REVIEW

Graphene oxide composite hydrogels for wearable devices

Senxuan Tang[1] · Zhihan Liu[1] · Xu Xiang[1]

Abstract

For graphene oxide (GO) composite hydrogels, a two-dimensional GO material is introduced into them, whose special structure is used to improve their properties. GO contains abundant oxygen-containing functional groups, which can improve the mechanical properties of hydrogels and support the application needs. Especially, the unique-conjugated structure of GO can endow or enhance the stimulation response of hydrogels. Therefore, GO composite hydrogels have a great potential in the field of wearable devices. We referred to the works published in recent years, and reviewed from these aspects: (a) structure of GO; (b) factors affecting the mechanical properties of the composite hydrogel, including hydrogen bond, ionic bond, coordination bond and physical crosslinking; (c) stimuli and signals; (d) challenges. Finally, we summarized the research progress of GO composite hydrogels in the field of wearable devices, and put forward some prospects.

Keywords Wearable devices · Graphene oxide · Composite hydrogels · GO composite hydrogels

1 Introduction

Two-dimensional materials have unique physical and chemical properties, so they have attracted extensive attention from researchers. Graphene, graphene oxide (GO), molybdenum disulfide (MoS_2), boron nitride (BN), MXene and other two-dimensional materials have been well studied as well as developed in the frontier fields [1–6]. Graphene is the first successfully-prepared two-dimensional material, which has many excellent properties, such as a large specific surface area, strong mechanical properties, an ultra-high conductivity and so on [7]. However, the poor material compatibility of graphene limits its development in some research fields. GO is a product of stripping after graphite oxidation. Compared with graphene, GO contains richer oxygen-containing functional groups, whose chemical properties are more active [8]. Therefore, it has been widely used in many fields, such as electrode materials [9], capacitors [10] and so on. In addition, GO has a good water dispersibility [11], which can be used to prepare a variety of composite materials, such as hydrogels.

Hydrogels have a good biocompatibility, flexibility and easy preparation, which are ideal materials for wearable devices [12–14]. There are two problems when hydrogels are used in wearable devices. One is that their mechanical properties are poor, which are not enough to support the application of wearable devices, the other is that their electrical properties cannot provide sensitive responses for wearable devices. The special structure of GO makes itself and its complexes have a stimulus-response ability, which can be used to prepare various sensors [15, 16]. Meanwhile, the high dispersibility of GO in water provides conditions for the in-situ preparation of GO composite hydrogels. Compared with traditional hydrogels, those containing GO component have better mechanical and electrical properties, which make up for the deficiency of traditional hydrogels in stress-strain response. Therefore, GO composite hydrogels are an excellent sensor material that can be used to monitor exercise and some physiological indicators. For wearable devices, when GO composite hydrogels deform during limb movement, their electrical properties will change. By monitoring the resistance, current or capacitance between the electrodes, the deformation can be expressed in the form of electrical signals. With the gradual deepening of research, more and more GO composite hydrogel materials have been successfully prepared and applied to wearable devices, which, based on GO composite hydrogels, can be used to monitor a variety of activities of users, such as the movement of fingers,

✉ Xu Xiang
  xxiang1984@sina.com

[1] School of Materials Science and Engineering,
   Chongqing Jiaotong University, Chongqing 400074,
   People's Republic of China

elbows and knees, etc. Moreover, GO composite hydrogels also have a good response to some weak but high-frequency vibrations, such as those generated by pulses, breathing and talking, etc.

The research on GO composite hydrogels in the field of wearable devices in recent years is collected in this review, the achievements of related works are summarized from the aspect of mechanical properties, stimuli and signals, and the challenges faced by research in this field are put forward.

1.1 Structure of GO

As a two-dimensional material, the unique molecular structure of GO has attracted a lot of attention from researchers [8, 17, 18]. The exact structural formula of GO is inconclusive, which is usually expressed in terms of layer number and oxygen content, and is further described in terms of size, defects as well as acidity, etc. [8, 19]. It is generally believed that the structure of GO includes a planar structure composed of carbon and oxygen-containing functional groups on the surface [8, 19, 20]. In graphene, the planar structure of carbon is sp^2 hybrid, while in GO, due to the destruction of C=C bonds caused by oxygen-containing groups, the types of carbon hybrids include not only sp^2 hybrids, but also a large number of sp^3 hybrids. The oxygen-containing groups on the GO sheet surface mainly include hydroxyl and epoxy groups, while carbonyl and carboxyl groups are at the edge of the GO sheet (Fig. 1) [21]. The structure of GO ensures that it has a better dispersibility in water than graphene, which provides support for the preparation of GO composite hydrogels. RGO is prepared by the reduction of GO, and the reduction methods include chemical reduction, thermal reduction and microwave reduction, etc. [22]. However, it is difficult to remove all oxygen through reduction, and most of rGO contains residual oxygen-containing groups. Therefore, rGO is essentially GO, with a lower oxygen content, a larger conjugated structure area and better electrical properties.

1.2 Mechanical properties of GO composite hydrogels

Hydrogels contain a large amount of water, which makes it has a good biocompatibility and advantages in the preparation of wearable devices [23]. However, the high content of water also leads to the weak mechanical properties of hydrogels, which cannot meet the requirements of wearable devices. Therefore, hydrogels can be better supported as materials for wearable devices by improving their mechanical properties [24]. GO can greatly improve the mechanical properties of hydrogels [25], so GO composite hydrogels are suitable materials for wearable devices. The enhancement effect of GO on the mechanical properties of hydrogels benefits from its rich oxygen-containing functional groups and large specific surface area. Through crosslinking, hydrophilic polymers form the network structure of hydrogels. The hydrophilic groups on polymer chains can form hydrogen bonds with the oxygen-containing groups on GO to enhance its mechanical properties. At the same time, a larger specific surface area provides more sites for the interaction between GO and hydrophilic polymers, further improving its mechanical properties. The bond energy of hydrogen bonds is low but can be recovered quickly after disconnection. Therefore, GO enhances the mechanical properties of composite hydrogels without sacrificing their tensile properties. On the other hand, single-layer and few-layer GO have a good flexibility. When the hydrogels are deformed, they can well adapt to the deformation of themselves and form new hydrogen bonds with the matrix polymer, which improves the tensile strength of the hydrogels [25]. Figure 2 and Table 1 show the mechanical properties of GO composite hydrogels used in wearable devices in recent years. It can be found that in most studies, the tensile-fracture properties of hydrogels have been the focus of researchers, which is because the
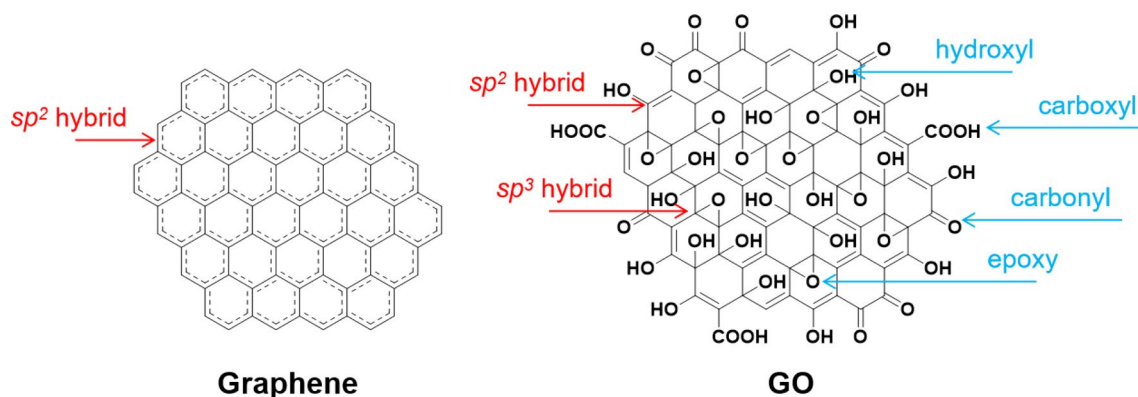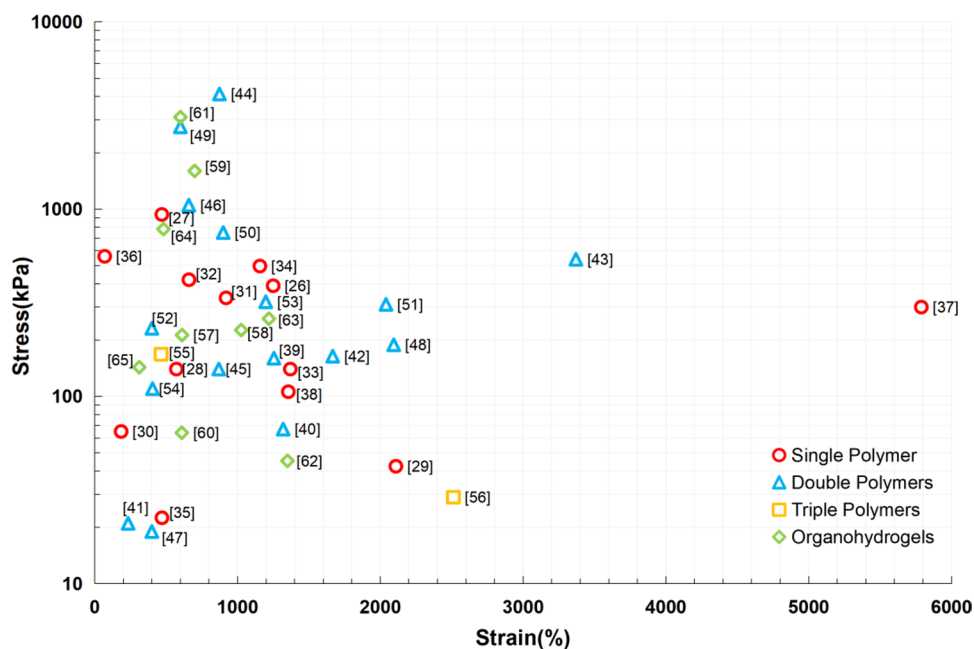

Fig. 1 Structure of graphene and GO

Fig. 2 Mechanical properties of GO composite hydrogels used for wearable devices reported in the references

hydrogels of most wearable devices are deformed due to the users' movement state, and then electrical signals are generated. In these studies, most of them used one or two matrix polymers to compound with GO, some of whom prepared organohydrogels. Except water, other organic solvents such as ethylene glycol, DMSO and glycerol were added to the organohydrogels. From the test results, organohydrogels will not significantly affect the mechanical properties, but can lower the evaporation water loss rate of hydrogels. On the whole, the mechanical strengths and tensile-fracture properties of the hydrogels of two-matrix polymers are better than those of single-matrix polymers. However, compared with ordinary hydrogels, the mechanical properties of GO composite hydrogels are significantly enhanced, which are embodied in the following aspects.

