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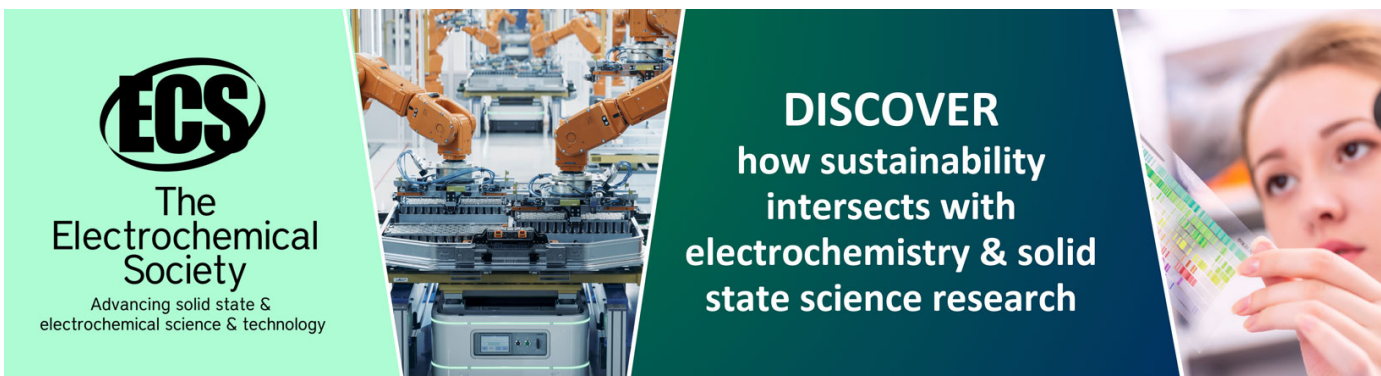
Flexible and wearable sensor based on graphene nanocomposite hydrogels

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Flexible and wearable sensor based on graphene nanocomposite hydrogels

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Abstract

Flexible and wearable sensor based on nanocomposite hydrogels has been proposed for monitoring the human large-scale, small-scale movements and several physiological signals. The nanocomposite hydrogel, prepared from graphene oxide (GO), polyvinyl alcohol (PVA) and polydopamine (PDA), exhibits excellent mechanical and electrical properties with tensile stress of 146.5 KPa, fracture strain of 2580%, fracture energy of 2390.86 KJ m⁻³, and the conductivity of 5 mS cm⁻¹. In addition, it possesses other merits including good self-healing with the electrical self-healing efficiency of 98% of its original resistance within 10 s, and strong self-adhesion onto a variety of surfaces of materials. This self-adhesive, self-healing, graphene-based conductive hydrogel can further assembled as wearable sensors to accurate and real-time detect the signals of human large-scale motions (including bending and stretching fingers joints, wrists joints, elbows joints, neck joints and knees joints) and small-scale motions (including swallowing, breathing and pulsing) through fracturing and recombination of reduced graphene oxide (rGO) electrical pathways in porous structures of hydrogel networks. Furthermore, the hydrogel can also be used as self-adhesive surface electrodes to detect human electrophysiological (ECG) signals. Therefore, the hydrogel-based wearable sensor is expected to be used for long-term and continuous monitoring human body motion and detecting physiological parameters.

Supplementary material for this article is available [online](#)

Keywords: hydrogel, graphene, self-adhesion, self-healing, wearable sensor

(Some figures may appear in colour only in the online journal)

1. Introduction

Continuous and long-term monitoring of human physiological parameters or kinetism plays an important role in chronic disease management, rehabilitation training and postoperative monitoring. Therefore, increasing attention has been paid to wearable medical devices [1–4]. However, traditional wearing

devices based on metal or semiconductor are difficult to be widely accepted by users due to wearing uncomfortable or the data obtained from devices is not accurate enough. Fortunately, the recent emergence of flexible wearable sensors has a great improvement in wearing comfort compared with the traditional wearing sensor [5, 6]. However, there remains some issues to be addressed, such as poor self-healing, needing for bandages to adhere to the skins, and low sensitivity. Therefore, it is still a great challenge to exploit a flexible wearable sensor with high sensitivity, self-adhesiveness and self-healing [7, 8].

Hydrogels [9–11] have become a promising candidate for flexible wearable devices due to their mechanical properties



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are similar to that of human tissues, particularly the unique self-healing properties have potential to blur the boundary between devices and human body. To enhance the electrical conductivity of hydrogels, conducting polymers such as polypyrrole [12], polyaniline [13], and poly (3, 4-ethylenedioxythiophene) [14] have recently gained increasing popularity in hydrogel as attractive materials. Darabi *et al* prepared a conductive self-healing hydrogel by chemical polymerization of acrylic monomers onto PPy grafted to double-bonded modified chitosan [15]. However, it was found that when a conducting polymer monomer was used as the conductive component in hydrogel solution, its self-polymerization characteristic resulted in aggregation, which was detrimental to the conductivity of the hydrogel. In addition, incorporating the carbon nanomaterials such as graphene and carbon nanotubes as conductive fillers into polymer networks is a new method to prepare flexible sensors in recent years [16–19]. This method can effectively endow the composite hydrogel with good electrical and mechanical properties as well as self-healing capability. Liao *et al* presented a conductive, self-adhesive, hybrid network hydrogel with conductive functionalized single-walled carbon nanotubes. The resulting hydrogel exhibits fast self-healing ability (within 2 s), high self-healing efficiency (99%) [20]. It shows that self-healing can extend the service life and enhance the anti-fatigue during their lifetime by repairing themselves as well as recovering to their original networks and function states. Although a variety of hydrogels based flexible sensors have been proposed, synergistic combination of mechanical and electrical properties has not yet been solved well [21–23].

