An Overview of Graphene-Based Nanomaterials in Electronic Skin Biosensing

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Abstract Skin, the largest organ in the body, is capable of detecting and reacting to a variety of external stimuli. The development of electronic skin (E-skin) for the imitation of the human sensory system has recently gained a lot of attention due to its potential applications in wearable human health monitoring and care systems, advanced robotics, artificial intelligence, and human–machine interfaces. Electronic skin sensing devices have accelerated due to graphene's capacity to achieve unique functionality using a variety of assembly processable processes. Consequently, the use of graphene and the components that make it up in biomedicine is growing. This review focuses on high-performance electronic skin that has been developed for biosensing applications through a number of research projects. Additionally, a brief discussion of electronic skin's production processes, research obstacles, and future prospects was included.

Keywords Graphene-based nanomaterials · Electronic skin · Biosensing · Nanotechnology

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Introduction

One of the effective and interesting in flexible electronics is the flexible electronic skin, or e-skin [1]. E-skins imitate the entire spectrum of human sensing abilities, and because they are flexible and biocompatible, they have a wide range of potential applications in touch-sensor technologies, artificial intelligence systems, personal healthcare monitoring, and human–machine interfaces [2, 3]. The e-skin resembles a stretchable, flexible skin with intelligent processing abilities [4]. Pressure sensing, which turns pressure into electrical signals and processes them appropriately, is the main purpose of the system. Traditionally, an electrocardiogram (ECG) measures the electrical potential on the body's surface, which is created when the action potential of the heart muscle cells changes in a person. The e-skin transmits energy and signals in order to remotely get the ECG signal. In contrast, the e-skin can reach the goal of real-time monitoring of human health and disease prevention by meeting the requirements of anytime, anywhere detection and long-term monitoring in the prevention and treatment of cardiovascular diseases [5].

Nanomaterials are a component of a commercial revolution that has given rise to an explosion of hundreds of new products as a result of their varied physico-chemical properties, which allow for their use in a wide variety of creative applications [6–23]. For the purpose of developing flexible and stretchable sensors, nanomaterials such as nanoparticles, metal nanowires, carbon nanotubes, graphene, and porous silicon are commonly selected as sensing materials [24, 25]. Among the materials mentioned above, graphene is a promising 2D material in many applications due to its extraordinary multiple properties, including having a high electrical conductivity, being extremely light, ultrahigh carrier mobility, excellent electrical conductivity, superior thermal conductivity, large theoretical specific surface area, high optical transmittance, high Young's modulus, and having exceptional mechanical strength and outstanding mechanical flexibility [26, 27]. The powerful bonds and van der Waals interactions between layers in particular give graphene a tendency to aggregate [28], creating a regulated self-assembly structure that distinguishes it from other materials and makes it a great option for e-skin. Because of graphene's exceptional biosensor capabilities, including its high electron transfer rate, wide potential window, and large specific surface area, receptors including enzymes, antibodies, and deoxyribonucleic acid (DNA) can be effectively immobilized on its surface [29, 30]. These graphene bioelectrodes also had excellent durability up to 5,000 compression cycles, super stretchability with a maximum strain of 150%, and a low sheet resistance of about 1.5 k per square, which indicated the potential for low-cost processing and rage-scale application for future wearable electronic skin [30].

This review focuses on new, high-performance electronic skin that has been developed for biosensing applications through a number of research projects. Additionally, a brief discussion of electronic skin's production processes, research obstacles, and future prospects was included.

The Fabrication Methods of Electronic Skin

Electronic interface-based bionics, or e-skin, is flexible. Similar to a biological skin, it can detect changes in pressure, touch, humidity, and temperature as well as identify the various shapes and textures of outside materials [31, 32]. Typically, flexible sensing electronic devices have a four-part structure. [5]: (i) Elastic and flexible substrates (ii) electrodes that conduct, (iii) sensing materials, and (iv) encapsulation materials. The conducting electrodes are used to transmit electrical signals, the sensing materials transform environmental stimuli into detectable electrical signals, and encapsulation material is used to protect the sensing material from potential external damage. A flexible substrate supports the e-skin and fits it to the biological skin. Therefore, choosing stretchy materials with an elastic modulus is necessary for the preparation of e-skin. Stretchable materials, hard materials with breaks in them, and bendable and flexible materials are the three options that are typically offered. The wavy flexible device, which has a particular amount of stretchability, is produced by combining the flexible device with the elastic substrate that has already been pre-stretched. While the wavy structure also affects the contact tightness, some electrical materials' characteristics change as they are bent. Although it is currently very challenging to obtain a high match between the tensile qualities and the device layout density, stretchable devices can be made employing discontinuous rigid components on flexible substrates. This technology can use existing high-performance devices. Also necessary for e-skin are superior mechanical and electrical qualities for stretchable materials. One remedy is to inject electroactive fillers into the insulating elastomer, but doing so too often will compromise the material's stretchability. Lowering the percolation threshold and constructing two-dimensional networks of one-dimensional materials, such as carbon nanotube networks, can increase the conductivity and stretchability. Altering the chemical and physical characteristics of polymeric electronic materials is another approach. One example is the addition of nonionic plasticizers to PEDOT: PSS, which has strong electrical conductivity but poor stretchability.