1.3 Hydrogen bonding

The structure of GO is different from that of graphene. GO contains abundant oxygen-containing groups, which makes it have a good water dispersibility, material compatibility and reactivity. The oxygen-containing functional groups of GO mainly include hydroxyl, carboxyl, carbonyl and epoxy groups, among which stable hydrogen bonds are easily formed with the functional groups of matrix polymer through hydroxyl and carboxyl groups, so as to improve the mechanical properties of hydrogels. As typical secondary bonds, the bond energy of hydrogen bonds is much lower than that of covalent bonds. Therefore, during the deformation process of hydrogels, they can be broken and formed continuously to adapt to the new deformation state [25].

Due to the reversibility of hydrogen bonds, the total number of them between GO and matrix polymer is basically unchanged, which provides support for the improvement of mechanical properties of composite hydrogels. PAA, PAM and PVA are three kinds of matrix polymers commonly used in GO composite hydrogels. Figure 3 shows the hydrogen bonding between the functional groups of matrix polymers and the GO sheet. Among these matrix polymers, all of the carboxyl, amide and alcohol groups on the carbon chains can form hydrogen bonds with oxygen-containing functional groups of the GO sheet. The mechanical properties of hydrogels are significantly improved under the action of a large number of hydrogen bonds.

1.4 Ionic bonding and coordination bonding

In addition to the hydrogen bonding, metal cations have been introduced in some studies to enhance the interaction between matrix polymers and GO, such as Ca^{2+} and Fe^{3+} (Fig. 3). High-charged Ca^{2+} and Fe^{3+} can form ionic bonds with carboxyl groups in the matrix polymer, thus forming new crosslinking points in hydrogels. Because GO also contains carboxyl groups, such ionic bonding exists not only with the matrix polymer, but also between the matrix polymer and GO as well as between GO and GO. Except the ionic bonding, coordination bonding is also used to enhance the mechanical strength of GO composite hydrogels. Through chemical reactions, dopamine was grafted to the GO sheet and catechol functional groups were introduced. Catechol has a strong coordination effect with Fe^{3+}, each of which can form coordination bonds with three catechol groups, thus greatly improving the crosslinking strength of hydrogels. In

**Table 1** Mechanical properties of GO composite hydrogels

GO composite hydrogel	Abbreviation	Matrix	Max strain (%)	Max stress (kPa)	Ref
Ag nanoparticle/coated Tannic acid@ graphene oxide/polyacrylamide hydrogel	Ag-TA@ GO-PAM	Single polymer	1250	390	[26]
Graphene oxide/adsorbed liquid metal nanodroplets/polyacrylic acid hydrogel	LMNPs@ rGO-PAA	Single polymer	470	937	[27]
Polyacrylamide/reduced graphene oxide composite hydrogel	PAM-rGO	Single polymer	572	140	[28]
Polyacrylamide/N, N'-isopropyl di-acrylamide/graphene oxide nanocomposite hydrogel	PAM-BIS-GO NC	Single polymer	2109.37	42.4	[29]
Polyvinyl alcohol/graphene oxide hydrogel	PVA-GO	Single polymer	185	65	[30]
Polyacrylic acid/graphene oxide hydrogel	PAA-GO	Single polymer	920	336	[31]
Polyacrylic acid/reduced graphene oxide nanocomposite hydrogel	PAA-rGO	Single polymer	660	420	[32]
Poly(acrylic acid)-Liquid metal/reduced graphene oxide hydrogel	PAA-LM-rGO	Single polymer	1370	140	[33]
Poly(acrylamide-co–N-(3-aminopropyl) methyl acrylamide hydrochloride/ reduced graphene oxide-polydopamine) hydrogel	poly(AAm-co-APMA-rGO-PDA)	Single polymer	1156.9 ± 110.1	496.8 ± 5.9	[34]
Reduced-graphene-oxide-doped poly(2-acrylamido-2-methyl-1-propanesulfonic acid-co-acrylamide) hydrogel	rGO-Poly(AMPS-co-AAm)	Single polymer	471	22.5	[35]
Ag nanowires/graphene composite/poly-dimethylsiloxane Strain Sensor	Ag NWs-G-PDMS	Single polymer	70	560	[36]
Reduced graphene sheets/ Polyacrylamide/N, N-methylenebi-sacrylamide/calcium hydroxide nano-spherulites hydrogel	GS-PAM-MBA-CNS	Single polymer	5789	300	[37]
Laponite/poly(2-acrylamido-2-meth-ylpropane sulfonic acid-co-acrylic acid-co–N-[3-(dimethylamino)propyl] methacrylamide)/graphene oxide-LiCl hydrogel	Laponite-poly(AMPS-co-AA-co-DMAPMA)GO-LiCl	Single polymer	1356	106	[38]
Polydopamine reduced graphene oxide/ sodium carboxymethyl cellulose/poly-acrylamide double network conductive hydrogel	D-rGO-CMCNa-PAM	Double polymers	1255	160	[39]
Reduced graphene oxide/sodium car-boxymethylcellulose/polyacrylic acid composite conductive hydrogel	rGO-CMCNa-PAA	Double polymers	1320	66.9	[40]
Polyvinyl alcohol/polydopamine/par-tially reduced graphene oxide hydrogel	PVA-PDA-pRGO	Double polymers	234	21	[41]
Sodium caseinate/reduced graphene oxide/polyacrylamide hydrogel	SC-rGO-PAAm	Double polymers	1667	164	[42]
Alginate nanofibril network/graphene oxide/polyacrylamide nanocomposite double network hydrogel	SNGP	Double polymers	3370	540	[43]
Graphene oxide@ poly(2-(dimethylamino)ethyl methacrylate) Janus NS/guar gum/poly(acrylic acid) self-healing nanocomposite hydrogels	GO@ PPy-PDMAEMA Janus NS-GG-PAA hydrogel	Double polymers	873.8	4120	[44]
Polyvinyl alcohol borax/tannic acid/ coated cellulose nanocrystals/pRGO hydrogel	PB-TA@ CNC-pRGO	Double polymers	869	140	[45]

Table 1 (continued)

GO composite hydrogel	Abbreviation	Matrix	Max strain (%)	Max stress (kPa)	Ref
Polyacrylic acid/reduced graphene oxide/ Polyaniline hydrogel	PAA-RGO-PANI	Double polymers	659 ± 5.78	1050 ± 8.34	[46]
Gelatin/graphene oxide/ferric ion cross-linked poly(acrylic acid) hydrogel	Gel-GO-PAA-Fe^{3+}	Double polymers	400	19	[47]
Gum arabic/reduced graphene oxide/ polyacrylamide hydrogels	GA-rGO-PAM	Double polymers	2094.3	188.7	[48]
Polyacrylic acid/graphene oxide/ferric cation/chitosan hydrogel	PAA-GO-Fe^{3+}-CS	Double polymers	600	2750	[49]
Polyacrylamide/Polydopamine reduce graphene oxide/sodium alginate hydrogel	PAM-prGO-SA	Double polymers	900	750	[50]
Graphene oxide / polyvinyl alcohol / polyacrylamide double network hydrogel	GPPD	Double polymers	2040	310	[51]
Poly(N-(3-sulfopropyl)-N-methacroyloxyethyl/N, N-dimethyl-ammonium betaine-acrylamide)/poly-vinyl alcohol/rGO porous composite hydrogel	Poly(SBMA-AM)-PVA-rGO	Double polymers	400	231	[52]
Graphene oxide@ Dopamine/Alginate/ Poly(acrylic acid-co-acrylamide) hydrogel	GO@ DA-Alginate-P(AAc-co-AAm)	Double polymers	1198	320.2	[53]
Sandwich-like Polypyrrole/Reduced Graphene Oxide Nanosheets Integrated Gelatin Hydrogel	PPy-rGO-PPy-Gel	Double polymers	405	110	[54]
Polyacrylamide/silk fibroin/poly(3,4-eth-ylenedioxythiophene): poly (4-styrene sulfonate)/graphene oxide composite hydrogel	PAM-SF-PEDOT: PSS-GO	Triple polymers	465	168	[55]
Graphene oxide/poly(N-isopropy-lacrylamide)/poly(3,4-ethylenediox-ythiophene): poly(styrenesulfonate) hydrogel	GO-PNIPAM- PEDOT: PSS	Triple polymers	2512	29	[56]
MXene-graphene oxide/cellulose nanofibrils/sodium alginate/poly(vinyl alcohol) hydrogel	MX-GO-CNFs-SA-PVA	Organohydrogel	611.5	212.7	[57]
Polyacrylic acid/chitosan/graphene oxide/Glycerol hybrid gel	PAA-CS-GO-Gly	Organohydrogel	1025.9 ± 102.7	226.2 ± 30.05	[58]
Poly(vinyl alcohol)/glycerol/cationic chitosan/GO hydrogel	PVA-glycerol-CC-GO	Organohydrogel	700	1600	[59]
Phenylboronic acid grafted alginate/ poly(vinyl alcohol)/polyacrylamide/ reduced graphene oxide organohydro-gel	Alg-PBA-PVA-PAM-rGO	Organohydrogel	610	64	[60]
Poly(vinyl alcohol)/dimethyl sulfoxide/ reduced graphene oxide/graphene oxide organohydrogel	PVA-DMSO-rGO-GO	Organohydrogel	600	3100	[61]
polydopamine/reduced oxide graphene/ poly(3,4-ethylenedioxythiophene): poly(4-styrene sulfonate) (PEDOT: PSS)/polyacrylamide composite organohydrogel	PCOH	Organohydrogel	1350	45.3	[62]
Silver nanoparticles@ reduced graphene oxide/poly(vinyl alcohol)/polyacryla-mide dual-network organohydrogel	Ag@ rGO-PVA-PAM	Organohydrogel	1220	260	[63]

Table 1 (continued)

GO composite hydrogel	Abbreviation	Matrix	Max strain (%)	Max stress (kPa)	Ref
Poly(acrylamide-co–N-(3-amino propyl) methacrylamide)/graphene oxide/glycerol organohydrogel	Poly(AAm-co-APMA)-GO-Gly	Organohydrogel	481.4 ± 46.9	782.9 ± 38.6	[64]
Polydopamine/reduced graphene oxide/sodium alginate/polyacrylamide composite organohydrogel	PDA-rGO-SA-PAM	Organohydrogel	312	143.2	[65]