Graphene is considered to be a desirable 2D material in the preparation of high strength and multifunctional hydrogels owing to its high modulus (1.1 TPa), high strength (125 GPa), large specific surface area (2630 m² g⁻¹), good electrical conductivity and biocompatibility [24, 25]. GO is synthesized by replacing of partial double bonds on graphene sheet surface by oxygen-containing functional groups such as hydroxyl group (–OH), carboxyl group (–COOH) and epoxy group (–C–O–C) [26, 27]. GO, compared with graphene, can be more stably dispersed in a variety of organic solvents because of the oxygen-containing groups, provide the conditions for the preparation of high-strength multifunctional composite hydrogels [24]. Annabi *et al* successfully proved that graphene-based nanocomposite hydrogels can synergistically combine high electrical conductivity and mechanical property with good *in vitro* as well as *in vivo* biocompatibility [28]. Typically, graphene-based hydrogels are prepared by the selection of a suitable chemical reducing agent using aqueous solution of GO dispersion as the precursor. However, since the reduction process is usually accompanied by the removal of oxygen-containing functional groups, avoiding the agglomeration of reduction products needs to be addressed [29–31].

In addition, the differences in stretchability and bending stiffness between human tissues or skins and the materials of sensors can cause mechanical failures such as delamination, which can seriously affect the reliability of the measured data of sensors [32]. Although sticking an adhesive such as polyacrylate adhesives, scotch tapes, and bandages to human skins

can partly improve the reliability of data acquisition of sensors, it does not fundamentally solve the conformal contact between the skins of human body and the surfaces of sensors [33, 34]. The PDA, featured with strong adhesiveness as well as durability and stability, has a molecular structure similar to that of mussel adhesion proteins [35, 36]. Dopamine (DA) monomer is easily self-polymerized to construct the PDA under oxygen and alkaline conditions (pH > 7.5), which has strong binding force with not only various metals but also nanomaterials [37]. It further promotes the uniform dispersion of nanofillers in the hydrogel networks, thus enhancing the electrical and mechanical stability of the hydrogels [38]. Moreover, Yang *et al* reported that, in the process of oxidative self-polymerization to form PDA, DA molecules can simultaneously reduce the GO to rGO, which is uniformly dispersed in a solvent [39]. Han *et al* obtained a conductive composite hydrogel with excellent self-adhesiveness and electrical conductivity by partially converting GO through PDA reduction to partially reduced GO (prGO), which successfully constructed perfect conductive pathways in the hydrogel networks, but the resulting hydrogel was relatively fragile [40].

In this work, we proposed a hydrogel-based wearable sensors by using conductive rGO, biocompatible and highly elastic PVA, and adhesive PDA. The prepared hydrogels possess excellent mechanical and electrical properties with tensile stress of 146.5 KPa, fracture strain of 2580%, and conductivity of 5 mS cm⁻¹, respectively. Moreover, the hydrogels can adhere to almost all types of inorganic and organic surfaces with durability and repeatability, such as copper foils, glasses, rubbers, and human skins. In addition, the fast and excellent self-healing properties without any other external factors is another attractive feature of the proposed flexible sensors. Furthermore, the hydrogel based sensors can be mounted directly on human joints and tissues to detect large-scale motions (e.g. bending movements of the knuckles, knees, and necks) and small-scale motions (e.g. swallowing, breathing, and pulsing) due to their excellent biocompatibility and self-adhesiveness.

2. Materials and methods

2.1. Materials

GO was purchased from Nanjing Jicang Nanotechnology Co., Ltd PVA (average polymerization degree: 1750 ± 50) was obtained from Aladdin. DA and sodium borate decahydrate were provided by Sangon Biotech (Shanghai) Co., Ltd Cell Counting kit-8 (CCK-8) was obtained from Dojindo Molecular Technologies and phosphate buffered saline (pH = 7.2–7.6) was purchased from Boster. Fetal bovine serum, Dulbecco's modified eagle medium and 0.05% trypsin ethylenediaminetetraacetic acid were obtained from Solaribio (US). All the solvents in this work were analytical grades.

2.2. Preparation of hydrogels

The PVA-prGO-PDA hydrogels were prepared by the following procedures. Firstly, PVA was dissolved in the deionized water (DI) under vigorous stirring at 95 °C until the

homogeneous solution formed, then 30 mg of GO was dispersed in DI water and sonicated for 60 min. 20 mg of DA and tris-HCl solution were added, and the pH of the mixture was adjusted to 11 using 1 mm NaOH solution followed by sonication for 10 min in an ice bath. The mixture solution was stirred vigorously at 65 °C for 2 h until the solution became deep black and homogeneous. After the PVA solution was added into the mixing solution and the mixture was stirred magnetically at 65 °C for about 6 h, the borax solution and PVA-prGO-PDA solution were mixed by stirring at 60 °C until the formation of hydrogels.

2.3. Characterization

Scanning electron microscope (SEM) images, Fourier transform infrared spectra, and microscopic images were obtained from Hitachi s-3000 N FE-SEM, Nicolet iS50 NIR, and Leica DMI8 inverted fluorescence microscope, respectively. The conductivity and the real-time resistance variation of hydrogels were measured with an electrochemical analyzer (CHI604E, CH Instruments Inc. China) by two-probe method. Tensile adhesion testing was used to measure the adhesive strength of the PVA-prGO-PDA, PVA-GO, PVA-PDA, and PVA hydrogel to porcine skin. The hydrogels were utilized to the surface of the sample with the width of 30 mm and the length of 20 mm. The adhesion tests were immediately suspended once the hydrogels were pulled to failure by a universal testing machine (E43-104, MTS, US). The tensile measurement tests were performed with the same universal testing machine under the surrounding conditions with a cross-head speed of 50 mm min⁻¹. The real-time ECG signals of the flexible sensors were obtained by a medical instrument laboratory system. The CCK-8 assay was used to test the cytotoxicity of the hydrogel to NIH3T3 fibroblasts.

