Design and Applications of Graphene-based Flexible and Wearable Physical Sensing Devices

Xinming Li[1,*] and Yang Chai[2,*]

1. Guangdong Provincial Key Laboratory of Nanophotonic Functional Materials and Devices, School of Information and Optoelectronic Science and Engineering, South China Normal University, Guangzhou, 510006, China. Email: xmli@m.scnu.edu.cn

2. Department of Applied Physics, The Hong Kong Polytechnic University, Hung Hom, Hong Kong, China. Email: ychai@polyu.edu.hk

Abstract: The rise of human-machine interaction and the Internet of Things technology requires the development of functionalized sensors that are mechanically flexible and fabulously wearable. Therefore, the emergence of new materials and devices is particularly important for technology design and development. Graphene has the atomically level thickness, mechanical flexibility, lightweightness, and high conductivity and transparence. Especially, the large specific area of graphene enables the perception of external stimuli with high sensitivity, which is expected to be used in flexible sensor technology. In this review, we will introduce the research progress of graphene in flexible physical signal sensors, including the device structure design and the applications of these devices in wearable technology. We will overview the development of new directions of sensors, such as miniaturization, intelligence, and multi-modal. We will also focus on the latest technical progress of related sensing devices and point out the challenges and directions of future development of wearable sensors.

Keywords: Graphene; Two-dimensional materials; Physical sensing technologies; Flexible devices

1. Introduction

Flexible sensing technology is extremely important for the development of the emerging Internet of Things and Intelligent Systems.[1-4] Sensor is a device that can sense other external information or changes, and convert it into electrical signals and transfer them to other devices or systems. Essentially, sensors can be used to detect and learn the interaction of a system with the surrounding environment.[5] Therefore, the sensor is a key component in the perception system. Flexible sensors will release the flexibility of the device, which can be designed as a wearable product. With the development of healthcare technology and the demand for the human-machine interaction, research on flexible and wearable sensors has become more important.

The type of flexible sensor typically requires the device with flexibility, stretchability, lightweight, non-toxicity, and excellent device performance. Graphene is a two-dimensional (2D) layered material with very unique structural characteristics and electrical, optical, and thermal characteristics.[6-9] These properties make graphene to be potentially used in flexible electronic devices, film coatings, and other fields.[9-12] In particular, graphene possesses the material characteristics required for the new flexible and wearable technology, and has become an important material candidate for the development of lightweight and flexible sensing devices.[13-16]

In this review, we will mainly focus on the design of flexible graphene structures and devices for physical signal sensing (**Figure 1**). There are many types of sensors according to the different target signals. Generally, graphene-based sensors can be divided into physical sensors (such as light, electricity, force, and temperature), chemical sensors (such as gas, molecule, ion, PH value, chemical potential, etc.), biosensors (such as biological tissues, microorganisms, biomolecules, proteins, bacteria, viruses, etc.) and related integrated intelligent systems according to the application of the measured signal. In this paper, we will review how to design a flexible physical signal sensor that implements a specific function and demonstrate its related performance and applications. The core technology of physical sensors is to design electronic devices around the measured physical signals, to control the carriers transport and related electrical device properties, so as to achieve the specific sensing functions. Based on this device design technologies, this article introduces the current graphene-based flexible physical sensing devices, and will also identify open questions, challenges, and future directions in this area.

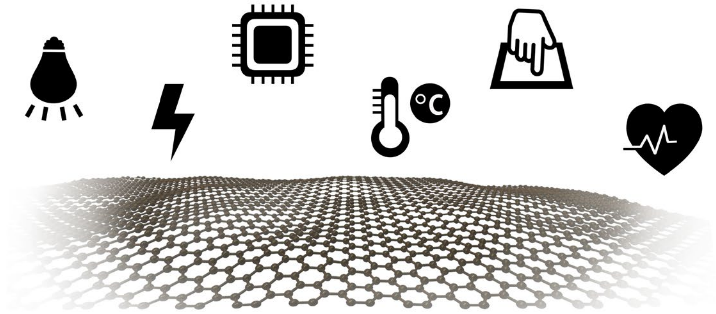

Figure 1 Graphene-based devices for physical signal sensing.

2. Graphene-based Optoelectronic applications

The photodetector is the optoelectronic device that converts light energy into an electrical signal, which detects light signals of a specific wavelength and obtains fast and highly responsive electrical signals. Flexible photodetectors have very important application prospects in wearable devices. For example, the flexible photodetector can simulate the function of the human eyes and detect the different light signal information; or the device can also be used as the wireless signal receiver to achieve the light information conversion for specific functions. Traditional photodetectors often use conventional semiconductor materials (Si, Ge, GaAs, etc) to detect the optical signals, but these semiconductors are hard and brittle that are difficult to be integrated with wearable devices. Graphene has excellent flexibility and optoelectronic characteristics and has the application potential in flexible photodetectors.[17]

Graphene has high carrier mobility and is also a broadband light-absorbing material, which helps it to be used in a flexible or wearable photodetector. However, when graphene absorbs light, its photo-generated carrier lifetime is short and the dark current is too high, which will limit its photoelectric conversion performance. Therefore, we need to design different structures to realize the adjustment of the graphene carrier transport behavior to realize its application in flexible photodetection. For example, graphene can be combined with traditional semiconductors, such as Si,[18] GaAs,[19] to form photodiodes. When the size of these conventional semiconductors is reduced to the nanoscale, their degree of flexibility is significantly improved, so that the flexible photodiode can be realized. For example, Liu *et al.* studied the effect of strain modulation on the performance of the graphene/ZnO nanorods film Schottky junction flexible photodetectors. The photoresponse is enhanced by 17% under the displacement of the atoms of −0.349% strain, which can be ascribed to the strain-induced piezo-potential for the facilitation of electron-hole separation.[20] Similarly, silicon can also be thinned to make it flexible. Schneider *et al.* reported a multispectral photodetector integrated with graphene and amorphous silicon (**Figure 2 (a)**). Compared with traditional devices (coated by aluminum-doped zinc oxide (ZnO: Al) electrodes), the photodetector of graphene/a-Si: H can achieve the 440% enhancement of photoresponse in the 320nm UV region.[21]

In another working mode, the graphene-based phototransistors can exhibit a higher light response. Although graphene has a broad spectral response, its weak light absorption and low gain limit its optoelectronic applications. Therefore, the combination the light-sensitive materials with graphene can realize the transfer of photo-generated carriers of the photosensitive layer to graphene, and increases the photoconductive gain of graphene. For example, the stacking heterostructure based on MoS_2/graphene gated by polymer electrolytes can be fabricated on a flexible substrate, and the internal responsivity of 570A/W can be achieved under ~0.1nW/μm^2 at 642 nm (**Figure 2 (b)**). Meanwhile, the photocurrent remains unchanged under bending conditions (**Figure 2 (c)**).[22] Researchers use different materials and graphene heterostructure to build photodetectors with different spectral responses. For example, Son *et al.* fabricated UV photodetector based on colloidal ZnO quantum-dot (QD) and graphene. The UV photodetector at a 365 nm wavelength showed a ratio of the photocurrent to the dark current with 1.68 after bending.[23] Liu *et al.* reported all-carbon hybrid films for flexible graphene-based photodetector with a high photoresponse of ~ 51 A/W and a response time of ~ 40 ms under the visible range.[24] Methylammonium lead iodide perovskite can also be added as a photosensitive material in the graphene channel, and the hybrid device has a response of 115 A/W at 515 nm.[25] Besides, the flexible graphene/MoS_2/Si can be fabricated when the silicon is thinned to 200 nm and the photoresponse is ~1×10^3 A/W under 168 nW light power with the response time of 0.2 s.[26] Liu *et al.* report a flexible photodetector based on graphene and gold oxide (AuOx) with an excellent light response in different spectral ranges (**Figure 2 (d))**. Due to the good gas impermeability of graphene, it can effectively prevent the oxygen desorption of AuOx, thereby achieving good stability of AuOx. In this structure, the hot electrons generated by graphene are separated at the AuOx–graphene heterojunction (**Figure 2 (e))**, and the photodetector has a high responsivity of 3100A/W at 310 nm (UV), 58 A/W at 500 nm and 9 A/W at 1550 nm under a very low applied bias of 0.1 V (**Figure 2 (f)**).[27]

Meanwhile, the rGO can also be widely used to construct flexible photodetectors. For example, defects and functional groups play a very important role in graphene-based photodetectors. Chang *et al.* reported that rGO exhibits the infrared light responsivity of ~0.7 A/W by oxygen defect engineering.[28] Further, Trung *et al.* reported an improved graphene-based infrared (larger than 800 nm) photodetectors in which the poly(vinylidenefl uoride-cotrifl fluoroethylene) (P(VDF-TrFE)) can increase the infrared response by a factor of two. Importantly, this device can respond sensitively to the infrared radiation from human body, so the photodetectors have a very unique application prospect in medical detection and health monitoring.[29]

