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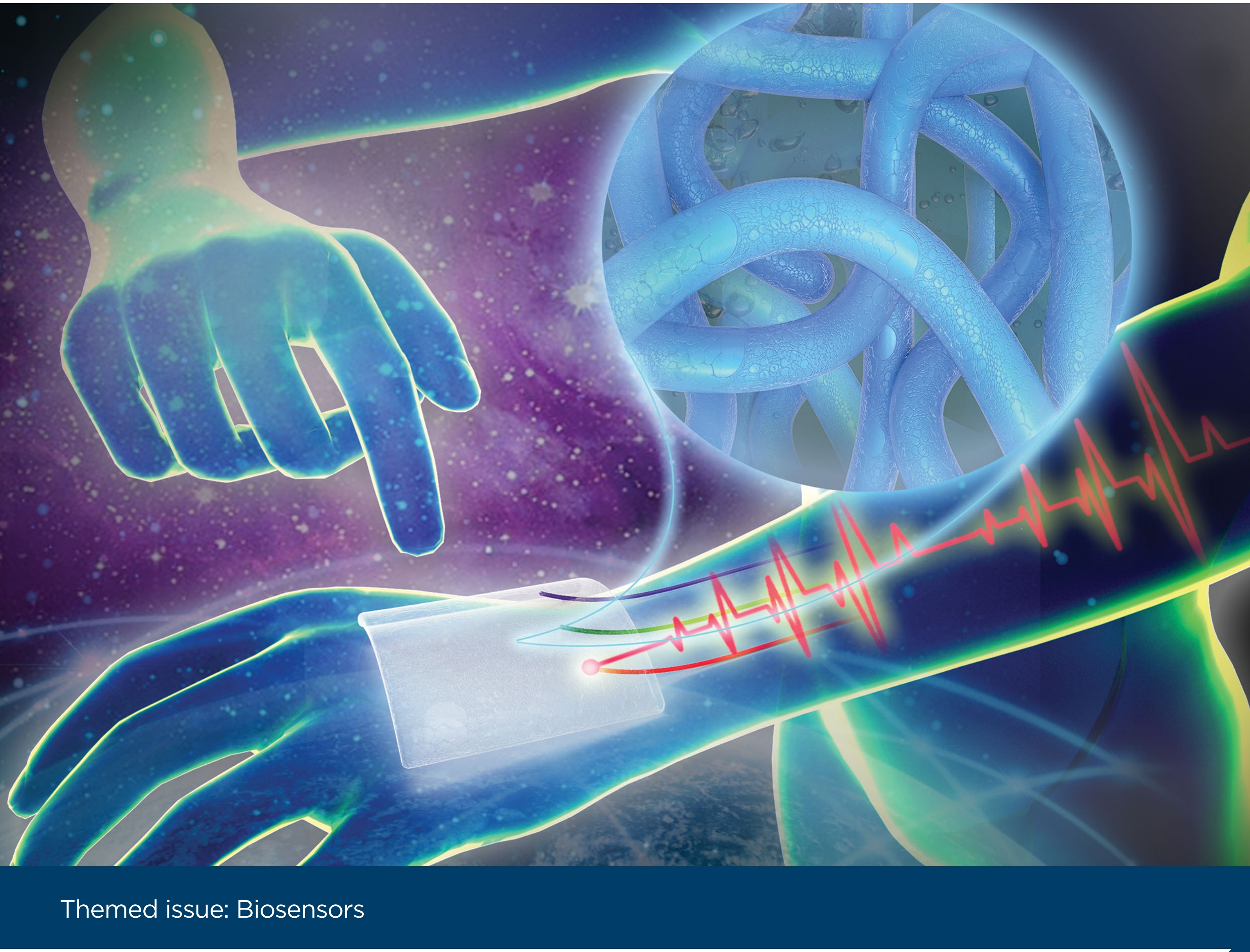

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From design to applications of stimuli-responsive hydrogel strain sensors

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Stimuli-responsive hydrogel strain sensors that synergize the advantages of both soft-wet hydrogels and smart functional materials have attracted rapidly increasing interest for exploring the opportunities from material design principles to emerging applications in electronic skins, health monitors, and human–machine interfaces. Stimuli-responsive hydrogel strain sensors possess smart and on-demand ability to specifically recognize various external stimuli and convert them into strain-induced mechanical, thermal, optical, and electrical signals. This review presents an up-to-date summary over the past five years on hydrogel strain sensors from different aspects, including material designs, gelation/fabrication methods, stimuli-responsive principles, and sensing performance. Hydrogel strain sensors are classified into five major categories based on the nature of the stimuli, and representative examples from each category are carefully selected and discussed in terms of structures, response mechanisms, and potential medical applications. Finally, current challenges and future perspectives of hydrogel strain sensors are tentatively proposed to stimulate more and better research in this emerging field.

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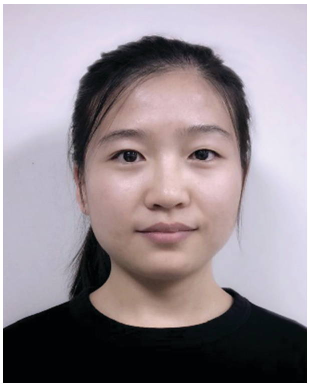
1. Introduction

Hydrogels, as classical soft-wet materials containing high water content in flexible 3D networks possess many structural and functional advantages such as hierarchical porous structures, viscoelasticity, transparency, stretchability, and biocompatibility. Particularly, recent studies have pushed the potential applications and development of hydrogels from the traditional fields of cell/tissue scaffolds,[1–4] drug/gene delivery carriers,[5–7] wound dressings,[8–11] and contact lenses[12–14] to the emerging



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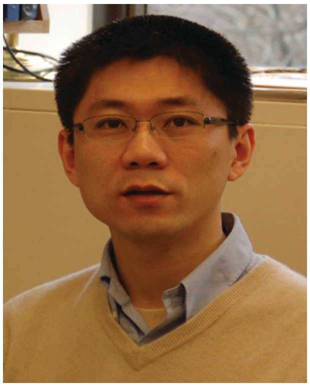

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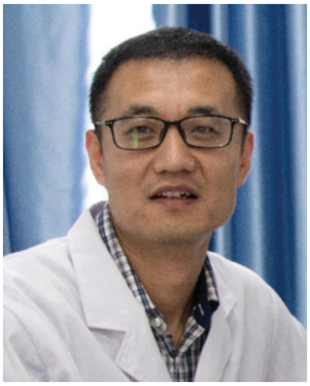
fields of wearable devices,[15–17] electronic skins,[18,19] soft robotics,[20–22] and artificial intelligence sensors.[23] Among them, stimuli-responsive hydrogel strain sensors have attracted significant and continuous interest to be developed as a promising platform for disease diagnosis, health monitoring, damage/motion detection, water/food safety, and environmental monitoring[24–28] because of their intelligent and programmable features capable of changing their shape/size/volume and probably other functional properties (*e.g.* conductivity, permeability, viscosity, and mechanics) in response to different stimuli. Different from dry sensors made of elastomers, polymer/inorganic composites, and carbon-based materials without water, the presence of a high water content enables hydrogel strain sensors to exhibit intrinsic swelling-induced viscoelastic, mechanical, self-recovery, self-healing properties,[29–36] rendering them great potential to realize a variety of sensing actuations under different stimuli.[37] On the other hand, due to the high hydration nature of hydrogel sensors, all their stimuli-responsive polymers, crosslinkers, and incorporated entities need to be compatible with and workable under wet and aqueous conditions. Another major difference is that the mismatched mechanics among polymer networks, additives, and crosslinkers with different swelling extents at polymer/water interfaces will create asymmetric forces that drive shape and volume changes of hydrogels even under subtle stimuli.

From a materials design viewpoint, all stimuli-responsive hydrogels can be potentially fabricated into hydrogel-based sensors. Several general design strategies have been proposed and demonstrated for the successful development of a wide variety of hydrogel sensors. Firstly, the most commonly used design is to directly fabricate stimuli-responsive materials into hydrogels, which are sensitive to external stimuli of temperature, pH, salt, light, and electric field. Several typical stimuli-responsive hydrogels include thermo-responsive poly(*N*-isopropylacrylamide) (polyNIPAM) and poly(vinyl methyl ether) (polyVME) hydrogels, pH-responsive carboxylated poly(2-hydroxyethyl methacrylate–ethylene glycol dimethacrylate) (poly(HEMA–EDMA)) hydrogels,[38] electronic-responsive conducting polymer-based (CPs) hydrogels,[39]



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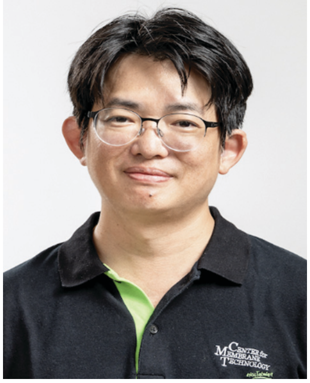

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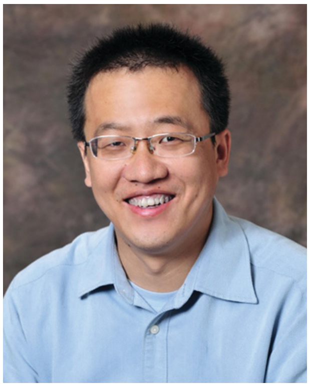

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light/force-sensitive poly(acrylamide (AAm)-*co*-methyl acrylate (MA))/spiropyran (SP) hydrogels,[40,41] and salt-responsive poly([2-(methacryloyloxy)ethyl]-trimethylammonium chloride)-[*N*-(2-hydroxyethyl) acrylamide]/poly(3-(1-(4-vinylbenzyl)-1*H*-benzo[*d*]imidazol-3-ium-3-yl)-propane-1-sulfonate) (poly(METAC-HEAA)/polyVBIPS) and polyNIPAM/polyVBIPS hydrogels.[42,43] Advances in chemical synthesis and gelation methods also allow the copolymerization or sequential polymerization of different stimuli-responsive monomers and crosslinkers to achieve dual or multiple stimuli-responsive properties embedded in hydrogel sensors, including polyAAm/poly(vinylalcohol)(PVA) humidity sensor,[44] poly(β-cyclodextrin (β-CD)-methacrylic acid (AAc)) hydrogels,[45] and Eu^{3+}/Tb^{3+}–polyNIPAM hydrogel.[46] In general, since hydrogels are typically isotropic materials, the use of pure polymer materials or simple polymer structures is unlikely to selectively trigger site-specific swelling or contraction, leading to shape adaptability and transformation in a controllable manner. To overcome this issue, another common design strategy is to incorporate stimuli-responsive entities (*i.e.* nanoparticles, nanoclay, nano-crystalline cellulose, and functional polymer chains) into hydrogel networks to create in-homogeneous structures, which in turn help to readily generate mismatch stress for a strain sensing under the desired stimuli. These stimuli-responsive entities can be either physically added to or chemically crosslinked with hydrogel networks, both of which require a controlled orientation and spatial distribution to achieve well-defined network architectures and to maximize the sensibility to external stimuli. However, the additional difficulty of snug-filling small entities into the porous structures of hydrogels needs to be addressed for preventing dead zones upon deformation.