3. Results and discussion

Figure 1 shows the schematic preparation procedure for Graphene-based, stretchable, self-healing, self-adhesive, conductive hydrogel through facile two-step method. Firstly, GO was dispersed in deionized water using ultra-sonication to form the GO dispersion solution. Then the DA was added into the prepared GO suspension at 65 °C for 2 h under alkaline condition. Once the DA was added to the alkaline solution, the pH-triggered oxidation self-polymerization of DA monomer occurred immediately resulting in reduction and surface functionalization of the GO sheets (figure 1(a)). The brown GO suspension gradually turned black as time escaping. Finally, the PVA solution was added into the mixing solution, in the presence of borate, the supramolecular crosslink effects among prGO, PVA and PDA in solution, such as hydrogen bonding, π - π stacking, and dynamic equilibrium interaction between hydroxyl (—OH) group of PVA and borate ion, resulted in the formation of PVA-prGO-PDA hydrogel (figure 1(b)) [41]. The PDA modified rGO was uniformly dispersed in the solution, imparting excellent electrical conductivity to the nanocomposite hydrogel (figure 1(c)). SEM images indicated that the nanosheets were decorated with the PDA microfibers

and interwoven in the PVA-prGO-PDA hydrogel crosslinked network (figure 2(d)). As shown in figure 3, determining the degree of reduction of GO in the system by analyzing the change in the content of oxygen-containing functional groups of GO. The absorption peaks at 1113 cm⁻¹ and 1365 cm⁻¹ are the vibrational absorption peaks of C—O—C and O—C = O, respectively. C = O stretching vibration characteristic peak on the carboxyl groups of GO at 1741 cm⁻¹. The reduction rate of the oxygen-containing functional groups of graphene oxide is 43.5% (table 1), suggesting that GO was successfully partially reduced by PDA. The existence of prGO is beneficial to improve the conductivity of hydrogels. Furthermore, the unreduced GO in the solution formed a strong hydrogen bond with the —OH groups in the PDA and PVA networks, ensuring good mechanical properties of the hydrogel system (figure 1(d)). The non-covalent bond among the PDA and dynamic equilibrium cross-linkages were not fixed but broken and reformed easily, resulting in the reliability of self-healing (figure 1(e)). In addition, the residual catechol group of PDA chains endow the PVA-prGO-PDA hydrogel with excellent self-adhesion (figure 1(f)), which ensure a stable and conformal adhesion onto the human skins without any other adhesive attachments.

The initial contents of GO and DA in the reaction system are crucial to the physical properties of the prepared PVA-prGO-PDA hydrogels. We compared the electrical conductivity of our nanocomposite hydrogels prepared with different contents of GO (the contents of DA in the system was 20 mg). As shown in figure 5(a), the electrical conductivity of the hydrogels increased with the content of GO in system increased. The conductivity of hydrogels could reach 5 mS cm⁻¹ at the content of 30 mg. However, the change of the conductivity was tiny when the content of GO exceeded 30 mg. In addition, the hydrogels were mounted on a rubber plate and stretched by bending the rubber plate at the same bending curvature. We also explored the sensing response of the conductive hydrogels with different GO contents to detect the tensile strain. The results indicated that the sensing response of the PVA-GO (40 mg)-PDA hydrogel did not increase sharply compared with the PVA-GO (30 mg)-PDA (figure 5(b)). Considering the conductivity, the sensing response and the cost of the prepared hydrogel, the PVA-GO (30 mg)-PDA hydrogel was selected in the following experiments.

We further observed the effect of the reaction system containing different contents of DA on the adhesive performance of prepared hydrogels. The results demonstrated that the adhesive strength and elongation of the hydrogel increased with the contents of DA increasing (figure 5(c)). The hydrogels are too sticky to be separated from the surface when the contents of DA exceeded 30 mg. Therefore, considering the adhesive strength and elongation of the prepared hydrogel, the 20 mg of PDA was used in the following experiments. In addition, we evaluated the effect of the contents of borax on the self-healing performance of the hydrogels because this performance of hydrogels is depended on the effective interaction between the cross-linking agent borax and PVA. The results showed that the prepared hydrogel could be obtained in 3:1 (figure 4). And the self-healing performance was evaluated in figures 5(j) and 7.

