Review

Carbon Future

Graphene-based sensors for human-machine interaction

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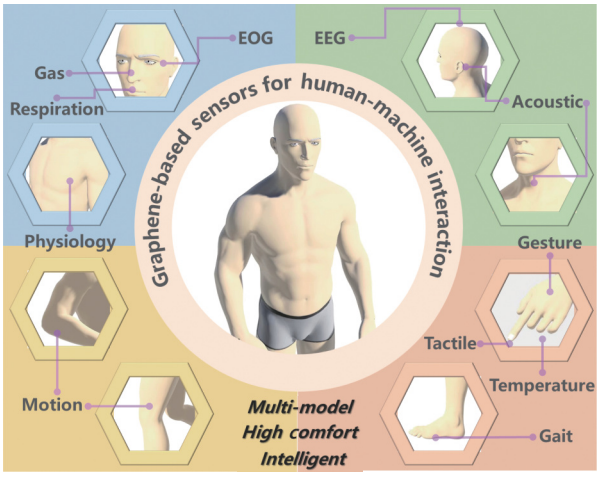
ABSTRACT: Since 2021, the concept of the metaverse has gained significant popularity and attention, not only among the general public but also among researchers who are interested in novel technologies and human-machine interfaces. Sensors, a critical component of human-machine interaction, have seen rapid advancements in recent years, particularly graphene-based sensors. These sensors offer a number of benefits, including flexibility, lightweight, ease of integration, and outstanding electrical properties. Over the past decade, our research team has focused on developing advanced graphene sensors for use in human-machine interaction and wearable healthcare. In this review, we showcase our team's efforts by presenting the design, manufacturing process, and performance of various graphene-based sensors, focusing on their suitability for diverse human-machine interaction needs across the human body. Additionally, we discuss potential future directions for the development of graphene-based sensors in human-machine interaction and share our insights.



KEYWORDS: graphene, flexible sensor, wearable electronics, human-machine interaction, metaverse

1 Introduction

The metaverse has been one of the driving forces behind the new global economy since 2021, influencing many things from technological innovation to the stock market[1,2]. This field integrates various cutting-edge technologies such as extended reality (XR), cloud computing, and human-machine interaction (HMI), which has sparked significant research into its related technologies and devices[3]. Of particular interest is the sensor, which plays a crucial role as the basic device of information interaction and a key component of HMI.

Graphene is a two-dimensional material with excellent characteristics including high flexibility, lightweight, good mechanical properties (42 $N \cdot m^{-1}$ breaking strength), high electron mobility (10,000 $cm^{-2} \cdot s^{-1}$ at room temperature), superior thermal conductivity (> 5000 $W \cdot mK^{-1}$ for a single-layer sheet), and excellent chemical inertness (due to its covalent bond structure)[4–6]. These properties make it an ideal choice for the creation of wearables, flexible, lightweight, easy-to-integrate sensors which are in high demand due to the rapid development of HMI[7,8]. Based on the various excellent properties of graphene, and the high-performance requirements as well as the massive demand for advanced sensors in the field of HMI, our research team has dedicated to the development of advanced graphene-based sensors for over a decade, with a focus on expanding their use in HMI and wearable healthcare[9–11].

The human body is a "delicate machine" full of sensors and actuators that constantly produces a multitude of diversified signals[12]. Based on this understanding, many parts of the human body, from head to toe, have the potential to be developed into human-machine interfaces. Brain, eyes, ears, nose, mouth, throat, fingertips, skin, joints, and feet can all be used as HMI interfaces based on electroencephalogram (EEG)[13,14], electromyography (EMG)[15,16], electrooculogram (EOG)[17], eye movement[18], light[19,20], breathing[21,22], voice[23–25], touch[26,27], temperature[28,29], movement[30–32], gait[33–35], and other physiological information. To effectively utilize these diverse HMI interfaces, it's important to design graphene-based sensors specifically for the types of signals being collected and the intended application location.

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In this review, we present an overview of some of our research team's efforts to create graphene-based sensors for human-machine interfaces. These sensors, designed for use on various parts of the human body, will be introduced with a focus on their target signals, design, manufacturing process, and performance features. Additionally, we delve into potential future developments for graphene-based sensors, including multi-modality, improved comfort, and intelligence.

2 Graphene-based sensors for HMI

2.1 Brain: graphene-based EEG sensors

Continuous and long-term monitoring of EEG signals plays a critical role in gaining insight into a person's physical, and psychological characteristics. This information can then be utilized for identifying illnesses, tracking health, and developing brain-computer interfaces. To meet the requirements of daily life EEG monitoring for human-machine interaction, graphene-based sensors must possess two key attributes: the capability to gather high-quality EEG signals, and the comfort and stability for prolonged wear.

In response to these needs, Qiao et al. developed a graphene nanomesh e-skin EEG sensor with high comfort[14]. In this work, a thin, soft, breathable, and stretchable polyurethane nanomesh substrate was prepared by electrospinning and coated with the graphene oxide (GO) solution. After that, by laser direct writing, the laser-scribed graphene (LSG) based EEG sensor was produced. As shown in Fig. 1a, the obtained LSG nanomesh e-skin can be attached to the human skin conformally. With good breathability, ultra-lightweight (~ 0.3 mg), and good stretchability and durability (remaining stable after over a thousand cycles of 40% stretching), the nanomesh EEG sensor can be worn continuously for at least 24 h. Furthermore, due to its good fit with the human skin, the sensor has a much lower skin contact impedance and higher signal-to-noise ratio than the commercial gel electrode, which allows it to collect high-quality EEG signals and reduce noise interferences. By combining the graphene nanomesh sensors with an intelligent algorithm, the wearer's concentration level was classified, which demonstrated its potential application in the field of HMI such as automatic driving.

2.2 Eyes: EOG, light, and eye pressure sensors

As a "window" for people to interact with the outside world, the eye can be equipped with a diverse set of sensors for HMI. Here, we will demonstrate graphene-based EOG/eye movement sensors, light detectors, and contact lens-type eye pressure sensors.

Xu et al. proposed an EOG and tactile perception collaborative interface for three-dimensional (3D) HMI[17]. Based on EOG sensors and a tactile sensing array consisting of honeycomb LSG (Fig. 1b), nine different eye movements and complex tactile information can be precepted. The whole system can realize eight directions of control and complex movement of trajectories. It allows for eight directions of control and more complex movements, freeing the wearer's hands and providing an immersive experience in the virtual world. This graphene-based 3D HMI also offers a better life for disabled individuals, allowing them to interact with both people and machines.