Fig. 3 Interaction of GO sheet with hydrogel polymer matrixes and ions
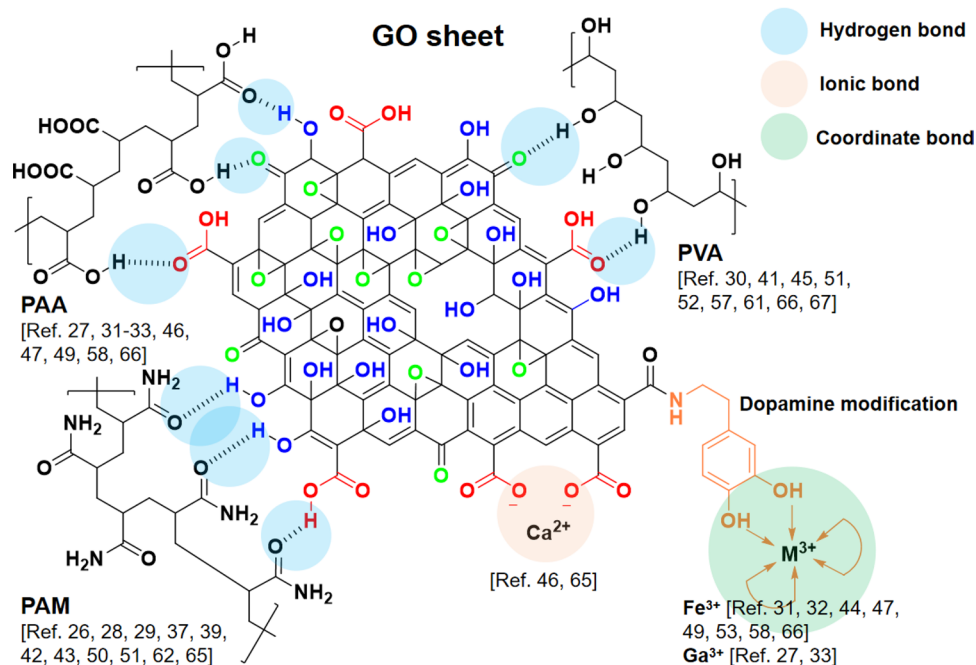
addition to the commonly used Ca^{2+} and Fe^{3+}, Ga^{3+} is also used to construct new crosslinking points in hydrogels [27, 33]. Using the liquid metal nanoparticles of Ga (gallium), GO is reduced in situ during the preparation of hydrogels to form a crosslinking system with the participation of Ga^{3+}. Through this method, the mechanical and electrical properties of GO composite hydrogels are improved at the same time, providing support for the preparation of highly sensitive wearable devices.

Enhancing the crosslinking system of hydrogels by adding metal ions is an effective method to improve their mechanical strength. However, it is worth noting that too high ion concentration may lead to the agglomeration of GO. Therefore, in the preparing process of this kind of hydrogels, the design of preparation methods as well as the concentration control of GO and ions will affect the performance of composite hydrogels. On the other hand, both ionic and coordination bonds provide new crosslinking points for hydrogels, which will not only greatly improve the mechanical strength of hydrogels, but may also affect their maximum tensile-fracture length.

1.5 Physical crosslinking

Physical crosslinking is one of the forms of hydrogel crosslinking networks, especially in composite hydrogels, the physical entanglement between matrix polymers and nano materials can improve their mechanical properties [27, 30, 43, 52, 54]. As a two-dimensional material, GO has a large specific surface area, which makes it so obvious that it is entangled by matrix polymers. Figure 4a shows the interaction between GO sheets and polymer chains in the case of single network. When hydrogels are deformed by external force, it will be resisted by the hydrogen bonds between the hydrogels and GO. At the same time, as a junction in hydrogels, GO will also exert force on the polymer chains, which will hinder their deformation and improve their mechanical properties (Fig. 4a'). In the case of double-crosslinking networks, GO will interact with two matrix polymers at the same time, producing an interaction similar to that of single networks, meanwhile improving the mechanical properties of hydrogels (Fig. 4b and b'). With the participation of metal ions, in addition
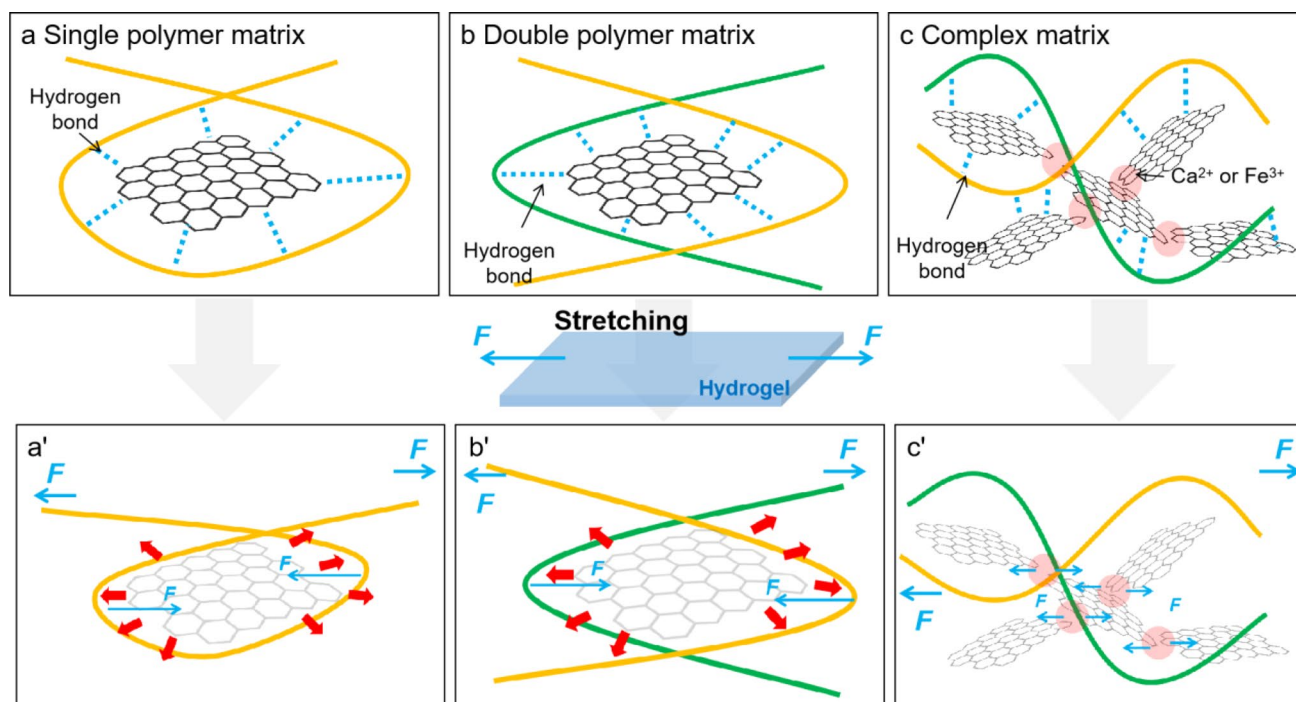
Fig. 4 The physical entanglement among GO sheets, matrix polymers and the interaction force during stretching. **a** and **a**' Single-polymer matrix, **b** and **b**' double-polymer matrix, **c** and **c**' complex matrix

to the interaction of physical entanglement of polymer chains, the contribution of ionic bonds and coordination bonds to the improvement of mechanical properties cannot be ignored. With metal ions as the media, a crosslinking network can be formed through GO, which can also form a crosslinking network with matrix polymers (Fig. 4c). Multiple-crosslinking networks make the physical entanglement between among networks more complex and stable, thus enhancing the mechanical strength of hydrogels (Fig. 4c').

It should be noted that the change of GO in the deformation process of hydrogels is different from that of other materials. As a two-dimensional material, GO has the characteristic of flexibility, which ensures that the GO sheets can be adjusted with the deformation of hydrogels during the stretching process. During the deformation of hydrogels, the GO sheets will deflect and rotate according to their interaction with matrix polymers as well as the direction of external force, so as to adapt to the deformation of hydrogels (Fig. 5). At the same time, because the interactions between GO and matrix polymers are mainly hydrogen bonds, the reversibility of hydrogen bonds ensures that the structure between GO and matrix polymers will not be destroyed with the deformation of hydrogels. The GO sheers play a role of a "2D flexible crosslinking point" in composite hydrogels, which is also one of the important reasons that GO can improve the mechanical properties of composite hydrogels [25].

1.6 Stimuli and signals

The improvement of mechanical properties of composite hydrogels by GO ensures the application of composite hydrogels in the field of wearable devices, which require materials to respond to environmental stimuli, such as temperature [33, 35, 43, 57, 61, 62, 65, 66], pH [39, 40, 67], light [68] and strain. Environmental stimulation will lead to changes in the structure and properties of matrix polymers. By detecting these changes, the intelligence of hydrogels can be realized.