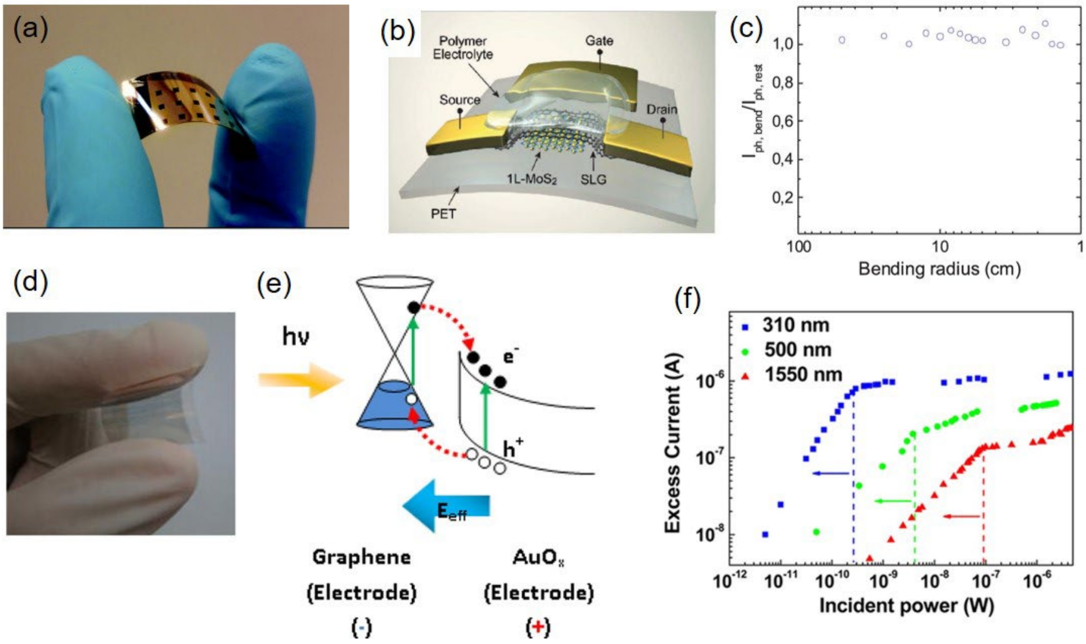

Figure 2 (a) The flexible graphene/Si photodetector.[21] Reprinted with permission from, copyright Royal Society of Chemistry. (b) The schematic illustration of the flexible graphene/MoS2 photodetector gated by polymer electrolyte. (c)The photocurrent as a function of bending radius.[22] Reprinted with permission from, copyright American Chemical Society. (d) The flexible graphene/ AuOx photodetector The scale bars represent 10 mm. (e) The schematic illustration of the band diagram of the graphene/AuOx heterojunction under the illumination. (f) The logarithmic scale of the excess current related to the incident power under various wavelengths of light.[27] Reprinted with permission from, copyright American Chemical Society.

According to the material selection and device design, the flexible photodetectors can be further extended to some relevant specific applications, such as sensors, positioners, etc.[30-36] For example, Kang *et al.* reported a stretchable photodetector based on crumpled graphene. This crumpled photodetector showed the 400% enhancement of photoresponsivity compared with the flat one, and photoresponsivity can be modulated by 100% under 200% applied strain.[30] Furthermore, the graphene/gold stretchable structure can be integrated on the surface of a contact lens, exhibiting a high signal-to-noise ratio under laser irradiation, which is expected to be used in wearable optical detection devices (**Figure 3 (a)**).[31] Moon *et al.* designed simple and flexible self-powered photodetectors based on monolithic rGO papers. When the laser is irradiated at different positions, the graphene film exhibits different voltages, so that the device can realize the position-sensitive detection (**Figure 3(b)**).[32] Importantly, Polat *et al.* designed a wearable photodetector based on graphene and quantum dots that can monitor heart rate, arterial oxygen saturation (SpO2), and respiratory rate (**Figure 3(c)**). Based on the reflectance mode and transmission mode, the graphene-based photodetector can measure the photoplethysmogram (PPG) signal on the wrist (**Figure 3(d)-(f)**).[36]

In addition to this light detection and perception function, graphene-based heterostructures can also be constructed as image sensors and memories.[37] For example, Jang *et al.* reported a photodetector device with a non-volatile memory function for storing photonic signals, which is constructed of pentacene organic semiconductors, gold nanoparticles (AuNPs) and graphene (**Figure 3(g)**). In this structure, the organic semiconductor functions as a light-sensitive material, and photo-generated holes are injected into the graphene, meanwhile, the AuNPs can trap the carriers and generate the plasmonic enhancement effects. Photocurrent can be retention under different light intensity with pulse light, and light information can also be erased by applying −20 V gate voltage (**Figure 3(h)**).[33] Besides, graphene/perovskite photodetectors can be synthesized by a two-step CVD method and the flexible photodetector array (24 × 24 pixels) can be designed (**Figure 3(i)**). The flexible image sensor with color discrimination can be achieved under three wavelength filters (380, 633, and 750 nm) (**Figure 3(j)**).[34] Inspired by the human visual system, the graphene-perovskite quantum dots phototransistors can realize the photonic synaptic behavior through the light-assisted memory effect, and can be used to realize the facial recognition with the aid of machine learning.[38] In addition, the related structural systems can also realize the image sensing and memory functions, and is expected to improve the processing efficiency and image recognition rate.[39-44]

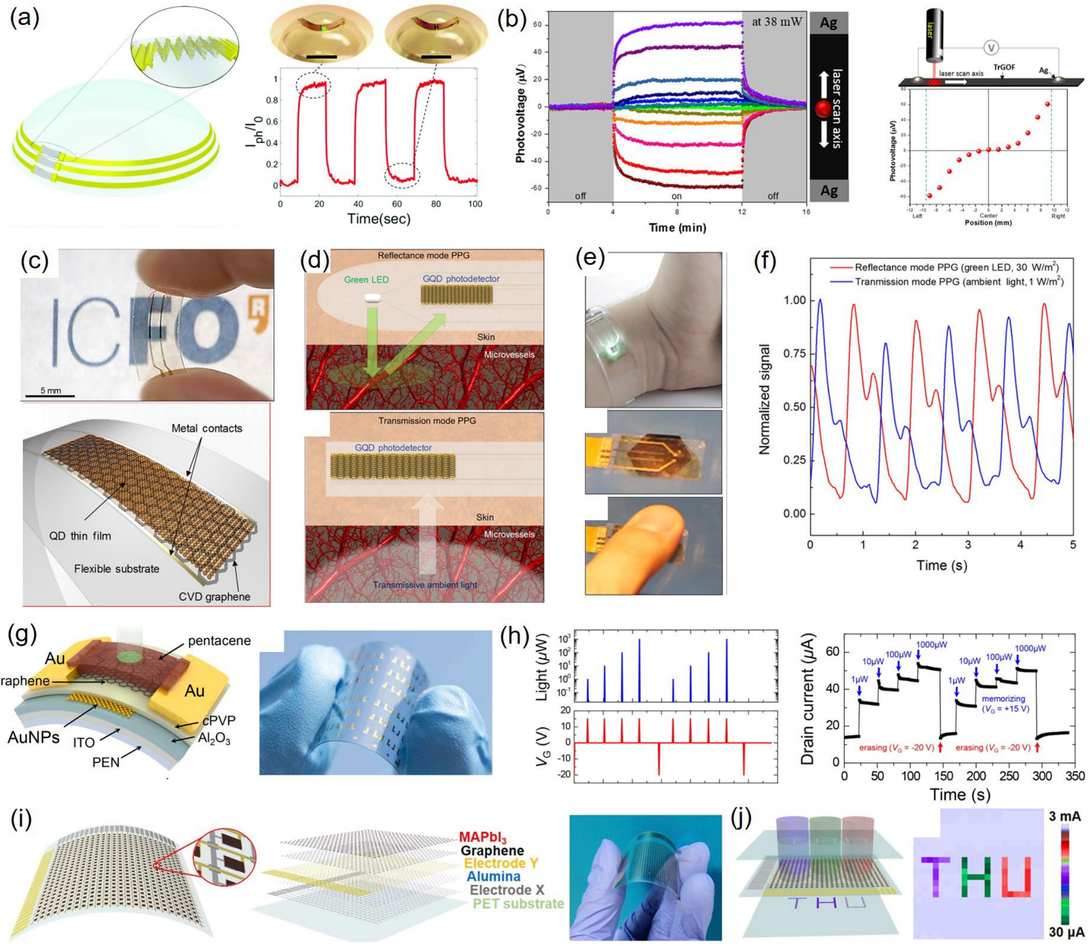

Figure 3 (a) The wrinkled graphene-AuNPs hybrid structure based photodetector integrated on contact lenses and its photoresponse.[31] Reprinted with permission from, copyright Royal Society of Chemistry. (b) When the laser spot illuminates the area of rGO between the electrodes, a photovoltaic response occurs and is related to the position of the laser spot.[32] Reprinted with permission from, copyright Springer Nature Limited. (c) The photographic image and schematic illustration of the flexible graphene sensitized with semiconducting quantum dots photodetector. (d) The schematic illustration and (e) photographic image of photoplethysmogram (PPG) in reflectance mode and transmission mode PPG based on the photodetector. (f) Normalized PPG results for transmission and reflectance modes of the photodetector.[36] Reprinted with permission from, copyright American Association for the Advancement of Science. (g) The schematic illustration and photographic image of the flexible graphene photodetector which is constructed of pentacene organic semiconductors, gold nanoparticles (AuNPs). (h) Memory performance of graphene photodetector.[33] Reprinted with permission from, copyright American Chemical Society. (i) The schematic illustration and photographic image of the flexible graphene/ perovskite photodetector array (24 × 24 pixels). (j) The schematic illustration and corresponding output image of the flexible graphene/ perovskite photodetector image sensor for color discrimination.[34] Reprinted with permission from, copyright Science China Press.