High-Performance Electronic Skin Preparation Using Novel Techniques

Researchers have been creating new techniques to make it simpler and more practical to prepare electronic skin with excellent performance in order to introduce it into people's lives as soon as possible. For example, Chen et al. [33] developed a wearable touch sensor that can simultaneously sense external pressure and has object recognition capabilities after being inspired by the superior electrical properties of electron-induced perpendicular graphene (EIPG) materials. The sensor employs a layered structure with elastic dielectric layers as electrodes (the dielectric layers are only 50 nm thick), and it primarily employs an electron cyclotron resonance (ECR) sputtering system to prepare vertically aligned graphene sheets, which results in a more efficient electron transfer channel and a quick and sensitive response in a broad pressure sensing range. Based on the drop in capacitance when approaching and leaving, this capacitive sensor can distinguish between neighboring objects. This is an efficient way to create vertical graphene nanosheets in flexible sensor electrodes, which take on a new function in the creation and advancement of e-skin.

Electronic skin with long-term stability, rapid response and high sensitivity has great worth in biomedicine, robotics, and in other fields. In terms of sensitivity and response time, electronic skin still faces difficulties. Lü et al. [34] presented flexible electronic skin based on piezoresistive graphene films with high sensitivity and quick reaction to address this issue. The SEM micrograph and image of the manufactured graphene film are shown in Fig. 1. A pressure sensor array made up of a 4 × 4 tactile sensing unit made up the electronic skin. The underlying substrate (polyimide substrate), the middle layer (graphene/polyethylene terephthalate film), and the upper substrate bump were all present in each sensing unit (polydimethylsiloxane). The dimensions of the electronic skins and a photo of an electronic skin made from produced graphene films are shown in Fig. 1. The flexible electronic skin achieved a positive resistance characteristic in the range of 0–600 kPa, a sensitivity of 10.80/kPa in the range of 0–4 kPa, a loading response time of 10 ms, and a spatial resolution of 5 mm, according to the results of the measurement and analysis experiments designed in this paper. Additionally, using a regular-shaped object and a change in the resistance value of each unit, the electronic skin is an accomplished form of detection. The high sensitivity flexible electronic skin developed in this work has significant potential applications in artificial intelligence, medical diagnosis, and other domains (Fig. 2).

Researchers are drawn to practical pressure sensing devices, especially tactile sensors, because graphene foam (GF) infiltrated with polymer. However, given a particular kind and level of pressure, the interface between the polymer and the graphene plays a crucial function. Tang et al. examined the effects of static and

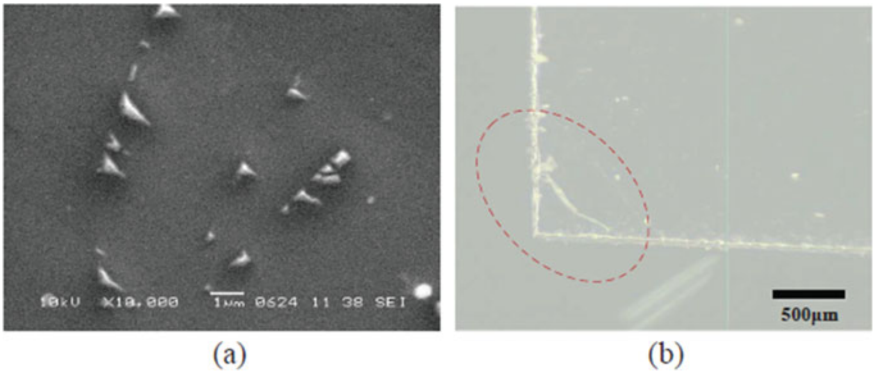


Fig. 1 **a** SEM micrograph; and **b** photograph of the graphene film [34]

