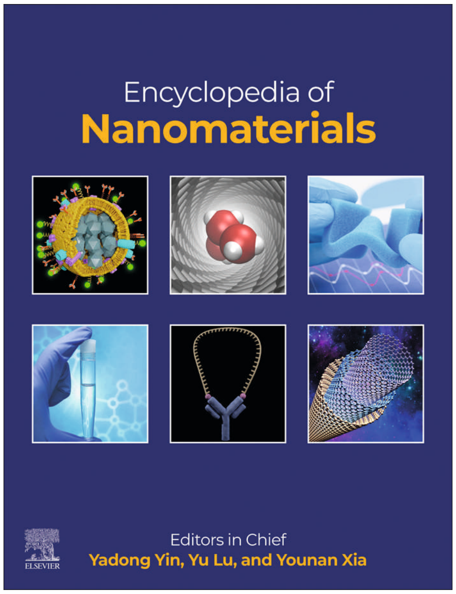

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Nanomaterials for soft wearable electronics

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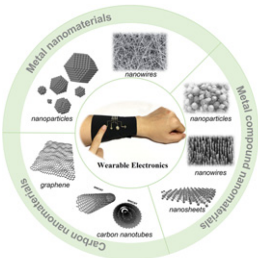
Abstract

Nanomaterial-enabled soft wearable electronics have seen tremendous progress in recent years with a wide range of applications such as monitoring of personal health, human-machine interfaces, and electronic skin for robotics and prosthetics. Nanomaterials, compared with bulk materials, can better accommodate the flexibility and stretchability requirements for soft wearable electronics. Here, several major types of nanomaterials used for soft wearable electronics are reviewed including metal nanomaterials, metal compound nanomaterials, carbon nanomaterials, and hybrid nanomaterials. For each type of nanomaterials, representative soft wearable electronic devices are presented. Finally, the outlook and perspective in this emerging field of soft wearable electronics are summarized.

Graphical Abstract



Key Points

- Mechanical consideration when incorporating nanomaterials into soft wearable devices is discussed.
- Four types of nanomaterials including metal, metal compound, carbon nanomaterials, and their hybrid for soft wearable devices are reviewed.
- The material properties, manufacturing techniques, and representative applications of these nanomaterials in soft wearable devices are presented.
- A brief discussion of fabrication strategies of nanomaterials for soft wearable devices is discussed.

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Introduction

In the past decades, soft wearable electronics have emerged as an alternative to conventional rigid wearables. Flexibility and stretchability in soft wearable devices are critical to achieving high signal-to-noise ratios, low motion artifacts, and comfortable wear (Yao *et al.*, 2020; Rogers *et al.*, 2010; Kim and Rogers, 2008; Liu *et al.*, 2021; Ghaffari *et al.*, 2021; Wang *et al.*, 2021a; Kim *et al.*, 2021). For example, the curvilinear and dynamic surfaces of human skins require wearable devices to accommodate strains caused by repeated movements in daily activities. The epidermis of human skin has Young's modulus of 140–600 kPa while the dermis has Young's modulus of 2–80 kPa (Kim *et al.*, 2011). Human skin can be stretched elastically with linear behavior up to ~15% strain and with nonlinear behavior up to ~30% strain (Kim *et al.*, 2011; Arumugam *et al.*, 1994). The strain introduced by joint motions can reach as high as 100% with the help of the wrinkles and creases (Webb *et al.*, 2013; Yao *et al.*, 2018b; Yao and Zhu, 2014). In addition to flexibility and stretchability, wearable devices should be thin, lightweight, low-cost, and with low power consumption. To achieve these features, materials that are used as the building blocks in soft wearable devices should meet the required electrical and mechanical properties.