3. Graphene-based Temperature Sensing

Temperature sensors are widely used in daily life and industrial production. In particular, we need to get timely feedback on temperature changes in nature and the human body in the fields of human-machine interaction and medical health. A wearable flexible temperature sensor that can realize real-time monitoring is one of the important solutions. The temperature sensors are mainly realized through various mechanisms, such as thermoelectric effect, thermal resistance effect, thermal expansion, infrared radiation, etc.

For graphene applications in temperature sensors, the change in resistance of graphene with temperature (Temperature coefficient of resistance, TCR) can reflect its related performance. Davaji *et al.* studied the effect of temperature sensing of single-layer graphene on different substrates. Due to the temperature-dependent electron mobility and electron-phonon scattering, the resistance of graphene will increase with the increase of temperature. Meanwhile, compared with the Si substrate and the suspended layer, graphene exhibits a more pronounced temperature response on the SiN substrate due to low thermal mass.[45]

On the other hand, the resistance of rGO films often exhibits negative TCR. The thermal excitation of the carriers in rGO is the main inducement factor, that is, as the temperature increases, the tunneling and hopping ability of carriers between nanosheets are increased, thereby reducing the resistance of the sensor. Liu *et al.* used rGO to make real-time monitoring temperature sensors, and the sensor responds quickly with the time of 0.654 s and the results are the same as those of a commercial thermocouple, which shows the healthcare potential of this sensor application.[46] Furthur, Zeng *et al.* studied the electrical conductivity of rGO film with the annealing temperature from 1000 to 3000 K and found that the rGO film reduced at 3000 K is promising for the temperature detection for a wide range (from 10 to 3000 K).[47] Yan *et al.* further studied the temperature response of graphene/PDMS under tensile conditions. By designing a curved graphene channel, the device can still maintain the tendency of the resistance to decrease with increasing temperature when the device is stretched at a maximum of

50%. This makes it possible to use the device in a stretchable environment.[48] Another common temperature sensing structure is based on a graphene-based composite. For example, Zu *et al.* reported the sensors based on rGO/polyorganosiloxane aerogel array that can detect the temperature (20−100 °C), which can be used for temperature sensing of glass cup with hot water (**Figure 4 (a)-(c)**).[49] Liu *et al.* also found the mixture of carbon black (CB) and rGO on a paper substrate can sense the temperature with the sensitivity of 0.6%/°C, while the working mechanism of sensor proposed is that small molecule (O_2, H_2O, etc.) will be eliminated from the sensing layer under high temperature, resulting in the improvement of conductive pathways.[50]

Recent advances have shown that graphene-based temperature sensors can be used to detect human skin temperature. Vuorinen *et al.* reported a graphene/PEDOT: PSS temperature sensor printed on a skin-conformable polyurethane adhesive bandage. It is found that the sensor exhibits a stable temperature response of 0.06%/°C in a protective argon atmosphere, but due to the mixed electronic and ionic conductive mechanism, the response capability will be reduced in the atmospheric environment.[51] This means that in the future design of flexible and adhesive temperature sensors, special attention should be paid to the impact of the external environment (including the skin surface) on device performance. Further, a casting solution rod casting method can be used to achieve the large-area ferroelectric PVDF/rGO composite films, and can realize the arrays (18 × 12 pixels) by placing the electrode on the top and bottom of the film (**Figure 4 (d)**). The resistance of this sensor reflects a negative TCR, which is mainly due to the contact resistance of rGO nanosheets and the intrinsic behavior of rGO. When the hand is placed on the surface of the sensor array, an image of the entire hand temperature change can be displayed (**Figure 4 (e)**). Besides, through the design of the interface interlocked micro dome array structure (**Figure 4 (f)**), the sensitivity of the sensor unit can be increased from 1.58%/°C to 2.93%/°C (**Figure 4 (g)**). This is because the thermal expansion of PVDF will increase the contact area of the interlocked micro dome array structure and increase the electrical conductivity.[52] Besides, the performance of temperature sensors can be integrated with the mechanical sensors

Figure 4 (h). It can be seen that the integrated sensor attached to the human neck can simultaneously measure skin temperature changes caused by drinking hot water and skin degeneration information caused by drinking water (**Figure 4 (i)**).[53]

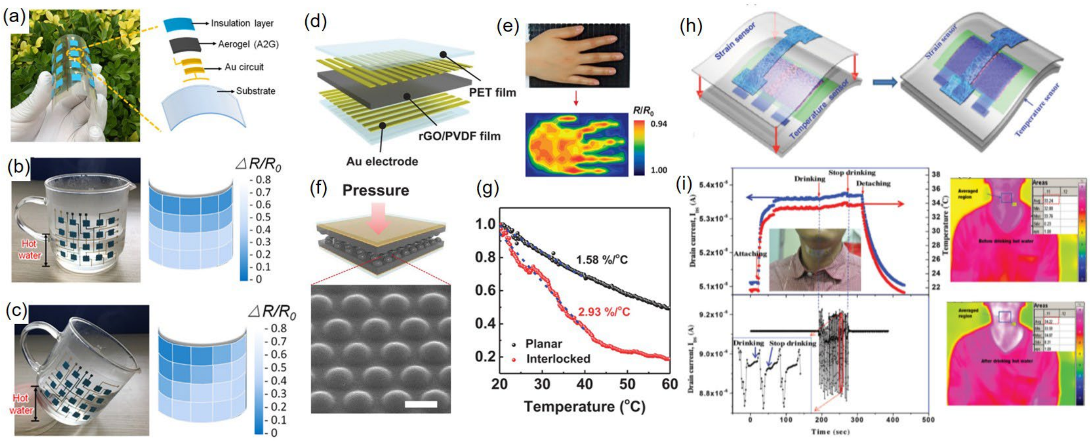

Figure 4 (a) a flexible temperature sensor array with 4 × 5 pixels based on rGO/polyorganosiloxane aerogel. (b) and (c) The sensor array attached to the surface of the glass filled with hot water and its associated temperature signal response.[49] Reprinted with permission from, copyright American Chemical Society. (d) The schematic illustration of the PVDF/rGO sensor array which is sandwiched by the electrode (18 × 12 pixels). (e) Resistance change mapping for the human palm temperature distribution. (f) The schematic illustration and SEM of the interlocked micro dome array structure. Scale bar, 10 μm. (g) Relative resistance change and sensitivity of planar PVDF/rGO sensor and interlocked micro dome structure sensor for 1 wt % rGO.[52] Reprinted with permission from, copyright American Association for the Advancement of Science. (h) The schematic illustration of integrated temperature and strain sensors. (i) Simultaneously monitor the resistance changes in skin temperature and muscle movements of the neck when drinking hot water. Infrared imaging shows the results of changes in neck temperature before and after drinking hot water.[53] Reprinted with permission from, copyright John Wiley & Sons, Inc.

4. Graphene-based Strain and Tactile Applications

Mechanical measurements are extremely widely used in flexible and wearable devices. Many efforts have been made to develop flexible strain and tactile sensors for human motion monitoring, human-computer interaction, electronic skins, and new-type robots. Based on the regulation of geometric deformation, defects, cracks, and carrier transport regulation of materials, this sensor can monitor tiny movements or deformations and record relevant information in various applications, such as human breathing, running, heartbeat, or remote control of the other devices.

Due to its thinness, lightness, and excellent electrical properties, graphene can be used for the applications on the nanoelectromechanical (NEMS) system and the strain or tactile sensing devices.[54-55] In particular, the study of graphene in NEMS systems provides a foundation and application-type bridge for its application in pressure sensors. The related resistance of suspended graphene will increase and the related gauge factor can be from 1.9 to 4.33, after introducing a small strain on it (**Figure 5 (a)**).[56-57] By comparing the electrical behavior with and without the cavity structure, the piezoresistive phenomenon of graphene is proven (**Figure 5 (b) and (c)**). Besides the device is independent of the crystal orientation, and can provide a direct electrical readout to reflect the relevant strain.[57] On the other hand, for suspended multi-layer graphene, a positive piezoconductive effect is observed, that is, the conductance increases with the increasing pressure, and the gauge factor of the tri-layer device was larger than that of the single-layer graphene. This may be due to the strain-induced competition between interlayer coupling and intralayer transport in the multilayer structure.[58] These suspended graphene NEMS devices have very unique monolayer structures and corresponding performances, and have been applied to pressure sensors[56-57, 59-60], accelerometers (**Figure 5 (d)**)[55, 61], electromechanical resonators[62-64], NEMS switches[65], microphones[66-67] (**Figure 5 (e)**) and speakers[68] (**Figure 5 (f)**).