At present, wearable devices are mainly used to monitor the movement of users, such as running, walking and other body movements, as well as their physiological conditions, such as heart rate, blood pressure and so on [69] (Fig. 6). Therefore, hydrogels mainly used in wearable devices are strain response hydrogels, which have an intelligent response to pressure, tension and vibration. When composite hydrogels are deformed by different types of external forces such as tension, compression, bending, folding or vibration, their corresponding physical properties will change significantly. On the other hand, as a wearable device, electrical signals are the most direct and useful signal output mode of sensors, which can be directly displayed on mobile phones, watches and other electronic devices, thus providing users with visual graphic information.
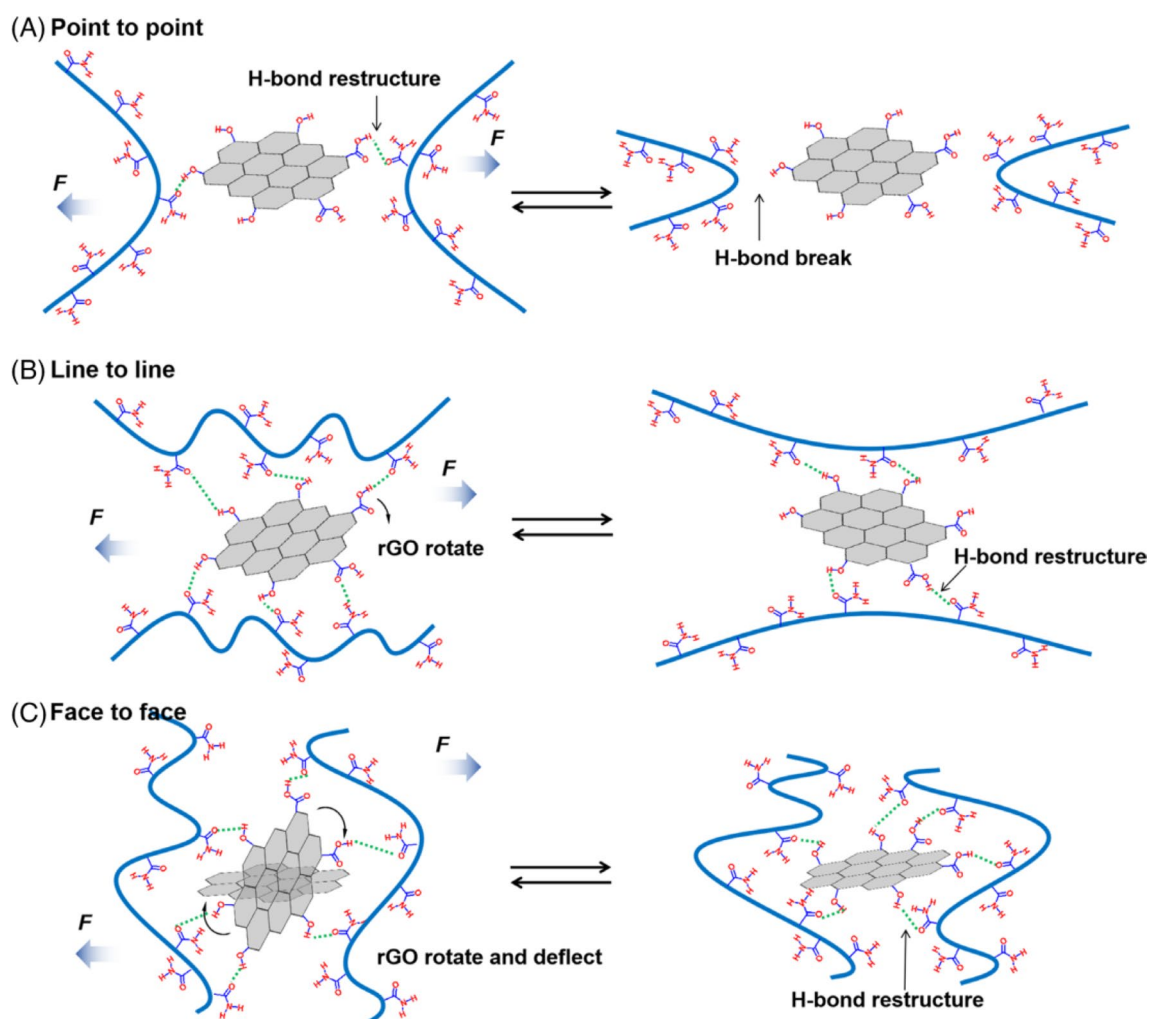
Fig. 5 The action mechanism of "2D flexible crosslinking point" in composite hydrogels **a** point to point, **b** line to line, **c** face to face. Reprinted with permission from reference [25]. Copyright 2021 Society of Plastics Engineers

According to different output signals, the ways to assemble wearable devices with hydrogels are also different. For resistance and current wearable devices, just connect electrodes at both ends of the hydrogels, and the assembly method is simple (Fig. 6a and b). For capacitive wearable devices, they need to be assembled into a sandwich structure, and the components need to be charged in use [29, 33, 70] (Fig. 6c). The assembly method of capacitive wearable devices is more complex.

GO can not only improve the mechanical properties of wearable devices, but also affect the electrical properties. Although many conjugated structures of GO are destroyed in the oxidation process, some of them are remained, which provide a small amount of conductivity for GO. When the amount of GO gradually increases, the conductivity of hydrogels will slowly increase [30, 31, 47, 71]. In some studies, reduced graphene oxide (rGO) is used instead of GO to prepare composite hydrogels [28, 40, 60]. In the process

of GO reduction, the conjugated structure of GO sheets is restored, so the conductivity of rGO is much greater than that of GO. RGO can improve the conductivity of composite hydrogels, thereby improving the sensitivity of stimulus response. By adding appropriate GO/rGO, the conductivity of hydrogels can be improved. The high content of GO/rGO may have a negative impact on the conductivity of composite hydrogels because it is easy to agglomerate due to π-π interaction, electrostatic interaction and other reasons. Therefore, the amount of GO/rGO needs to balance with the conductivity, mechanical properties and dispersion of GO/rGO.

1.7 Resistance and current type

Resistance and current are the most-easily detected electrical signals [43, 61, 72]. At the same time, the assembly process of resistance/current wearable devices is relatively simple, which is the first choice of most researchers. When
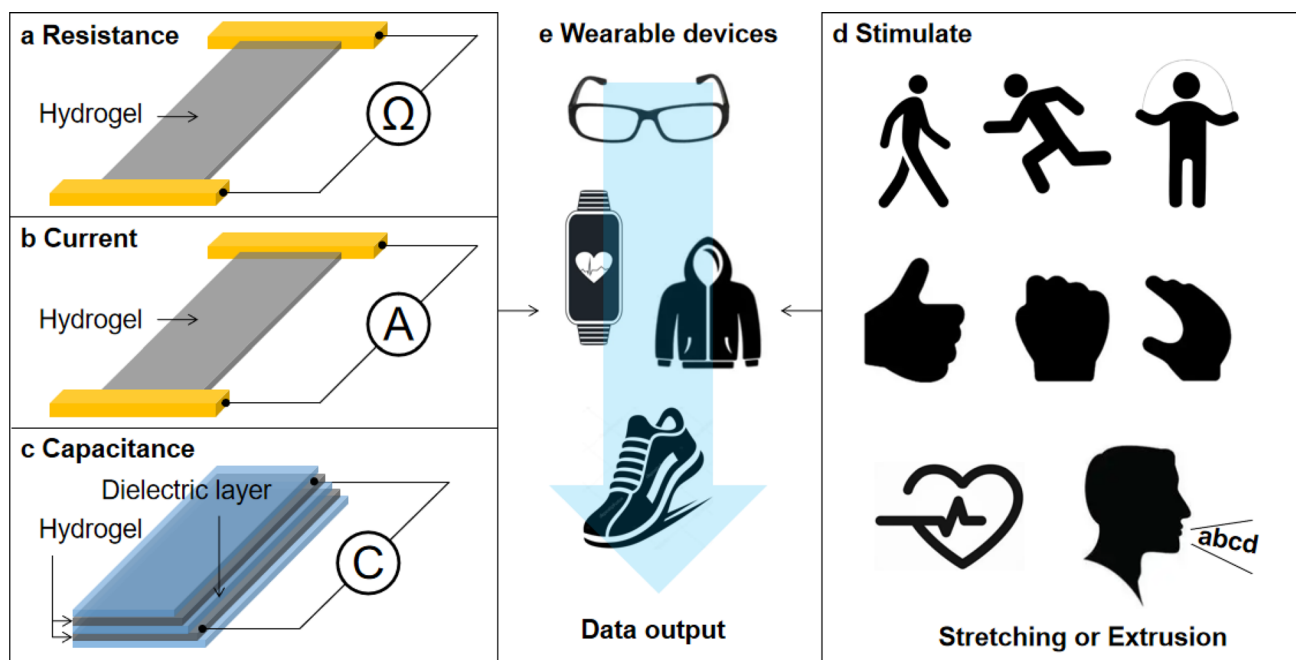
Fig. 6 Assembly of wearable devices and some types of stimulation

the hydrogels are deformed by external force, the distance between electrodes changes, resulting in changes in resistance and current, so as to monitor the users' state. According to the calculation formula of resistance: $R = \rho l / S$, the resistivity (ρ) is constant; when the hydrogels are stretched, the distance (l) between the electrodes increases, while its cross-sectional area (S) decreases, so the resistance (R) increases. On the other hand, the stretching of the hydrogels will increase the distance among the GO sheets, thereby increasing the resistance of the composite hydrogels. For example, *Z. Deng* et al. [44] reported a composite hydrogel (GO@ PPy /PDMAEMA) for the assembly of wearable devices. By detecting the changes in resistance caused by the deformation of hydrogel, the users' joints were monitored in real time (Fig. 7).

1.8 Capacitive type

Compared with resistance and current, although the assembly of capacitive wearable devices is more complex, they have more advantages than resistance/current. The biggest advantage is that in addition to being sensitive to tensions, capacitive wearable devices also have a good response to pressure [29, 33], and can cope with different limb changes [70] (Fig. 8). Through assembly, two layers of hydrogels form a capacitor, which takes different charges after charging (Fig. 8a). When the capacitor is stretched or extruded, the two hydrogels deform, the distance between which changes, and the charge is redistributed, leading to the

change of capacitance. On the other hand, capacitive wearable devices need to be encapsulated with polymer materials, which greatly slows down the evaporation of water in hydrogels and makes them maintain a long-term stable stimulus responsiveness.

1.9 Other

Further, in addition to the simple monitoring functions of sports and physiological activities, researchers have also developed additional and recognitive functions of wearable devices. For example, *H. Zhang* et al. [55] reported a composite hydrogel with structural colors, which could not only monitor the actions in real time, but also display different colors according to the size of hydrogel deformation (Fig. 9). *F. B. Kadumudi* et al. [73] reported a self-healing hydrogel with super tensile properties, through which action recognition was realized by combining different signals (Fig. 10). More functions provide a broader range of applications for wearable devices. In the latest research, wearable devices are also used in many fields, such as the detection of COVID-19 [74], automatic real-time detection of diseases, health monitoring [75, 76], temperature adjustable clothes [77] as well as implantable and wearable bioelectronic devices [78].