From a structural design viewpoint, constructing hybrid network structures in hydrogel sensors, such as double-network structure, bilayer/multi-layer structure, interpenetrating structure, nanofilled structure, gradient structure, and other heterostructures, also allows the creation of mismatch stress in the polymer networks (*e.g.* softer *vs.* stiffer, swelling *vs.* non/less-swelling, and hydrophobic *vs.* hydrophilic) for achieving different sensing actuations in response to various external stimuli. Moreover, the network structure of hydrogel sensors can be cross-linked using either irreversible or reversible crosslinkers/additives.[47,48] The incorporation of single or multiple reversible crosslinkers/additives, in most cases, enable different components or regions of hydrogels to undergo different extents of volume expansion/contraction in certain positions, directions, and even swelling rates, thus leading to programmable shape changes by bending, expanding, contracting, twisting, and fast buckling.

In all these designs, searching for new responsive sensing materials/components and constructing well-defined network structures are equally important for the rational design of highly sensitive and robust hydrogel sensors. However, although this sounds simple and straightforward, the appropriate integration of sensing materials and entities into hydrogel networks still remains a challenge mainly due to the incompatibility between materials, structures, and gelation methods. Moreover, on one hand, many hydrogel strain sensors exhibit a certain degree of reversible and multiple sensing ability due to their intrinsic stimuli-responsive nature; on the other hand, since the exposure of hydrogels to external stimuli often damages their irreversible bonds and crosslinkers, hydrogel sensors also suffer from a loss in their original sensing ability. Thus, the introduction of self-healing functions in hydrogel sensors becomes necessary to rapidly recover their original network structure and to sustain their sensing ability through the dynamic reformation of reversible noncovalent interactions (*e.g.* hydrogen bonds, π–π stacking, hydrophobic interactions, host–guest interactions, and metal–coordination interactions) and/or reversible covalent bonds (*e.g.* imine bonds, disulfide bonds, acylhydrazone bonds, and boronate ester bonds).[49–51] Again, the cooperative and compatible fabrication of both self-healing and stimuli-responsive materials/entities in the same hydrogel may also face additional challenges. Similarly, hydrogel strain sensors must exhibit sensing-appropriate physical, mechanical, and biological properties. Generally, hydrogel strain sensors usually require high mechanical strength, toughness, and stretchability for transducing mechanical deformation into electrical/chemical/optical signals.[52–55] Specifically, *in vivo* injectable hydrogel strain sensors must be biodegradable and biocompatible, and their degraded products also need to be biocompatible with minimal inflammatory responses.

All of these challenges from both materials and structural viewpoints highlight the importance of fabrication methods. Hydrogel stain sensors can be rationally designed and fabricated from a large dataset of stimuli-responsive monomers and crosslinkers *via* different polymerization methods, such as living cationic and anionic polymerization, reversible addition–fragmentation chain-transfer (RAFT) polymerization, atom-transfer radical polymerization (ATRP), self-assembly, microfabrication, and laser writing, all of which can well control the chemistry and structure in hydrogel networks.[56–61] In some cases, the polymer networks in hydrogel sensors can be readily functionalized with different stimuli-responsive pendent groups and additives even before gelation, and the resulting hydrogels can be directly used for sensing applications without the need for additional substrates and matrixes to serve as sensor platform.

As shown in Table 1 and Fig. 1, we summarized and classified some classical hydrogel sensors with different types of sensing ability. This review aims to highlight the most important and recent works on hydrogel sensors in the past five years, instead of a comprehensive review. Specifically, this review mainly covers the design principles and synthesis strategies of hydrogel sensors from both materials and structural viewpoints, selectively highlights some classical and interesting hydrogel sensors for different sensing purposes and applications, and finally presents some of the current scientific/technological barriers and the future research directions that should be undertaken to overcome these barriers.

2. General design strategy and categories of hydrogel strain sensors

To better understand the structure–function relationship of hydrogel strain sensors, here we classified hydrogel strain sensors into five basic groups of nanoparticle-incorporated, conducting polymer-incorporated, polyelectrolyte-incorporated,

Table 1 Summary and classification of the different types of hydrogel strain sensors

Classification	Hydrogel	Signal	GF or LoD	Stretchability (%)	Other	Ref.
Conductive polymer-incorporated	PEDOT:PSS–polyAAm array	Electric	GF = ∼20	∼15	Long-term sensing (∼2 months), detect subtle force of 0.02–4.49 N	62
	PANI–poly(AAm-*co*-HEMA)	Electric	GF = 11	∼300	Detect wrist bending, speaking; biocompatible	39
	PANI/PSS–UPyMA	Electric	GF = 3.4	∼300	Self-healing (<30 s); detect pulse beating, speaking, finger bending	63
	polyNIPAAm/PANI	Electric	GF = 3.92	∼200	Stable performance (∼350 cycles)	64
Polyelectrolyte-incorporated	ACC–polyAAc/sodium alginate (SA)	Capacitance	LoD = ∼1 kPa	∼55	Self-healing; detect speaking, blood pressure, and finger bending	65
	Poly(MAA-*co*-DMAPS)	Electric	LoD = 0.6–0.7% °C^{-1}	>10 000	Self-healing; Temperature-responsive sensor (10–80 °C)	66
Nanoparticles-incorporated	Chitosan–AuNPs	Color	LoD = 2.4 μM of H$_2$O$_2$	—	Wide detection range (8.0 μM–15 mM)	67
	PNIPAM–AuNPs	Color	LoD = 0.2 °C	>100	Color-temperature sensor range (25–40 °C); detecting time (∼1 s)	68
	Fe$_3$O$_4$–poly(AAc-*co*-AAm)	Magnetic	LoD = 0.1 pH unit	—	Repeatable, reversible detection; detecting time (<60 min)	69
	Platinum nanoparticles (PtNPs)–PANI array	Electric	LoD = ∼0.2 mM of glucose	—	High selectivity; signal-to-noise ratio of ∼3	70
	PtNPs–PANI	Electric	LoD = ∼0.7 μM of glucose	—	High selectivity; detecting time (<3 s)	71
	AgNWs–polyAAm	Electric	GF = 0.71	>22 000	Wide working condition (−20 to 80 °C); detect breathing, speaking, and finger bending	72
	Nano barium ferrite (BaFe$_{12}$O$_{19}$)–polyAAc	Electric	—	>40	High ionic conductivity (1.22 × 10^{-2} S cm^{-1}); good self-recoverability	73
	PDA–CNTs–poly(AAc-AAm)	Electric	—	∼700	Self-adhesive; wide working condition (−20 to 60 °C); detect arm bending and wrist pulse	74
	MXene (Ti$_3$AlC$_2$)–polyvinyl alcohol (PVA)	Electric	GF = 25	>3400	Self-healing; self-adhesive; detect human speaking, writing, and smiling	75
	PC/rGO–PVA	Electric	GF = 14.14	>5000	Self-healing (∼3 s); detect finger bending, smiling, speaking, and wrist pulse	76
	F-SWCNT–polyPDA–PVA	Electric	—	>100	Self-healing; self-adhesive; detect bending and relaxing of walking, chewing, and pulse	77
	SWCNT/PVA	Electric	GF = 1.51	>1000	Self-healing; detect bending and relaxing of knee/finger (>1000 cycles)	78
Inorganic electrolyte-incorporated	TA/sodium alginate (SA)–polyAAm	Electric	GF = 2.0	>2100	Self-healing; detect smiling, finger bending, and wrist pulse	79
	KCl–(κ-carrageenan)/polyAAm	Electric	GF = 0.63	∼1000	Self-healing; detect finger bending	80
	Cu^{2+}–polyAAm	Electric	LoD = ∼0.005% strain	>1500	Detect finger bending, speaking, and wrist pulse	81
	NaCl–gelatin/PVA	Electric	GF = ∼0.4	∼715	Lower working condition (−20 °C); detect finger and elbow bending, speaking	82
	LiCl–poly(acrylated thymine–AAm)	Electric	—	∼2000	Self-healing; self-adhesive; detect breathing, speaking and finger bending	83
	NaCl–polyAAm	Capacitance	—	>590	Detect finger bending (>4000 cycles) and location of touch	84
	LiCl–polyAAm	Capacitance	—	>1000	Detect location of touch (work as smart screen)	85
	NaCl–SA/polyAAm	Electric	GF = 2.0	∼3120	Detect speaking, finger bending, and wrist pulse	86
	NaCl/SDS-regenerated silk fibroin(RSF)/HPAAm	Electric	GF = 2.0	>1900	Detect finger bending and location of touch	87
	LiCl–polyAAm	Capacitance	GF = 4.0	>500	Adhesive; stable performance (over 1500 cycles)	88
	PIL–BF$_4$/PEDGA	Electric	—	>1400	Wide working condition (−75 °C to 340 °C); detect finger bending (>10 000 fatigue cycles)	89
Biomolecule-incorporated	PVA/RSF/borax	Electric	—	∼5000	Track leg, knee bending and different gestures	90
	Poly(AAm-*co*-aptamer)	Color	LoD = 0.0128HAU of H5N1	—	<30 min detection time	91
	DNA-based	Cell	—	—	Detect live circulating tumor cells	92

Note: Gauge Factor (GF) and Limit of Detection (LoD).

