well known that many nanomaterials, such as carbon nanotubes, are often modified with carboxylic groups on the surface by the mixed acid treatment and the diazonium reaction, to improve the water solubility and their miscibility with polymers.[65] Third, PAAm polymers can be post-treated with alkali (such as hydrolysis) so that the functional groups of amides can be further changed to carboxylate groups (−COONa) after post-treatment. With post-treatment, the hydrogels swelled substantially to contain more water (>98%) and the response time to strain changes was significantly improved as the mechanical properties change from viscoelastic to elastic.[17] Finally, PAAm could form various types of copolymers (e.g., PAAm-co-PAAc) through free radical polymerization, providing a variety of polymer matrices for hydrogel-based strain sensors.

5.2 PAAc-based polymer matrix

PAAc is another major type of polymer matrix used in hydrogels for strain sensors. The numerous carboxylic groups on the polymer chains promote the ability of PAAc to swell extensively in water.[66] The swelling behavior can be modified by solution conditions (such as ion concentration), which provide an opportunity to build various polymer matrices with controllable conductivity, stretchability, and stimuli-responsiveness. First, the carboxyl groups of PAAc can form abundant intramolecular/intermolecular H-bonds, which is essential for the design of self-healing hydrogels.[67] In particular, PAAc is often selected for carbon-based conductive hydrogels. For example, Zheng *et al.* prepared a hydrogel comprised of oxidized cellulose nanofibers-graphene nanocomposites and PAAc. The resulting hydrogels demonstrated a stretchability (850%), mechanical strength (compression strength of 2.54 MPa, tensile strength of 0.32 MPa), electrical conductivity (0.25 S cm^{-1}) and healing efficiency of 96.7 % within 12 h.[68] The good self-healing capability and high stretchability of the hydrogel were ascribed to the formation of H-bonded networks between PAAc chains and graphene nanosheets.

In addition to H-bonds, the self-healing PAAc hydrogels can also be formed by the physical cross-linking between the carboxyl groups and metal ions.[69] For example, carboxyl groups of PAAc can form coordination interaction with Fe^{3+}, which improve the mechanical and electrical properties of the prepared hydrogels.[70] The reversible ionic coordination interactions between carboxyl groups of the PAAc and metal ions, significantly contribute to the self-healing property.

5.3 PVA-based polymer matrix

PVA is a water-soluble linear polymer (molecular weight: 20,000 to 400,000 g/mol) produced by the hydrolysis of poly (viny acetate), which is not synthesized by polymerization of its structural monomer (i.e., vinyl alcohol).[71] PVA polymers are often used to prepare hydrogel-based strain sensors due to the low toxicity, good biocompatibility, and relatively low cost. PVA hydrogels can be synthesized either by physical crosslinking of H-bonds or chemical crosslinking. For physical crosslinking, H-bonds lead to the entanglement of PVA polymer chains and eventual formation of PVA hydrogels.[72] For chemical crosslinking, the chemically active part reacts with the hydroxyl groups of the PVA chains, leading to entanglement between the PVA chains.[73] Compared with the aforementioned polymer matrices, PVA has relatively poor water retention and low ionic conductivity. This is mainly because the hydroxyl groups on the PVA chains do not have a strong ability to attract water molecules, and the long carbon chains weaken the interaction between the polymers and the water molecules.[52]

PVA is often selected for the conductive polymer-based hydrogels, because PVA could form robust structure through the hydroxyl groups with the boronic acid groups of conductive polymers, such as PANI.[74] In the case of PEDOT:PSS polymers, PVA can also form robust polymer network with them to improve the mechanical properties and stretchability of PEDOT:PSS without compromising the electrical properties.[53] Especially, H-bonds among PVA chains and chain entanglement among long PVA and/or PSS chains contribute to the mechanical and electrical properties. In addition to conductive polymers, carbon-based nanomaterials, can also be introduced into the PVA matrix. Liao *et al.* reported a nanocomposite hydrogel comprised of single-wall carbon nanotube, PVA, and polydopamine. The prepared hydrogels are non-cytotoxic and can promote cell adhesion and proliferation, indicating the potential application of wearable soft strain sensors with reliable biocompatibility.[75]

Furthermore, the cross-linked polymer networks can also be formed by the interaction of hydroxyl groups of PVA with other functional groups, such as tetrafunctional borate ion, to reversibly form borate-diol ester bonds, promising PVA a candidate for fabrication of self-healing conductive hydrogels.[76]

5.4 PNIPAAm-based polymer matrix

PNIPAAm is a type of thermoresponisve polymer, with excellent biocompatibility and thermoresponisve switching between hydrophilicity and hydrophobicity. They can undergo a reversible volume phase transition around the physiological temperature, and thus confer thermal responsiveness to the strain sensors. For example, Feng *et al.* prepared functional hydrogel-based flexible sensors by integrating N-isopropylacrylamide (NIPAAm) into conductive dual-network hydrogel based on PVA-graphene oxide and PAA-Fe^{3+}, and tuned the temperature sensing point to near 37 °C.[77] In addition to conductive hydrogels, PNIPAAm or their copolymers has also been widely used in the synthesis of photonic hydrogels for visualize mechanical strains.[20]

5.5 PSBMA-based polymer matrix

Poly (2-(methacryloyloxy)ethyl dimethyl-(3-sulfopropyl)ammonium hydroxide) (PSBMA) is a commonly used zwitterionic polymers, having sulfonic acid groups as the negative groups and ammonium cation groups as the positive groups on the polymer chains. The positive and negative charges in zwitterionic polymers give the hydrogels a high dipole moment. Such polymer structures contribute to the self-adhesive ability of the zwitterionic hydrogels owing to the dipole-dipole moment between the hydrogels and the substrates, which is an important requirement for wearable strain sensors.[38,78]

6. Mechanical properties of the nanocomposite hydrogels for strain sensing

As strain-sensing materials, mechanical properties (e.g., fracture strength, toughness, viscoelasticity, hysteresis, and fatigue resistance) are important for their ability to detect strains. The mechanical properties of hydrogels significantly affect their strain-sensing capabilities, including sensitivity, accuracy, reversibility, and durability.