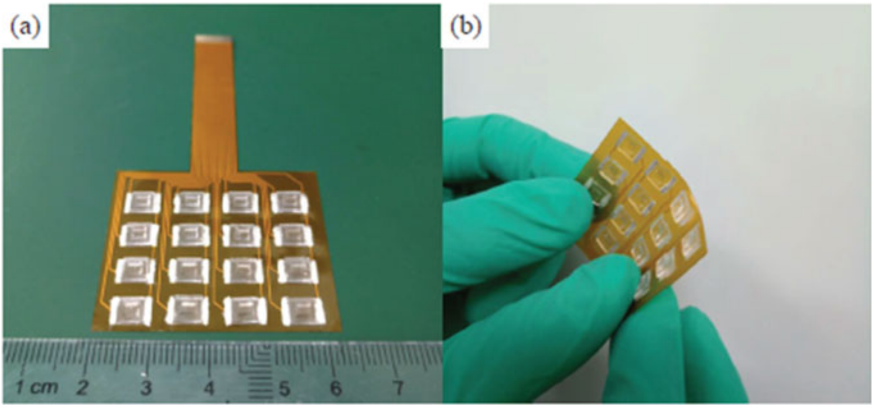
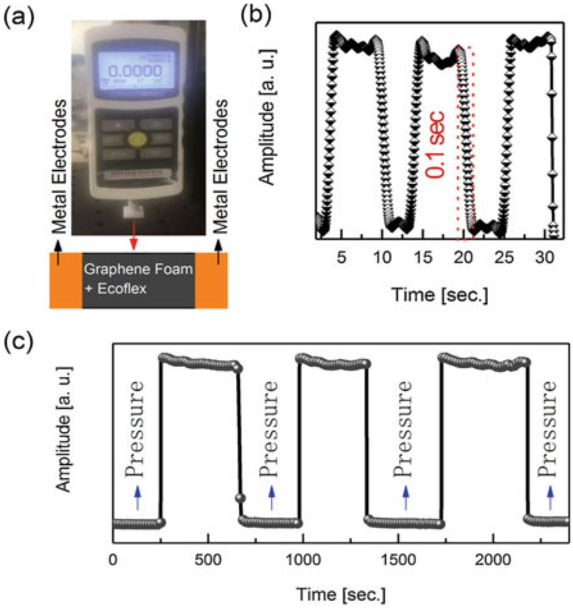


Fig. 2 The size of the electronic skins (**a**); Photograph of the electronic skin (**b**) [34]

dynamic pressure on GF-polymer in this study [35]. During static pressure measurement, a recovery time of $\sim$0.1 s is noted (Fig. 3). GF-polymer samples, in contrast to static pressure, are extremely sensitive to dynamic load, and a short recovery period of 0.06 s is noted. When compared to the static pressure sensor's sensitivity of $\sim 2.1 \pm 0.3\ \mathrm{Pa}^{-1}$, the dynamic pressure sensor's sensitivity of $\sim 1.17 \pm 0.1\ \mathrm{Pa}^{-1}$ is almost two times higher. In terms of technology, pressure is exerted via tactile sensors in a variety of ways, and GF-polymer is a great material to use for creating pressure sensors. The authors advised that dynamic pressure-based sensors, such as microphones for the measurement of sound pressure levels of GF-polymer, are preferable for applications involving the technological sensitivity of the sensors and are therefore more suitable for GF-based pressure sensors.

A prior study [36] presented a revolutionary self-assembly technology based on the Marangoni effect to manufacture large-area ultra-thin graphene films. It was inspired by the fish scale structure of the predecessors, which used the tunneling effect to alter the resistance of graphene sheets. In response to this impact, graphene films are quickly generated on the liquid/gas interface, layered on top of one another via π–π interaction, and the films thus obtained have high transparency (86–94% at 550 nm), a thickness of 2.5–5.0 nm, tunable sheet resistance, and structural homogeneity. Due to the tunneling effect, a suitable dense film exhibits a GF of 1037 at 2% strain, demonstrating extremely high sensitivity. The fabrication of high-sensitivity strain sensors using this versatile and straightforward approach of self-assembling graphene films is ideal for e-skin applications and industrial production. In addition to the technique of electronically inducing the vertical arrangement of graphene sheets and the Marangoni effect, high-sensitivity sensors can also be made by deoxidizing graphene oxide through flame heating in a small area. For instance, Song et al. [37]'s confinement of the graphene oxide film to two quartz plates and swift thermal reduction of the graphene oxide's gas resulted in the film's rapid expansion into a 2D porous structure. By adjusting the sp^2 and sp^3 domains in graphene oxide, it is possible to fabricate a variety of controllable microstructures that can be used

Fig. 3 Static pressure unit and schematic of metal electrodes on the GF-polymer sample (**a**). The change in the resistance of GF-Ecoflex sample with a recovery time ~ 0.1 s (**b**). The stability test was performed under periodic pressure for a long period of time which shows good stability (**c**) [35]



to create flexible pressure sensor arrays, voice recognition technology, and pulse detection technology in addition to high-performing conventional pressure sensors.