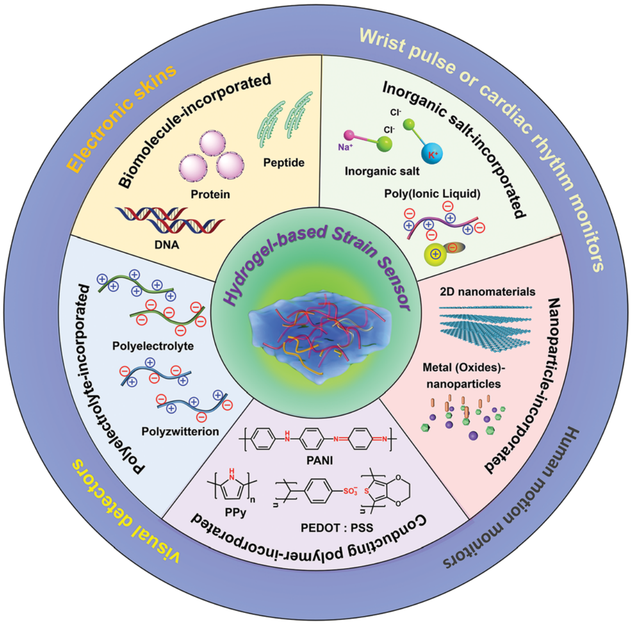

Fig. 1 Overview of the five typical types of hydrogel strain sensors.

inorganic salt-incorporated hydrogel, and biomolecule-incorporated hydrogel strain sensors, mainly depending on their structural-dependent network structures, gelation methods, and sensing mechanisms, not depending on the nature of the stimuli. Since these five types of hydrogel strain sensors possess anisotropic structures, a general working principle is to create non-uniform spatial stress in different regions/parts of the hydrogels to realize different stimuli-responsive sensing properties *via* strain-induced shape-adaptive actuations. On the other hand, while these five types of hydrogel strain sensors can achieve similar or the same stimuli-responsive sensing capacity (*i.e.* thermo, light, electric, and pH sensing capacity),[93] they are driven by different sensing mechanisms strongly depending on their network structures (Fig. 1). Inorganic nanofiller-incorporated hydrogel sensors are constructed by adding nanofillers or crosslinkers in polymer networks to create hybrid polymer/inorganic structures, where both materials contribute unique and beneficial properties to the hydrogel sensor, while the latter hydrogel sensors are likely constructed by different polymers to create an overall heterogeneous polymer/polymer structure. All these network structures allow the creation and maximizing the stress mismatch for strain-induced actuation upon external stimuli, and the sensing mechanics of these hydrogel sensors can further be tuned and optimized by changing the compositions in their hydrogel matrix, tailoring them for specific applications.

2.1. Nanoparticle-incorporated hydrogel sensors

Due to the unique and superior optical, conductive, and magnetic properties of nanoparticles (NPs), the physical incorporation or chemical crosslinking of nanoparticles into the polymer network enables the construction of hybrid inorganic NPs/polymer network structures, which help to translate the conformational change of the surface bound stimuli-responsive polymers into observable optical property changes and improve their mechanical property, making them promising materials for desirable sensing applications.[72,73,76,94] Several of the widely used one/two/three-dimensional NPs of gold nanoparticles (AuNPs), gold nanorods (AuNRs), silver nanoparticle (AgNPs), carbon nanotubes, graphene oxide, and MXenes, and magnetic nanoparticles (Fe_3O_4) have been incorporated into polyAAm, polyNIPAM, PVA, poly(NIPAM-*co*-AAm), and poly(AAc-*co*-AAm) to produce several representative strain sensors for pH, temperature, color and human movement.[95] As a typical example, AuNPs are often incorporated into a hydrogel matrix containing –SH, –CN and $-NH_2$ groups *via* the formation of dynamic covalent bonds, which are responsive to different stimuli in different strain sensors. AuNPs were readily self-assembled on the amine-rich chitosan hydrogel matrix through horseradish peroxidase (HRP) immobilization, producing s AuNP-incorporated hydrogel biosensor,[67] which showed a wide range of response to H_2O_2 (8.0–15 μM), a low detection limit of 2.4 μM, and long-term sensing stability of up to 4 weeks. Moreover, AuNPs can also be modified by plasmonic nanostructures with different plasmonic materials, cluster structures, and geometries, and the modified AuNPs are used as real-time colorimetric indicators for hydrogel color sensors. As shown in Fig. 2a, combining plasmonic AuNPs with thermo-responsive polyNIPAM hydrogels, a smart colorimetric stretchable hydrogel biosensor was able to achieve rapid, reversible, and strain-insensitive color shifts between red and grayish violet in response to a temperature change from 25–40 °C within 1 s.[68] Besides the conventional AuNPs, AgNPs, and plasmonic nanoparticles, the incorporation of magnetic Fe_3O_4 nanoparticles into poly(AAc-*co*-AAm) hydrogels enabled the production of a pH sensor, which displayed a repeatable and reversible response to pH change, with a sufficient sensitivity to detect a 0.1 unit change in pH (Fig. 2b).[69]

Carbon nanotubes (CNTs) and reduced graphene oxide (rGO) as typical two-dimensional carbon materials have been widely integrated and used as different electronic sensing devices made of elastomers, polymers, and organics due to their superior electronic properties of Fermi velocity, thermal conductivity, charge carrier mobility, and mechanical strength.[96–99] However, the strong hydrophobicity and poor solubility of CNTs and rGO make it very challenging to directly incorporate them into highly hydrophilic hydrogels (70–90% water content) and to avoid their aggregation in the hydrogel matrix while still retaining their electronic sensing functions. To overcome this issue, a common strategy is to modify these hydrophobic carbon materials with hydrophilic/charge phenol hydroxyl, carboxylic acid, epoxide, and oxide groups to not only improve the dispersion of CNTs and strengthen their interfacial interactions with the polymer network, but also enhance their mechanical and electrical properties *via* electrostatic and π–π interactions, and hydrogen bonding. Consequently, different modified CNTs have been incorporated into polyAAm, polyAAc, polysaccharides, gelatin, collagen, and polyethylene glycol (PEG) hydrogels *via* hydrothermal and solvothermal reactions, cross-linking gelation, doping methods, and template-based synthesis. Among the different CNTs-incorporated hydrogel strain sensors, flexible and wearable strain sensors have

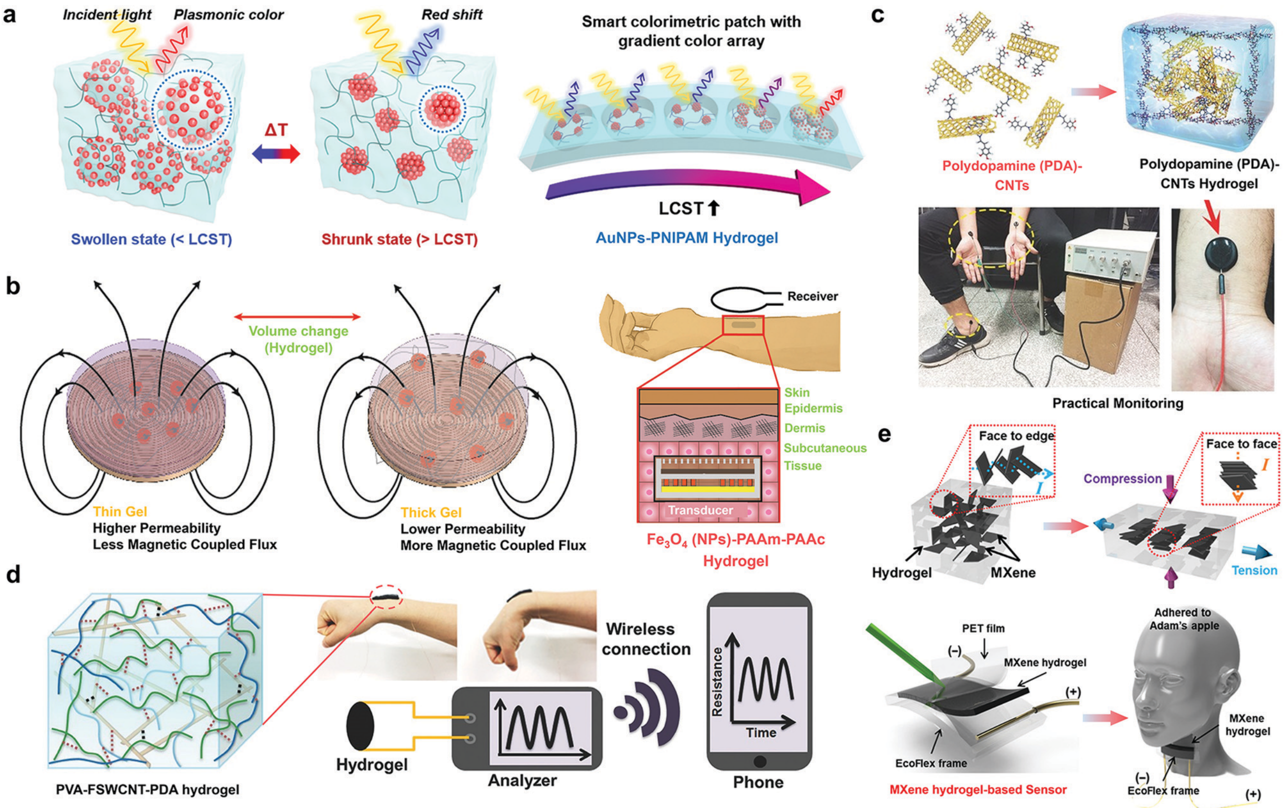

Fig. 2 Nanoparticle/polymer hydrogel sensors. (a) Thermo-responsive colorimetric plasmonic AuNPs–PNIPAM hydrogel sensors, exhibiting rapid and reversible color shifts from red to violet in response to a temperature change (25–40 °C) within 1 s. [Adapted with permission from ref. 68, Copyright 2018 Nature Publishing Group]. (b) Wireless iron oxide nanoparticle (Fe_3O_4NP)-embedded poly(AAc-*co*-AAm) hydrogel sensors that can detect subtle pH changes (0.1 unit) in response to a magnetic field. [Adapted with permission from ref. 69, Copyright 2014 Elsevier]. (c) Mussel-inspired PDA–CNTs-incorporated poly(AAm–AAc) hydrogels with highly adhesive, tough, and conductive properties for detecting human wrist pulse even at −20 °C. [Adapted with permission from ref. 74, Copyright 2018 Wiley-VCH]. (d) Automatous healable and adhesive PVA–FSWCNT–PDA hydrogel strain sensor for the real-time detection of human motions of finger bending and relaxing, walking, chewing, and pulse with fast self-healing ability (within 2 s) in response to external changes of stress or strain.[77] (e) MXene–PVA hydrogel sensors for monitoring electromechanical signal changes with a gauge factor of 25. [Adapted with permission from ref. 75, Copyright 2018 American Association for the Advancement of Science].