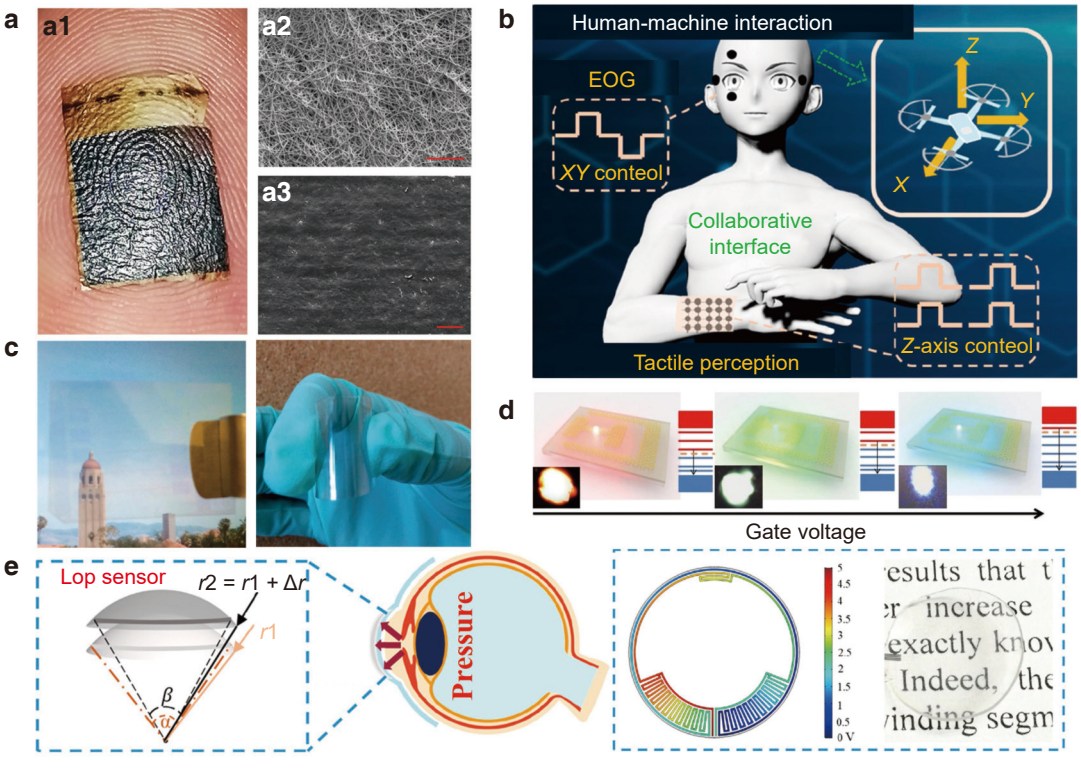

Fig. 1. Graphene-based sensors for the brain and eyes. a, EEG sensor made of graphene nanomesh e-skin. Reproduced with permission from Ref.[14], © Wiley-VCH GmbH 2021. **b**, EOG and tactile perception collaborative 3D HMI. Reproduced with permission from Ref.[17], © American Chemical Society 2022. **c**, The fully transparent and flexible graphene-based infrared photodetector. Reproduced with permission from Ref.[19], © American Chemical Society 2014. **d**, The all-graphene-based flexible field-effect light-emitting device. Reproduced with permission from Ref.[20], © Macmillan Publishers Limited 2015. **e**, The fully transparent and flexible graphene-based infrared photodetector. Reproduced with permission from Ref.[36], © American Chemical Society 2020.

The limitations of the human eye in perceiving visible light can now be overcome with the use of graphene sensors. As demonstrated by Liu et al., the first fully transparent and flexible graphene-based infrared photodetector was created by doping chemical vapor deposition (CVD)-grown graphene with P-N junctions (Fig. 1c). The sensor was prepared by chemical doping CVD grown graphene into P-N junctions. With both good photoresponsivity and bending capability, this graphene sensor shows the potential to broaden one's visual sensing ability and be applied to optical HMI. In addition to light detection, light emission is also crucial in realizing complete optical HMI. Wang et al. invented a spectrally tunable all-graphene-based light-emitting device[20]. The graphene oxide/reduced-graphene oxide interface demonstrated a bright spectrally tunable electroluminescence from blue (~ 450 nm) to red (~ 750 nm) (Fig. 1d). This flexible graphene LED not only represents a method for emission wavelength tuning but also has the potential in forming a closed loop of ocular optical HMI. Additionally, it offers the potential for creating a flexible display in a more straightforward manner.

Contact lenses, which are light, flexible, and can integrate a number of sensors, are excellent platforms for a variety of eye-machine interfaces[18, 36, 37]. Xu et al. proposed a highly transparent and sensitive graphene sensor for 24 h continuous intraocular pressure (IOP) monitoring (Fig. 1e)[36]. The graphene wheatstone bridge, consisting of multilayer graphene embedded in the contact lens, is capable of detecting changes in radial expansion and contraction of the cornea caused by variations in IOP with high sensitivity. The lightweight and transparent graphene contact lens sensor minimizes interference with normal vision while effectively sensing eye deformations. It also holds great potential for eye movement HMI.

2.3 Ears: acoustic sensors and earphones

Although the ear is primarily responsible for collecting sound, it can perform additional functions when equipped with graphene sensors. This section provide an overview of graphene-based acoustic sensors and vocal devices, such as earphones, which capitalize on the exceptional sound-receiving and generating capabilities of graphene. Additionally, we explore how other two-dimensional materials like MXene can be used to create acoustic sensors.

Complete acoustic interaction functions can be achieved through the use of multi-model acoustic devices that have both sound receiving and generating functions. Sun et al. have proposed a graphene-based dual-function acoustic transducer for human-robot interfaces to meet the demand for intelligent HMI. (Fig. 2a)[38]. This acoustic interface includes an artificial ear that works through the triboelectric acoustic sensing mechanism and an artificial mouth

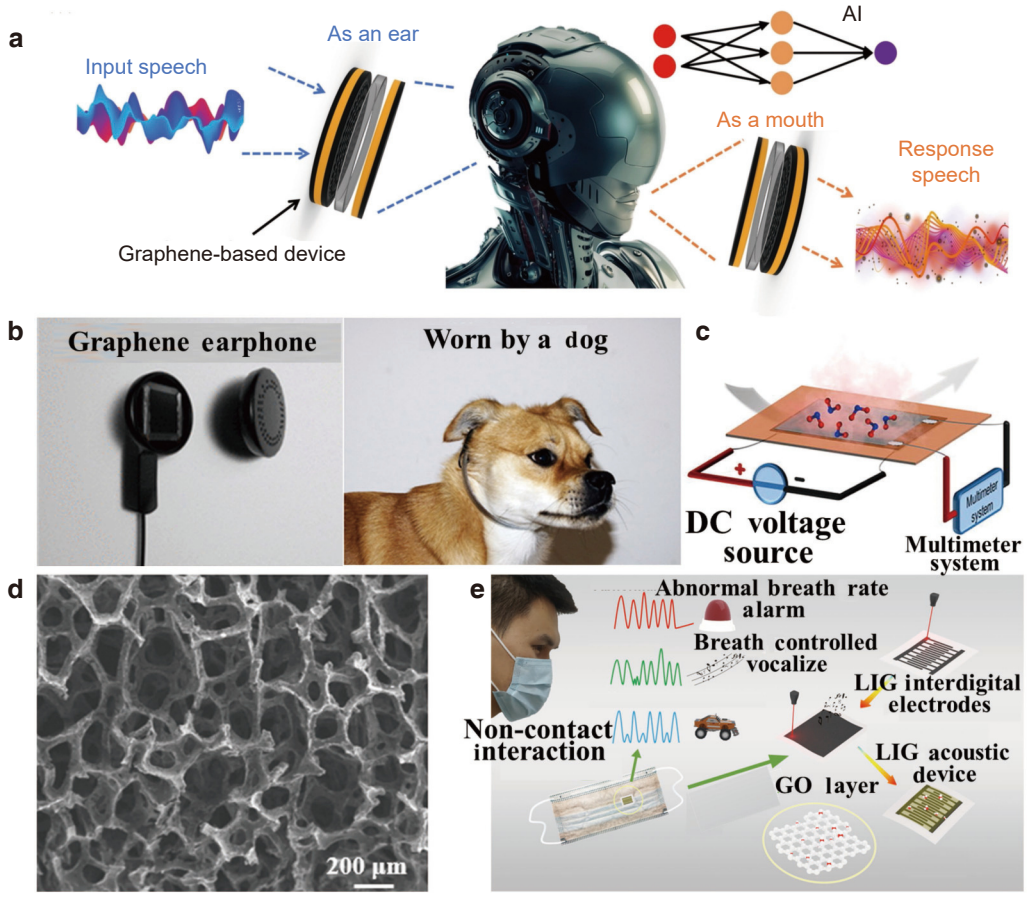

Fig. 2. Graphene-based sensors for ears, nose, and mouth. a, Graphene-based dual-function acoustic transducers for human-robot interfaces. Reproduced with permission from Ref.[38], © Sun, H. et al. 2022. **b**, Graphene earphone. Reproduced with permission from Ref.[40], © American Chemical Society 2014. **c**, The LIG-based multi-functional gas sensor. Reproduced with permission from Ref.[42], © American Chemical Society 2022. **d**, The SEM image of the porous graphene network for humidity sensing. Reproduced with permission from Ref.[21], © Elsevier B.V. 2018. **e**, The humidity-based HMI system. Reproduced with permission from Ref.[22], © American Chemical Society 2022.

that works through the thermoacoustic sound emission mechanism, both of which are made from laser-induced graphene (LIG). The interface has a high sensitivity (4500 $mV \cdot Pa^{-1}$) and can withstand a large number of cycles (1,000,000 cycles and 60 days). With the help of machine learning, this interface can achieve speech recognition, emotion analysis, content processing, and more, making it ideal for intelligent robotic communication. In addition, the graphene-like two-dimensional material is also suitable for high-performance acoustic sensors. Gou et al. proposed an ultrasensitive MXene-based intelligent artificial eardrum[39]. With two-stage amplification of acoustic signals, the artificial eardrum shows an ultrahigh sensitivity of 62 kPa^{-1} with a very low detection limit (0.1 Pa), showing great potential in wearable acoustical healthcare and HMI.