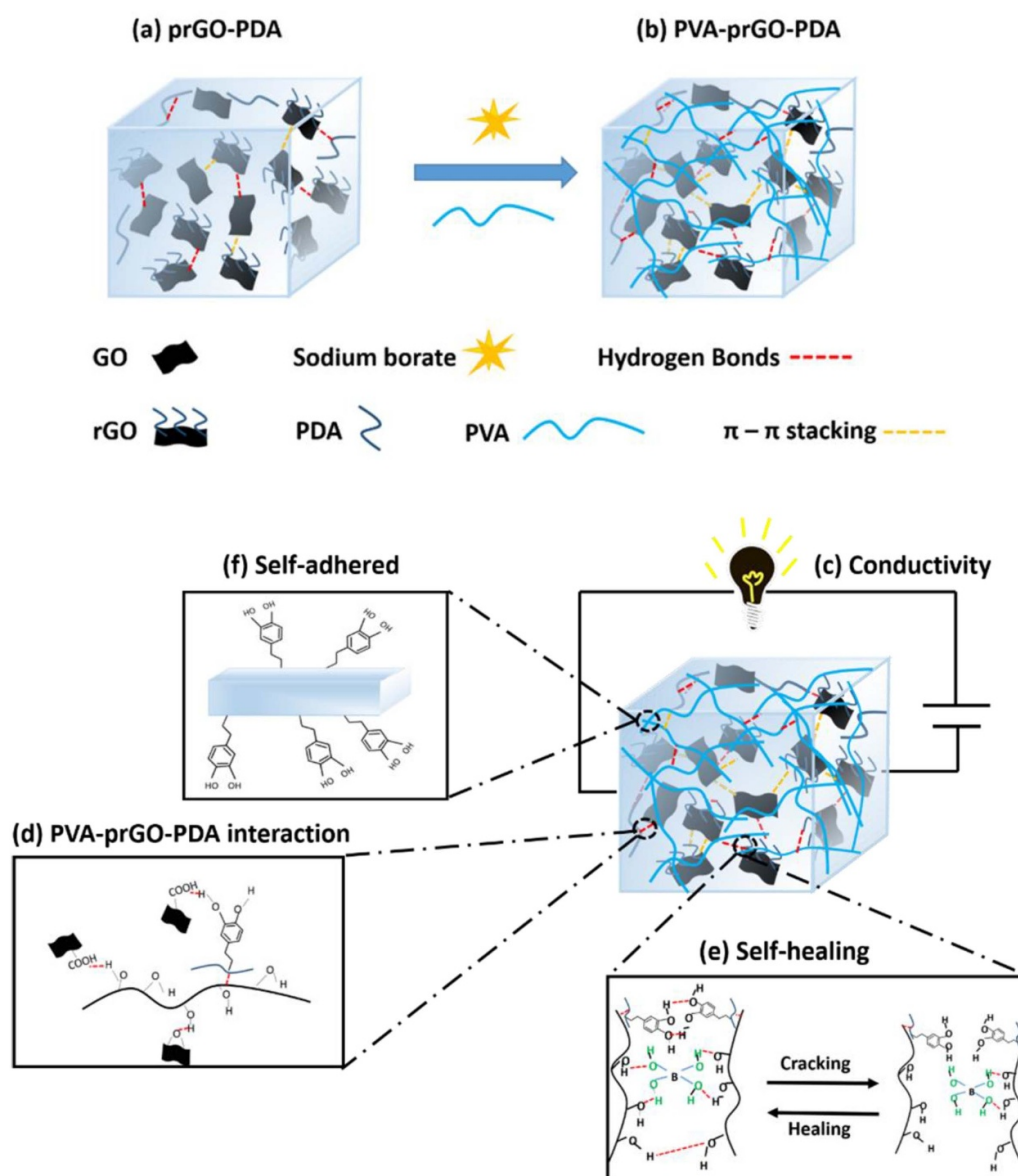


Figure 1. The schematic preparation procedure for Graphene-based, self-healing, self-adhesive conductive hydrogel sensors fabricated via facile two-step method. (a) The conductive prGO cross-linked network via supramolecular crosslinks was formed, (b) the PVA solution was added into the system of the a mixing solution, in the presence of borate, the Graphene-based, self-healing, self-adhesive conductive hydrogel was fabricated, (c) the rGO imparted the hydrogel with conductivity, (d) the interaction between partially unreduced GO with PDA and PVA enhanced the mechanical properties of hydrogels, (e) reversibly self-healing was enabled, (f) the residual catechol group of PDA chains endowed the PVA-prGO-PDA hydrogel with self-adhesion property.

Mechanical properties of materials are one key to fabrication of flexible wearable sensors. Here, we compared among PVA, PVA-PDA, PVA-GO, PVA-prGO-PDA hydrogels using a universal tester with a loading rate of 100 mm min^{-1} at room temperature. All the hydrogels specimens were cut into the width of 20 mm, gauge length of 5 mm and depth of 3 mm. To avoid water volatilization of the test hydrogels specimens were wrapped by the polyethylene film. As demonstrated in figure 5(d), compared with pure PVA hydrogel, PVA-prGO-PDA had a tensile strength of 146.5 KPa, which was almost 8 times of the PVA. In addition, the measured elongation was 2580%, with fracture energy of $2390.86 \text{ KJ m}^{-3}$. In contrast, the PVA-PDA hydrogel had high elongation of 1748%

but low tensile strength of 33 KPa, with fracture energy of 410.82 KJ m^{-3} , and the PVA-GO hydrogel had large tensile strength 250 KPa but low elongation of 987%, with fracture energy of $1836.76 \text{ KJ m}^{-3}$. The typical tensile stress-strain curves of the hydrogels (PVA, PVA-PDA, PVA-GO, and PVA-prGO-PDA) are listed in figure 5(e). These data confirmed that the appropriate addition of GO and DA dramatically improved the mechanical property of the hydrogels. The PVA-prGO-PDA hydrogel obviously obtained both high tensile strength and elongation. The 3D porous cross-linking network formed by PVA, prGO, PDA endowed PVA-prGO-PDA hydrogel with excellent mechanical properties.