become immensely important for ultrasensitive human–machine interaction, healthcare monitoring, and human motions. As a classical example, conductive functionalized single-wall carbon nanotubes (FSWCNT), biocompatible polyvinyl alcohol (PVA), and polydopamine (PDA) were crosslinked to form a healable and adhesive PVA–FSWCNT–PDA hydrogel with dynamic supramolecular structures, which exhibited fast self-healing ability (within 2 s), high self-healing efficiency (99%), and robust surface adhesion on different substrates (Fig. 2c). The PVA–FSWCNT–PDA hydrogel was further fabricated into a wearable soft strain sensor for the real-time detection of different human motions of finger bending and relaxing, walking, chewing, and pulse (Fig. 2d).[77] To expand this idea, single wall carbon nanotubes (SWCNT), GO, and silver nanowires (AgNWs) were separately incorporated into PVA hydrogels to produce different types of NP/PVA hydrogels as conductive strain sensors, all of which achieved fast and high self-healing efficiency (98%) of electric signals within ∼3 s, sustained extreme elastic strain of 1000% with a gauge factor of 1.51, and effectively monitored and distinguished multifarious human motion of the finger, elbow, knee, and neck.[78] More impressively, MXene ($Ti_3C_2T_x$) as another 2D carbonitride/metal carbide was incorporated into the PVA hydrogel to obtain an MXene-based hydrogel strain sensor, which could detect various body motions (hand gestures and facial expressions) and monitor vital signals (human pulse and vocal sound) (Fig. 2e).[75] This high strain-induced electromechanical sensitivity is largely attributed to the 3D network structure of the MXene nanosheets embedded within the hydrogel matrix. Large deformation reduces the spacing between the nanosheets, but increases the surface contacts between the MXene nanosheets, and transforms the face-to-edge interconnections into face-to-face interconnections, all of which decrease the M-hydrogel resistance, and thus increase the detection sensitivity. It should be noted that besides the above-mentioned examples, most of nanoparticle/polymer hydrogel strain sensors can only be stretched to a strain of ∼200%, thus the breaking of the conducting contacts during deformation usually leads to a poor and irreversible detection sensitivity in these strain sensors. Thus, additional efforts should be made to increase the stretchability and self-healing properties of nanoparticle/polymer hydrogel strain sensors.

2.2. Conducting polymer-incorporated hydrogel strain sensors

Conducting polymers such as polypyrrole (PPy), polyaniline (PANI), and poly(3,4-ethylene-dioxythiophene) (PEDOT) are π-conjugated polymers with alternating single and double covalent bonds for conducting electrons.[100–102] The use of conducting polymers to make hydrogel strain sensors possesses several intrinsic advantages as follows: (1) conductive polymers themselves are able to carry solvent ions for electrical conduction with fairly low resistance in the hydrogel networks. (2) In contrast to rigid NPs and metallic additives in polymer hydrogels to empower electronic conductivity, conductive polymer chains are very flexible and compatible with other polymers in the same hydrogel systems for empowering their high stretchability. Less structural disparity in network length scales can reduce the source of inhomogeneity in mechanical and electrical properties. (3) Obviously, conductive polymers can be simply fabricated with other elastomeric and polymer substrates into electronic sensors, and accordingly the conjugated polymers inside hydrogel networks possess high conductivity and thermal stability for piezoresistive strain sensors.[64,103–105]

A general working mechanism for conducting hydrogel strain sensors is the delocalization of π-bonded electrons over the conjugated backbone structures. Thus, PANI, as firstly described in the mid-19th century by Henry Letheby, has been widely used for hydrogel bioelectronics.[106,107] As an interesting example, a triplex biosensor made of nano-patterned PANI hydrogels and different enzymes was able to detect triglycerides, lactate, and glucose, with good selectivity and high sensitivity in both PBS and human serum samples.[70] The hydrophilic porous microstructure of the PANI hydrogel was favorable for the transportation of electrons generated from hydrogen peroxide in the enzymatic reactions. To enhance the electronic conductivity of hydrogel strain sensors, another common design strategy is to integrate conducting polymers with various ionic or organic dopants and nanoparticles. The incorporation of such dopants enables not only the creation of free radicals that can pair with the dopant to form polarons, but also oxidizes (p-doping) or reduces (n-doping) the conducting polymer at the π-bond sites by removing or donating electrons, both of which offer additional paths for charge carriers along the conducting polymer backbone.[108] Following this working principle, the incorporation of platinum nanoparticles (PtNPs) into the PANI hydrogel allowed the construction of a glucose sensor, with an ultrahigh low detection limit of 0.7 μM glucose within 3 s (Fig. 3a).[71] The significant increase in glucose detection ability of PtNPs/PANI hydrogel sensors is attributed to their enhanced electrocatalytic activity for oxidizing H_2O_2. Similarly, polyAAm hydrogel doped with poly(3,4-ethylenedioxythiophene):polystyrene sulfonate (PEDOT:PSS) conductive polymers was used create a classical piezoresistive tactile sensor (Fig. 3b).[62] However, additional challenges still remain. Simple doping/mixing of additives into the conductive polymer network usually does not necessarily enhance mechanical strength, but instead make the hydrogels become more brittle due to the over-crosslinking effect.[63] To address this issue, the rational design of an interpenetrating network (IPN) between conducting polymers and other polymers is a possible solution to minimize the potential trade-offs between mechanical and electrical properties *via in situ* polymerization or in-growth polymerization. Using this design strategy, several conducting hydrogels with IPN have been developed using polymerizing conducting polymers (*e.g.*, PEDOT:PSS and PANI) with other polymers (*e.g.* polyAAm, polyAAc, and gellan gum). All of these conductive IPN hydrogel sensors displayed enhanced mechanical properties compared to conductive hydrogel sensors without IPN, while still retaining high sensing ability (*e.g.* gauge factor = 11) (Fig. 3c).[39] Moreover, considering that the double-network (DN) structure is a special case of IPN, several recent attempts have fabricated two different polymers to form conductive hydrogels with a DN structure for imparting both high electronic conductivity and mechanical toughness, including PEDOT:PSS/PDMAAm DN hydrogels[109] and polyAAc/PEDOT:PSS DN hydrogel.[110]

2.3. Polyelectrolyte-incorporated hydrogel strain sensors

Polyelectrolytes (*e.g.* poly(2-acrylamido-2-methylpropane-1-sulfonic acid) [poly(AMPS)], polyAAc, and polyzwitterions) are among the most studied macro-ionic molecular systems. Considering the ionic nature of polyelectrolytes, polyelectrolyte hydrogels copolymerizing these electrolyte monomers into hydrogel networks possess intrinsic ionic groups and counterions, and thus they commonly undergo volume shrinking or swelling in response to pH and ionic changes.[111–113] Compared to neutral polymer hydrogels, polyelectrolyte hydrogels usually exhibit "polyelectrolyte effect" behavior, *i.e.* the polyelectrolyte chains undergo fully stretching (swelling) conformation in water, but adopt a collapsed (shrinking) conformation in salt solution simply because the introduction of polyelectrolytes will screen the long-range electrostatic repulsions and short-range-excluded volume interactions between and within the polyelectrolyte chains, leading to ion/pH-responsive volume changes.[114,115] Moreover, the strong binding of counterions to the polyelectrolyte chains creates additional crosslinks in the hydrogel network, which influence the mechanical properties of polyelectrolyte hydrogels and even cause gel collapse in some cases. Moreover, the presence of charged groups allows the introduction of metal ion coordination and supramolecular interactions in the hydrogel networks, and thus to promote the hydroelectric separation and conductivity of the hydrogels.

Copolymerization of charged monomers [*e.g.* AMPS, AAc, 2-(methacryloyloxy)ethyl]dimethyl-(3-sulfopropyl) ammonium hydroxide (SBMA), 3-dimethyl (methacryloyloxyethyl) ammonium propanesulfonate (DMAPS) with neutral acrylamide monomers [*e.g.* AAm, NIPAM, HEMA][66,116–118] and the introduction of these charged monomers into the hydrogel network (*e.g.* PVA, P2VP, SA and polyAAm) to form IPN structures[119–123] are two common strategies to construct polyelectrolyte hydrogel sensors. The copolymerization strategy can achieve poly(SBMA-*co*-HEMA) hydrogel sensor polyelectrolyte hydrogels with a high fracture elongation of 2000–10 000%, fracture strength of 0.2–0.6 MPa, and gauge factor of more than 1.5 (Fig. 4a). Another zwitterionic poly(MAA-*co*-DMAPS) hydrogel sensor with UCST and LCST characteristics displayed ultrastretchability of up to 10 000%,