In a recent work, Yin et al. demonstrated a high-performance, skin-like pressure sensor and sensing arrays based on graphene/polyamide composite interlocking fabric [38]. They did this using a straightforward but effective, affordable, and large-scale capable approach. By attaching CRG to interlocking fabric, flexible G/IF may be made instantly. The entire fabrication procedure can be scaled up without the use of cumbersome, expensive machinery or traditional wafer-based methods. Due to the special microstructure of the composite fabric, the pressure sensor that is produced has extremely high sensitivity (2.34 kPa^{-1}), an extremely low detection limit (<1.38 Pa), a wide sensing range (1.38 Pa–80 kPa), and outstanding stability for 10,000 loading and unloading cycles (Fig. 4). The pressure sensor may also be used for various deformation detection methods, such as sinusoidal vibration, bending, and stretching. It is possible to detect high-fidelity human physical signals, such as respiration, wrist pulse, and vocal recognition, using the flexible G/IF pressure sensor since it can be conformally contacted onto uneven surfaces while also having superior sensing capability. The G/IF pressure sensor, which has the potential to be used in electronic skin and real-time biomedical monitoring, was made using chemically reduced graphene and interlocking fabric, both of which are commercially available materials.

The substrates used in today's wearable sensors limit their flexibility, comfort, and stretchability and cause interface mismatches. The substrate is also damaging to the health of the skin and stops sweat from evaporating. Electronic skin (e-skin) with multifunction was made possible by Qiao et al. using substrate-free laser scribed

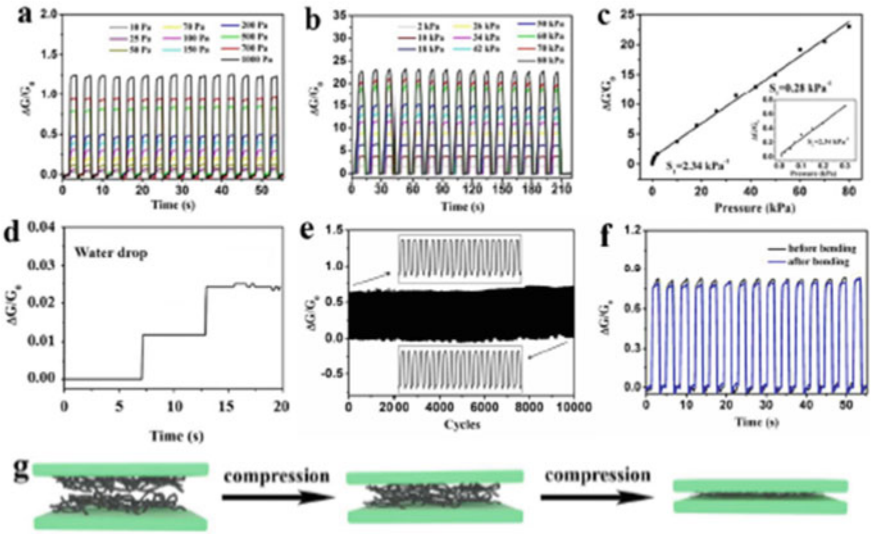


Fig. 4 **a** Relative change in conductance under applied pressures ranging from 10 to 1000 Pa. **b** The change in conductance under applied pressures ranging from 2 to 80 kPa. **c** Relative conductance variation of a G/IF sensor as a function of pressure, showing a sensitivity of 2.34 kPa^{-1} within 0–0.3 kPa and 0.28 kPa^{-1} in higher pressure region. **d** Relative conductance variation of the G/IF pressure sensor upon water droplets. **e** Performance of the pressure sensor at 300 Pa for 10 000 loading–unloading cycles. **f** Pressure sensing performance of the sensor at 500 Pa before and after bending for 500 cycles. **g** Schematic illustration of the pressure sensing mechanism [38]

graphene (SFG) [39]. The SFG offers good gas permeability, low impedance, and flexibility when compared to the e-skin with substrate. Almost any object, even silicon and human skin, may be transferred using only water, and the SFG can even be suspended. The SFG is breathable because it can develop numerous through-holes similar to stomas in leaves. The gauge factor (GF) of graphene electronic skin (GES), which may be customized after the pattern, can be used as a strain sensor. Electrocardiogram (ECG) and breathing are a few examples of physiological signals that can be picked up. Additionally, the hanging SFG has a great sensitivity to vibrations. The substrate-free construction significantly reduces the resistance between SFG e-skin and the human body. Last but not least, a real-time body status monitoring ECG detecting system has been developed based on the GES. A convolutional neural network (CNN) was successfully constructed and trained to evaluate the ECG data automatically. In the area of health telemonitoring, this approach has a lot of potential.