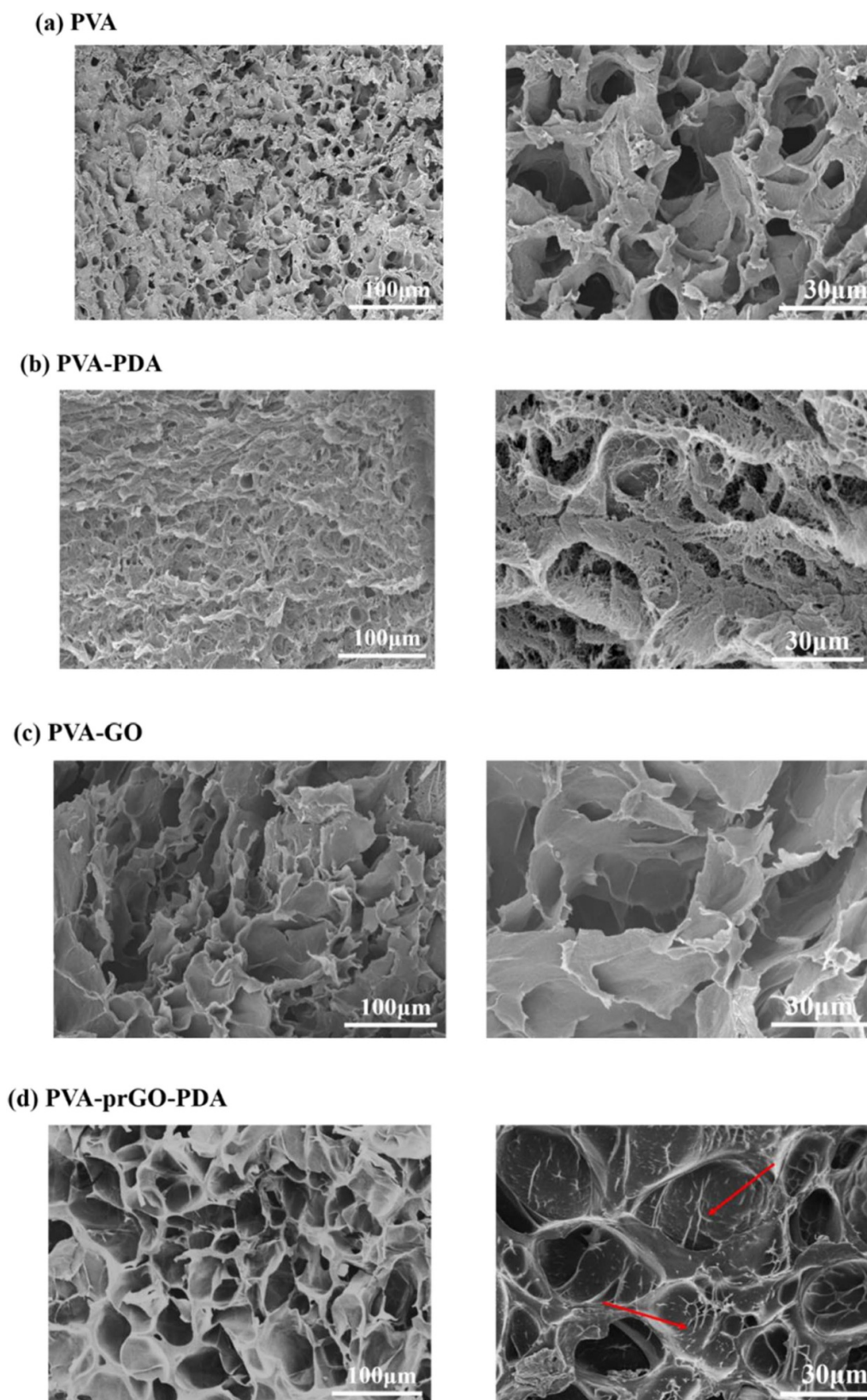


Figure 2. The SEM images of PVA, PVA-PDA, PVA-GO and PVA-prGO-PDA hydrogels. The red arrows in (d) showed the microfibril structures of PDA chains.

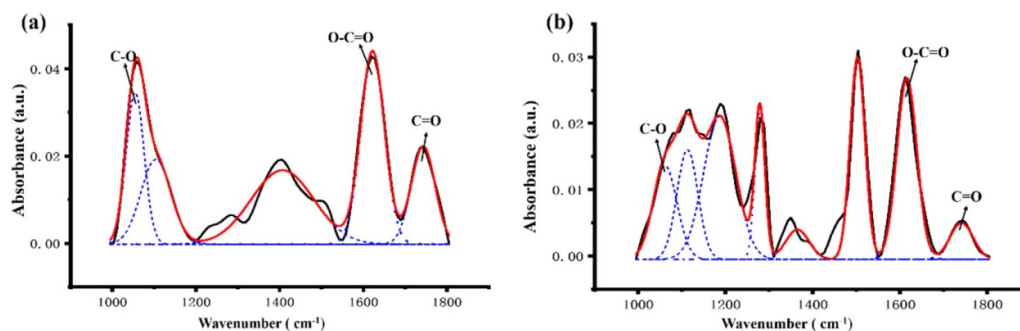


Figure 3. The peaks of the oxygen-containing functional groups of the Fourier transform infrared spectrum of pure GO and the peaks of the oxygen-containing functional group of the Fourier infrared spectrum of PDA-prGO. The black line is the Fourier spectrum curve, the red curve is the fitting curve, and the blue dotted line is the peak fitting curve.

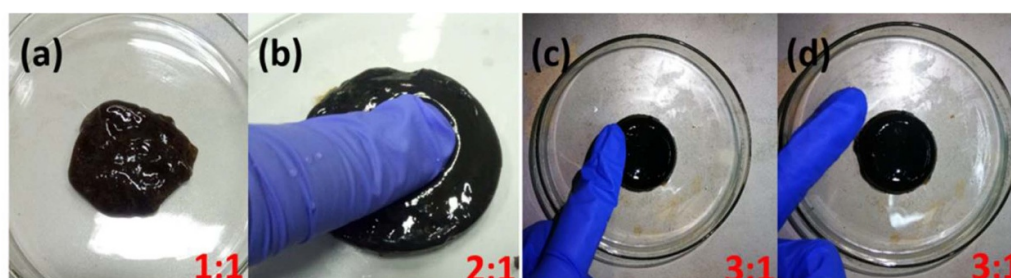


Figure 4. The PVA-prGO-PDA hydrogels were fabricated at the volume ratio of borax to PVA was 1: 1, 2:1, 3:1, respectively. The hydrogel could not be formed when the volume ratio of borax to PVA was 1: 1, and were too soft to be applied to the wearable sensors, and the prepared hydrogel possessed good self-healing performance when the volume ratio of borax to PVA was 3: 1.

Table 1. Peaks fitting results for Fourier transform infrared spectrum of pure GO and PDA-prGO.

Functional groups	Pure GO (%)	PDA-prGO (%)
C = O	13.70484	4.44645
C—O	16.25685	11.90906
O—C = O	27.16966	15.94333
Total oxygen functional groups	57.17491	32.29884
Reduction degree		43.50871

Electrical property of materials is another key to fabrication of flexible wearable sensors. As shown in figure 5(f), pure PVA hydrogel and PVA-GO hydrogel were almost non-conductive. After adding DA, the PVA-prGO-PDA hydrogel had excellent conductivity and the conductivity reached a value of 5 mS cm^{-1} . The excellent electrical properties of PVA-prGO-PDA hydrogels were owing to the presence of rGO. DA monomer was added to the solution, leading to most of the oxygen-containing functional groups of GO were reduced during the self-polymerization process, which obtained rGO with excellent conductivity and good dispersibility. Therefore, the electrical paths were successfully constructed inside the hydrogel to form the PVA-prGO-PDA hydrogel with piezoresistive effect, which provided the foundation for sensitive flexible strain sensors [42]. Furthermore, as shown in figure 5(g), the hydrogel based sensors successfully responded to the rubber board bending. Among the results, the variation of the relative resistance change in the PVA-prGO-PDA hydrogel

reached the value of 300%. Hence, compared with the PVA-PDA hydrogel, the PVA-prGO-PDA hydrogel had stronger effective sensing response. Thus, we believe that the PVA-prGO-PDA hydrogel with sensitive sensing response could efficiently detect the motions of human.