It generally requires hydrogel-based materials with high strength and toughness, that can withstand sufficient stretching or compression without breaking easily. Simultaneously, hydrogel-based materials are required to respond immediately or in real-time to strain changes. The response speed is closely related to the time-dependent mechanical performance of hydrogels, which are typically viscoelastic. The rearrangement and reversible crosslinking of the polymer chains contribute to the viscoelasticity, and the relaxation time is related to the shear viscosity and shear modulus of the gels. Covalent cross-linking in polymer networks leads to the formation of an elastic network, whereas physical bonds among the polymer chains contribute to viscoelastic properties. Hydrogels containing dynamic physical bonds often have long relaxation times owing to their high shear viscosity. Thus, in order to achieve a high-speed response, including more covalent cross-linking with high water content and less physical bonding/interactions may play a role. The fabrication of an elastic hydrogel with high water content by chemical crosslinking is not difficult to achieve. However, the challenge is to incorporate photonic crystal structures or conductive components in hydrogels while maintaining the elastic nature of the composites.

Covalently cross-linked high-water-content hydrogels primarily behave as elastic solids and exhibit fast response to mechanical deformation. However, unlike physical bonds, which can exhibit self-healing properties, covalent bonds are easily broken without self-recovery, which may result in hydrogel-based strain sensors with low durability. The long-term durability of hydrogel-based sensors under continuous strain or stress loading is crucial. One way to estimate the long-term durability of hydrogel-based sensors is to detect optical or electrical drift over a long time under a constant strain. Another commonly used method is to observe the reversible sensing ability over 10,000 repetitive cycles of stretching/releasing or compression/relaxation cycles. If there is an optical shift or increase in the resistance after repeated cycles, irreversible cracks may form in the hydrogels. To avoid this, modifications of mechanical properties, such as self-healing or self-recovery properties, including effective energy dissipation domains and fabrication of hydrogels with high fatigue resistance through physical bonds between the nanostructures and polymer networks, need to be considered.

7. Summary and Perspectives

With the development of nanomaterials and advanced synthetic methods, hydrogels have developed significantly in terms of mechanical properties, self-healing, stimulus sensitivity, adhesiveness, biocompatibility, and 3D printability. For example, conventional hydrogels such as polyelectrolyte hydrogels usually have weak mechanical strength and are prone to permanent breakage. This inherent mechanical weakness makes hydrogels unsuitable for strain sensors that need to undergo repeated mechanical stretching or compression. Strategies such as polyampholytes, double networks, interpenetrating polymer networks, and various physical bonding (H-bonds) techniques can be applied to improve the mechanical properties of hydrogels.

The development of hydrogel strain sensors faces several challenges: (1) Stretchability and sensitivity. High stretchability requires a hydrogel to maintain structural integrity under large strains, while high sensitivity requires large structural changes at small strains.[79] (2) Maintaining the long-term stability of the hydrogels under working conditions. For example, one challenge is the development of non-water-evaporable hydrogels. The environmental stability of hydrogels is important for their daily use as strain-sensors. Unlike traditional elastomer-based materials, hydrogels that contain a large amount of water easily lose water in open environments. In this case, hydrogel-based sensors lose their functionality, which makes them unsuitable for practical applications. Thus, a number of organohydrogels have been developed using a facile solvent displacement, which exhibit anti-drying property.[80] Although there are various strategies, such as solvent replacement, surface modification, or packaging, long-term stable hydrogels that can be repeatedly used in daily life is highly required. (3) Can nanocomposite hydrogels avoid causing damage or inflammation to biological tissues when in conformal contact with them? Hydrogels themselves appear to be nontoxic and biocompatible for skin sensors, blood pressure sensors, or implanted devices. However, doped materials such as carbon nanotubes have been reported to be toxic in various cell lines and animal models.[81]

Although some excellent reviews on conductive hydrogels as strain sensors exist in the literature,[82,83] there is still no comprehensive minireview on nanocomposite hydrogels as strain sensors from the perspective of strain-optical and strain-electrical conversions.

The future of strain sensors based on hydrogels mainly focuses on the novel design and efficient utilization of optical and electrical signals, as well as the balance between optical/electrical and mechanical properties, functionality, and stability. Simultaneously, for current practical applications, there is an urgent need to develop photonic or conductive hydrogels with multiple shapes for flexible optical and bioelectronic devices. The combination of 3D/4D printing methods with photonic or conductive hydrogels at different scales to fabricate various designed structures will further facilitate their applications in flexible optical/electrical sensors or devices.

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Conflicts of interest

There are no conflicts of interest to declare.

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