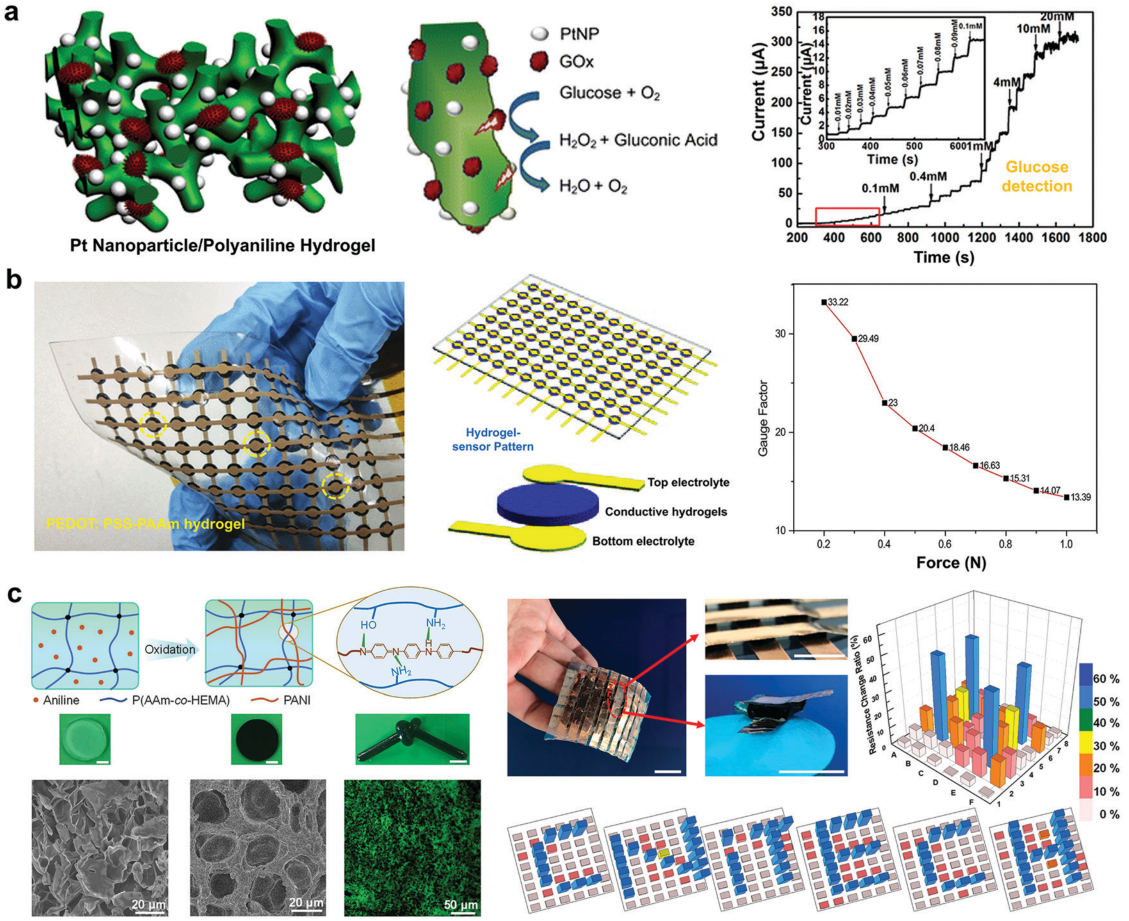

Fig. 3 Conductive polymer hydrogel strain sensors. (a) PtNPs/PANI hydrogels as a glucose sensor with high sensitivity of 96 μA mM^{-1} cm^{-2}, low detection limit of 0.7 μM, and fast response time of ~3 s. Such unprecedented sensitivity is largely attributed to the synergistic integration of a PtNP catalyst into the 3D-microstructured PANI hydrogel matrix for promoting the electro-oxidation of hydrogen peroxide. [Adapted with permission from ref. 71, Copyright 2013 American Chemical Society]. (b) PolyAAm/PEDOT:PSS piezoresistive hydrogel sensors with a high gauge factor of 13–33 in response to a subtle force of 0.2–1.0 N. The blending of PEDOT:PSS in PAM hydrogels allows the output impedance (kΩ) to be reduced to promote piezoresistive sensitivity. Adapted with permission from ref. 62, Copyright 2018 IEEE. (c) Conductive PANI–poly(AAm-*co*-HEMA) hydrogel sensors with high sensitivity (GF = 11) at low strain and outstanding linearity at high strain for detecting human motions of speaking, wrist pulse, and hand writing. [Adapted with permission from ref. 39, Copyright 2018 American Chemical Society].

which can converse forces to electrical signals for tracking human movements and perceiving changes in external temperature (Fig. 4b).[66] IPN structures can slightly enhance the tensile strength or sensitivity of hydrogel sensors by improving the integrity of the network structures. For instance, polyAAc/SA, polyAAc/PAAm, and polySBMA/PVA strain sensors exhibited 300–1000% stretchability, ~0.6 MPa tensile strength, and high detection capacity for multiple-time elbow/finger bending. Moreover, polyAAc/SA hydrogel sensors also exhibited high and sustainable sensitivity for detecting a wide range of pressure between 0.17–1000 kPa (Fig. 4c).[65]

2.4. Inorganic salt-incorporated hydrogel strain sensors

Different from polyelectrolyte hydrogel strain sensors, electrolyte hydrogels are typically prepared *via* the direct physical or chemical doping of sodium chloride (NaCl), lithium chloride (LiCl), and cupric ions (Cu^{2+}) into polyAAm, HPAAm, and PVA hydrogels. Direct physical doping of NaCl, LiCl, and Cu^{2+} salts into polyacrylamide (polyAAm) hydrogels provides a simple and straightforward route to form ion-doped electrolyte hydrogels.[84,85] The resultant NaCl/polyAAm, LiCl/polyAAm, and Cu^{2+}/polyAAm hydrogel strain sensors are not only highly stretchable by up to 15-times of their original length and stable to sustain over 1000 pressure-capacitance cycles, but also detect any subtle human motion such as smiling, speaking, and finger bending. Moreover, these electrolyte hydrogels can also be fabricated into epidermal touch panels for use as human skin-machine interfaces for writing words, and playing games and a piano. (Fig. 5a). To enhance the mechanical properties, doping of different salts in the DN network structure produces different hydrogel strain sensors, including (i) κ-carrageenan/polyAAm DN hydrogel doped with KCl to achieve great strain sensitivity with a gauge factor of 0.63 at the strain of 1000%, elastic modulus of 280 kPa, and a fracture energy of 6150 J m^{-2};[80] (ii) PVA/gelatin doped with NaCl to achieve fracture stress of 1 MPa, fracture strain of 715%, elastic modulus of 157 kPa, and toughness of 3605 kJ m^{-3}[82];

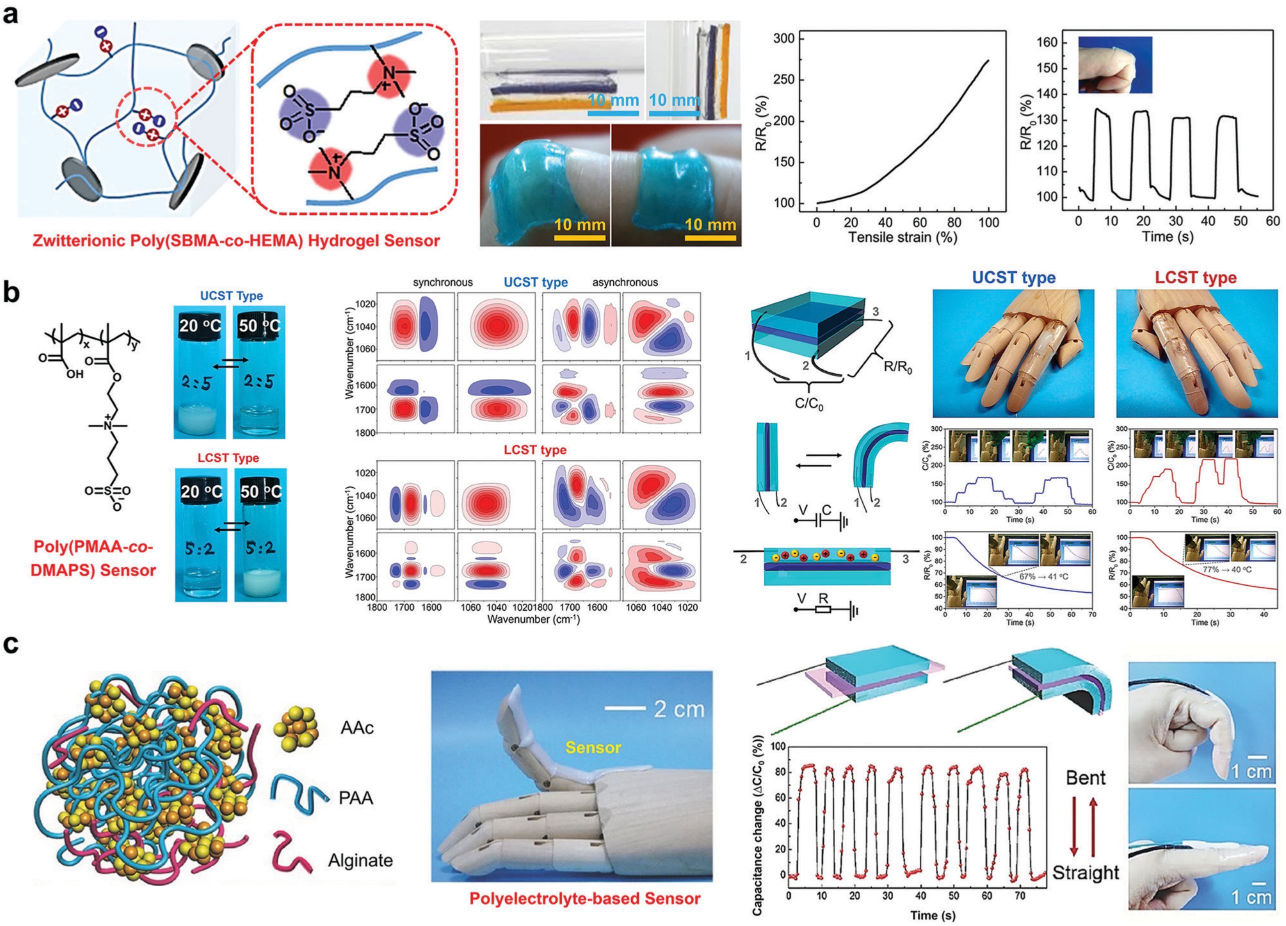

Fig. 4 Polyelectrolyte-incorporated hydrogel strain sensors. (a) Poly(SBMA-*co*-HEMA) hydrogel sensors with high strain conductivity and sensitivity (gauge factor = 1.8), high fracture toughness of 2.45 MJ m^{-3}, fracture strain of 2000%, fracture strength of 0.27 MPa, and compliant adhesion on different surfaces (hydrogels, skin, glass, silicone rubber, and nitrile rubber) [adapted with permission from ref. 116, Copyright 2019 American Chemical Society]. (b) Zwitterionic poly(MAA-*co*-DMAPS) hydrogel sensors to detect the movements of a wooden prosthetic finger and environmental temperature changes by tuning UCST and LCST for conversing forces to electrical signals. [Adapted with permission from ref. 66, Copyright 2018 American Chemical Society]. (c) PolyAAc/SA hydrogels used as skin-mimic sensors for detecting subtle pressure changes as induced by a gentle finger touch, human motion, or small water droplets. [Adapted with permission from ref. 65, Copyright 2017 Wiley-VCH].

and (iii) SA/tannic acid (TA)/polyAAm hydrogel with ternary ionic multi-bond network with high sensitivity (GF of 2.0) (Fig. 5b).[79] All of these salt-doped hydrogels were fabricated into stretchable wearable strain sensors for the real-time monitoring of various human motions and physiological signals. However, although physical doping can achieve the regeneration of electrical conductivity by simple re-soaking methods, the strong dependence of salt concentration on ion-doped polyelectrolyte hydrogels will also affect the sensitivity and specificity of hydrogel sensors in real-world applications.