Self-adhesion is essential to ensure the accuracy of measurement data from wearable sensors, furthermore, it can also greatly improve the convenience for wearable sensors. The catechol group on the PDA chains endowed our PVA-prGO-PDA hydrogel with special self-adhesive performance. On the one hand, the free phenol hydroxyl in hydrogel could interact with the $-\text{NH}_2$ groups, $-\text{SH}$ groups and other groups on skin tissue, on the other hand, a cation- π , π - π interaction could be formed between the catechol groups and the contact surfaces. The two kinds of molecular interactions allow the hydrogel to adhere to different material surfaces [43]. As shown in figure 6, the hydrogel adhered firmly to the copper foil, glass, rubber, and human skins surfaces without any other attachments. The adhesive strength of the hydrogels was quantified by tensile adhesion experiment, and porcine skin was employed to simulate the test surface of human skin tissue. The adhesion strength is defined as the ratio of maximum load to interface contact area. The test results indicate that the adhesion strength of pure PVA hydrogel and PVA-GO hydrogel were 2.8 KPa and 2.64 KPa, respectively. In contrast, the incorporation of DA into the PVA-GO hydrogel could adhere effectively to the skins with a strong adhesion strength of 13.04 KPa (figure 5(h)), and the elongation of the material was significantly enhanced by increasing

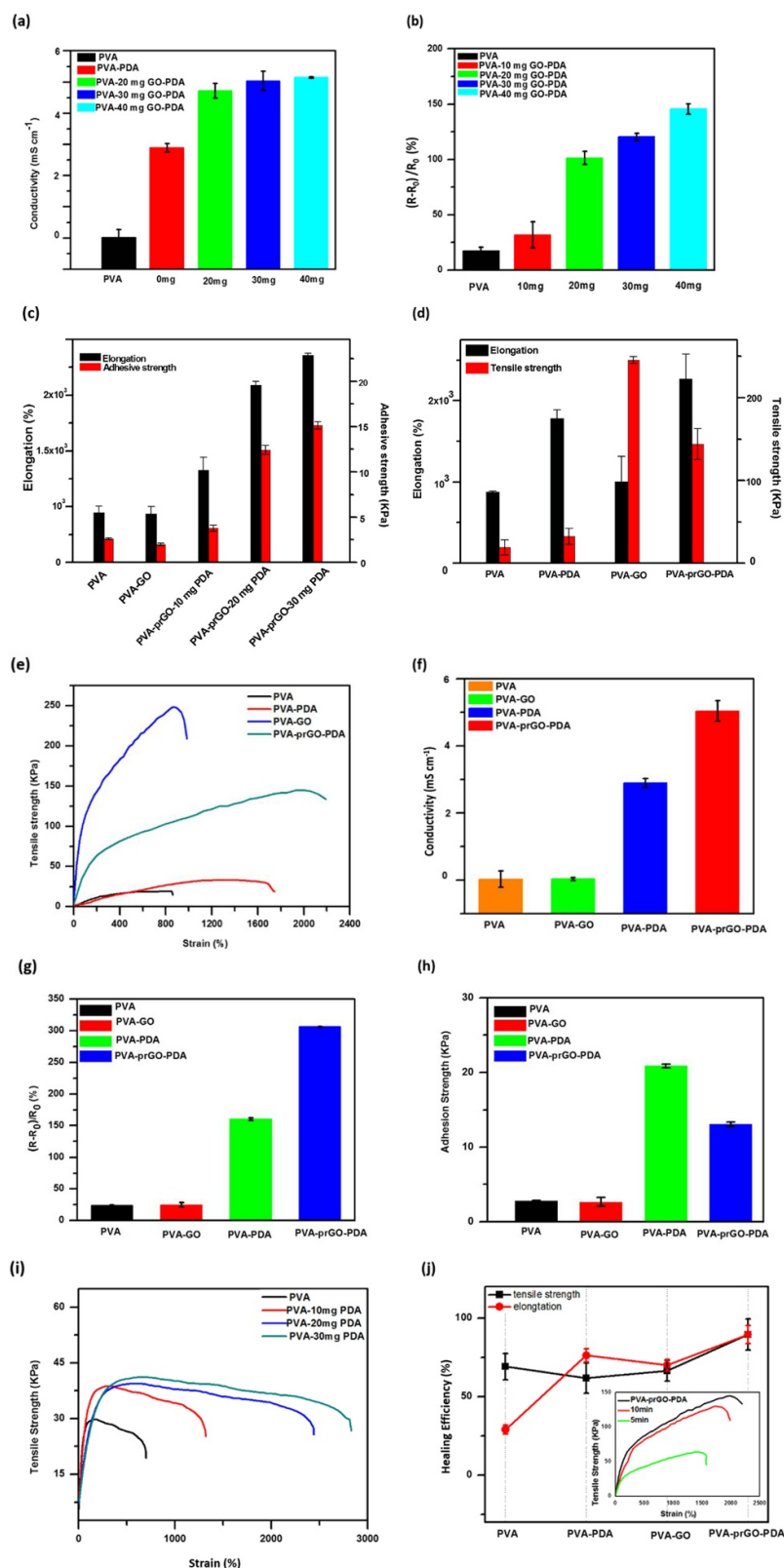


Figure 5. Characterization of conductive, self-adhesive, mechanical and self-healing properties of PVA-prGO-PDA hydrogel. (a) Conductivity with different GO contents of the hydrogel, (b) the sensing response of the hydrogels based sensors with different GO contents utilizing the bent rubber board at the same bending curvature, (c) the adhesive strength and elongation of the hydrogel with different PDA contents, (d) tensile strength and elongation of the hydrogels, (e) typical tensile stress–strain curves of the PVA, PVA-PDA, PVA-GO, and PVA-prGO-PDA hydrogels, (f) conductivity of the PVA, PVA-GO, PVA-PDA and PVA-prGO-PDA hydrogels, (g) the sensing response of the PVA, PVA-GO, PVA-PDA and PVA-prGO-PDA hydrogels based sensors to bend rubber board at the same bending curvature, (h) the adhesive strength of the PVA, PVA-GO, PVA-PDA and PVA-prGO-PDA hydrogels, (i) typical tensile stress–strain curves of the PVA hydrogels with different contents of PDA, (j) the self-healing efficiency of the PVA, PVA-GO, PVA-PDA and PVA-prGO-PDA hydrogels, and inset is the stress–strain curves of the original PVA-prGO-PDA hydrogel and self-healing the hydrogel repaired after 5 min and 10 min.

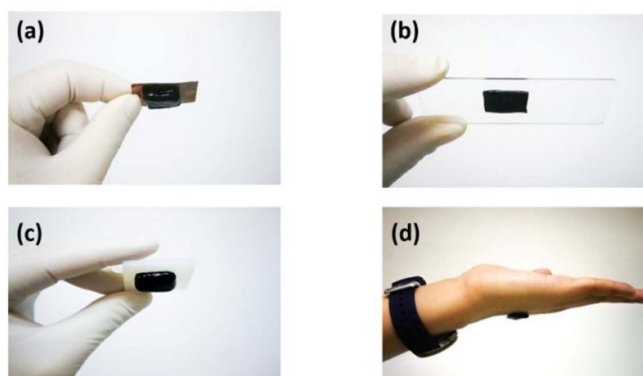


Figure 6. Self-adhesive performance of the PVA-prGO-PDA hydrogels. The PVA-prGO-PDA hydrogels stably adhered to different surfaces. (a) Copper foil, (b) glass, (c) rubber plate, and (d) human skins.