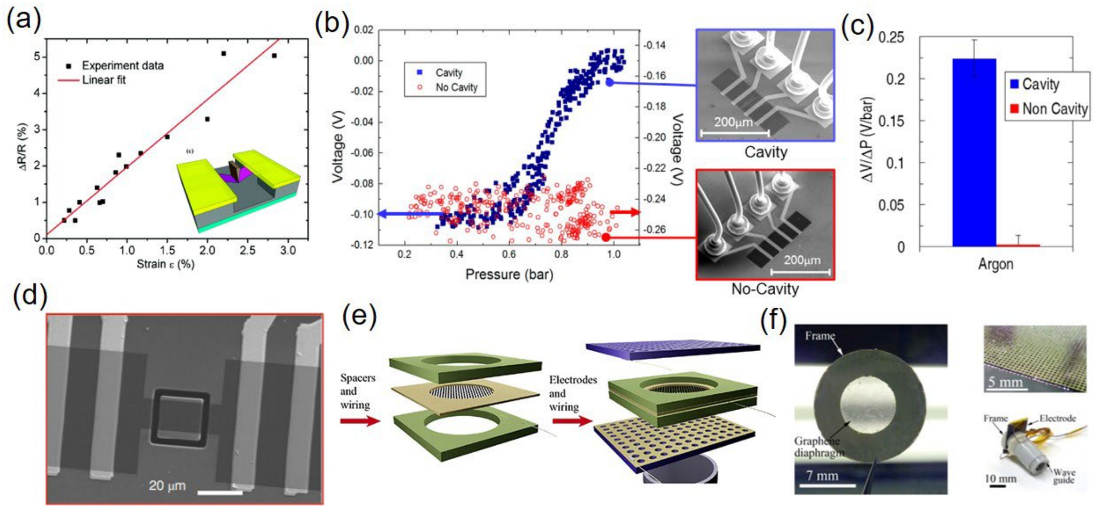

Figure 5 (a) The plot of the relative change in resistance as a function of strain for suspended graphene; Inset: The schematic illustration of the suspended graphene NEMS device.[56] Reprinted with permission from, copyright American Chemical Society. (b) Pressure and voltage

measurements for the suspended graphene NEMS devices with cavity and without a cavity. (c) The voltage change relative to the pressure for the two kinds of devices.[57] Reprinted with permission from, copyright American Chemical Society. (d) The accelerometers of suspended graphene with attached proof masses.[55] Reprinted with permission from, copyright Springer Nature Limited. (e) The schematic illustration of the construction of the microphone.[66] Reprinted with permission from, copyright National Academy of Sciences. (f) Suspended Graphene diaphragm and assembled speaker. [68] Reprinted with permission from, copyright AIP Publishing LLC.

In addition to the suspended graphene-based NMES systems, flexible sensors based on graphene/flexible substrates are more widely studied. The simple flexible device is constructed by transferring graphene to a flexible substrate（such as, PDMS or PEI）. Here, the flexible elastomer can serve both as a support and as a function of transferring deformation to graphene, thereby finally realizing the change of electrical properties of graphene. Park. *et al.* reported graphene-based conformal devices with a 70 nm thickness, which shows very low bending stiffness (**Figure 6 (a)**). In this way, the device can provide excellent coverage on an uneven surface without the need of other adhesives, and its electrical characteristics are also very stable.[69] Besides, Li *et al.* have also developed a simple technique based on by Marangoni self-assembly method for preparing large-area graphene films. Since the graphene film obtained by this method has only a nanometer-scale thickness, it can have a very high resistance change response (more than 1000 of gauge factor (GF) under 2% strain) under a small stretching condition.[70] After that, Park *et al.* used the same method to transfer the patterned graphene on a pre-formed substrate. The sticky patterned graphene can better contact with the rough skin and avoid delamination between the device and the skin without the aid of medical tape (**Figure 6 (b)**).[71] Furthermore, Luo *et al.* directly verified the changes in the conductive path caused by the slip of the graphene nanosheet through *in-situ* TEM characterization technology. It is found that the resistance changes of graphene are linear to the relative contact area of the nanosheets, and the tunnel current can be observed when the nanosheets are separated.[72]

To improve the stretchability and responsive properties of the sensor, different processing techniques and device design were introduced.[73-84] For example, through patterning techniques, CVD-grown graphene has non-monotonic piezoresistive properties with a tensile strain of 7.1%. This non-linear behavior may depend on the defects, disorders, and microcracks of graphene.[75] Besides, the graphene mesh network structure can be realized by adjusting the surface tension, which shows very stable tensile properties.[76] Furthermore, the technology of large-area patterning has been greatly developed. Oren *et al.* used the negatively patterned PDMS as a template, and the drop-casted graphene can be transferred by tape after molding. Patterned graphene has a feature size of only a few micrometers and can be arrayed or shaped according to the needs of the application (**Figure 6 (c)**).[78] Smart gloves based on patterned graphene can sense changes in force in time and space distribution. Cracks on the surface of the material can significantly enhance the sensitivity of the sensor. Based on this mechanism, rGO/PDMS structure with bio-inspired micro-cracks and hierarchical surface textures was designed (**Figure 6 (d)**). The sensor has super-hydrophobic properties and exhibits super-high sensitivity with the gauge factor of 8699.[79] To achieve stretchability while achieving high sensitivity, graphene planar structures have proven to be an effective trade-off structure.[80]

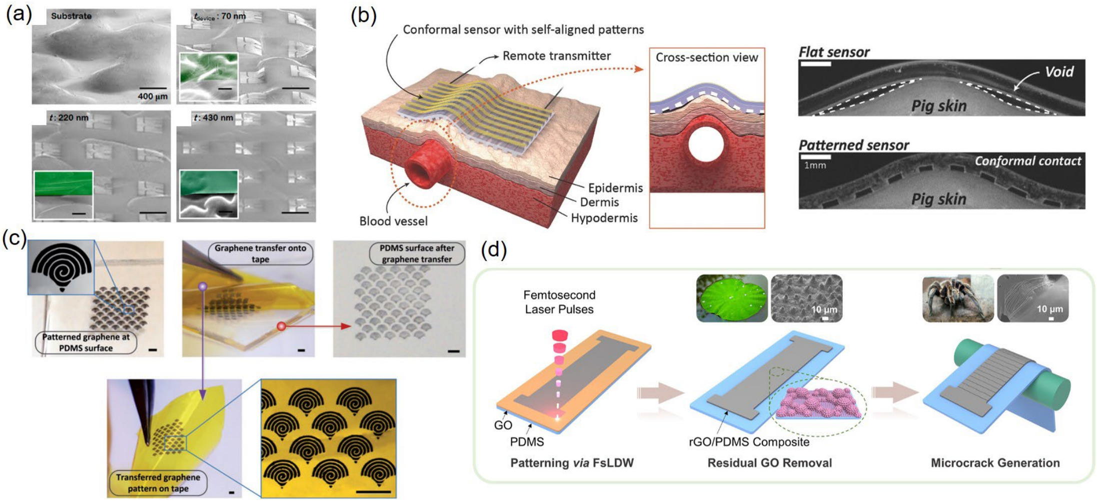

Figure 6 (a) The relationship between the adhesion degree to the skin and the thickness of the device. SEM images show the substrate before transferring the ultrathin graphene field effect transistors and the transferring devices of different thicknesses (70nm, 220nm, 430nm).[69] The inset images hows the graphene field effect transistors with the scale bar of 100 μm. Reprinted with permission from, copyright American Chemical Society. (b) The schematic illustration of the sensor array on the skin.[71] Compared to flat sensors, patterned sensors can better contact the skin. Reprinted with

permission from, copyright John Wiley & Sons, Inc. (c) Formation of patterned graphene design on tape.[78] The scale bars represent 1 mm. Reprinted with permission from, copyright John Wiley & Sons, Inc. (d) The schematic illustration of the formation of the rGO/ PDMS structure with micro-cracks and hierarchical surface textures.[79] Reprinted with permission from, copyright American Chemical Society.

On the other hand, graphene can be combined or hybridized with other materials to optimize its mechanical properties. For example, the liquid exfoliated graphene can be injected into the rubber, and the gauge factors can still be as high as 35 at over 800% strain, and these sensors can still work well at 160 Hz vibration frequency.[85] Boland *et al.* further analyzed the deformation mechanism and performance relationship of embedding graphene nanosheets in the low-viscosity polymer matrix, and studied the electrical response behavior under deformation conditions, so that the sensor can detect motion, blood pressure or other minor pressures.[86] Another common method is to achieve the structure of graphene textiles.[87-89] For example, Karim *et al.* developed a scalable and cost-effective method for producing the wearable e-textiles with graphene using a simple pad-dry technique at commercial production rates of 150 m/min (**Figure 7 (a)**).[90] Furthermore, the dyeing of textile yarns with graphene-based inks is improved by high-speed yarn dyeing technology. Based on this processing method, the conceptual intelligent clothing with RFID technology can monitor the temperature and other physiological signals (**Figure 7 (b)**).[91] Using a scalable pad-dry-cure method, along with rolling and packaging of graphene sheets, graphene-based textiles still have high conductivity after laundry washing cycles.