The thermoacoustic effect of graphene allows it to produce sounds that are beyond the range of human hearing, making it useful not only for humans but also for interacting with machines and animals. Tian et al. developed a graphene earphone using a one-step laser scribing technology[40]. The graphene earphone can play sounds ranging from 100 Hz to 50 kHz, a range of frequencies wider than commercial earphones. They demonstrate that a dog wearing the graphene earphone could be trained and controlled by 35 kHz sound waves (beyond the hearing range of the human) (Fig. 2b). The MXene sound source was developed after the graphene sound source. Gou et al. used the Ti_3C_2 MXene films to realize an MXene earphone[41]. The fabricated earphone has a higher sound pressure level (68.2 dB at 15 kHz) and shows excellent performance for acoustic HMI.

2.4 Nose: graphene-based gas sensors

Compared to our own noses, "electronic noses" may be able to sense odors more accurately, and even analyze gas types and concentrations. Gas sensors have great application prospects in environmental protection, healthcare, safety, HMI, and other fields. With a large specific surface area and strong adsorption ability, graphene has been widely used in gas-sensing applications. In one study, Peng et al. created a LIG-based NO_2 sensor with a self-alarm function using molybdenum disulfide (MoS_2) as the sensitive material in conjunction with LIG electrodes[42]. The sensor reaches 86.81% high sensitivity for 5 ppm NO_2, and 22.9% for 250 ppb. Additionally, the sensor can trigger an alarm when detecting an exceeded gas concentration, thanks to the thermoacoustic effect of the graphene. Mi et al. from our team developed a graphene sensor to monitor the ethanol gas concentration[43]. In contrast to the afore mentioned work, in this research, reduced graphene oxide (rGO) was used as the sensitive material and was fabricated into a sensor array, demonstrating significant promise in the fields of medicine and environmental science.

2.5 Mouth: respiration sensors

Respiration is one of the most essential physiological behavior of people, which can be used to assess the health condition or used in non-contact interaction. Pang et al. proposed a humidity sensor based on porous graphene for respiration monitoring[21]. A nickel foam was used as a template for the CVD graphene in order to form a 3D porous conductive structure (Fig. 2d), which is suitable for humidity monitoring. To enhance the sensing performance, surface modification of graphene was achieved through the use of GO, poly(3,4-ethylene dioxythiophene)-poly(styrene sulfonate) (PEDOT: PSS), and Ag colloids. The humidity sensor exhibits the excellent capability of distinguishing between different breathing states. Zou et al. created a humidity-based HMI system that utilizes LIG. The respiratory sensor of the system contains interdigital electrodes and a drop-casted GO layer, with a LIG acoustic device located on the opposite side[22] (Fig. 2e). The respiratory sensor consists of LIG interdigital electrodes and the drop-casted GO layer, with a LIG acoustic device on the reverse side. Integrating a humidity sensor and a thermoacoustic device into the facial-mask HMI system allows it to monitor respiration signals in real time and issue an acoustic alert if the signal is abnormal. Moreover, the system delivers a new method for non-contact remote control, which can be applied to various HMI applications during a pandemic.

2.6 Throat: graphene artificial throats

In 2011, our team demonstrated that graphene can emit sound[44]. This finding led to an expansion in the use of graphene in the field of acoustics. Since then, several sensors and actuators for HMI have been created, utilizing graphene's acoustic effects. One particularly promising development is the artificial throat, which shows potential for use in healthcare and HMI[15,16,23–25,45]. In this section, we present a chronological overview of notable graphene-based artificial throats that have been developed since our initial discovery.

In 2017, based on the thermoacoustic effect and strain-sensing ability of the graphene, an intelligent artificial throat with sound-sensing ability was developed[24]. The artificial throat was made of LIG on a flexible thin-film polyimide (PI) substrate, which was low-cost and can be attached to the human throat conformally (Fig. 3a). Compared with traditional sound sources and sound detectors, the graphene artificial throat can realize both sound emitting and sound sensing using a single device. It can detect the throat vibrations from a mute person and convert them into comprehensible sound, and help the disabled regain the ability to communicate verbally.

In 2019, for more comfortable wearable requirements and to improve speech classification ability, a skin-like ultra-sensitive graphene artificial throat was proposed[25]. A thin-film poly (vinyl alcohol) with LSG was obtained using a water-assisted transferring process. The skin-like artificial throat had a better attachment to the human skin (Fig. 3b), which resulted in a high detection sensitivity not only for the vibrations of the throat but also for the subtle movement of muscles of the throat. Besides, the LSG gave the artificial throat an excellent sound-emitting ability (75 dB at 0.38 W power, 2 mm distance). Moreover, a customized circuit system realized the *in-situ* acoustic signals' monitoring and processing. Its lightweight and soft nature made it more suitable for daily long-term wear than previous artificial throats.

A new artificial throat called the EMG-strain synergetic intelligent artificial throat (Fig. 3c) was developed in 2022 to improve signal detection accuracy and long-term wear comfort[22]. This throat is made of polyurethane (PU) nanomesh and LSG, making it breathable and flexible for comfortable long-term use. Furthermore, this work innovatively combined the laryngeal EMG signals with the strain signals to increase voice distinguishing accuracy. With a synergetic convolution neural network for processing EMG signals and a two-layer convolution neural network for processing strain signals, the artificial throat can realize 98.9% accuracy in language classification, which is expected to help people with voice loss rebuild their ability to communicate.