Different from direct physical doping, the chemical doping strategy helps to enhance the mechanical properties of hydrogels. Usually, the chemical doping strategy drives free salt ions to strongly bind to the polymer network *via* metallic ion coordination and supramolecular assembly. These interactions reduce the possibility of salting-out phenomena and blockage of the network channels of hydrogel sensors, both of which cause the failure of hydrogel sensors. Chemical doping of sodium dodecyl sulfate (SDS)/NaCl into the polyAAm or HPAAm hydrogel network containing sodium alginate (SA) nanofibrils, regenerated silk fibroin (RSF), and cellulose derivatives improves the mechanical properties. NaCl/SA/polyAAm[86] and NaCl/SDS–RSF/HPAAm[87] hydrogel sensors (0.65–1.2 MPa) possessed 2–7 times higher tensile stress than pristine hydrogel sensors (0.1–0.2 MPa), with maximum stretchability of up to 1800–3100%. For the NaCl/SDS–RSF/HPAAm DN gel (Fig. 5c), SDS not only promoted the rapid formation of the first SDS–RSF network, but also formed SDS micelles to act as cross-linking points for a second HPAAm network. All of these tough electrolyte hydrogels can also be used as touchable or resistance-sensitive devices for sensing electronic signals.

Ionic liquids (ILs) are composed of cation–anion pairs of an organic ion and inorganic counterion. Different from inorganic salts, ILs are hydrophobic and difficult to add to hydrophilic hydrogels directly. Using click reaction and solvent exclusion methods, PIL–BF$_4$,[89] 1-butyl-3-methylimidazolium chloride (BMIMCl),[124] and [C16mim]$^+$Cl$^-$ [125] were encased or cross-linked into PEDGA/PETA, xanthan gum, and bulk g-C$_3$N$_4$ hydrogels.

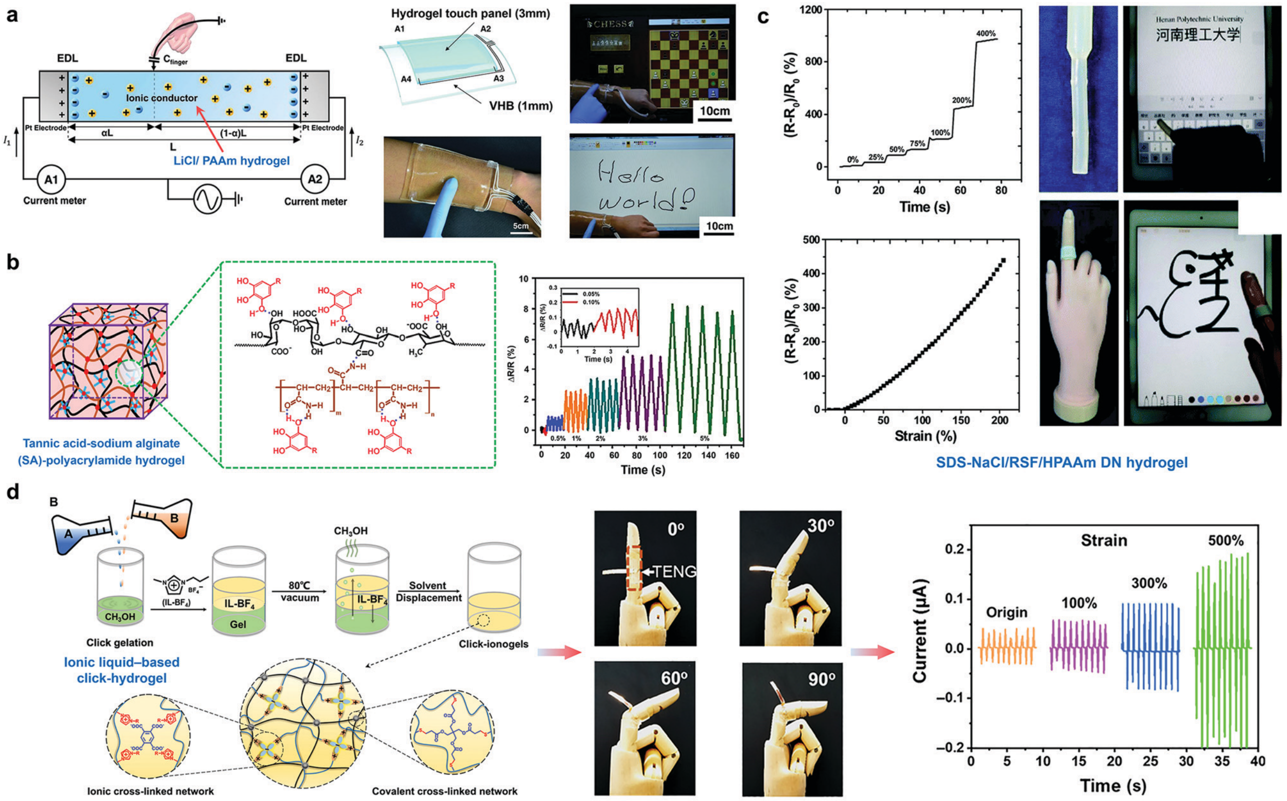

Fig. 5 Inorganic salt-incorporated hydrogel strain sensors. (a) LiCl–polyAAm hydrogel sensor with high stretchability and transmittance, which was further used as an ionic touch panel for writing words and playing games and a piano. [Adapted with permission from ref. 85, Copyright 2016 American Association for the Advancement of Science]. (b) SA/TA/polyAAm hydrogel sensor with a high gauge factor of 2–9 at a subtle (<100%) and large (2100%) strain for monitoring large limb motions of the human body (finger, knee, and elbow) and subtle muscle movements (smiling, chewing, and wrist pulse). Adapted with permission from ref. 79, Copyright 2019 American Chemical Society. (c) NaCl/SDS–RSF/HPAAm hydrogel sensor with high compression/tensile strength of 122/1.2 MPa, large extensibility of 19, high toughness of 1769 J m^{-2}, and high ionic conductivity of 0.012 S cm^{-1}. High ionic conductivity and strain sensitivity enable the RSF/HPAAm DN gel to be used as a touching screen pen and electronic skin. [Adapted with permission from ref. 87, Copyright 2019 Royal Society of Chemistry]. (d) Ionic liquid-based PIL–BF_4/PEDGA hydrogel strain sensor with high ionic conductivity (0.83 S m^{-1} at room temperature) for detecting finger bending under a wide temperature range (−75 to 340 °C) over 10 000 fatigue cycles. [Adapted with permission from ref. 89, Copyright 2019 American Association for the Advancement of Science].

The resultant hydrogel sensors possessed high tensile strength of 1.5–2.2 MPa, stretchability of 1500% up to 10 000 fatigue cycles, and broad thermal responsive range of −75 to 340 °C. Due to the high ionic conductivity (0.83 S m^{-1} at room temperature), hydrogel sensors can retain their high and stable electric sensitivity even at extremely low (−60 °C) and high (200 °C) temperatures (Fig. 5d). Furthermore, ILs are promising for preparing anti-freezing organogel and hydrogel sensors, which require further research efforts.

2.5. Biomolecule-incorporated hydrogel strain sensors

Hydrogels integrated with biomolecules (*e.g.* proteins, peptides, antibody, DNAs, and enzymes) present many general bio-related advantages of biodegradability, biocompatibility, and biostability, and specific bio-functionalities of drug/gene carriers, bioactivity, and bioimaging.[126–128] From a fabrication viewpoint, biomolecule-based hydrogels are often fabricated *via* a variety of methods, including click chemistry, Michael addition of cysteine residues to vinyl sulfones or maleimides, UV-initiated cross-linking, native chemical ligation, nonspecific amine-carboxylic acid couplings, and self-assembly.[129–131] Functionally, biomolecule-based hydrogels achieve their specific functions *via* simple sequence design, but stimuli-responsive sensing properties from the coupling of functional biomolecules with the physical properties of polymers. The incorporation of biomolecules into synthetic polymer networks can also influence the swelling, structure, and mechanical properties of hydrogels. Practically, most biomolecule-based hydrogels are dominantly used for tissue engineering and extra-cellular matrix (ECM) mimics, but stimuli-responsive biomolecule-based hydrogels have attracted increasing interests for next-generation bioelectronic interfaces over the last decade due to their similarities to biological tissues and versatility in electrical, mechanical, and biofunctional engineering.