For thin graphene film, nanowires, nanotubes or nanoparticles can play a good structural strengthening role or function expansion. [92-97] Shi *et al.* designed a hybrid structure of graphene and carbon nanotubes, which can inhibit the buckling phenomenon of carbon nanotubes, so that the electrical response of the hybrid structure can be better matched with the strain.[92] Ren *et al.* have developed a method for electrostatic spinning the surface of graphene films, which can realize the independent transfer of films (**Figure 7 (c)**). The related structure can be used in pressure sensors, which demonstrate the high sensitivity (44.5 kPa^{-1} within 1.2 kPa) and low operating voltage (0.01–0.5 V).[93] Other functional materials combined with graphene can also be extended to achieve different types of detection functions. For example, Chen *et al.* combine graphene with piezoelectric nanowires, and use the polarization potential of the piezoelectric material to convert the impulse response into a continuous static response under pressing conditions (**Figure 7 (d)**).[94] Zaretski *et al.* deposited different island-like metals on the surface of graphene, and realized a highly sensitive and versatile sensor under the synergistic effect of tunneling effect and crack effect of metals and graphene.[95] Based on a similar mechanism, the same group developed a graphene/palladium nanoislands composite structure that can monitor the swallowing function in patients with cancer of the head and neck post-radiation therapy (**Figure 7 (e)**).[96]

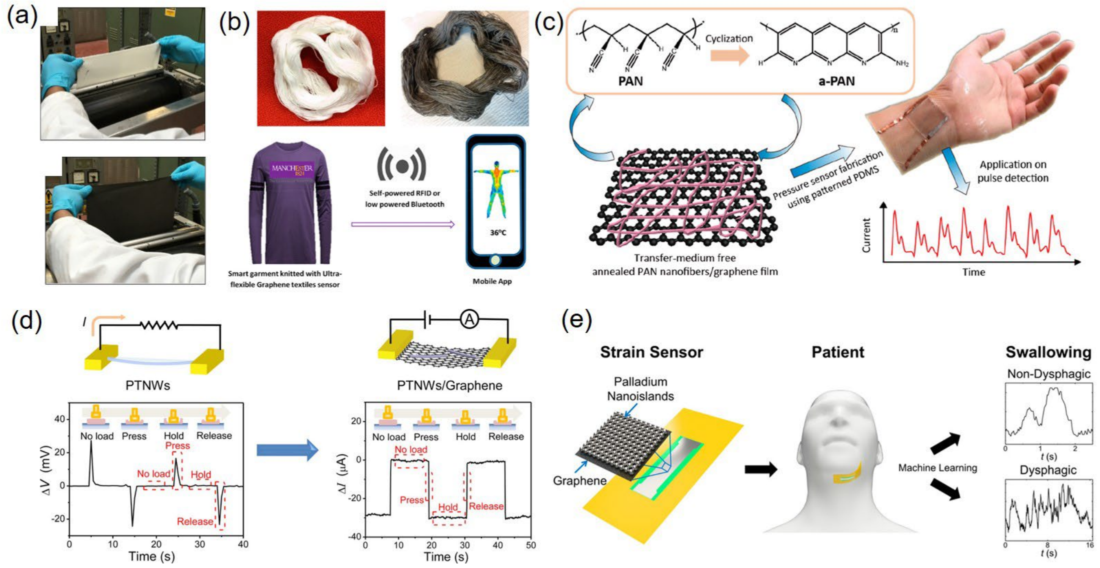

Figure 7 (a) The untreated cotton fabric and the cotton fabric filled with rGO through padding bath and rollers.[90] Reprinted with permission from, copyright American Chemical Society. (b) Unwashed cotton yarn and cotton yarn dyed with rGO. Based on such cotton yarn, the concept of smart clothing can be proposed.[91] Reprinted with permission from, copyright American Chemical Society. (c) The schematic illustration and characterizations of the a-PAN/graphene films which can be used for pulse detection.[93] Reprinted with permission from, copyright American Chemical Society. (d) The schematic illustration and characterizations of the piezoelectric nanowires and piezoelectric nanowires/graphene sensors for press detection.[94] Reprinted with permission from, copyright American Chemical Society. (e) The schematic illustration and characterizations of the palladium nanoisland /graphene sensors and its application for monitoring the swallowing function in patients.[96] Reprinted with permission from, copyright American Chemical Society.

5. Graphene-based Electrophysiological Signal Monitoring Applications

Long-term monitoring of electrophysiological signals is extremely important in the medical and healthcare field, especially for signals such as electrocardiogram (ECG) signal, electromyogram (EMG) signal, and electroencephalograph (EEG) signal. The equipment is often large, which is not conducive to real-time wearing the human body. Besides, intermittent measurements can only be performed in fixed places such as hospitals, so continuous monitoring of human electrophysiological signals cannot be achieved. Therefore, the flexible and lightweight wearable non-invasive medical sensor is expected to realize long-term use without being limited by the space of time.

Graphene with good mechanical properties, electrical properties, and electrochemical stability provides the possibility of measuring the electrophysiological signals as bioelectrodes. After verifying the graphene composite structure without any biocompatibility issues, Yun *et al.* constructed a composite structure of porous PDMS with graphene for monitoring the electrophysiological signals of ECG, EEG, and EMG on the human skin surface.[98] For example, the graphene-based electrodes are attached to the left and right forearms and left legs, as shown in **Figure 8 (a) and (b)**. The graphene electrode gave a very clear and standard ECG signals without significant baseline drift. At the same time, it was found that the ECG signal is similar to the signal measured by a commercially Ag/AgCl electrode using adhesive hydrogel (**Figure 8 (c)**). For EEG monitoring, the graphene-based electrodes are attached to the forehead and earlobe (**Figure 8 (d)**). By detecting the alpha-rhythm evoked from the brain during the eye blinking, there are red trips at about 10 Hz when the eyes closed from the spectrograms (**Figure 8 (e)**). Meanwhile, the power spectrum density at 10 Hz when the eyes are closed was higher than the ones when eyes are open (**Figure 8 (f)**). For the measurement of EMG signals, electrodes are placed at different positions on the arm to record muscle information (**Figure 8 (g) and (i)**). Obvious signals were observed when the muscle was in motion, which was comparable to results from commercial electrodes (**Figure 8 (h) and (j)**).

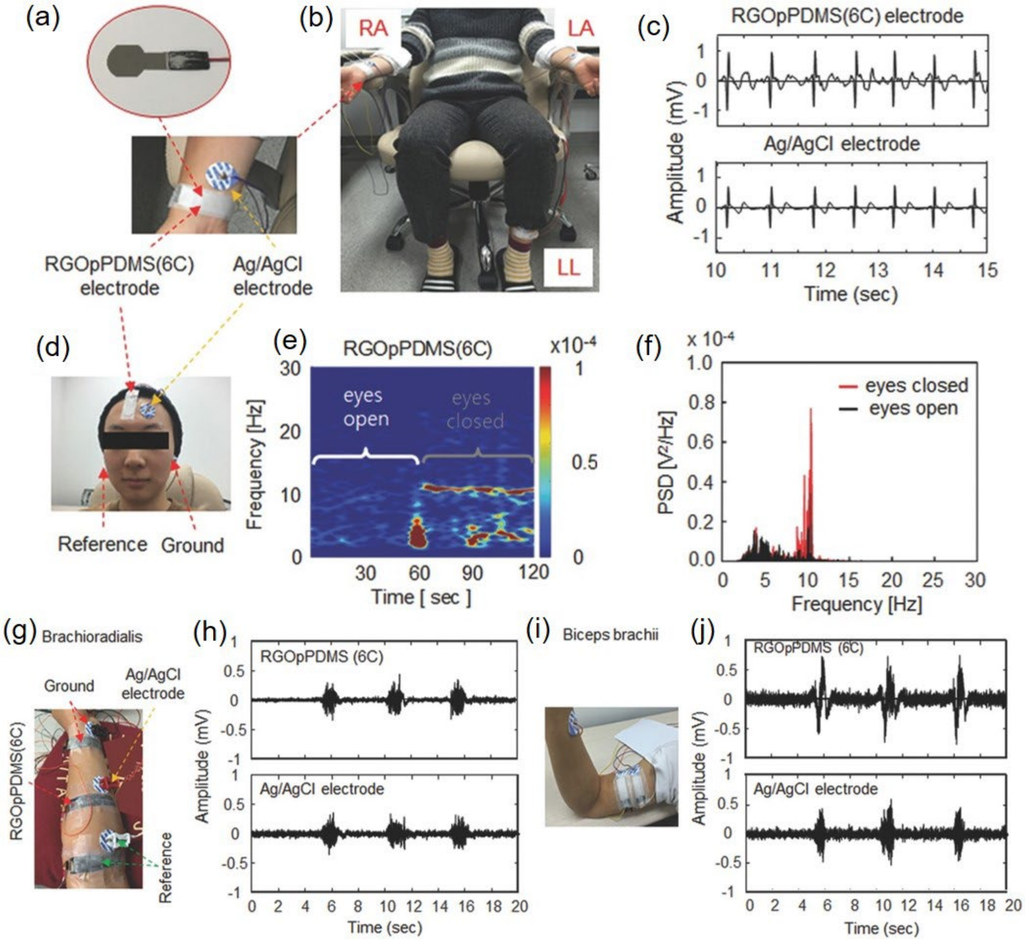

Figure 8 (a) and (b) the graphene-based electrodes attached to the left and right forearms and left legs. (c) the ECG signal measured by graphene-based electrodes and a commercially Ag/AgCl electrode using adhesive hydrogel. (d) the graphene-based electrodes attached to the forehead and earlobe. (e) the EEG signal measurement by detecting the alpha-rhythm. (f) The power spectrum density when the eyes are closed and open. (g) and (i) the graphene-based electrodes attached to the different positions on the arm. (h) and (j) the EMG signals for the graphene-based electrodes and commercial Ag/AgCl electrodes.[98] Reprinted with permission from, copyright John Wiley & Sons, Inc.