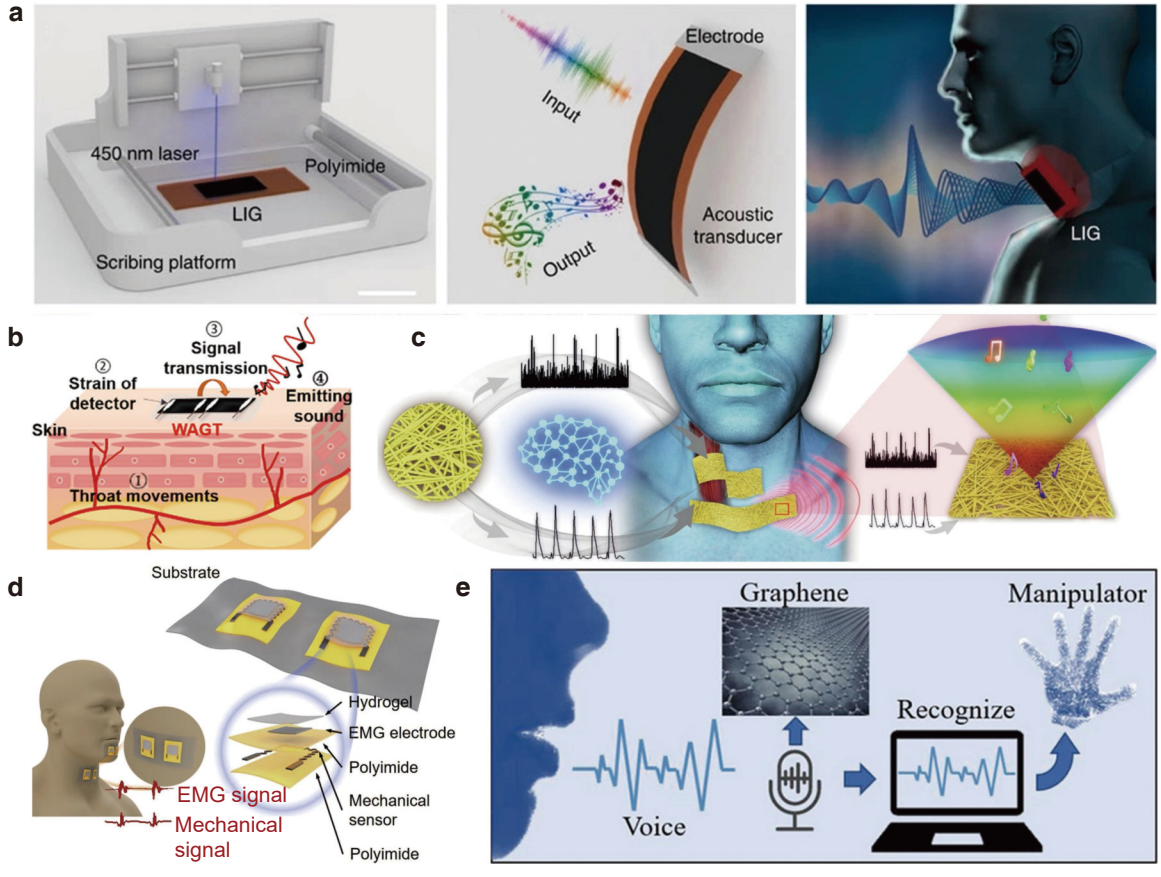

Fig. 3. Graphene artificial throats. a, The intelligent artificial throat with sound-sensing ability. Reproduced with permission from Ref.[24], © Tao, L.-Q. et al. 2017. **b**, The skin-like ultra-sensitive graphene artificial throat. Reproduced with permission from Ref.[25], © American Chemical Society 2019. **c**, The EMG-strain synergetic intelligent artificial throat. Reproduced with permission from Ref.[15], © Elsevier B.V. 2022. **d**, The bioinspired dual-channel. Reproduced with permission from Ref.[16], © Tian, H. et al. 2022. **e**, The deep learning enabled graphene flexible microphones. Reproduced with permission from Ref.[45], © American Chemical Society 2022.

At the end of 2022, the dual-channel artificial throat has shown improved speech recognition ability. Tian et al. developed a dual-channel speech recognition system utilizing graphene-based EMG and mechanical sensors (Fig. 3d), inspired by the facial expression signal transmission process from the brain to the muscle[16]. Considering that the energy of EMG is mainly concentrated in the high-frequency part and the energy of mechanical signals is concentrated in the low-frequency part, the fusion of two different kinds of physiological signals for speech recognition can reduce the number of electrodes worn and improve the recognition accuracy. The whole artificial throat system consists of two patches, each of which outputs one channel of EMG signal and two-channel mechanical signals. The study achieved a 100% recognition rate in a dataset comprising numbers 0–9 and a recognition accuracy of 96.85% in an extended dataset of 71 words, which is the highest accuracy on the largest single-word dataset to date. In addition, this artificial throat has excellent anti-noise characteristics and can achieve a recognition accuracy of over 95% in a 70 dB noise environment. These results demonstrate that bioinspired dual-channel fusion sensors have a promising future for speech recognition in complex environments.

Recently, further advances have been made in intelligent speech recognition based on graphene (Fig. 3e) [45]. Zhang et al. used deep learning to realize speech command recognition using graphene microphones. The human voices of numbers 0–9 and 20 sentences were collected and constituted a dataset with 1,500 samples. These findings showcase the potential for implementing the graphene-based artificial throat in real-world applications.

2.7 Hands: gesture recognition and tactile sensing

As an excellent sensitive material, graphene is a good choice for realizing high-performance strain and pressure sensors which are suitable for gesture recognition and tactile sensing.

Owing to the trade-off between the gauge factor and the detecting range of traditional strain sensors, when facing the hand's movements with both large and small motions, their sensitivity to gesture recognition is challenged. To address this issue, Tan et al. devised a laser-programming approach for customizing the performance of graphene-based strain sensors[46]. By optimizing the parameters of the laser scribing process during the fabrication of the LSG-based strain sensor, they were able to adjust the gauge factor with high sensitivity, spanning 2 orders of magnitude. As shown in Fig. 4a, a pair of finger-attached graphene sensors can recognize a variety of gestures and distinguish the signals used to operate a computer mouse. This study sheds light on the potential of customizing sensing performance to meet specific HMI requirements.

Although the exposed strain sensors usually have a better gauge factor, encapsulation can increase the strain range and the durability of sensors. Lv et al. used ultrathin encapsulation to realize a large strain range of the strain sensor while maintaining high sensitivity[47]. The fabrication process is straightforward. First, drop-