Pencils and paper are widely used for writing and sketching in modern society because they are easy to use, affordable, readily available, and disposable. Their applications in cutting-edge skin-interfaced health monitoring and therapeutics haven't been thoroughly examined yet. A few of the on-skin electronic devices reported by

Xu et al. [40] modified thermoregulators, humidity energy harvesters, sweat biochemical (pH, uric acid, glucose) sensors, and biophysical (temperature, biopotential) sensors. These devices use office copy paper as flexible supporting substrates, while graphite patterns made with a pencil serve as conductive traces and sensing electrodes The modified devices are capable of performing real-time, continuous, and high-fidelity monitoring of a variety of vital biophysical and biochemical signals from human bodies, such as skin temperatures, electrocardiograms, electromyograms, alpha, beta, and theta rhythms, instantaneous heart rates, respiratory rates, and sweat pH, uric acid, and glucose, as well as delivering preprogrammed thermal stimulations. Notably, the recorded signal quality is equivalent to those assessed using traditional techniques. Additionally, to prepare humidity energy harvesters, a gradient distribution of oxygen-containing groups is made between pencil-drawn electrodes on copy paper. From ambient humidity, a single device (0.87 cm^2) can produce a sustained voltage of up to 480 mV for more than two hours. As another example of on-skin chemical intervention, a self-powered on-skin iontophoretic transdermal drug-delivery device is created. Additionally, research is being done on antennas made of pencil paper, two- and three-dimensional (3D) circuits with LEDs and batteries, reconfigurable assemblies, and biodegradable electronics (based on water-soluble papers).

In summary, numerous other great studies that were not emphasized exist in addition to the many study techniques for creating e-skin that were discussed in this paper. The main reason why e-skin hasn't completely taken over peoples' daily lives yet is that it hasn't been prepared well, leaving out several flaws in the ways described. More thorough and in-depth research is required to address these problems that prevent its broad use and make it fully serve people's health and lives.

Research Challenges and Perspectives

It would be possible to employ sensors more effectively in their intended application areas, such as monitoring the quality of drinking water, detecting blood sugar levels in real time, or analyzing urine, by converting them into sensing systems [41]. Additionally, due to the special characteristics of graphene, it is ideally suited as a multipurpose sensor to measure variables with widespread consumer applications, such as strain, pressure, or magnetic field with just one sensor. However, such applications require a complete sensing system. Graphene is a desirable material for biosensing applications such detecting glucose or cholesterol due to its enormous surface area, exceptionally low levels of contamination (chemical purity), and relatively simple functionalization [41]. More research is needed, nevertheless, to guarantee that the biosensor system is cost-competitive, non-toxic, compact, durable, and resilient for use in these applications [42]. Additionally, concerns regarding biocompatibility must be carefully taken into account [43]. To show selectivity or specificity as a biosensor, more research is required. A significant reaction to the target analyte or biomolecule and a negligible or nonexistent response to any interfering species are the ideal characteristics for graphene sensors. This necessitates additional work, including enhanced functionalization layers, and probes that "catch" the targets to be felt. Practically speaking, manufacturing and fabrication processes that can create sensors with consistent and reproducible features are required [44]. This means that novel sensor device designs and operating modes are required in order to minimize sensor variation or produce consistent performance. Additionally, more research into the integration of signal conditioning and processing circuitry to counteract performance changes, enhance sensitivity, selectivity, and operating lifetime would be extremely beneficial for real-world uses or graphene-based sensors.

Conclusions

An overview of recent advancements in graphene e-skins has been provided in the current study, with an emphasis on the efforts of many research scientists to build high-performance electronic skin with stretchable, flexible, pressure-sensitive sensors and multifunctional e-skins. By utilizing the special physical and chemical properties of incorporating graphene-based nanostructured materials into electronic skin, such as high electrical conductivity, high thermal conductivity, high mechanical strength, and lightweight, the reviewed studies successfully constructed electronic skin with good biosensing capabilities. Flexible electronic skin made of graphene has been developed more recently and is now widely employed in the biomedical industry. To ensure that the biosensor system is trustworthy, small, sturdy, and cost-competitive, as well as non-toxic and simple to manufacture, more work must be done.

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