A lighter and thinner graphene electronic tattoo was designed by the "wet transfer, dry patterning" method, which can be directly laminated on the skin conforms by van der Waals forces. This kind of electrode always maintains the structure when it is stretched or compressed, and keeps good contact with the skin (**Figure 9 (a)**). Besides, to demonstrate the sensing performance, the graphene electronic tattoo has been successfully used to measure EEG, ECG, EMG signal of alpha rhythm during eye blinking, the heart and electromyogram measurement of muscle activity, and its results are similar to the ones from commercially electrodes (**Figure 9 (b)-(d)**).[99] Further, Ameri *et al.* designed a graphene sensor system with an imperceptible electrooculogram (**Figure 9 (e)**). Through the movement of the eye, the graphene sensor can realize different voltage signals (**Figure 9 (f)**), and the signal processing can remotely control the motion of objects such as airplanes (**Figure 9 (g)**).[100]

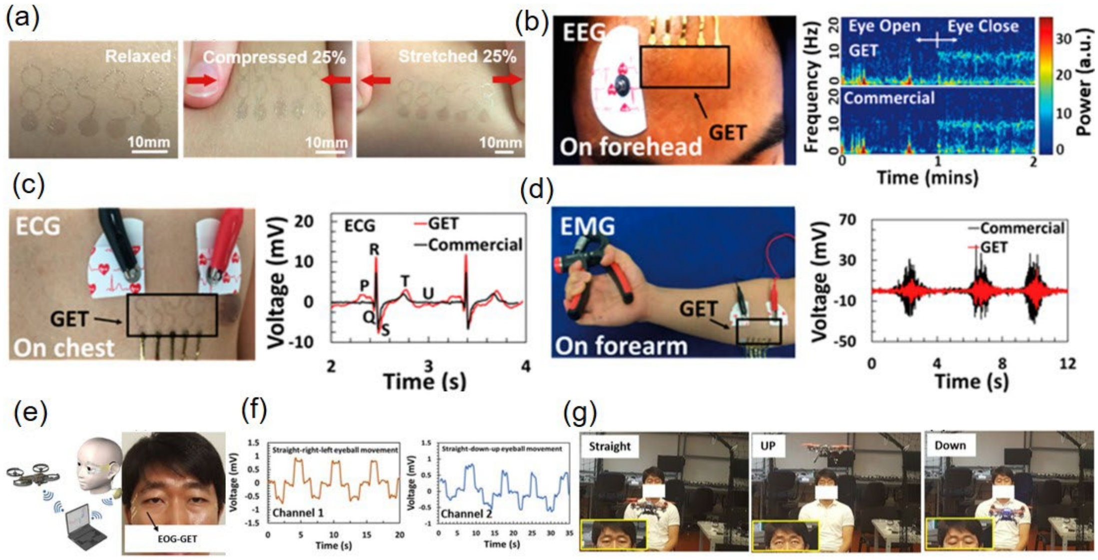

Figure 9 (a) the graphene-based electrodes attached to the skin when it is stretched or compressed. (b)-(d) Measurement of the EEG, ECG, EMG signal of the alpha rhythm, the heart and muscle activity, and its results are similar to the ones from commercial electrodes.[99] Reprinted with permission from, copyright American Chemical Society. (e) a graphene sensor system with an imperceptible electrooculogram and (f) the different voltage signals through the movement of the eye. (g) The motion control of the airplanes remotely.[100] Reprinted with permission from, copyright Springer Nature Limited.

Because of its transparency and high conductivity, graphene can be used for electroretinogram (ERC) measurements in combination with contact lens. Here, the soft graphene can make good contact with the eyeball and avoid tear films or air gaps that may form at its interface. Besides, good contact is also conducive to achieving better spatial resolution signals. Since graphene is a highly transparent material, it is possible to collect ERG responses from the entire corneal surface. Yin *et al.* reported this electrode structure and realized that the array was attached to the surface of rabbit eyes (**Figure 10 (a)-(c)**).[101] **Figure 10 (d)** and **(e)** present a record response showing the ERG from different channels at a stimulus intensity of 0.3 cd s m^{-2}. The amplitudes of a and b waves in the central region are significantly higher than those in the peripheral region. Meanwhile, the location correlation of a and b wave amplitudes does not correlate with electrode impedance distribution. The spatial distribution of b-wave amplitude at different stimulus intensities was shown in **Figure 10 (e)** and the amplitude decreased with increasing distance from the center of the cornea. This change reflects the regional differences in the retina's response to stimulation.

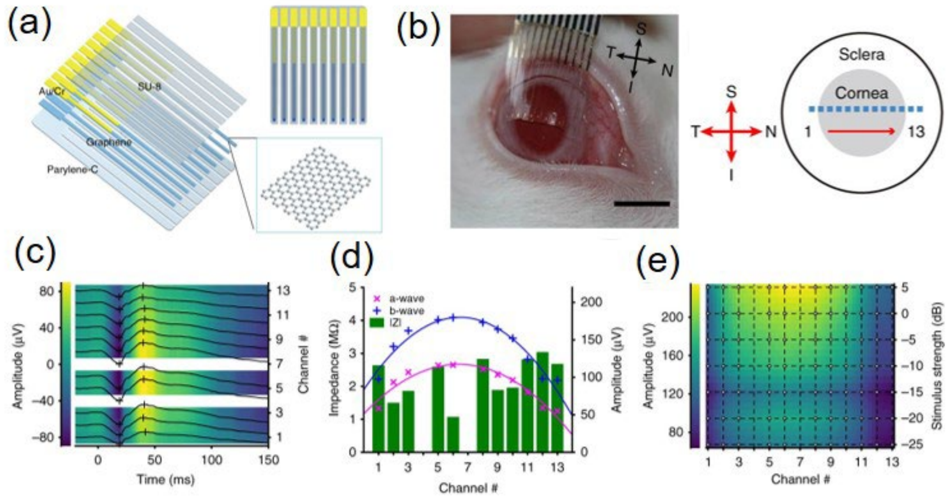

Figure 10 (a) The schematic illustration of the graphene multi-electrode array. (b) Graphene electrode array in contact with the rabbit eyes. Scale bar, 5 mm. (c) The position of the recording channel on the rabbit's eye. (c) A set of multi-electrode ERG responses under stimulus intensity with 0.3 cd s/m^2. Cross marked with a and b wave locations. (d) The electrode impedance values at 100 Hz and a and b wave amplitudes of EEG signals recorded on different channels related to c. (e) Spatial distribution of b-wave amplitude at different stimulus intensities. 0 dB corresponds to 3.0 cd s/m^2.[101] Reprinted with permission from, copyright Springer Nature Limited.

6. Summary and perspective

In short, graphene-based flexible sensors have great potential applications and will face important development opportunities with the advent of intelligent society and the Internet of Things. Focusing on graphene electrical properties and structural characteristics, the targeted device design is the focus of research in this field. This review introduces the physical signals sensing by graphene-based flexible devices, with emphasis on the design and function of device structures. Despite the rapid development of research on flexible sensor devices, there are still many challenges to its practical application, which is also an opportunity for future development in this field. Based on the current research progress, here we propose several aspects that should be paid attention to in the future: (1) The signal accuracy of the sensor. At present, the accuracy of flexible graphene-based sensors is still difficult to compare with traditional sensors. After all, the development of new technologies is still in the preliminary stage, which requires follow-up research and more attention should be paid to signal acquisition and analysis. (2) Wireless communication of sensors. The important applications of flexible sensing technology are wearable and portable sensors and devices. The realization and application of wireless communication technology are essential. This includes not only the information interaction between different sensors but also the information communication between sensors and other devices. At present, radio frequency (RF) wireless technology has been widely used in wearable sensors, and there is still room for improvement in the integrated development of new type flexible sensors. (3) The energy supply of the sensor. At present, many sensors are powered by batteries. Although many advances have been made in flexible batteries in recent years, there are still many shortcomings with commercial battery performance which limits the application of flexible sensors. These issues involve multiple disciplines such as material devices, electronic circuits, information communication, and energy management. At the same time, the development in this field also requires a more active interaction between academia and industry. As technology advances, graphene-based flexible sensors are expected to achieve greater breakthroughs.

Acknowledgments

Prof. Xinming Li acknowledges the financial support from South China Normal University start-up fund and the Guangdong Provincial Key Laboratory of Nanophotonic Functional Materials and Devices**.**

References

[1] Kim Y *et al.* 2018 A bioinspired flexible organic artificial afferent nerve *Science* **360** 998-1003

[2] Rogers J A, Someya T and Huang Y 2010 Materials and mechanics for stretchable electronics *Science* **327** 1603-1607

[3] Hammock M L, Chortos A, Tee B C K, Tok J B H and Bao Z 2013 25th anniversary article: the evolution of electronic skin (e-skin): a brief history, design considerations, and recent progress

Advanced Materials **25** 5997-6038

[4] Gao W *et al.* 2016 Fully integrated wearable sensor arrays for multiplexed in situ perspiration analysis *Nature* **529** 509-514

[5] Silva C W D, *Sensor Systems: Fundamentals and Applications*. CRC Press, Taylor & Francis Group: 2016.