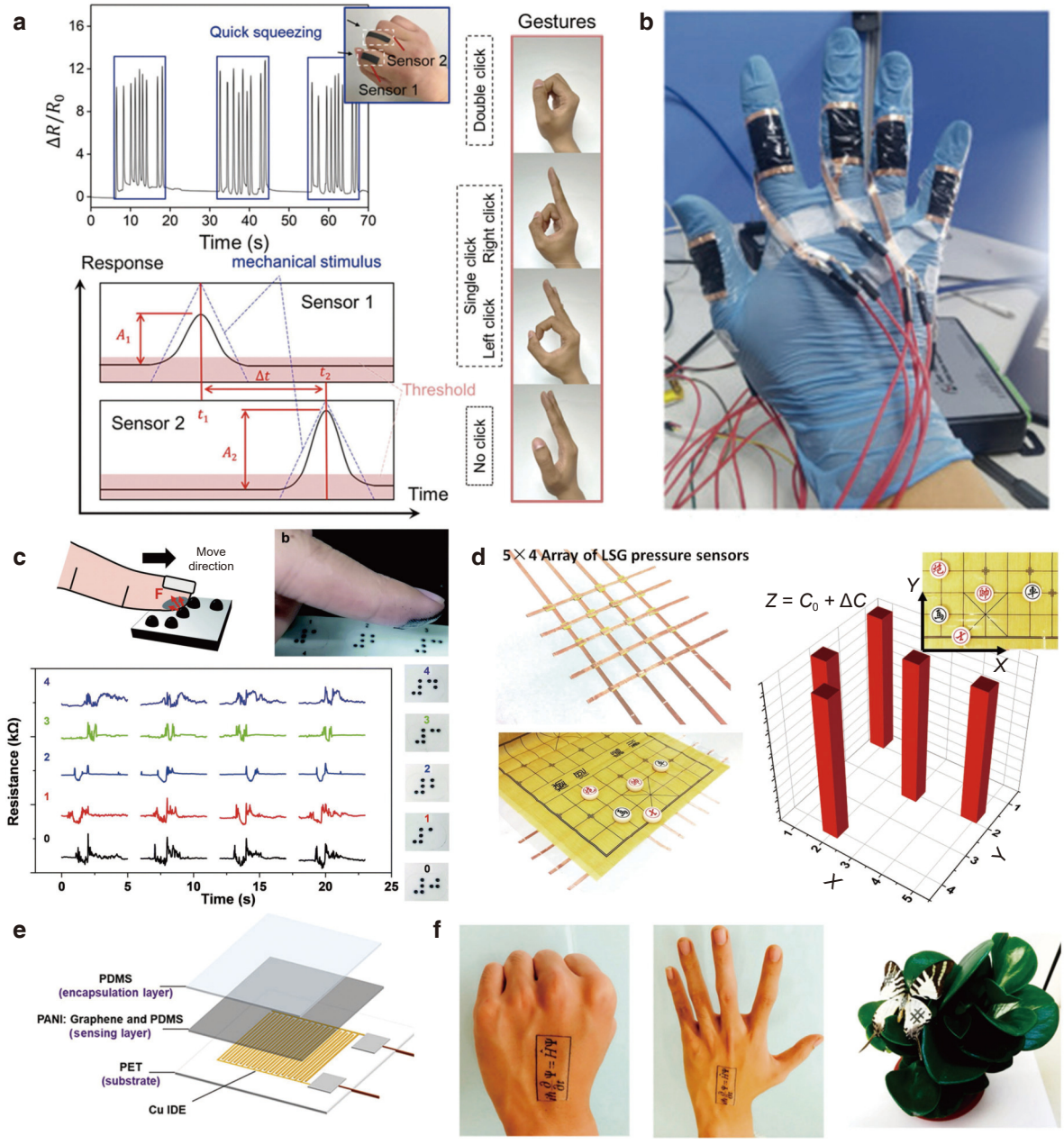

Fig. 4. Graphene-based gesture, tactile, temperature, and physiological sensors. a, The hand gesture classification for HMI. Reproduced with permission from Ref.[46], © American Chemical Society 2021. **b**, Ultrathin encapsulated strain sensors for gesture recognition. Reproduced with permission from Ref.[47], © Elsevier B.V. 2022. **c**, The intelligent tactile sensor for distinguishing convex Braille numbers. Reproduced with permission from Ref.[26], © The Royal Society of Chemistry 2022. **d**, A 5 × 4 array of pressure sensors integrated into a chess board. Reproduced with permission from Ref.[27], © Tian, H. et al. 2015. **e**, The ultrahigh linear sensitive temperature sensor. Reproduced with permission from Ref.[28], © American Chemical Society 2022. **f**, The multilayer graphene epidermal electronic skin. Reproduced with permission from Ref.[48], © American Chemical Society 2018.

casting GO on the ultrathin PU substrate and acquired rGO sensitive layer by laser scribing GO. Second, adding copper interaction on two sides of the rGO. Finally, using another PU film to encapsulate the sensor. As shown in Fig. 4b, by applying five strain sensors on five joints on the right hand, multiple-channel signals can accurately distinguish different hand movements with high sensitivity.

Pressure sensors with high sensitivity and flexibility are suitable for wearable tactile sensing. Besides, if sensors can be worn senselessly, it can minimize the impact on people's actual feelings when wearing such sensors. Qiao et al. inspired by the structure of the reinforced concrete realized a nanomesh-reinforced graphene pressure sensor[26]. The PU nanomesh skeleton helps the pressure sensor undertake higher pressure, which increases the measuring range and stability of the sensor. Moreover, because of the positive resistance response coming from the porous graphene structure, the sensor has a higher sensitivity under higher pressure. This positive resistance response can be suitable for applications such as helping blind people to learn Braille or helping disabled people reconstruct tactile sensations. When touching the Braille with a small area, the pressure sensor should sustain a high pressure of up to 1 MPa, which exceeds the detecting range of most flexible sensors. The

sensor reinforced with nanomesh, as depicted in Fig. 4c, can sensitively detect pressure changes when affixed to the forefinger and touched against Braille numbers. Additionally, when integrated with a convolutional neural network, the sensor can achieve the autonomous classification of the touched information.

Achieving high sensitivity and a large measuring range is a major challenge in developing pressure sensors for tactile sensing applications. In 2015, Tian et al. discovered a way to produce a graphene pressure sensor with remarkable sensitivity across a wide range[27]. The LSG made on the PI substrate has a loosely V-shaped stacked structure. When two LSG-PI films are arranged in a face-to-face stack to create a pressure sensor, the loose stack graphene becomes denser as pressure increases, causing a significant resistance change that results in high sensitivity. To accurately detect pressure distribution for HMI applications, the sensor can be converted into an array. Fig. 4d displays a Chinese chess board integrated with a 20-pixel pressure array. This array can detect the chess piece layout in real time by analyzing the pressure distribution.

2.8 Skin: temperature sensing and physiological signal monitoring

Human skin is a vital conduit for humans to interact with the environment. It comprises numerous innate sensors and actuators that can detect external stimuli like temperature and transmit various physiological signals. In this section, we highlight two specific areas-temperature sensors and heaters for thermal interactions[28,29] and electronic skin for monitoring multiple physiological signals[48,49].

One of the most significant indicators of both the body's state and the surroundings is temperature. In their research, Liu et al. introduced a temperature sensor that exhibits rapid response, high sensitivity, and excellent linearity, relying on a graphene-polydimethylsiloxane (PDMS) composite (Fig. 4e)[28]. To give the sensor good linear characteristics, this work creatively used polyaniline (PANI) with unique negative temperature characteristics to compensate for the graphene composite. The proposed sensor has a 1.6%/°C sensitivity, a 0.3 °C accuracy, and a 0.7 s response time. By combining with a read-out circuit, it shows capabilities including monitoring real-time skin temperature, ambient temperature, respiratory rate, etc. Zhang et al. developed a super flexible and custom-shaped graphene heater[29]. The heater can be fabricated with customized patterns through a one-step laser scribing process and can reach 247.3 °C in just 20 s. Moreover, this work shows its application scenarios including flexible warmers and displays, which apply to the thermal HMI.

The graphene electronic skin (e-skin) is a kind of epidermal sensing platform. It is very suitable for the monitoring of various epidermal physiological signals. Qiao et al. created a versatile e-skin sensor using multilayer LSG technology that can be easily attached to a range of surfaces (Fig. 4f)[48]. When encapsulated using Ecoflex, the e-skin sensor shows high sensitivity for both subtle and large epidermal physiological signals including pulse, respiration, joint movement, and finger bending, which shows potential in daily healthcare and long-term physical indicators monitoring.

2.9 Limbs: motion sensing and interaction

Limbs are the natural medium of HMI, which can produce a lot of motion information. On the one hand, a variety of body language can be obtained through motion sensors by recording people's movements[30,32]. On the other hand, motion actuators (bionic soft robots) can be used as an extension of human limbs to achieve non-contact HMI or perform more diverse tasks[31,50,51].

The motion range of the human body can be large or small, which means that the motion sensor needs a high sensitivity in a large linearity range to accurately identify the human motion signal. Inspired by the human skin whose reticular layer contains force receptors that have high sensitivity even under large external stimuli, Pang et al. proposed a graphene pressure sensor with randomly distributed spinosum (Fig. 5a)[30]. By using an abrasive paper with spinosum microstructure as the template and coating it with PDMS, the epidermis microstructure can be transferred to the PDMS film. Then, PDMS with microstructure was used as the substrate of the sensor and was coated with GO solution. After the drying and the thermal reduction processes, the conductive rGO was realized as the sensitive material. Finally, by coating another layer of PDMS to encapsulate the rGO layer, the sensor was made. According to structural analyses, the randomly distributed microstructure contributes to the high sensitivity and large linearity range of the sensor. The sensor performs well when detecting typical human motions such as pushups, walking, and arm bending.

As a daily necessity, clothing has many advantages, such as large areas, direct attachment to the human body, and flexibility, which make it suitable as the carrier of wearable motion sensors. Yang et al. facing the need for human motion monitoring proposed a graphene textile strain sensor with negative resistance variation[32] (Fig. 5b). Following a similar process used in industrial clothing production, the researchers utilized GO solution as a dye to color the polyester fabric and further reduced it at high temperatures. Most significantly, this strain sensor exhibits a distinctive negative resistance variation with increasing strain. Notably, this strain sensor exhibits unique negative resistance variation as the strain increases. With its exceptional performance, the graphene textile strain sensor can be seamlessly integrated into clothing to detect both subtle and significant human motions, showing promising prospects for daily HMI applications.