For protein/polymer hydrogels formed by native proteins (*e.g.* collagen, gelatin, silk fibroin, bovine serum albumin, β-lactoglobulin, YajC-CT, and ovalbumin) and synthetic polymers, they achieve stimuli-responsive proprieties mainly through conformational changes between native and denaturation states upon external simulation.[132] For example, a series of PEG/protein hydrogels were synthesized with different proteins of soy protein,

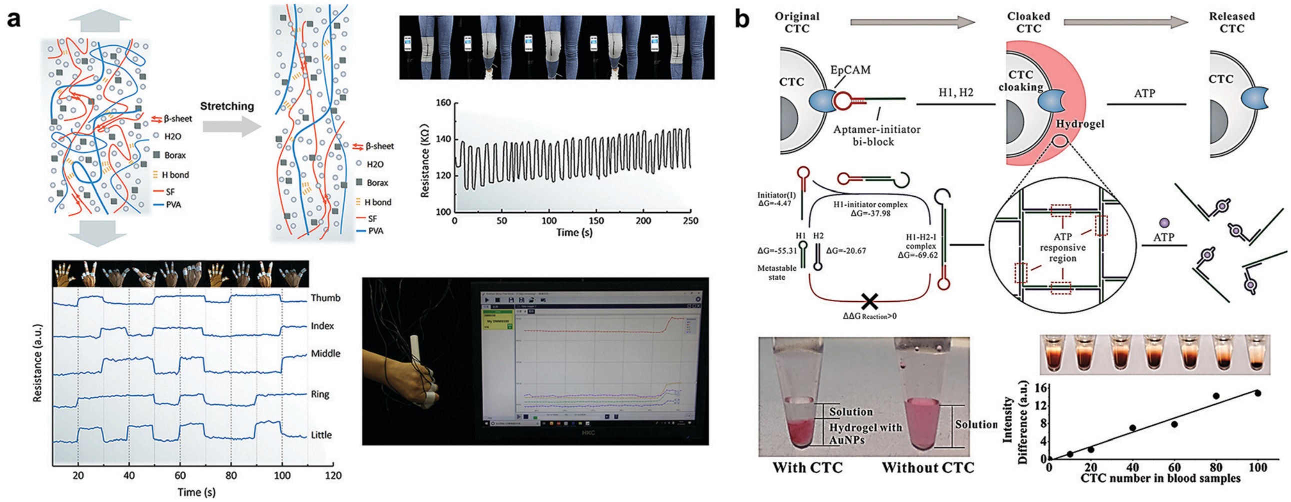

Fig. 6 Biomolecule-incorporated hydrogel strain sensors. (a) Protein-based PVA/RSF/borax hydrogel sensor with high stretchability, self-healing, and adhesion properties, which was integrated and used as a physical sensing platform for human motion detection [adapted with permission from ref. 90, Copyright 2019 American Chemical Society]. (b) ATP-aptamer-responsive DNA hydrogel sensor for capturing and releasing of circulating tumor cells with high cell viability. [Adapted with permission from ref. 92, Copyright 2019 American Chemical Society].

hydrolyzed soy protein, pea albumin, and casein.[133–135] Due to the polyelectrolyte nature of proteins, the resultant PEG/protein hydrogels exhibited pH responsiveness for drug release, but their mechanical properties were weak. Besides PEG, other polymers including PVA, poly(propylene oxide) (PPO), polyAAc, and polyHEMA have been used with RSF to produce polymer/RSF hybrid hydrogels, which can be regulated by heating, variation of pH,[136] alcohol, ions,[90] and shearing. RSF usually adopts β-sheet structures and favors the aggregation of these β-sheets into crystalline fibrous structures in polymer networks, and these hierarchical structures are the main contributors to the excellent mechanical properties of polymer/RSF hybrid hydrogels. Some PVA/RSF/borax hydrogels can also be used as a physical sensor to detect the bending and release of the leg, hand, and finger (Fig. 6a). However, the low stability and easy denaturation of proteins make protein/polymer hydrogels vulnerable in their sensing, mechanical, and biological properties. As an alternative, small peptides can mimic the main function of natural proteins, be facilely synthesized and precisely modified by simply controlling their sequences at large-scale and low-cost level *via* gene expression in bacteria, recombinant technology, and solid-phase peptide synthesis, and be easily incorporated with polymers *via* click chemistry. One design strategy to introduce stimuli-responsive properties for these peptides is to control their conformational changes *via* sol-to-gel transition by various environmental stimuli. For instance, an FEFEFKFK/polyNIPAAm and POG8/PEG hydrogel exhibited thermo-responsive properties, which could be tuned by the self-assembly of FEFEFKFK into β-sheet-rich fibers and POG8 into triple-helix fibers.[137,138]

Compared to proteins and peptides, DNA possesses enhanced stability under intensive heating, pressure, and chemical processing. DNA-functionalized hydrogels are still a relatively new field. The most important property of DNA hydrogels is that nucleobases including adenine (A), thymine (T), guanine (G), cytosine (C), and uracil (U) serve as programmable microscopic motifs or crosslinkers for constructing predictable secondary structural networks from the bottom up *via* base-pairing hybridization.[139] The hybridization of complementary DNA strands leads to the crosslinking of polymer chains and hydrogel formation with precise structural control and specific responses. DNA generally behaves like a long linear polymer and forms a hydrogel *via* physical entanglement or by chemical crosslinking of other polymers, composites, and small molecules. When nucleobases were independently introduced into polyacrylamide, DNA–polyAAm hydrogels exhibited a greatly enhanced elastic modulus of 0.43 MPa by several orders of magnitude,[140] and surface adhesion (220–780 N m^{-1}) on various solid materials, including polytetrafluoroethylene, plastics, glass, rubbers, metals, and wood.[141] This mechanical enhancement in DNA (linear)-polyacrylamide hydrogels is largely attributed to their DNA-self-assembled supramolecular structures. Besides linear DNAs, DNA can also be precisely designed and self-assembled into different branched architectures of X, Y, T, and cross-shapes, which were further self-assembled into large-scale, three-dimensional hydrogel structures *via* efficient, ligase-mediated reactions, but these branched DNA hydrogels were very weak with a tensile modulus in the range of 1–50 kPa and tensile strain of 0.42–0.57.[139]

Different from pure DNA hydrogels, due to the negatively charged nature of DNA, DNA is also used to form responsive complexes through physical interactions with cationic polyelectrolytes, nanocomposites and chemical grafting onto synthetic polymers.[142,143] Physical DNA–polymer and DNA–composite hydrogels undergo reversible and switchable changes in their morphology, color, and size in response to external stimuli, and thus are often used as smart sensors and devices to monitor binding events. However, physical DNA–polymer hybrid hydrogels usually suffer from weak mechanical strength and poor sensitivity, presumably due to the lack of precise control to form ordered structures. Instead of using physical interactions, DNA can be covalently grafted onto or crosslinked with polymers and proteins to construct programmable structures in hydrogel networks. For example, co-assembly with an octapeptide and DNA allowed the fabrication of DNA-inspired hydrogel sensors, which could selectively hybridize DNA and generate fluorescence, with a detection limit of 22 pM. DNA–poly(phenylenevinylene) hybrid hydrogels (DNA/SP–PPV) have been developed for monitoring drug release driven by the pH of the medium.[144] An ATP-responsive DNA hydrogel could achieve the controllable capture and release of cells by incorporating an ATP aptamer in the clamped HCR (aptamer-triggered clamped HCR, atcHCR) (Fig. 6b).[92] Overall, the unique properties of DNA-based hydrogels are particularly useful for biomolecule detection under biologically-relevant conditions, minimizing unwanted probe–probe interactions, and have the potential to hugely increase the analyte storage capacity of devices. In addition, unlike traditional reversible crosslinks such as DHEBA, DNA-crosslinks do not require the addition of an initiator-catalyst system to re-establish dissociated crosslinks.

3. Applications of hydrogel-based sensors

3.1. Hydrogel strain sensors as electronic skins

Human skin as a naturally evolved product combines multiple sensations and excellent mechanical properties, which offers a perfect example for developing biomimetic devices with stimuli-responsive properties for human–machine interactions. ''Hydrogel electronic skins'' are considered artificially intelligent skins, which marry the mutual advantages of hydrogels (*e.g.* high-water content, soft, biocompatibility, and high stretchability/toughness) and electronic devices (*e.g.* high electrical conductivity and stimuli-responsiveness).[15,145,146] A transparent LiCl–polyAAm hydrogel sensor was designed as an electronic skin, which could sense a 0–1 kPa pressure change and subtle human movements up to more than 1000 times.[84] Following a similar design strategy, bioinspired mineral ACC/polyAAc/alginate hydrogel electronic skins were developed to sense gentle human motion (*e.g.* finger touch, speaking and laughing) with very high pressure sensitivity of up to 1 kPa.[65] Zwitterionic poly(MMA-*co*-DMAPS) hydrogel strain sensors by integrating a parallel-plate capacitor with an ionic resistor enabled the mechanoreceptor (detection for 10 000 strain change) and thermo-receptor in human skin to be mimicked (10–80 °C) due to temperature-sensitive transition between LCST and UCST.[66] However, challenges still remain, where most of hydrogel-based electronic skins still possess relatively unitary functions. Accordingly, more precise and multifunctional electronic skins, LiCl–PVA hydrogel strain sensors (> 2000% of biaxial strain), combined with a flexible unit bearing power supply, control, readout and communication units, stretchable transducer batch, and printed circuit board was successfully constructed as an intelligence electronic skin.[147] This LiCl–PVA hydrogel electronic skin could real-time monitor and control the human body temperature, collect some health information (*e.g.* blood pressure and heart beat), and trigger drug release for treatment whenever abnormal physiological signals were received. This electronic skin outperforms most of the current hydrogel electronic skins, which is worthy of further exploration for potential bioelectronic applications.