[6] Novoselov K S, Geim A K, Morozov S V, Jiang D, Zhang Y, Dubonos S V, Grigorieva I V and Firsov A A 2004 Electric field effect in atomically thin carbon films *Science* **306** 666-669

[7] Zhu Y, Murali S, Cai W, Li X, Suk J W, Potts J R and Ruoff R S 2010 Graphene and graphene oxide: synthesis, properties, and applications *Advanced Materials* **22** 3906-3924

[8] Bonaccorso F, Sun Z, Hasan T and Ferrari A 2010 Graphene photonics and optoelectronics *Nature Photonics* **4** 611

[9] Jang H, Park Y J, Chen X, Das T, Kim M S and Ahn J H 2016 Graphene-based flexible and stretchable electronics *Advanced Materials* **28** 4184-4202

[10] Novoselov K S, Fal V, Colombo L, Gellert P, Schwab M and Kim K 2012 A roadmap for graphene *Nature* **490** 192-200

[11] Secor E B, Lim S, Zhang H, Frisbie C D, Francis L F and Hersam M C 2014 Gravure printing of graphene for large-area flexible electronics *Advanced Materials* **26** 4533-4538

[12] Zhao G *et al.* 2017 The physics and chemistry of graphene-on-surfaces *Chemical Society Reviews* **46** 4417-4449

[13] Kim H and Ahn J-H 2017 Graphene for flexible and wearable device applications *Carbon* **120** 244-257

[14] Chen S, Jiang K, Lou Z, Chen D and Shen G 2018 Recent Developments in Graphene-Based Tactile Sensors and E-Skins *Advanced Materials Technologies* **3** 1700248

[15] Huang H, Su S, Wu N, Wan H, Wan S, Bi H and Sun L 2019 Graphene-Based Sensors for Human Health Monitoring *Frontiers in chemistry* **7** 399

[16] Yu X, Cheng H, Zhang M, Zhao Y, Qu L and Shi G 2017 Graphene-based smart materials *Nature Reviews Materials* **2** 1-13

[17] Chen X *et al.* 2019 Graphene Hybrid Structures for Integrated and Flexible Optoelectronics *Advanced Materials* **32** 1902039

[18] Yu T, Wang F, Xu Y, Ma L, Pi X and Yang D 2016 Graphene Coupled with Silicon Quantum Dots for High-Performance Bulk-Silicon-Based Schottky-Junction Photodetectors *Advanced Materials* **28** 4912-4919

[19] Li X *et al.* 2016 Graphene/h-BN/GaAs sandwich diode as solar cell and photodetector *Optics Express* **24** 134-145

[20] Liu S, Liao Q, Lu S, Zhang Z, Zhang G and Zhang Y 2016 Strain modulation in graphene/ZnO nanorod film schottky junction for enhanced photosensing performance *Advanced Functional Materials* **26** 1347-1353

[21] Schneider D S, Bablich A and Lemme M C 2017 Flexible hybrid graphene/a-Si: H multispectral photodetectors *Nanoscale* **9** 8573-8579

[22] De Fazio D *et al.* 2016 High responsivity, large-area graphene/MoS2 flexible photodetectors *ACS Nano* **10** 8252-8262

[23] Son D I, Yang H Y, Kim T W and Park W I 2015 Transparent and flexible ultraviolet photodetectors based on colloidal ZnO quantum dot/graphene nanocomposites formed on poly (ethylene terephthalate) substrates *Composites Part B: Engineering* **69** 154-158

[24] Liu Y, Liu Y, Qin S, Xu Y, Zhang R and Wang F 2017 Graphene-carbon nanotube hybrid films for high-performance flexible photodetectors *Nano Research* **10** 1880-1887

[25] Dang V Q, Han G-S, Trung T Q, Jin Y-U, Hwang B-U, Jung H-S and Lee N-E 2016 Methylammonium lead iodide perovskite-graphene hybrid channels in flexible broadband phototransistors *Carbon* **105** 353-361

[26] Tao L, Chen Z, Li X, Yan K and Xu J-B 2017 Hybrid graphene tunneling photoconductor with interface engineering towards fast photoresponse and high responsivity *npj 2D Materials and Applications* **1** 19

[27] Liu Y-L, Yu C-C, Lin K-T, Yang T-C, Wang E-Y, Chen H-L, Chen L-C and Chen K-H 2015 Transparent, broadband, flexible, and bifacial-operable photodetectors containing a large-area graphene–gold oxide heterojunction *ACS Nano* **9** 5093-5103

[28] Chang H *et al.* 2013 Regulating infrared photoresponses in reduced graphene oxide phototransistors by defect and atomic structure control *ACS Nano* **7** 6310-6320

[29] Trung T Q, Ramasundaram S and Lee N E 2015 Infrared detection using transparent and flexible field-effect transistor array with solution processable nanocomposite channel of reduced graphene oxide and P (VDF-TrFE) *Advanced Functional Materials* **25** 1745-1754

[30] Kang P, Wang M C, Knapp P M and Nam S 2016 Crumpled Graphene Photodetector with Enhanced, Strain-Tunable, and Wavelength-Selective Photoresponsivity *Advanced Materials* **28** 4639-4645

[31] Kim M, Kang P, Leem J and Nam S 2017 A stretchable crumpled graphene photodetector with plasmonically enhanced photoresponsivity *Nanoscale* **9** 4058-4065

[32] Moon I K, Ki B, Yoon S, Choi J and Oh J 2016 Lateral photovoltaic effect in flexible free-standing reduced graphene oxide film for self-powered position-sensitive detection *Scientific Reports* **6** 33525

[33] Jang S, Hwang E, Lee Y, Lee S and Cho J H 2015 Multifunctional graphene optoelectronic devices capable of detecting and storing photonic signals *Nano Letters* **15** 2542-2547

[34] Xia K *et al.* 2020 CVD growth of perovskite/graphene films for high-performance flexible image sensor *Science Bulletin* **65** 343-349

[35] Ali A *et al.* In *High-performance, flexible graphene/ultra-thin silicon ultra-violet image sensor*, 2017 IEEE International Electron Devices Meeting (IEDM), IEEE: 2017; p 17524777.

[36] Polat E O *et al.* 2019 Flexible graphene photodetectors for wearable fitness monitoring *Science advances* **5** eaaw7846

[37] Zhou F, Chen J, Tao X, Wang X and Chai Y 2019 2D Materials Optoelectronic Memory - Convergence of Electronic Memory and Optical Sensor *Research* **2** 9490413

[38] Pradhan B *et al.* 2020 Ultrasensitive and ultrathin phototransistors and photonic synapses using perovskite quantum dots grown from graphene lattice *Science advances* **6** eaay5225

[39] Zhou F *et al.* 2019 Optoelectronic resistive random access memory for neuromorphic vision sensors *Nature nanotechnology* **14** 776-782

[40] Chai Y 2020 In-sensor computing for machine vision *Nature* **579** 32-33

[41] Mennel L, Symonowicz J, Wachter S, Polyushkin D K, Molina-Mendoza A J and Mueller T 2020 Ultrafast machine vision with 2D material neural network image sensors *Nature* **579** 62–66

[42] Qin S, Wang F, Liu Y, Wan Q, Wang X, Xu Y, Shi Y, Wang X and Zhang R 2017 A light-stimulated synaptic device based on graphene hybrid phototransistor *2D Materials* **4** 035022

[43] Seo S *et al.* 2018 Artificial optic-neural synapse for colored and color-mixed pattern

recognition *Nature Communications* **9** 5106

[44] Tran M D, Kim H, Kim J S, Doan M H, Chau T K, Vu Q A, Kim J H and Lee Y H 2019 Two-Terminal Multibit Optical Memory via van der Waals Heterostructure *Advanced Materials* **31** 1807075

[45] Davaji B, Cho H D, Malakoutian M, Lee J-K, Panin G, Kang T W and Lee C H 2017 A patterned single layer graphene resistance temperature sensor *Scientific Reports* **7** 8811

[46] Liu Q, Tai H, Yuan Z, Zhou Y, Su Y and Jiang Y 2019 A High-Performances Flexible Temperature Sensor Composed of Polyethyleneimine/Reduced Graphene Oxide Bilayer for Real-Time Monitoring *Advanced Materials Technologies* **4** 1800594

[47] Zeng Y, Li T, Yao Y, Li T, Hu L and Marconnet A 2019 Thermally Conductive Reduced Graphene Oxide Thin Films for Extreme Temperature Sensors *Advanced Functional Materials* **29** 1901388

[48] Yan C, Wang J and Lee P S 2015 Stretchable graphene thermistor with tunable thermal index *ACS Nano* **9** 2130-2137

[49] Zu G, Kanamori K, Nakanishi K and Huang J 2019 Superhydrophobic ultraflexible triple-network graphene/polyorganosiloxane aerogels for a high-performance multifunctional temperature/strain/pressure sensing array *Chemistry of Materials* **31** 6276-6285

[50] Liu H, Xiang H, Wang Y, Li Z, Qian L, Li P, Ma Y, Zhou H and Huang W 2019 A Flexible Multimodal Sensor That Detects Strain, Humidity, Temperature, and Pressure with Carbon Black and Reduced Graphene Oxide Hierarchical Composite on Paper *ACS Applied Materials & Interfaces* **11** 40613-40619

[51] Vuorinen T, Niittynen J, Kankkunen T, Kraft T M and Mäntysalo M 2016 Inkjet-printed graphene/PEDOT: PSS temperature sensors on a skin-conformable polyurethane substrate *Scientific Reports* **6** 35289

[52] Park J, Kim M, Lee Y, Lee H S and Ko H 2015 Fingertip skin–inspired microstructured ferroelectric skins discriminate static/dynamic pressure and temperature stimuli *Science Advances* **1** e1500661

[53] Trung T Q, Ramasundaram S, Hwang B U and Lee N E 2016 An all-elastomeric transparent and stretchable temperature sensor for body-attachable wearable electronics *Advanced Materials* **28** 502-509

[54] Zheng Q, Lee J-h, Shen X, Chen X and Kim J-K 2020 Graphene-based wearable piezoresistive physical sensors *Materials Today* **36** 158-179

[55] Fan X *et al.* 2019 Graphene ribbons with suspended masses as transducers in ultra-small nanoelectromechanical accelerometers *Nature Electronics* **2** 394-404

[56] Huang M, Pascal T A, Kim H, Goddard III W A and Greer J R 2011 Electronic− mechanical coupling in graphene from in situ nanoindentation experiments and multiscale atomistic simulations *Nano Letters* **11** 1241-1246