In order to achieve movement, motion actuators are required in addition to motion sensing. Graphene's excellent thermal and electrical properties make it very suitable to be used as the core material of electrothermal actuators (ETA)[50]. Using selective laser reduction of GO on a thin film PI substrate, they created a U-shaped robotic hand. This method of manufacturing is highly efficient, making soft robots suitable for large-scale, low-cost applications[31]. The graphene ETA, when used as a robotic hand, is capable of achieving a distinctive large bending angle of 270° in just 8 seconds using a 65 V driving voltage, and takes 19 seconds to recover. Furthermore, it can hold objects that weigh over ten times its own weight (as shown in Fig. 5c). The graphene ETA exhibits great potential in fields such as biomimetic robotics, artificial muscles for healthcare, and HMI.

Artificial intelligence (AI) devices and systems typically rely solely on electrical input for self-training and function execution, while the human body possesses a unique ability to detect and process multiple types of signals for dynamic self-training and self-renewal. The nervous system, for instance, evolves through continuous learning, while human muscles can also enhance themselves through mechanical input, such as exercise. Through physical training, muscle fibers are first broken down and then rebuilt to become stronger. Building on this concept, Deng et al.

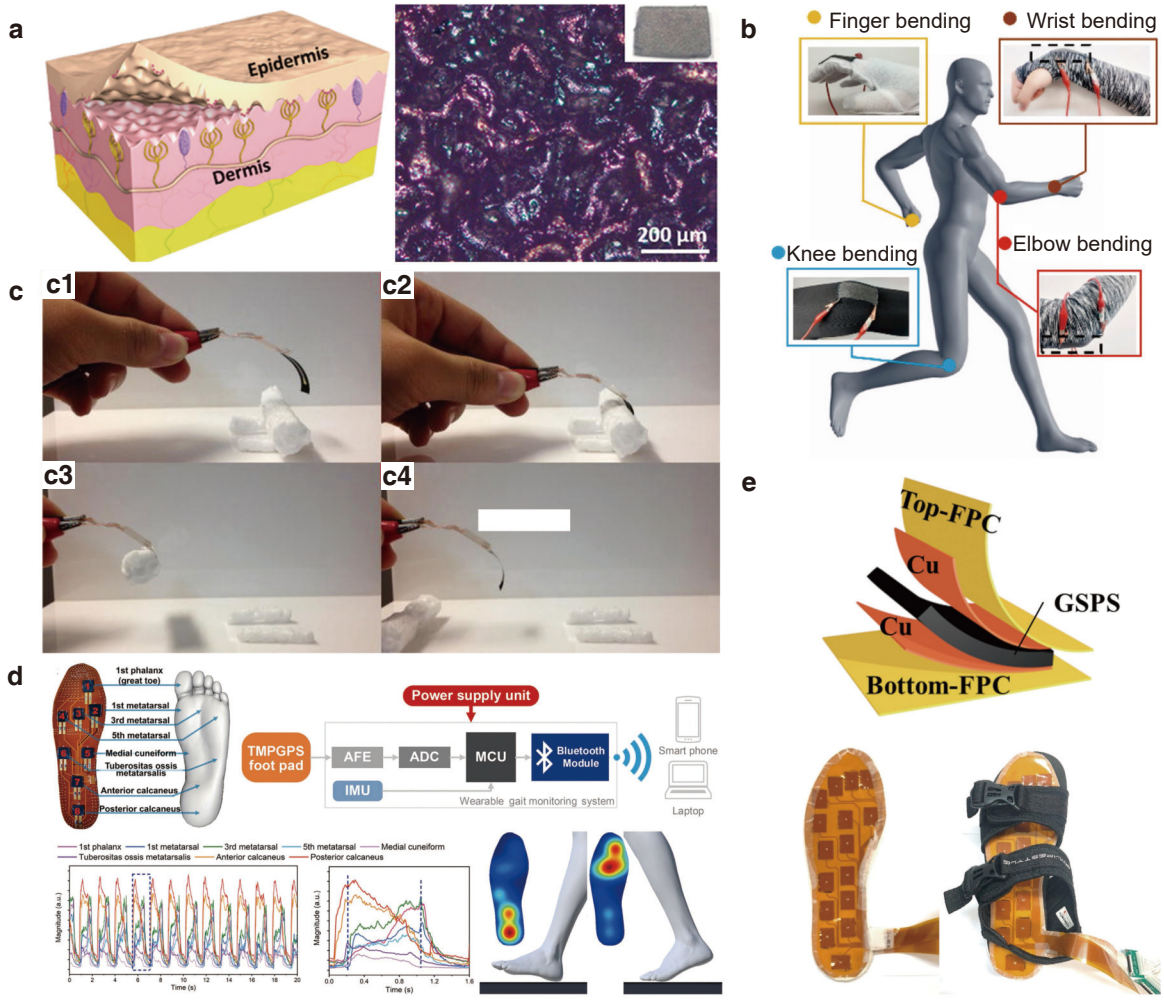

Fig. 5. Graphene-based motion sensors, actuators, and gait monitoring system. a, The epidermis microstructure inspired graphene pressure sensor. Reproduced with permission from Ref.[30], © American Chemical Society 2018. **b**, Graphene textile strain sensor. Reproduced with permission from Ref.[32], © American Chemical Society 2018. **c**, A simple robotic hand based on LSG. Reproduced with permission from Ref.[31], © AIP Publishing 2017. **d**, The triode-mimicking graphene pressure sensor. Reproduced with permission from Ref.[33], © American Chemical Society 2020. **e**, The in-shoe system based on the porous graphene pressure sensor. Reproduced with permission from Ref.[34], © Cui, T. et al. 2021.

created an artificial intelligence graphene muscle[51]. The multiple "muscle lines" consisting of graphene can be directly printed on a flexible substrate. After undergoing long-term strain training, the resistance of the graphene muscle lines displays three stages: decrement, enhancement, and stabilization, similar to human muscle behavior. This effect is derived from changes in the transverse and longitudinal overlapping areas of graphene nanosheets on a microscopic scale. Additionally, the electrical signal can be used to drive the graphene muscle, enabling it to stretch, relax, and contract, mimicking human muscle function due to the electrothermal properties of graphene. Based on the sensing and actuating performance of the graphene muscle line, they further proposed a new concept called "Force Learning", which reached a high accuracy of ~ 96%. This study highlights the potential for graphene-based devices to possess learning capabilities, which could lead to exciting possibilities in the development of intelligent HMI.

2.10 Foot: gait monitoring

Gait patterns can provide a lot of physiological and psychological information about people, and have been preliminarily applied in disease diagnosis, postoperative rehabilitation, behavior analysis, and other fields. Graphene-based pressure sensors offer excellent flexibility, a broad measurement range, and high sensitivity, making them an ideal choice for gait monitoring.

Almost all existing pressure sensors are based on negative resistance variations, making it difficult to remain a high sensitivity when subjected to significant pressure. This can pose a significant drawback when monitoring gait because the pressure on the sensor can reach human weight levels. To address this issue, Wu et al. developed a pressure sensor that exhibits a positive resistance-pressure response based on LSG[33]. The sensor has a unique triode-like structure, where the force surface of the sensor acts as the base of the triode, and static bias of the pressure-regulating triode enables mechanical signal detection and amplification. Within the measurement range, the sensor's sensitivity escalates as the pressure increases, making it apt for detecting substantial pressure signals with precision. Fig. 5d reveals that the gait system incorporating various pressure sensors based on graphene can record human gait information accurately, displaying its potential in healthcare, high-accuracy sensing, and HMI applications.