1.10 Challenge

GO composite hydrogels have a good development potential in the field of wearable devices, but they also face many
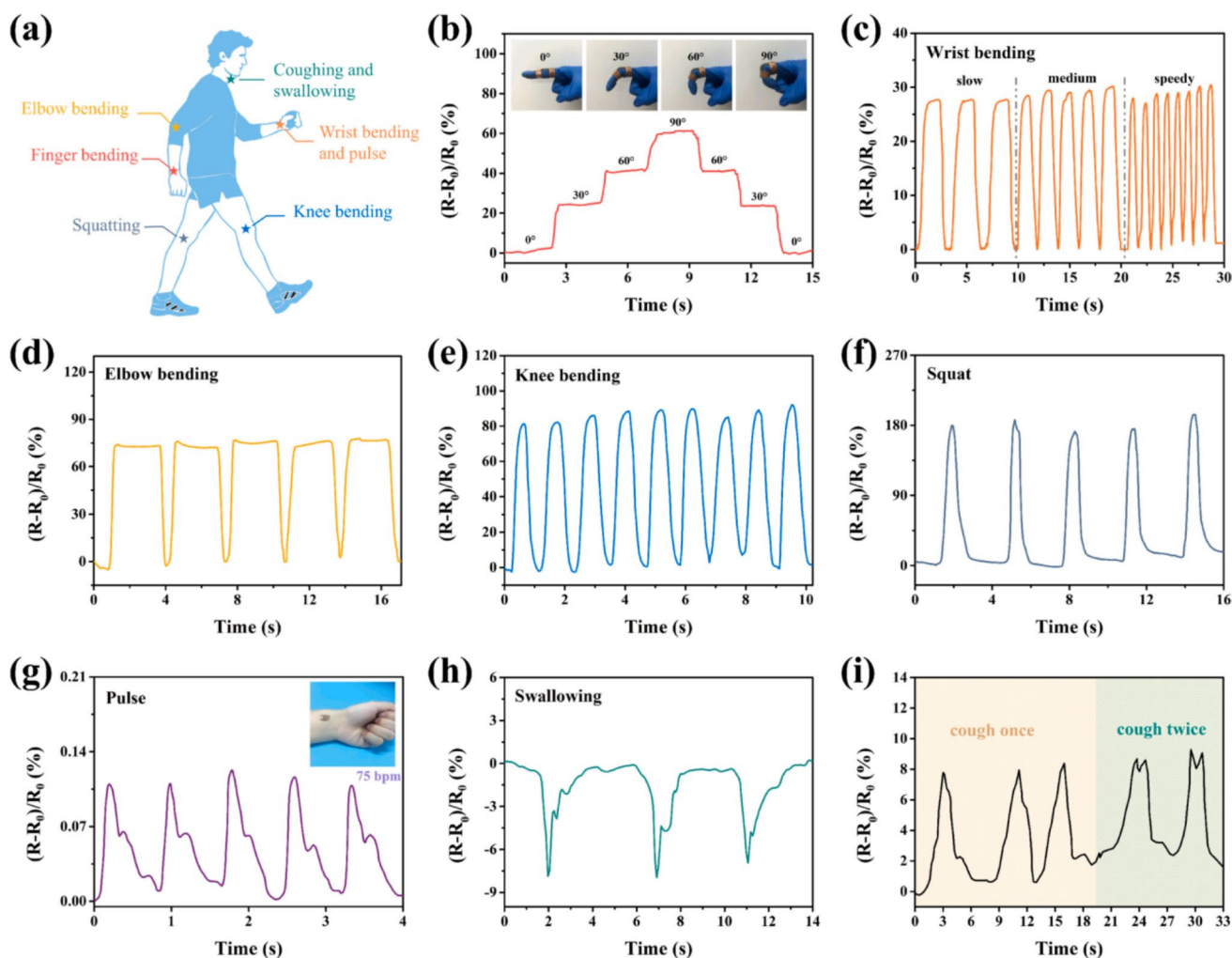
Fig. 7 **a** The self-healing GO@ PPy / PDMAEMA Janus NS nano-composite hydrogels as wearable sensors for detecting various human movements in real time, **b** finger bending at different angles, **c** wrist bending with different speeds, **d** elbow bending, **e** knee bending, **f** squatting, **g** pulse, **h** swallowing, **i** coughing. Reprinted with permission from reference [44]. Copyright 2021 Elsevier.

challenges, and more research is needed to solve these problems.

1.11 Mechanical properties

In many studies, the mechanical properties of hydrogels are the focus of researchers. At present, the mechanical strength of hydrogels is far from the level of human skin, while their tensile-fracture length is redundant. After a small range of statistics, the tensile deformation of wearable devices attached to the human joint surfaces during movement is not higher than 50%. Considering the redundancy of devices, 200% of the tensile-fracture length is sufficient to meet the requirements of all wearable devices. Therefore, in future research, researchers may pay more attention to the strength of hydrogels rather than their tensile-fracture length.

1.12 Water loss of hydrogels

The main component of hydrogels is water. Once water evaporates, wearable devices will not work. GO cannot solve the problem of water loss, so how to keep water is the key to the further application of GO composite hydrogels. At present, main water retention methods used by researchers include: (a) encapsulating hydrogels with polymer materials (VHB, PE or PET) [29, 43, 57], (b) replacing water with other solvents (such as glycerite [58, 59, 63–65, 72]), and (c) coating silicone oil on the surface of hydrogels [56]. According to the experimental results, hydrogel encapsulation method is more effective than the other two methods. For all that, the above methods cannot be used to completely solve the problem of water loss, and researchers need to do more research in the future.
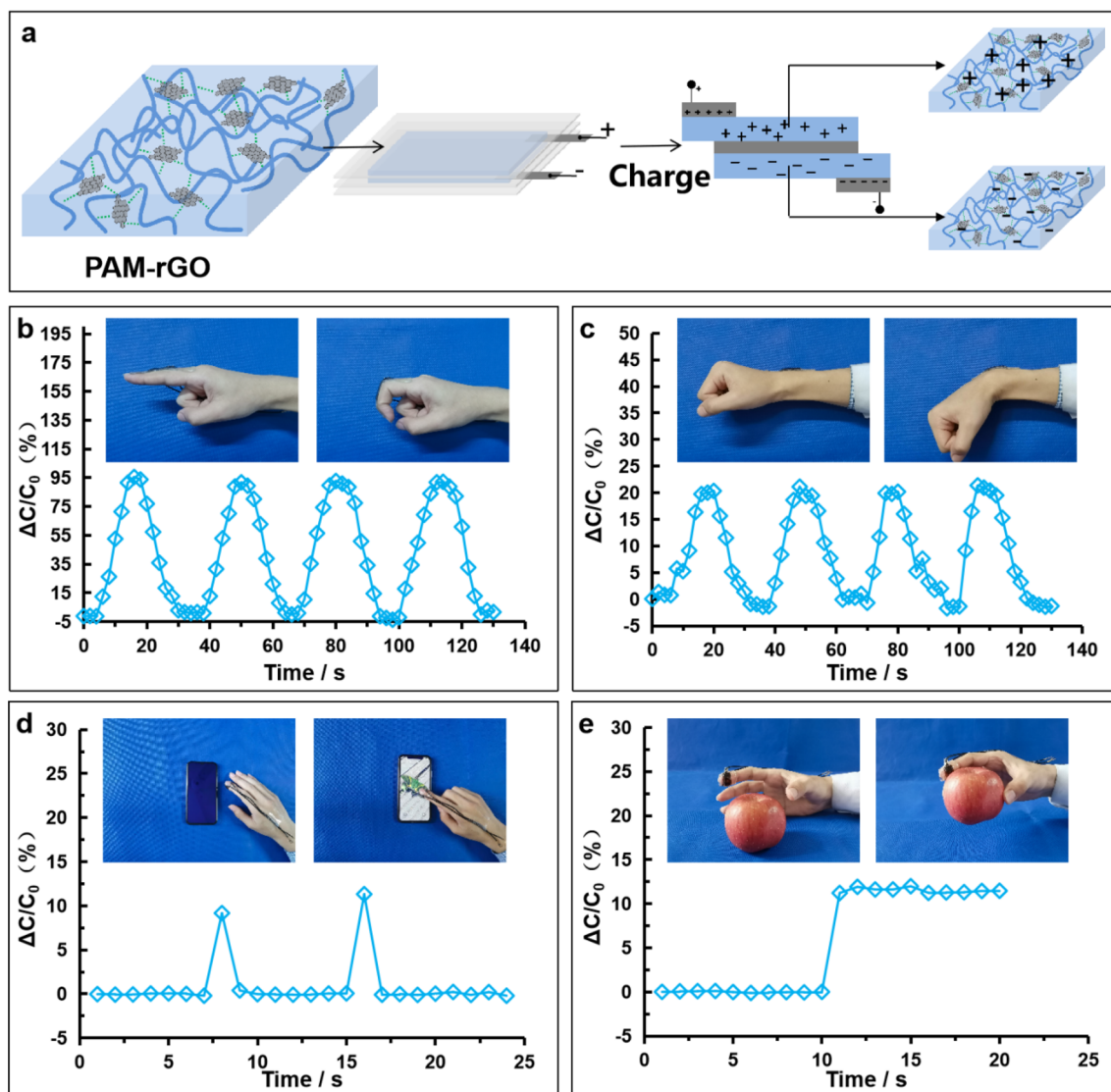
Fig. 8 **a** Capacitive wearable device assembled by PAM-rGO composite hydrogel. **b** The strain sensor in response to finger bending. **c** The strain sensor in response to wrist bending. **d** The pressure sensor in response to clicking on the phone screen. **e** The pressure sensor in response to picking up an apple. Reprinted with permission from reference [70]. Copyright 2022 Elsevier.

1.13 Self-healing performance

The self-healing performance of hydrogels is also a focus of researchers [79–81]. In the current research, most self-healing hydrogels have been able to meet the needs of wearable devices. However, it should be pointed out that the actual working environment or the water loss of hydrogels is not considered in most self-healing tests. For example, some hydrogels need one or more days to complete the self-healing process. Within one day, hydrogels may have lost a lot of water, which leads to the failure of wearable devices. Therefore, in the field of wearable devices, the self-healing performance of hydrogels needs to be considered together with the mechanical properties and water retention of hydrogels,

so as to ensure that wearable devices can still work normally during the self-healing process of hydrogels.