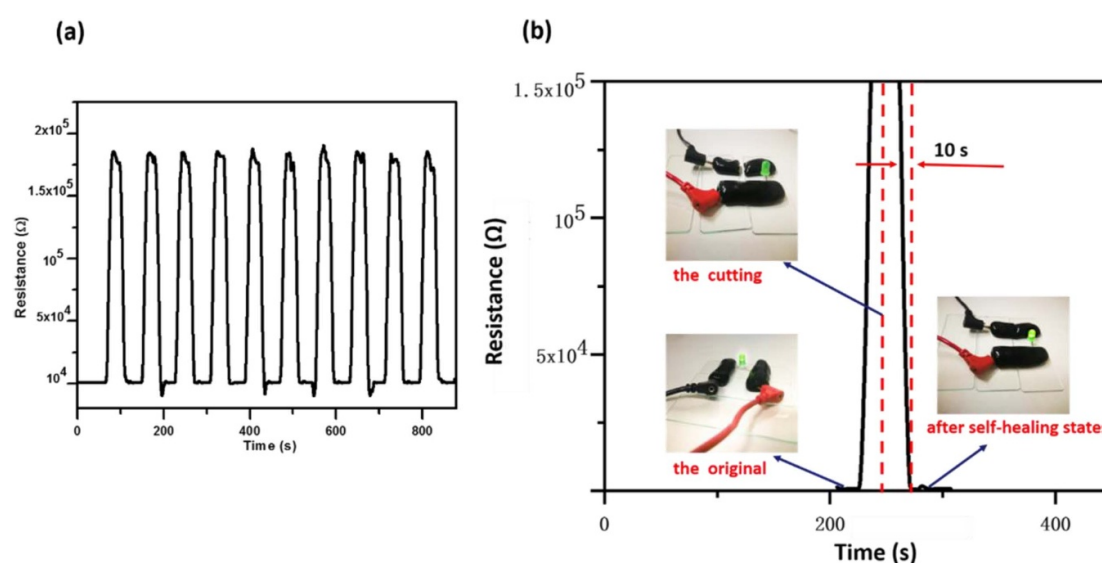


Figure 7. The electrical self-healing efficiency of the hydrogel. (a) The real-time resistance measurements by cutting and reconnecting the conductive PVA-prGO-PDA hydrogel. (b) The cycles of cutting and reconnecting at the same location and a magnified self-healing cycle. The conductive PVA-prGO-PDA hydrogel was connected in series with green LED lamp into the circuit. And three states of the original, the cutting, and after self-healing states were displayed, respectively.

the DA content (figure 5(i)). Therefore, the PDA-prGO-PDA hydrogel showed stable adhesion due to the maintained catechol groups in the PDA for firm adhesiveness. Moreover, the tensile properties of the hydrogel are enhanced, especially the elongation, due to the tangles of PDA and PVA chains in hydrogels.

The self-healing is an important index for the durability of wearable sensors. Due to the existence of non-covalent interaction forces such as hydrogen bonding and π - π interaction [44], the PVA-prGO-PDA hydrogel possessed good self-healing properties, and thus its mechanical and electrical properties could be restored after damaging. The mechanical self-healing efficiency, is defined as the ratio of the healed tensile strength to the original tensile strength and the ratio of the healed elongation to the original elongation, was used to quantify the self-healing properties of the hydrogel. After the fractured hydrogel recovered for 10 min, its maximum elongation decreased to 89.495%, and the healed tensile strength recovered to the initial 89.375% (figure 5(j)). The electrical

properties of PVA-prGO-PDA hydrogel could also be restored, verified by the cutting-connection test. The PVA-prGO-PDA hydrogel was completely cut into two pieces by a blade, afterward, the resistance tended to infinity compared with the initial hydrogel resistance of about 740Ω (figure 7(a)). Then, the resistance of the hydrogel immediately returned to the initial value while the hydrogel had been reconnected. The electrical self-healing efficiency of the hydrogel is defined as the ratio of electrical conductivity after recovery to initial conductivity. As demonstrated in figure 7(b), the hydrogel electrical self-healing efficiency could stably return to 98% within 10 s. Furthermore, in order to demonstrate the self-healing process of hydrogel more intuitively, it was connected in series with a green LED lamp into the circuit, as shown in figure 7(b). At first, the LED lamp was conducted by a direct current power with the voltage of 1.5 V. Then, the LED was extinguished due to the completely separated hydrogel by a blade. The two severed hydrogels were reconnected under the process of self-healing after the contact of two cutting surfaces, meanwhile,

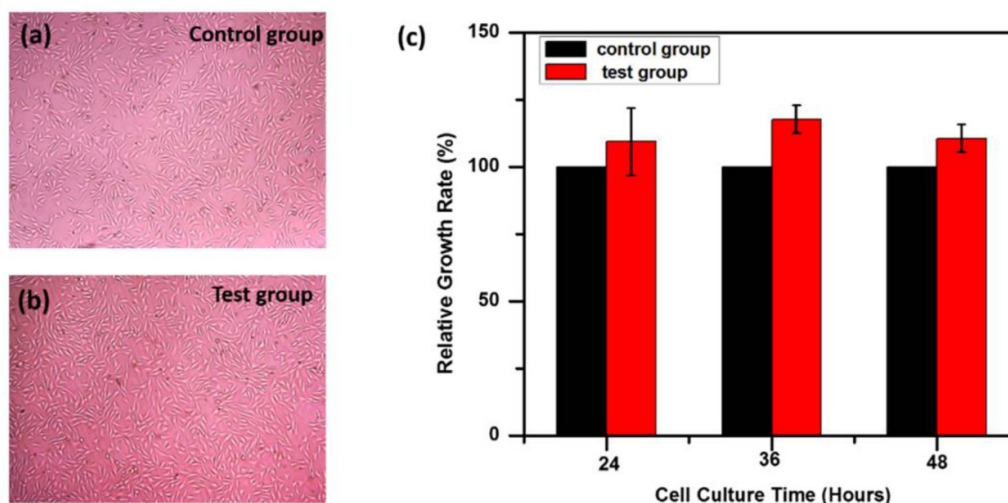


Figure 8. The *in vitro* cytotoxicity results. (a) Microscopic images of NIH3T3 fibroblasts cultured for 48 h in soaking solution substrates of (a) control group (b) test group utilizing the PVA-prGO-PDA hydrogel. (c) The optical density of NIH3T3 fibroblasts cultured for 24, 36, and 48 h.