To collect gait information more accurately and more comfortably, and make the graphene-based gait monitoring system suitable for long-term use in daily life, we further developed a flexible insole gait monitoring system (Fig. 5e)[34]. To achieve this, we used a biomimetic soft and porous styrene-butadiene rubber as the sensor's skeleton to provide a comfortable wearing experience. The graphene slurry was coated to the porous rubber skeleton to form a three-dimensional conductive network. According to the various stress points of the sole of the human foot, sixteen pressure sensors were integrated into the insole system to obtain complete gait information. The top and bottom of the insole were encapsulated using PI flexible circuit boards which were also used to realize the multi-channel sensor electrical connection. When used for gait monitoring, the insole system can accurately record gait information including walking, running, jumping, squatting, etc. In addition, the gait system can be customized to a person's foot size. Combined with the excellent performance of the graphene sensor and the flexible integration methods of the system, it is very suitable for long-term daily wear for gait information interaction.

3 Discussion

In the preceding sections, our team's recent research served as an example to summarize various graphene-based sensors for HMI applications in a comprehensive manner. The design ideas, fabrication methods, and performance parameters of selected graphene-based sensors are highlighted. As an important part of HMI interfaces and a possible extension of human ability, graphene-based sensors for HMI are expected to become more diverse and practical in the coming years. In the same part of the body, the human and machine can interact with different signals, or interact in many different ways. The human body and machines can interact in various ways and use different signals within the same body part. Our team is focusing on developing graphene-based sensors for HMI, which is just a small fraction of the expanding range of related research. We aim to inspire more researchers to explore this field using our work as a starting point.

Next, we will briefly summarize our proposed development directions of graphene-based sensors for HMI, which we believe are crucial research areas for future investigations.

3.1 Multi-model graphene-based sensors for HMI

Graphene-based sensors need to be designed to monitor diverse signals in various application scenarios, to meet a wide range of HMI requirements. The development of multi-modal sensors with adaptable signal acquisition capabilities for different application scenarios is a promising direction to pursue in this context of using graphene-based sensors.

In section 2.9, we introduce a graphene-based textile sensor for multiple physiological signal monitoring. To further achieve a multi-model garment with multifunctional graphene-based sensors, Wei et al. developed a wearable batch-prepared graphene-based textile based on LSG and thermal-transfer technology[52]. In order to address the limitations of conventional textile sensors that can only gather signals and are difficult to integrate with actuators to deliver instant feedback, the researchers developed a textile sensor utilizing graphene technology. This sensor is capable of detecting strain, pressure, physiological electrical signals, and emitting sound, and has been incorporated into clothing to work in tandem for health monitoring purposes (as shown in Fig. 6a). With the help of this graphene-based multi-modal sensor, the study has created smart clothing that can provide comprehensive multiple physiological signals monitoring-signal analysis-abnormal alarm function. Taking ECG monitoring as an example, the graphene-based ECG sensor on this cloth collected ECG signals and transmitted them to the processing unit in real time, when the abnormal ECG signal is detected, the multi-model graphene sensor becomes a sound emitter and gives the wearer health tips or warnings. This multi-modal graphene-based sensor combines a wide range of capabilities and is suitable for large-scale production and applications. It can greatly reduce the complexity of HMI system construction and has the promise to play an important role in HMI and daily healthcare.

Lightweight and flexible wearable electromagnetic shielding devices are of great significance to human health and the protection of wearable electronic devices[53]. If a multi-mode electromagnetic shielding armor can be realized, apart from its basic body protection function, it is also suitable for daily HMI functions including physiological information sensing. Xu et al. proposed a multi-model graphene armor that can realize both human electromagnetic interference (EMI) shielding and motion monitoring[54] (Fig. 6b). The multi-model sensor consists of laminated graphene film and hierarchical porous graphene foam with a film/foam/film sandwich structure. The laminated graphene film was made using a single-step laser induction process and the porous graphene foam was realized by dip-coating porous styrene-butadiene rubber into graphene suspended in the liquid. The sandwich sensor with a 2 mm thickness can realize a high EMI shielding effectiveness of up to 69.8 dB and can realize flexible adjustment by changing the thickness of the sensor. Furthermore, the porous graphene foam is attributed to the excellent strain-sensing performance of the sensor. The gauge factor can reach 258 with a 100% strain range. This multi-model graphene armor sensor offers stretchability, lightweight, and conformability. It ensures EMI shielding comfort e.g. for pregnant women without affecting the fetus and also protects the joints of the human body. Besides, with good strain sensing performance, it can detect multiple physical signals including pulse, voice, motions, etc. The graphene armor, which combines electromagnetic shielding and multi-physiological signal monitoring, has great potential in the field of wearable health monitoring and HMI.

3.2 High comfort graphene-based sensors for HMI

The advancement of HMI applications necessitates wearable sensors that are both lightweight and flexible. Since numerous physiological and psychological signals of the human body require continuous monitoring to obtain adequate information or to enable real-time information exchange, the comfort of wearing graphene-based sensors has become a critical area of research in addition to enhancing detection performance[7, 14, 55]. The important factors of comfort include improving the flexibility of the sensor to avoid wearing discomfort, making the sensor breathable to avoid sweat accumulation, and ensuring that the sensor is biocompatible and safe for contact with human skin. In general, the principle governing sensor wear is that it should have minimal impact on the wearer's daily life, and the wearer's daily activities should have minimal impact on the sensor's performance. In this section, we provide some techniques for creating high-comfort graphene-based HMI sensors using representative works from our team.

To avoid the influence of the sensor's substrate on the wearing comfort, Qiao et al. proposed a substrate-free multilayer graphene