1.14 Stability

In some research works, the working stability of GO composite hydrogels has been well verified. However, the environment in which most of the tests are conducted is stable and mild, which cannot reflect the working state of hydrogel-wearable devices in various extreme environments, especially when the hydrogels lose water. To ensure the working stability of wearable devices, researchers are required to comprehensively consider the water loss, fatigue resistance and so on.
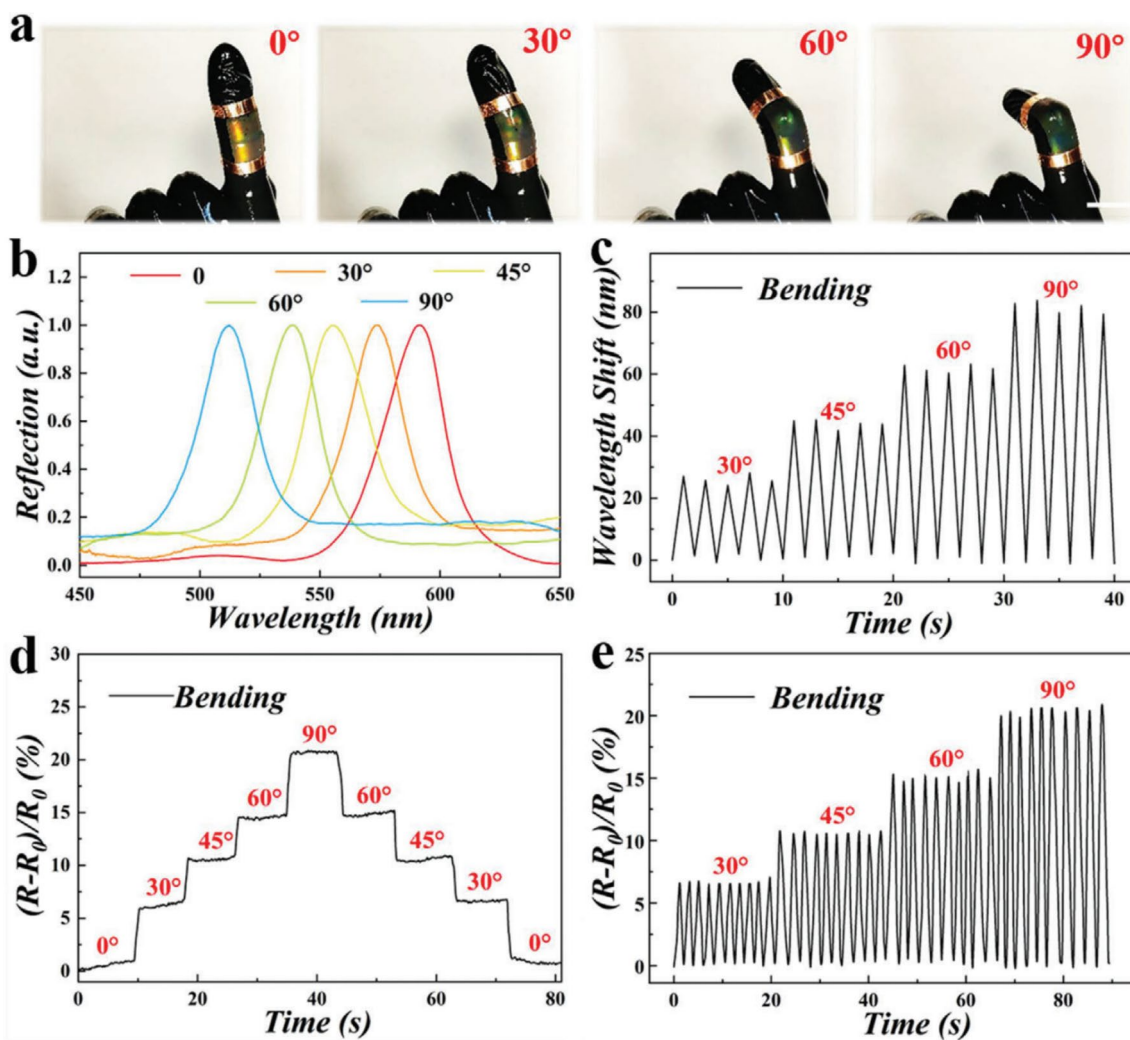
Fig. 9 The changes of optical and electrical signals of the composite hydrogel film during the bending process. **a** Photographs of the composite hydrogel structural color film with different bending angles of the finger. The scale bar is 1 cm. **b** The reflection peak wavelengths of composite hydrogel films with different bending angles. **c** The wavelength shift values of the composite hydrogel films with differ-ent bending angles of the finger. **d** The relative resistance changes of composite hydrogel film during the bending process of the finger. **e** The relative resistance variation of composite hydrogel film under cyclic changes of different angles. Reprinted with permission from reference [55]. Copyright 2021 Wiley-VCH GmbH

1.15 Corrosion of electrode

The corrosion of hydrogels to electrodes is a problem that is easily ignored. Oxygen, water, electrolyte as well as charging and discharging process will corrode the electrodes, which will affect the service life of wearable devices. GO/rGO contains charge, which can replace electrolyte to improve the electrical performance of hydrogels. Meanwhile, a stable network structure is formed in hydrogels with GO/rGO via hydrogen bonding and physical entanglement, so it will not move to the electrodes under the action of an electric field, which is conducive to reducing the corrosion of electrodes. GO and rGO are similar in structure and play the same role in reducing electrode corrosion [70]. GO and rGO cannot be

used to completely solve the problem of electrode corrosion, and further research is needed.

1.16 Summary and outlook

In conclusion, GO composite hydrogels have both advantages and challenges in the field of wearable devices. GO can significantly improve the mechanical and electrical properties of hydrogels, which is helpful for assembling better wearable devices. However, there are still many challenges to be solved in this field, such as the low mechanical strength, the rapid evaporation of water, insufficient self-healing ability to meet the actual needs and rapid electrode corrosion, etc. Nevertheless, we believe that GO composite
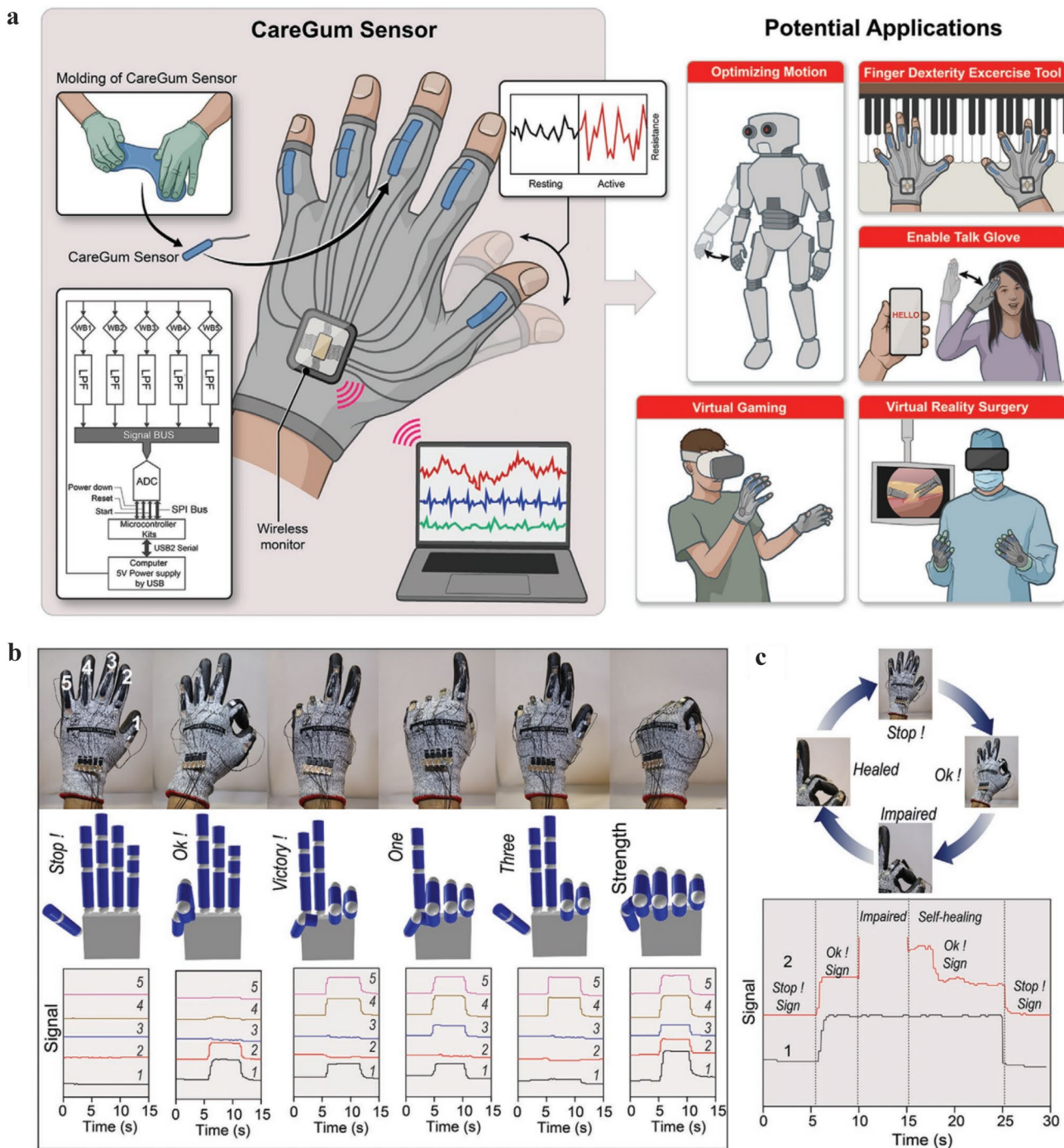
Fig. 10 A self-healable bionic E-glove. **a** Schematic diagram showing the E-glove fabrication and its potential applications. **b** Photos showing various hand gestures monitored with E-glove and the corresponding images of the virtual hand in different positions as well as the associated signal responses from five fingers (five channels). **c** Photos showing different hand gestures with E-glove and their corresponding signal changes. Reprinted with permission from reference [73]. Copyright 2021 Wiley-VCH GmbH

hydrogels can be applied to wearable devices in the future, through which not only the movement of the human body can be detected, but the pulse, blood pressure, respiration and other small signals of the human body can also be monitored in real-time with a high resolution. This can help us better understand our own physical conditions and respond to possible risks in advance. In conclusion, we hope that this review can provide some new ideas and inspiration for the further development of GO composite hydrogels in the field of wearable devices.

Declarations

Conflicts of interest There are no conflicts of interest to declare.