[57] Smith A *et al.* 2013 Electromechanical piezoresistive sensing in suspended graphene membranes *Nano Letters* **13** 3237-3242

[58] Xu K *et al.* 2015 The positive piezoconductive effect in graphene *Nature Communications* **6** 8119

[59] Dolleman R J, Davidovikj D, Cartamil-Bueno S J, van der Zant H S and Steeneken P G 2016 Graphene squeeze-film pressure sensors *Nano Letters* **16** 568-571

[60] Kumar M and Bhaskaran H 2015 Ultrasensitive room-temperature piezoresistive transduction in graphene-based nanoelectromechanical systems *Nano Letters* **15** 2562-2567

[61] Fan X, Forsberg F, Smith A D, Schro¨der S, Wagner S, O¨stling M, Lemme M C and Niklaus F 2019 Suspended Graphene Membranes with Attached Silicon Proof Masses as Piezoresistive Nanoelectromechanical Systems Accelerometers *Nano Letters* **19** 6788-6799

[62] Bunch J S, Van Der Zande A M, Verbridge S S, Frank I W, Tanenbaum D M, Parpia J M, Craighead H G and McEuen P L 2007 Electromechanical resonators from graphene sheets *Science* **315** 490-493

[63] Chen C, Rosenblatt S, Bolotin K I, Kalb W, Kim P, Kymissis I, Stormer H L, Heinz T F and Hone J 2009 Performance of monolayer graphene nanomechanical resonators with electrical readout *Nature Nanotechnology* **4** 861

[64] Miao T, Yeom S, Wang P, Standley B and Bockrath M 2014 Graphene nanoelectromechanical systems as stochastic-frequency oscillators *Nano Letters* **14** 2982-2987

[65] Sun J, Schmidt M E, Muruganathan M, Chong H M and Mizuta H 2016 Large-scale nanoelectromechanical switches based on directly deposited nanocrystalline graphene on insulating substrates *Nanoscale* **8** 6659-6665

[66] Zhou Q, Zheng J, Onishi S, Crommie M and Zettl A K 2015 Graphene electrostatic microphone and ultrasonic radio *Proceedings of the National Academy of Sciences* **112** 8942-8946

[67] Todorović D, Matković A, Milićević M, Jovanović D, Gajić R, Salom I and Spasenović M 2015 Multilayer graphene condenser microphone *2D Materials* **2** 045013

[68] Zhou Q and Zettl A 2013 Electrostatic graphene loudspeaker *Applied physics letters* **102** 223109

[69] Park Y J, Lee S-K, Kim M-S, Kim H and Ahn J-H 2014 Graphene-based conformal devices *ACS Nano* **8** 7655-7662

[70] Li X *et al.* 2016 Large-area ultrathin graphene films by single-step marangoni self-assembly for highly sensitive strain sensing application *Advanced Functional Materials* **26** 1322-1329

[71] Park Y, Shim J, Jeong S, Yi G R, Chae H, Bae J W, Kim S O and Pang C 2017 Microtopography-guided conductive patterns of liquid-driven graphene nanoplatelet networks for stretchable and skin-conformal sensor array *Advanced Materials* **29** 1606453

[72] Luo Z, Li X, Li Q, Tian X, Wang C, Wu X and Shen G 2020 In Situ Dynamic Manipulation of Graphene Strain Sensor with Drastically Sensing Performance Enhancement *Advanced Electronic Materials* **6** 2000269

[73] Wu Q *et al.* 2020 Triode-Mimicking Graphene Pressure Sensor with Positive Resistance Variation for Physiology and Motion Monitoring *ACS Nano* **14** 10104–10114

[74] Chang T-H *et al.* 2019 Stretchable graphene pressure sensors with Shar-Pei-like hierarchical wrinkles for collision-aware surgical robotics *ACS applied materials & interfaces* **11** 10226-10236

[75] Bae S-H, Lee Y, Sharma B K, Lee H-J, Kim J-H and Ahn J-H 2013 Graphene-based transparent strain sensor *Carbon* **51** 236-242

[76] Han J, Lee J Y, Lee J and Yeo J S 2018 Highly stretchable and reliable, transparent and conductive entangled graphene mesh networks *Advanced Materials* **30** 1704626

[77] Jin F-H *et al.* 2019 High performance and low-cost graphene vacuum pressure sensor based on one-step laser scribing *Appl. Phys. Lett.* **114** 081602

[78] Oren S, Ceylan H, Schnable P S and Dong L 2017 High-Resolution Patterning and Transferring of Graphene-Based Nanomaterials onto Tape toward Roll-to-Roll Production of Tape-Based

Wearable Sensors *Advanced Materials Technologies* **2** 1700223

[79] Dinh Le T-S, An J, Huang Y, Vo Q, Boonruangkan J, Tran T, Kim S-W, Sun G and Kim Y-J 2019 Ultrasensitive Anti-Interference Voice Recognition by Bio-Inspired Skin-Attachable Self-Cleaning Acoustic Sensors *ACS nano* **13** 13293-13303

[80] You X, Yang J, Wang M, Zhou H, Gao L, Hu J, Zhang X and Dong S-m 2020 Novel Graphene Planar Architecture with Ultrahigh Stretchability and Sensitivity *ACS Applied Materials & Interfaces* **12** 18913–18923

[81] Qiao Y *et al.* 2018 Multilayer graphene epidermal electronic skin *ACS nano* **12** 8839-8846

[82] Fan T *et al.* 2020 Analog sensing and computing system with low-power consumption for gesture recognition *Advanced Intelligent Systems* **2** 2000184

[83] Tian Y, Wang D-Y, Li Y-T, Tian H, Yang Y and Ren T-L 2020 Highly Sensitive, Wide-Range, and Flexible Pressure Sensor Based on Honeycomb-Like Graphene Network *IEEE Transactions on Electron Devices* **67** 2153 - 2156

[84] Xu J *et al.* 2020 Highly Transparent and Sensitive Graphene Sensors for Continuous and Non-invasive Intraocular Pressure Monitoring *ACS applied materials & interfaces* **12** 18375–18384

[85] Boland C S *et al.* 2014 Sensitive, high-strain, high-rate bodily motion sensors based on graphene–rubber composites *ACS Nano* **8** 8819-8830

[86] Boland C S *et al.* 2016 Sensitive electromechanical sensors using viscoelastic graphene-polymer nanocomposites *Science* **354** 1257-1260

[87] Yang Z, Pang Y, Han X-l, Yang Y, Ling J, Jian M, Zhang Y, Yang Y and Ren T-L 2018 Graphene textile strain sensor with negative resistance variation for human motion detection *ACS nano* **12** 9134-9141

[88] Wang H *et al.* 2020 Laser Writing of Janus Graphene/Kevlar Textile for Intelligent Protective Clothing *ACS nano* **14** 3219-3226

[89] Shi J *et al.* 2020 Smart Textile-Integrated Microelectronic Systems for Wearable Applications *Advanced Materials* **32** 1901958

[90] Karim N, Afroj S, Tan S, He P, Fernando A, Carr C and Novoselov K S 2017 Scalable production of graphene-based wearable e-textiles *ACS Nano* **11** 12266-12275

[91] Afroj S, Karim N, Wang Z, Tan S, He P, Holwill M, Ghazaryan D, Fernando A and Novoselov K S 2019 Engineering graphene flakes for wearable textile sensors via highly scalable and ultrafast yarn dyeing technique *Acs Nano* **13** 3847-3857

[92] Shi J *et al.* 2016 Graphene reinforced carbon nanotube networks for wearable strain sensors *Advanced Functional Materials* **26** 2078-2084

[93] Ren H *et al.* 2019 Transfer-medium-free nanofiber-reinforced graphene film and applications in wearable transparent pressure sensors *ACS nano* **13** 5541-5548

[94] Chen Z, Wang Z, Li X, Lin Y, Luo N, Long M, Zhao N and Xu J-B 2017 Flexible piezoelectric-induced pressure sensors for static measurements based on nanowires/graphene heterostructures *ACS Nano* **11** 4507-4513

[95] Zaretski A V, Root S E, Savchenko A, Molokanova E, Printz A D, Jibril L, Arya G, Mercola M and Lipomi D J 2016 Metallic nanoislands on graphene as highly sensitive transducers of mechanical, biological, and optical signals *Nano Letters* **16** 1375-1380

[96] Ramírez J *et al.* 2018 Metallic Nanoislands on Graphene for Monitoring Swallowing Activity in Head and Neck Cancer Patients *ACS nano* **12** 5913-5922

[97] Qiao Y *et al.* 2020 Multifunctional and high-performance electronic skin based on silver

nanowires bridging graphene *Carbon* **156** 253-260

[98] Yun Y J *et al.* 2017 Highly Elastic Graphene-Based Electronics Toward Electronic Skin *Advanced Functional Materials* **27** 1701513

[99] Kabiri Ameri S, Ho R, Jang H, Tao L, Wang Y, Wang L, Schnyer D M, Akinwande D and Lu N 2017 Graphene electronic tattoo sensors *ACS Nano* **11** 7634-7641

[100] Ameri S K, Kim M, Kuang I A, Perera W K, Alshiekh M, Jeong H, Topcu U, Akinwande D and Lu N 2018 Imperceptible electrooculography graphene sensor system for human–robot interface *npj 2D Materials and Applications* **2** 19

[101] Yin R *et al.* 2018 Soft transparent graphene contact lens electrodes for conformal full-cornea recording of electroretinogram *Nature Communications* **9** 2334