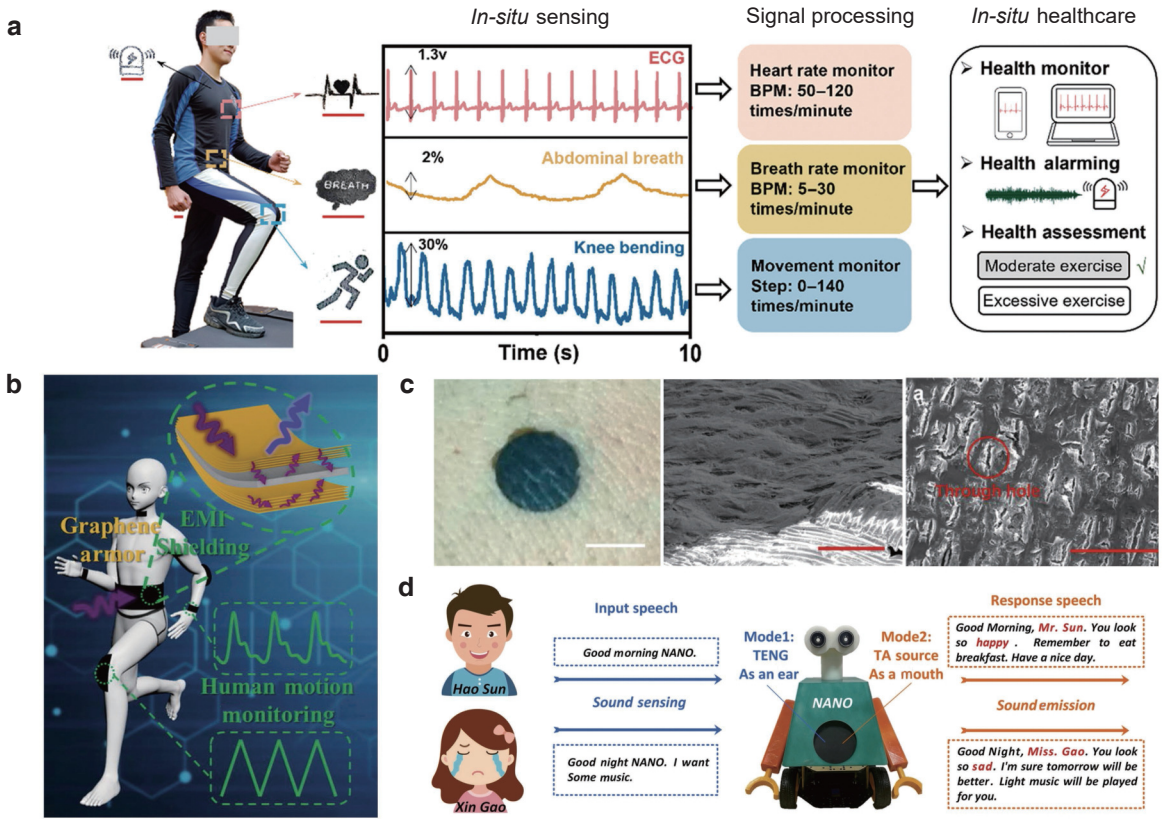

Fig. 6. Multi-model, high comfort, or intelligent graphene-based sensors for HMI. a, The "intelligent clothes" consists of multi-model graphene-based textile sensors. Reproduced with permission from Ref.[52], © American Chemical Society 2021. **b**, The electromagnetic interference shielding armor with strain sensing ability. Reproduced with permission from Ref.[54], © Elsevier B.V. 2021. **c**, The substrate-free multilayer graphene e-skin for physiological signal monitoring. Reproduced with permission from Ref.[55], © American Chemical Society 2020. **d**, The machine learning-assisted human-robot interfaces. Reproduced with permission from Ref.[38], © Sun, H. et al. 2022.

e-skin for physiological signal monitoring[55]. The manufacturing process is as follows: First, GO solution was mixed with tetrahydrofuran (THF) and was dropped onto a three-layer transfer paper with a supporting layer, a sacrificial layer, and a substrate layer and was dried to form a uniform film. Second, using a laser-scribing technique, the surface layer of the GO film was transferred onto the LSG film. Third, the sacrificial layer was dissolved in water and the substrate layer with the LSG film was transferred to the human skin. Finally, the LSG layer can be peeled off, leaving the substrate-free LSG layer on the skin. As shown in Fig. 6c, the substrate-free graphene e-skin shows good attachment to the human skin, and it fits closely to the skin lines. The substrate-free e-skin has better flexibility and stretchability than the sensor with a substrate. This e-skin fits well with the skin, improving the quality of the collected signal, such as the ECG acquisition. The electrode-skin contact impedance is reduced due to the skin conformal adhesion of the substrate-free e-skin, which greatly enhances the anti-interference ability during signal acquisition. It enables continuous signal acquisition without interrupting daily activities. Most significantly, compared to sensors with substrates, the substrate-free graphene e-skin has better breathability. As shown in Fig. 6c, like the stomata used by plants for transpiration, respiration, and photosynthesis, the LSG has holes on it to ensure the perspiration process of the wearer is not impeded. This creatively avoids the impact of sweat accumulation between the sensor and the skin on comfort, device life, and signal quality during long-term wear, and improves the wear experience.

Fiber sensors have been widely used in wearable sensors with high comfort in recent years due to their intrinsic breathability and flexibility. The graphene-based sensor with an electrospinning substrate proposed by Qiao et al. which we have mentioned in section 2.1 is a good example[14]. Biocompatible PU is spun into a nanoscale fiber network by electrospinning. As a sensitive material, graphene covers the surface of the fibers to form a three-dimensional conductive network. The weight of the sensor is at the milligram level and the soft material used to make it ensures that it is nearly imperceptible to the wearer. Additionally, the nanomesh serves as the sensor's framework, enhancing its durability and life while also allowing for good air permeability, enabling the sensor to be worn for multiple days. This high-performing sensor is ideal for continuous, long-term wearable signal monitoring and HMI applications due to its exceptional performance, strength, and wearable comfort.

3.3 Intelligent comfort graphene-based sensors for HMI

Endowing sensors with intelligence can enhance their information processing efficiency and expand their use in intelligent HMI. This trend toward intelligence is currently being observed in many wearable sensing systems, including those based on graphene-based sensors. In this section, we will provide a brief overview of some of the recent developments in the intellectualization of graphene-based sensors for HMI.

Based on the graphene-based dual-function acoustic devices mentioned in section 2.3, Sun et al. further developed a speech recognition system for human-robot interaction[38]. They combined the graphene sensor with machine learning to create an HMI

system that can sense speech, analyze emotions, recognize the identity, and actuate sound. To train and test their models, they gathered 78360 speech data and used the intelligent acoustic system on a robot called "NANO". As depicted in Fig. 6d, the robot's entire system activates when it detects the wake-up word "Nano" spoken by a human through the graphene-based acoustic sensor. Then, the person communicating with the robot is identified by voice. Further, by analyzing the emotional features of the speech, such as happy or sad, "NANO" can understand and respond in real-time with greetings or reassurance with the graphene-based acoustic actuator. The utilization of dual-mode graphene-based acoustic devices in this study highlights the potential and distinctive qualities of intelligent graphene-based sensors in acoustic HMI applications.

Coupled with intelligence, graphene-based sensors can realize more efficient HMI. In section 2.2, the three-dimensional HMI system invented by Xu et al. improves the efficiency of eye-movement recognition with the help of intelligent algorithms[17]. To make the machine recognize the nine kinds of eye movements made by humans (normal blinking, look up, look down, look right, look left, closed down, closed up, up blinking, down blinking), a machine-learning algorithm was designed and trained by the EOG dataset collected by the graphene-based EOG sensors. Although it is challenging to manually classify EOG signals that contain nine types of eye movement information, an intelligent algorithm can proficiently distinguish the variances between them. The nine eye movement signals can therefore control the UAV's movement in a two-dimensional plane. Nine kinds of eye movement signals can control the movement of the UAV in a two-dimensional plane. When combined with Z-axis control instructions identified by the graphene tactile sensor array, the entire graphene-based sensing system can attain a comprehensive and efficient three-dimensional HMI. This intelligent and multi-modal graphene-based sensor presents an astute solution and promises to meet the needs of future HMI applications.

As graphene-based wearable sensors become more multi-model, more comfortable, and more intelligent, they are expected to develop into "information organs" integrated with the human body with *in-situ* sensing, intelligent processing, and interaction abilities in the future.

The "information organ" should be able to realize the complete absence of interference to the wearer when wearing, realize independent thinking, and be able to assist the wearer from the outside and improve the wearer's ability like an organism with comprehensive capabilities. Besides, it may have the ability including self-repair function and self-update function to achieve long-term stable wearing and autonomous improvement. In terms of hardware composition, they may include a variety of sensors and other information acquisition/application modules, and should be integrated with the skin in many aspects including materials, structure, function, etc., with good flexibility and biocompatibility, and at the same time can achieve rich functions with high performance.

4 Conclusions

Based on the research of our team in recent years, in this review, we methodically introduce some typical graphene-based sensors for HMI applications. By summarizing their design methods, manufacturing process, and performance characteristics in a "people-oriented way", we demonstrate sensors with promising applications that can be adapted to a variety of HMI scenarios. These sensors show promise for various HMI scenarios, and we discuss potential directions for their further development. It is expected that graphene-based sensors for HMI will evolve into multi-modal, high-comfort, and intelligent devices capable of adapting to diverse application scenarios and prolonged daily use. They are also featuring enhanced information processing capabilities and may one day serve as an "information organ" with *in-situ* sensing, intelligent processing, and interaction abilities.

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Author contributions

T.C. wrote the original draft and played a leading role in the investigation. D.L. and Y.Q. contributed to the Section on graphene-based sensors for healthcare applications and revised the manuscript. T.H. and J.X. contributed to the Section on graphene-based sensors for intelligent human-machine interaction applications and revised the manuscript. The Section of graphene devices-related to sound sensing were provided by H.X. The whole work was done under the conceptualization and supervision of T.-L.R., Y.Y., and H.L.. H.T. provided some valuable suggestions for this paper and revised the manuscript. All authors have given approval of the final version of the manuscript.

Competing interests

The authors have no competing interests to declare that are relevant to the content of this article.

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