References

1. Cho B, Yoon J, Lim SK, Kim AR, Kim DH, Park SG et al (2015) Chemical sensing of 2D graphene/MoS₂ heterostructure device. ACS Appl Mater Interfaces 7(30):16775–16780. https://doi.org/10.1021/acsami.5b04541

2. Jiang J, Bai S, Zou J, Liu S, Hsu J-P, Li N et al (2022) Improving stability of MXenes. Nano Res 15(7):6551–6567. https://doi.org/10.1007/s12274-022-4312-8

3. Li N, Peng J, Ong W-J, Ma T, Arramel ZP et al (2021) MXenes: An emerging platform for wearable electronics and looking beyond. Matter 4(2):377–407. https://doi.org/10.1016/j.matt.2020.10.024

4. Sun J, Lu C, Song Y, Ji Q, Song X, Li Q et al (2018) Recent progress in the tailored growth of two-dimensional hexagonal boron nitride via chemical vapour deposition. Chem Soc Rev 47(12):4242–4257. https://doi.org/10.1039/c8cs00167g

5. Yu W, Sisi L, Haiyan Y, Jie L (2020) Progress in the functional modification of graphene/graphene oxide: a review. RSC Adv 10(26):15328–15345. https://doi.org/10.1039/d0ra01068e

6. Liu G, Shi Y, Wang L, Song Y, Gao S, Liu D et al (2019) Reduced graphene oxide/polypyrrole composite: an advanced electrode for high-performance symmetric/asymmetric supercapacitor. Carbon Lett 30(4):389–397. https://doi.org/10.1007/s42823-019-00108-x

7. Novoselov KS, Fal'ko VI, Colombo L, Gellert PR, Schwab MG, Kim K (2012) A roadmap for graphene. Nature 490(7419):192–200. https://doi.org/10.1038/nature11458

8. Dreyer DR, Park S, Bielawski CW, Ruoff RS (2010) The chemistry of graphene oxide. Chem Soc Rev 39(1):228–240. https://doi.org/10.1039/b917103g

9. Kumar MIS, Kirupavathy SS, Shalini S (2021) Exploration on reduced graphene oxide/strontium pyro niobate electrode material for electrochemical energy storage applications. Carbon Lett 31(4):619–633. https://doi.org/10.1007/s42823-020-00203-4

10. Mathew EE, Balachandran M (2021) Crumpled and porous graphene for supercapacitor applications: a short review. Carbon Lett 31(4):537–555. https://doi.org/10.1007/s42823-021-00229-2

11. Konios D, Stylianakis MM, Stratakis E, Kymakis E (2014) Dispersion behaviour of graphene oxide and reduced graphene oxide. J Colloid Interface Sci 430:108–112. https://doi.org/10.1016/j.jcis.2014.05.033

12. Sun JY, Keplinger C, Whitesides GM, Suo Z (2014) Ionic skin. Adv Mater 26(45):7608–7614. https://doi.org/10.1002/adma.201403441

13. Xia M, Pan N, Zhang C, Zhang C, Fan W, Xia Y et al (2022) Self-powered multifunction ionic skins based on gradient polyelectrolyte hydrogels. ACS Nano 16(3):4714–4725. https://doi.org/10.1021/acsnano.1c11505

14. Li W, Zhang J, Niu J, Jin X, Qian X, Xiao C et al (2022) Self-powered and high sensitivity ionic skins by using versatile organogel. Nano Energy. https://doi.org/10.1016/j.nanoen.2022.107359

15. Yang M, Chen Y, Wang H, Zou Y, Wu P, Zou J et al (2022) Solvothermal preparation of CeO₂ nanoparticles-graphene nanocomposites as an electrochemical sensor for sensitive detecting pentachlorophenol. Carbon Lett 32(5):1277–1285. https://doi.org/10.1007/s42823-022-00353-7

16. Yoon JH, Park HJ, Park SH, Lee KG, Choi BG (2019) Electrochemical characterization of reduced graphene oxide as an ion-to-electron transducer and application of screen-printed all-solid-state potassium ion sensors. Carbon Lett 30(1):73–80. https://doi.org/10.1007/s42823-019-00072-6

17. Compton OC, Nguyen ST (2010) Graphene oxide, highly reduced graphene oxide, and graphene: versatile building blocks for carbon-based materials. Small 6(6):711–723. https://doi.org/10.1002/smll.200901934

18. Amin I, Batyrev E, de Vooys A, van der Weijde H, Shiju NR (2022) Covalent polymer functionalization of graphene/graphene oxide and its application as anticorrosion materials. 2D Mater. 9:032002

19. Xiang X, Zhu Y, Yin M, Xia S, Guo C (2022) Study of microwave reduction of graphene oxide suspension: structure and functional groups. J Mater Sci 57(5):3280–3294. https://doi.org/10.1007/s10853-021-06748-7

20. Bu Y, Li K, Xiong Z, Liang Z, Zhang J, Bi Z et al (2022) Evolution of layer distance and structural arrangement of graphene oxide with various oxygen content and functional types in low temperature: a ReaxFF molecular dynamics simulation. Appl Surf Sci. https://doi.org/10.1016/j.apsusc.2021.151390

21. Gao W, Alemany LB, Ci L, Ajayan PM (2009) New insights into the structure and reduction of graphite oxide. Nat Chem 1(5):403–408. https://doi.org/10.1038/nchem.281

22. Xiang X, Zhu Y, Gao C, Du H, Guo C (2021) Study on the structure of reduced graphene oxide prepared by different reduction methods. Carbon Lett 32(2):557–566. https://doi.org/10.1007/s42823-021-00287-6

23. Ying B, Liu X (2021) Skin-like hydrogel devices for wearable sensing, soft robotics and beyond. iScience. 24(11):103174

24. Rahmani P, Shojaei A (2021) A review on the features, performance and potential applications of hydrogel-based wearable strain/pressure sensors. Adv Colloid Interface Sci 298:102553. https://doi.org/10.1016/j.cis.2021.102553

25. Xia S, Feng S, Deng Z, Liang Q, Xiang X, Xie H (2021) The role of reduced graphene oxide as a "2D flexible crosslinking point" in composite hydrogels. Polym Eng Sci 62(2):416–425. https://doi.org/10.1002/pen.25854

26. Chen K, Wang F, Hu Y, Liu M, Liu P, Yu Y et al (2022) Highly stretchable, sensitive, and durable Ag/tannic acid@graphene oxide-composite hydrogel for wearable strain sensors. ACS Appl Polym Mater 4(3):2036–2046. https://doi.org/10.1021/acsapm.1c01880

27. Su Y, Zhao J, Zhan W, Yuan H, Wu L, Sui G et al (2022) A multifunctional hydrogel fabricated via ultra-fast polymerization by graphene oxide-adsorbed liquid metal nanodroplets. Chem Eng J. https://doi.org/10.1016/j.cej.2022.135018

28. Xiang X, Li H, Zhu Y, Xia S, He Q (2021) The composite hydrogel with "2D flexible crosslinking point" of reduced graphene oxide for strain sensor. J Appl Polym Sci. https://doi.org/10.1002/app.50801

29. Wu G, Panahi-Sarmad M, Xiao X, Ding F, Dong K, Hou X (2021) Fabrication of capacitive pressure sensor with extraordinary sensitivity and wide sensing range using PAM/BIS/GO nanocomposite hydrogel and conductive fabric. Compos Part A Appl Sci Manuf. https://doi.org/10.1016/j.compositesa.2021.106373

30. Wei J, Wang R, Pan F, Fu Z (2022) Polyvinyl alcohol/graphene oxide conductive hydrogels via the synergy of freezing and salting out for strain sensors. Sensors. https://doi.org/10.3390/s22083015

31. Mao J, Zhao C, Li Y, Xiang D, Wang Z (2020) Highly stretchable, self-healing, and strain-sensitive based on double-crosslinked nanocomposite hydrogel. Compos Commun 17:22–27. https://doi.org/10.1016/j.coco.2019.10.007

32. Jing X, Mi H-Y, Peng X-F, Turng L-S (2018) Biocompatible, self-healing, highly stretchable polyacrylic acid/reduced graphene oxide nanocomposite hydrogel sensors via mussel-inspired chemistry. Carbon 136:63–72. https://doi.org/10.1016/j.carbon.2018.04.065

33. Zhang Z, Tang L, Chen C, Yu H, Bai H, Wang L et al (2021) Liquid metal-created macroporous composite hydrogels with self-healing ability and multiple sensations as artificial flexible sensors. J Mater Chem A 9(2):875–883. https://doi.org/10.1039/d0ta09730f

34. Zhao R, Xu X, Hu L (2021) Highly strong, stretchable, and conductive reduced graphene oxide composite hydrogel-based sensors for motoring strain and pressure. ACS Appl Polym Mater 3(10):5155–5161. https://doi.org/10.1021/acsapm.1c00898

35. Chen J, Wen H, Zhang G, Lei F, Feng Q, Liu Y et al (2020) Multifunctional conductive hydrogel/thermochromic elastomer hybrid fibers with a core-shell segmental configuration for wearable strain and temperature sensors. ACS Appl Mater Interfaces 12(6):7565–7574. https://doi.org/10.1021/acsami.9b20612

36. Niu S, Chang X, Zhu Z, Qin Z, Li J, Jiang Y et al (2021) Low-temperature wearable strain sensor based on a silver nanowires/graphene composite with a near-zero temperature coefficient of resistance. ACS Appl Mater Interfaces 13(46):55307–55318. https://doi.org/10.1021/acsami.1c14671

37. Wang Q, Li L, Lu Z, Hu X, Li Z, Sun G (2019) Highly dispersed graphene network achieved by using a nanoparticle-crosslinked polymer to create a sensitive conductive sensor. ChemElectroChem 6(19):5006–5013. https://doi.org/10.1002/celc.20190 1152

38. Li G, Li L, Zhang P, Chang C, Xu F, Pu X (2021) Ultra-stretchable and healable hydrogel-based triboelectric nanogenerators for energy harvesting and self-powered sensing. RSC Adv 11(28):17437–17444. https://doi.org/10.1039/d1ra02010b

39. Yin H, Li S, Xie H, Wu Y, Zou X, Huang Y et al (2022) Construction of polydopamine reduced graphene oxide/sodium carboxymethyl cellulose/polyacrylamide double network conductive hydrogel with high stretchable, pH-sensitive and strain-sensing properties. Colloids Surf A Physicochem Eng Asp. https://doi.org/10.1016/j.colsurfa.2022.128428

40. Wu L, Hu Y, Tang P, Wang H, Bin Y (2021) High stretchable, pH-sensitive and self-adhesive rGO/CMCNa/PAA composite conductive hydrogel with good strain-sensing performance. Compos Commun. https://doi.org/10.1016/j.coco.2021.100669

41. Wang M, Chen Y, Khan R, Liu H, Chen C, Chen T et al (2019) A fast self-healing and conductive nanocomposite hydrogel as soft strain sensor. Colloids Surf A Physicochem Eng Asp 567:139–149. https://doi.org/10.1016/j.colsurfa.2019.01.034

42. Wang Y, Gao G, Ren X (2021) Graphene assisted ion-conductive hydrogel with super sensitivity for strain sensor. Polymer. https://doi.org/10.1016/j.polymer.2020.123340

43. Hou W, Luan Z, Xie D, Zhang X, Yu T, Sui K (2021) High performance dual strain-temperature sensor based on alginate nanofibril/graphene oxide/polyacrylamide nanocomposite hydrogel. Compos Commun. https://doi.org/10.1016/j.coco.2021.100837

44. Deng Z, Lin B, Wang W, Bai L, Chen H, Yang L et al (2021) Stretchable, rapid self-healing guar gum-poly(acrylic acid) hydrogels as wearable strain sensors for human motion detection based on Janus graphene oxide. Int J Biol Macromol 191:627–636. https://doi.org/10.1016/j.ijbiomac.2021.09.051

45. Liu X, Ma Y, Zhang X, Huang J (2021) Cellulose nanocrystal reinforced conductive nanocomposite hydrogel with fast self-healing and self-adhesive properties for human motion sensing.