3.2. Hydrogel strain sensors as wrist pulse or cardiac rhythm monitors

Artery pulse in (pre)clinical practice is a significant indicator of arterial blood pressure, cardiac rhythm, and aged blood vessel, which often provides useful information for the real-time, non-invasive diagnosis of possible cardiovascular diseases.[148,149] Due to the highly flexible and soft matrix of hydrogel strain sensors, hydrogel sensors enable closely fitting the curvature and surface of human skin with blood vessels underneath it to precisely detect three symbolic pulse waves as induced by wrist pulse, blood-pressure, and cardiac rhythm: percussion wave (P or P_1 wave), tidal wave (T or P_2 wave), and diastolic wave (D or P_3 wave), where the P_1/P_2 waves are the early/late systolic peak pressure and the P_3 wave is the diastolic pulse waveform.[150,151] More interestingly, the P_1-wave/P_2-wave (radial artery augmentation index) and time delay between these two-wave peaks largely reflect the arterial stiffness level, which is highly related to the health condition of humans.[152] For instance, several hydrogel-based wrist pulse/blood-pressure sensors made of PANI–poly(AAm-*co*-HEMA),[39] F-polyNIPAM/PANI,[64] AgNWs–polyAAm–PVP,[72] SA/TA/polyAAm,[79] Cu^{2+}/polyAAm,[81] LiCl–PVA,[85] NaCl–SA/polyAAm,[86] PANI/PSS,[63] polySBMA/PVA,[123] Ca^{2+}–SA–polyAAc,[153] agar/NaCl/polyAAm,[154] chitosan–PPy–polyAAc,[155] and SA–CNC/polyAAc[156] possessed high sensitivity with a gauge factor of 0.5–3, allowing the tracking of irregular pulses, waves, and intervals within 0.1 s. However, the shape, frequency, and amplitude of the pulse signals collected by these hydrogel sensors were still rough, and in some cases, they cannot accurately distinguish percussion (P_1), diastolic (P_2) and tidal (P_3) waves, limiting their further practical applications.

To overcome this detection sensitivity issue, the incorporation of nanoparticles into hydrogels, *i.e.* PDA–CNTs–poly(AAm–AAc),[74] MXene–PVA,[75] FSWCNT–PDA–PVA,[77] PDA–PPy–polyAAm,[105] CNTs–Egg white,[157] and SWCNT/polyAAm,[158] allows the gauge factor to be increased to 3–80 to distinguish different pulse waves under 10 kPa. Meanwhile, these wrist pulse/blood-pressure hydrogel sensors enable the transmission of wireless signals to smart phones. Besides hydrogel detection sensitivity (gauge factor), the specific contact area between hydrogel sensors and the human body is also important for wrist pulse or blood-pressure. A wireless pressure-sensing device incorporating Ca^{2+}–SA[159] hydrogel microspheres could continuously detect and distinguish characteristic pulse waves from human skin (~25 Pa), consistent with the clinically used wrist pulse detectors. On the other hand, hydrogel strain sensors are still inferior to metal/organic-based (*e.g.* ZnO-, Pt-, CNT-, and Au nanowires-based) pulse sensors in terms of both detection sensitivity and specificity, presumably due to the irreversible fatigue and damage of the network structure in hydrogels. Thus, some reversible bonds or self-healable materials should be introduced in these hydrogel sensors.

3.3. Hydrogel strain sensors as visual sensors

Stimuli-responsive hydrogels (*e.g.* thermal-responsive, glucose-responsive, pH-responsive, and cocaine-responsive hydrogels) possess the general ability to induce volume/size changes (*i.e.* shrinking and swelling) upon external stimuli. Particularly, when incorporating photonic crystals (PCs) or nanoparticles (PNs) into a hydrogel network, it would alter the periodicity layout of PCs and PNs to trigger a transformation of bio-/physical-signals into colorimetric signals, eventually achieving optical changes.[160] Colorful poly(St-MMA-AAm) hydrogel sensor-embedded PCs were successfully designed to realize reversible color changes from transparent to violet, blue, cyan, green and red in response to different humidity conditions, and the color change almost covered the whole visible range.[161] Such real-time, optical visual change was determined by expanding or contracting the crosslinked networks upon an increase or decrease of external humidity. Similarly, when incorporating PCs into the Si structure, poly(HEMA-*co*-MMA)–PCs hydrogel sensors could achieve color changes in response to temperature (25–50 °C) due to swelling/shrinking-induced viscoelastic deformation. A hybrid protein kinase A (PKA)/PCs hydrogel sensor enabled color changes for the label-free detection of kinase (PKA), as evidenced by a red shift, with the lowest detection limit of 2 U μL^{-1} for 2 h.[162] Moreover, modified AuNPs with plasmonic nanostructures can also be incorporated into hydrogel sensors as real-time colorimetric visual detectors. A smart colorimetric stretchable plasmonic AuNPs–polyNIPAM hydrogel biosensor was able to achieve rapid, reversible, color changes between red and grayish violet in response to a temperature change from 25–40 °C within 1 s.[68] In general, PC/AuNP-incorporated hydrogel sensors hold great promise for wearable healthcare due to their visual signals by the naked eyes, without the need for any additional detection instruments. However, when applying these visual sensors to wearable healthcare devices, epidermal contortion and human activity often inevitably cause irreversible damage to the hydrogel networks and structures, which in turn compromise the performance of these hydrogel sensors. To overcome this issue, we propose the introduction of some pattern structures to minimize the stretching of PCs to protect their colorimetric response.

3.4. Hydrogel strain sensors as human motions

Like other strain sensors, hydrogel strain sensors enable different human motion activities to be captured or distinguished. Current commercial and representative organic/inorganic-based strain sensors made of a combination of CNT, GO, and other conductive materials with elastomeric substrates usually have limited stretchability of up to 200%, while hydrogel strain sensors tend to adopt more complex and considerable deformation detection due to their higher stretchability (300–10000%).[163,164] Multi-scale strains of human motions include large-scale motions (*e.g.*, bending and twisting of hands, arms, and legs with minimal strain of >200%) and small-scale motions (*e.g.*, subtle movements of face, chest, and neck during emotional expression, breathing, and speaking with a minor strain of <50%). Both single-network hydrogel strain sensors (*e.g.* Cu^{2+}–polyAAm[81] and MXene–PVA[75]) and double-network hydrogel sensors (*e.g.* KCl-κ-carrageenan/polyAAm,[80] NaCl–SA/polyAAm,[86] NaCl/SDS–RSF/HPAAm,[87] and SA/TA/polyAAm[79]) were able to detect these human motions. However, it is generally difficult for hydrogel networks to recover to their initial state immediately after undergoing large deformation, and thus re-calibration of signal baseline of many hydrogel sensors after several-round tests is required. Therefore, ultra-stretchable hydrogel sensors with good mechanical self-recovery are expected to improve the substantial detection ability for human motion. For instance, upon the incorporation of PVP-capped AgNWs into the polyAAm hydrogel, the resultant hydrogel sensor exhibited extremely high stretchability (>22 000%), autonomous self-healing, mechanical compliance, and an ultrawide linear response range from a gauge factor of 0.15 at 0–430% tensile strain to 0.71 at 430–18100% tensile strain.[72] Another typical MXene–PVA hydrogel sensor with 3400% stretchability and good self-recovery was designed to track facial expressions, hand gestures, finger bending, hand writing, and vocal motions. Due to its soft and sticky characteristics, the MXene–PVA sensor could readily adhere onto various positions of the human skin for stable and repeatable detection.[75] This sensor with great self-recovery can real-time and *in situ* track large body deformations, spatial gesture movements, and physiological signals for motion behaviors, and provide health evaluation without any calibration because the quick recovery of its cross-linked network will maintain the relatively stable signal baseline.

4. Conclusion and perspectives

This review provided an update on the current status and developments on hydrogel strain sensors, including their design strategies, preparation methods, functional properties, mechanical sensing mechanisms, and practical applications. However, despite the great advancements, functional hydrogel strain sensors with excellent electrical, magnetic, optical, and biocompatibility properties are still in their infancy. Current research still mainly focuses on the exploration of new stimuli-responsive hydrogel systems, but some fundamental issues need to be addressed.

Conventional hydrogels used for strain sensors usually possess limited mechanical strength and are prone to permanent breakage. This intrinsic mechanical weakness often renders hydrogel sensors vulnerable to damage under continuous and responsive actions, resulting in unstable sensitivity and specificity. Also, the lack of dynamic cues and mechanical strength within the hydrogels limit their sensing ability. To remedy this issue, on one hand, introducing self-healing properties to hydrogel sensors will repair structural damage, increase reusability and stability, and restore sensing ability to some extent. On the other hand, the use of tough hydrogels (*e.g.* double-network hydrogels and nanocomposite hydrogels) as a structural platform to integrate different sensing components is considered as alternative design strategy to enhance the sensing capacity of hydrogel sensors. However, it still remains a great challenge to design and construct self-healing hydrogel sensors, which combine several properties of rapid autonomous self-healing ability, high-mechanical healing efficiency, and easily distinguishable response signals. Also, retaining the long-time self-healing of hydrogel sensors is another critical concern particularly under harsh conditions (*e.g.* underwater, freezing temperature, and salt/acidic solution). When applying hydrogel strain sensors to wearable devices, besides ultra-stretchability and high mechanical strength, additional robust adhesion to various solid surfaces (*e.g.* metal, glass, ceramic, and wood) and soft materials (*e.g.* rubber, skin, tissue, and organs) is required to maintain high strain sensitivity and stability to detect large human motions and tiny physiological signals. Another research direction is to apply hydrogel sensors for bio-applications, and in this regard, hydrogels themselves must be also nontoxic and biocompatible for hydrogel-based skin sensors, blood-pressure sensors, and implanted/injectable sensors. From an experimental viewpoint, it is always unstoppable motivation for material researchers to explore cutting-edge chemistries and fabrication technologies for designing and synthesizing novel hydrogels with desired properties towards specific sensing applications. In addition, beyond hydrogels, it is also interesting to develop and study organic gels prepared in or containing non-water solvents of varying polarity to understand the solvent effect on different aspects of organic gels, to reveal the differences and similarities in the gelation process, mechanical properties, toughening mechanisms, and network structures between hydrogels and organic gels, and to expand completely different gel systems for different applications.

In parallel, future efforts should be made to use computational modeling and machine learning technologies, together with polymer chemistry, synthesis, and gelation, to better understand the component-structure-performance relationship of hydrogel materials and to improve the structural-based design of hydrogel sensors and their interactions with target components and exogenous stimuli. Multi-scale simulations should study the deformation and recovery of hydrogels at the atomic, molecular, and macroscopic levels to clarify these issues since very little work has been done on this front.

Conflicts of interest

There are no conflicts to declare.

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