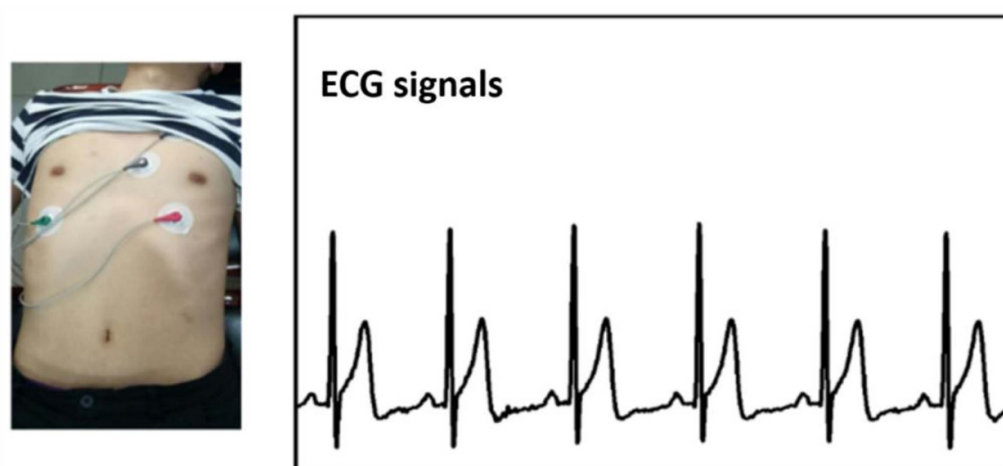


Figure 9. Detect the ECG signals via a medical instrument laboratory system.

the LED was lighted and gradually restored to the initial brightness after about 10 s. Therefore, the PVA-prGO-PDA hydrogel with repeatable and rapid self-healing abilities, and high self-healing efficiency makes it an intriguing material for wearable electronic devices or electronic skins.

The biocompatibility of the wearable PVA-prGO-PDA hydrogel-based sensor by an *in vitro* cytotoxicity test, and the cell toxicity of the hydrogels was evaluated by CCK-8 assay. The *in vitro* cytotoxicity test was performed by using NIH3T3 fibroblasts cultured for 24, 36, and 48 h. Figure 8 demonstrates the results of the *in vitro* cytotoxicity. As shown in figures 8(a) and (b), the NIH3T3 fibroblasts cultured for 48 h in soaking solution substrates of the PVA-prGO-PDA hydrogel exhibited the irregular triangle morphology, indicating that the fibroblasts were in healthy growth state. The quantitative proliferation of the fibroblasts was investigated by CCK-8 assay (figure 8(c)). The relative growth rate value was

higher than that of tissue culture plate after 48 h incubation, this result indicated that the hydrogels do not negatively affect cells adhesion and can even facilitate the fibroblasts growth. In summary, PVA-prGO-PDA hydrogels showed good biocompatibility *in vitro*, which could be used for wearable medical devices for portable and real-time health monitoring systems.

The self-healing, self-adhesive, conductive PVA-prGO-PDA hydrogel could be assembled as a flexible wearable sensor for the detection of several human activities. Figures S1(a)–(e) (available online at stacks.iop.org/SMS/29/075027/mmedia) illustrated the flexible sensors for detecting of several large-scale human motions. The conductive hydrogel was connected to the corresponding analyzer through copper wires to record the signals of resistance changes during the movement of human body. The hydrogels attached directly and firmly onto the joints of fingers, wrists,

elbows, neck and knees to detect the corresponding motion signals, respectively. The sensing response is defined as:

$$N = (R - R_0)/R_0 \cdot 100\%$$

Where R_0 is the initial resistance without applied strain force and R is the resistance with applied strain. In addition to detection of large-scale movements, the small-scale movements such as swallowing, breathing could be detected using our hydrogel-based sensor (figures S1(f)–(g)). Besides, the PVA-prGO-PDA hydrogel also could be used as self-adhesive surface electrodes. As shown in figure 9, the availability of detecting human ECG signals through the proposed electrodes could be compared with that of the commercial medical electrodes under the same parameters setting of the experimental instrument, revealing that the PVA-prGO-PDA hydrogel provides a potential applications in bioelectronics. Particularly, the hydrogel-based sensors adhered onto the skin for a period of time did not cause skin redness or discomfort.

4. Conclusion

In summary, a PVA-prGO-PDA hydrogel-based wearable sensor with outstanding mechanical and electrical properties, reliable self-healing, and self-adhesive performance was designed and successfully fabricated to detect the human large-scale and tiny-scale movements in this work. The PVA-prGO-PDA hydrogel exhibited high tensile strength of 146.5 KPa, adhesive strength of 23.04 KPa and elongation of 2580%. Moreover, the presence of rGO was the essential factor supporting the hydrogel with excellent conductivity, which ensured the sensitive motion sensing for engineering the flexible wearable sensors. The PVA-prGO-PDA hydrogel-based sensors could repeatedly, quickly and precisely detected the movements of human body joints, including fingers joints, wrists joints, elbows joints, neck joints and knees joints motions. Simultaneously, this conductive hydrogel sensor could efficiently monitor the small-scale motions such as swallowing, breathing and pulsing, which provided the feasibility of wearable sensors for portable, remote and real-time health monitoring systems. In addition, the PVA-prGO-PDA hydrogel could be applied as the surface electrodes to detect physiological signals of ECG. Therefore, the hydrogel-based wearable sensor is expected to be used for long-term and continuous detection of human motion and physiological parameters.

Acknowledgments

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Conflicts of interest

The author(s) declare that they have no competing interests.

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