46. Hsu HH, Zhang X, Xu K, Wang Y, Wang Q, Luo G et al (2021) Self-powered and plant-wearable hydrogel as LED power supply and sensor for promoting and monitoring plant growth in smart farming. Chem Eng J. https://doi.org/10.1016/j.cej.2021.129499

47. Li Y, Yang Y, Liu X, Chen C, Qian C, Han L et al (2021) Highly sensitive and wearable self-powered sensors based on a stretchable hydrogel comprising dynamic hydrogen bond and dual coordination bonds. Colloids Surf A Physicochem Eng Asp. https://doi.org/10.1016/j.colsurfa.2021.127336

48. Zheng X, Gao Y, Ren X, Gao G (2021) Polysaccharide-tackified composite hydrogel for skin-attached sensors. J Mater Chem C 9(9):3343–3351. https://doi.org/10.1039/d0tc05589a

49. Zhang Y, Li M, Han X, Fan Z, Zhang H, Li Q (2021) High-strength and highly electrically conductive hydrogels for wearable strain sensor. Chem Phys Lett. https://doi.org/10.1016/j.cplett.2021.138437

50. Zhang L, Jiang Q, Zhao Y, Yuan J, Zha X, Xie H et al (2022) Strong and tough PAm/SA hydrogel with highly strain sensitivity. J Renew Mater. 10:415

51. Dai X, Long Y, Jiang B, Guo W, Sha W, Wang J et al (2022) Ultra-antifreeze, ultra-stretchable, transparent, and conductive hydrogel for multi-functional flexible electronics as strain sensor and triboelectric nanogenerator. Nano Res 15(6):5461–5468. https://doi.org/10.1007/s12274-022-4153-5

52. Zhang Z, Zheng Z, Zhao Y, Hu J, Wang H (2021) Highly stretchable porous composite hydrogels with stable conductivity for strain sensing. Compos Sci Technol. https://doi.org/10.1016/j.compscitech.2021.108968

53. Jin X, Jiang H, Zhang Z, Yao Y, Bao X, Hu Q (2021) Ultrastretchable, self-adhesive, strain-sensitive and self-healing GO@DA/Alginate/P(AAc-co-AAm) multifunctional hydrogels via mussel-inspired chemistry. Carbohydr Polym 254:117316. https://doi.org/10.1016/j.carbpol.2020.117316

54. Yang X, Cao L, Wang J, Chen L (2020) Sandwich-like polypyrrole/reduced graphene oxide nanosheets integrated gelatin hydrogel as mechanically and thermally sensitive skinlike bioelectronics. ACS Sustain Chem Eng. https://doi.org/10.1021/acssuschemeng.0c01998

55. Zhang H, Guo J, Wang Y, Sun L, Zhao Y (2021) Stretchable and conductive composite structural color hydrogel films as bionic electronic skins. Adv Sci 8(20):e2102156. https://doi.org/10.1002/advs.202102156

56. Zhang H, Yue M, Wang T, Wang J, Wu X, Yang S (2021) Conductive hydrogel-based flexible strain sensors with superior chemical stability and stretchability for mechanical sensing in corrosive solvents. New J Chem 45(10):4647–4657. https://doi.org/10.1039/d0nj05880g

57. Hu K, Zhao Z, Wang Y, Yu L, Liu K, Wu H et al (2022) A tough organohydrogel-based multiresponsive sensor for a triboelectric nanogenerator and supercapacitor toward wearable intelligent devices. J Mater Chem A 10(22):12092–12103. https://doi.org/10.1039/d2ta01503j

58. Xia S, Song S, Li Y, Gao G (2019) Highly sensitive and wearable gel-based sensors with a dynamic physically cross-linked structure for strain-stimulus detection over a wide temperature range. J Mater Chem C 7(36):11303–11314. https://doi.org/10.1039/c9tc03475g

59. Yang T, Wang M, Jia F, Ren X, Gao G (2020) Thermo-responsive shape memory sensors based on tough, remolding and anti-freezing hydrogels. J Mater Chem C 8(7):2326–2335. https://doi.org/10.1039/c9tc05804d

60. Ma D, Wu X, Wang Y, Liao H, Wan P, Zhang L (2019) Wearable, antifreezing, and healable epidermal sensor assembled from long-lasting moist conductive nanocomposite organohydrogel. ACS

Appl Mater Interfaces 11(44):41701–41709. https://doi.org/10.1021/acsami.9b15412

61. Chen H, Huang J, Liu J, Gu J, Zhu J, Huang B et al (2021) High toughness multifunctional organic hydrogels for flexible strain and temperature sensor. J Mater Chem A 9(40):23243–23255. https://doi.org/10.1039/d1ta07127k

62. Xie Z, Chen Z, Hu X, Mi H-Y, Zou J, Li H et al (2022) Ultrastretchable, self-healable and adhesive composite organohydrogels with a fast response for human-machine interface applications. J Mater Chem C 10(21):8266–8277. https://doi.org/10.1039/d2tc00932c

63. Zhang J, Zhao X, Wang Z, Liu Z, Yao S, Li L (2022) Antibacterial, antifreezing, stretchable, and self-Healing organohydrogel electrode based triboelectric nanogenerator for self-powered biomechanical sensing. Adv Mater Interfaces. https://doi.org/10.1002/admi.202200290

64. Zhao R, Jiang L, Zhang P, Li D, Guo Z, Hu L (2022) Graphene oxide-based composite organohydrogels with high strength and low temperature resistance for strain sensors. Soft Matter 18(6):1201–1208. https://doi.org/10.1039/d1sm01655e

65. Xie Z, Li H, Mi H-Y, Feng P-Y, Liu Y, Jing X (2021) Freezing-tolerant, widely detectable and ultra-sensitive composite organohydrogel for multiple sensing applications. J Mater Chem C 9(31):10127–10137. https://doi.org/10.1039/d1tc02599f

66. Feng S, Li Q, Wang S, Wang B, Hou Y, Zhang T (2019) Tunable dual temperature-pressure sensing and parameter self-separating based on ionic hydrogel via multisynergistic network design. ACS Appl Mater Interfaces 11(23):21049–21057. https://doi.org/10.1021/acsami.9b05214

67. Wang T, Zhang X, Wang Z, Zhu X, Liu J, Min X et al (2019) Smart composite hydrogels with pH-responsiveness and electrical conductivity for flexible sensors and logic gates. Polymers. https://doi.org/10.3390/polym11101564

68. Gao Q, Pan P, Shan G, Du M (2021) Bioinspired stimuli-responsive hydrogel with reversible switching and fluorescence behavior served as light-controlled soft actuators. Macromol Mater Eng. https://doi.org/10.1002/mame.202100379

69. Chen Z, Chen Y, Hedenqvist MS, Chen C, Cai C, Li H et al (2021) Multifunctional conductive hydrogels and their applications as smart wearable devices. J Mater Chem B 9(11):2561–2583. https://doi.org/10.1039/d0tb02929g

70. Xiang X, He Q, Xia S, Deng Z, Zhang H, Li H (2022) Study of capacitance type flexible electronic devices based on polyacrylamide and reduced graphene oxide composite hydrogel. Eur Polym J. https://doi.org/10.1016/j.eurpolymj.2022.111200

71. He F, You X, Gong H, Yang Y, Bai T, Wang W et al (2020) Stretchable, biocompatible, and multifunctional silk fibroin-based hydrogels toward wearable strain/pressure sensors and triboelectric nanogenerators. ACS Appl Mater Interfaces 12(5):6442–6450. https://doi.org/10.1021/acsami.9b19721

72. Pan X, Wang Q, He P, Liu K, Ni Y, Chen L et al (2020) A bionic tactile plastic hydrogel-based electronic skin constructed by a nerve-like nanonetwork combining stretchable, compliant, and self-healing properties. Chem Eng J. https://doi.org/10.1016/j.cej.2019.122271

73. Kadumudi FB, Hasany M, Pierchala MK, Jahanshahi M, Taebnia N, Mehrali M et al (2021) The manufacture of unbreakable bionics via multifunctional and self-healing silk-graphene hydrogels. Adv Mater 33(35):e2100047. https://doi.org/10.1002/adma.202100047

74. Ates HC, Yetisen AK, Güder F, Dincer C (2021) Wearable devices for the detection of COVID-19. Nat Electron 4(1):13–14. https://doi.org/10.1038/s41928-020-00533-1

75. Lee SH, Kim YS, Yeo MK, Mahmood M, Zavanelli N, Chung C et al (2022) Fully portable continuous real-time auscultation with a soft wearable stethoscope designed for automated disease diagnosis. Sci Adv 8(21):12. https://doi.org/10.1126/sciadv.abo5867

76. Lin R, Kim HJ, Achavananthadith S, Xiong Z, Lee JKW, Kong YL et al (2022) Digitally-embroidered liquid metal electronic textiles for wearable wireless systems. Nat Commun 13(1):2190. https://doi.org/10.1038/s41467-022-29859-4

77. Hong S, Gu Y, Seo JK, Wang J, Liu P, Meng YS et al (2019) Wearable thermoelectrics for personalized thermoregulation. Sci Adv 5(5):11. https://doi.org/10.1126/sciadv.aaw0536

78. Jiang YW, Zhang ZT, Wang YX, Li DL, Coen CT, Hwaun E et al (2022) Topological supramolecular network enabled high-conductivity, stretchable organic bioelectronics. Science 375(6587):1411–1417. https://doi.org/10.1126/science.abj7564

79. Qin T, Liao W, Yu L, Zhu J, Wu M, Peng Q et al (2022) Recent progress in conductive self-healing hydrogels for flexible sensors. J Polym Sci. https://doi.org/10.1002/pol.20210899

80. Deng Z, Hu T, Lei Q, He J, Ma PX, Guo B (2019) Stimuli-responsive conductive nanocomposite hydrogels with high stretchability, self-healing, adhesiveness, and 3D printability for human motion sensing. ACS Appl Mater Interfaces 11(7):6796–6808. https://doi.org/10.1021/acsami.8b20178

81. Taylor DL, In Het Panhuis M (2016) Self-healing hydrogels. Adv Mater 28(41):9060–9093. https://doi.org/10.1002/adma.201601613