Considerable efforts have been devoted to developing flexible and stretchable wearable devices using advanced materials. These devices can be categorized into physical sensors (e.g., temperature, hydration, electrophysiology, strain, pressure, and tactile) (Li *et al.*, 2017; Trung and Lee, 2016; Kenry *et al.*, 2016; Myers *et al.*, 2015; Cui *et al.*, 2014), biochemical sensors (e.g., electrochemical and gas sensors) (Seesaard *et al.*, 2015; Windmiller and Wang, 2013; Bandodkar *et al.*, 2016a), wearable actuators (e.g., soft robotics, heaters, and drug delivery devices) (Amjadi *et al.*, 2018; Tharmatt *et al.*, 2021; Chen *et al.*, 2021b; Zhou *et al.*, 2016; Zhao *et al.*, 2015a), wearable power sources (e.g., batteries, supercapacitors, and energy harvest devices) (Chong *et al.*, 2019; Zamarayeva *et al.*, 2017; Fan *et al.*, 2020; Zhou *et al.*, 2014; Xue *et al.*, 2017; Liu *et al.*, 2019; Wang *et al.*, 2021b), and wearable displays (e.g., wearable organic light emitting diode (OLED)) (Kim *et al.*, 2017; Park *et al.*, 2011; Ig Mo *et al.*, 2008; Lee *et al.*, 2017). Based on their unique properties, nanomaterials have been widely exploited for these devices as different components, including electrodes, dielectrics, semiconductors, and interconnects. Beyond single devices, wearable systems enabled by integrating multiple devices are emerging (Xu *et al.*, 2019; Xie *et al.*, 2019; Zhao *et al.*, 2015b; Yao and Zhu, 2014; Yao *et al.*, 2019). Compared to single devices, such systems can provide more functions, often times in a self-powered and wireless fashion, which is of great significance for healthcare, fitness, human-machine interactions, among other applications (Chen and Pei, 2017; Yeo and Lim, 2016; Trung and Lee, 2016). On the other hand, these systems are more complex, where the combination of different nanomaterials demands more comprehensive and deeper investigation into the properties and compatibility of these nanomaterials for wearable device applications.

In this article, advanced nanomaterials that have emerged in the past decades and their applications in soft wearable electronics are summarized. Metal, metal compound, carbon, and their hybrid nanomaterials will be described in detail. These nanomaterials are categorized according to their dimensions, including zero-dimensional (0D) (nanoparticles (NPs)), one dimensional (1D) (e.g., nanowires (NWs) and nanorods (NRs)), and two dimensional (2D) (e.g., nanosheets and nanoflakes) nanomaterials. This book chapter starts with mechanical consideration when incorporating these nanomaterials into soft wearable devices. Next, we present the application of four types of nanomaterials, metal, metal oxide, carbon nanomaterials, and their hybrid, for wearable devices. For each type of nanomaterials, we discussed their materials properties, related manufacturing techniques, and their applications in soft wearable devices. Then, a brief discussion of fabrication methods for nanomaterial-enabled soft wearable electronics is provided. Last, the outlook and perspective for the application of nanomaterials in soft wearable devices are briefly summarized.

Mechanical Consideration

Conventional materials for soft wearable devices meet a major challenge due to the fact that most bulk metal and semiconducting materials lack flexibility or stretchability (Choi *et al.*, 2016; Yao *et al.*, 2018a; Park *et al.*, 2014). For wearable devices especially on-skin electronics, the situation can be mitigated using two strategies – material and structure. For the material strategy, intensive studies have been recently reported focusing on nanomaterials and nanocomposites for soft wearable devices (Wang *et al.*, 2018a; Jayathilaka *et al.*, 2019). A major method for constructing nanomaterial-enabled wearable devices is depositing functioning nanomaterials into the matrix of structural materials like polymers and fabrics to form nanocomposites. The nanomaterials are typically in the form of a percolation network, where individual nanomaterials can slide, bend, and/or rotate to accommodate mechanical deformation of the nanocomposites. Such intrinsic flexibility and stretchability of the nanocomposites have been discussed in previously published review papers (Yao *et al.*, 2018a; Yao *et al.*, 2020; Yao and Zhu, 2015).

For the structure strategy, researchers have proposed several mechanical designs, such as wavy structures (**Fig. 1**(a and b)), kirigami patterns (**Fig. 1**(c)), serpentine shapes (**Fig. 1**(d)), fractal designs (**Fig. 1**(e and f)), and porous structures including porous films and foams (**Fig. 1**(g)) (Khang *et al.*, 2006; Xu *et al.*, 2012; Li *et al.*, 2019; Fan *et al.*, 2014; Zhou *et al.*, 2020; Pan *et al.*, 2017; Cui *et al.*, 2019). For example, the wavy structure can be generated by two approaches, prestrain-release-buckling (**Fig. 1**(a)) and stretching-release-buckling (**Fig. 1**(b)) (Yao and Zhu, 2015). In the former approach, a stress-free film is transferred onto a prestrained soft substrate, which is subsequently released (Jiang *et al.*, 2007; Xu *et al.*, 2012). In the latter approach, both thin film and soft substrate are stretched and released, with the film either on top of or embedded below the surface of the soft substrate; upon stretching, individual nanomaterials within the film slide against each other (Xu and Zhu, 2012; Zhu and Xu, 2012). The horseshoe-shaped pattern has been utilized in both metal thin films and nanomaterial films to reduce local stresses. Experimental and theoretical studies have been carried out to show the stretchability and electrical stability under stretching (Li *et al.*, 2019; Han *et al.*, 2016). Kirigami patterns have attracted

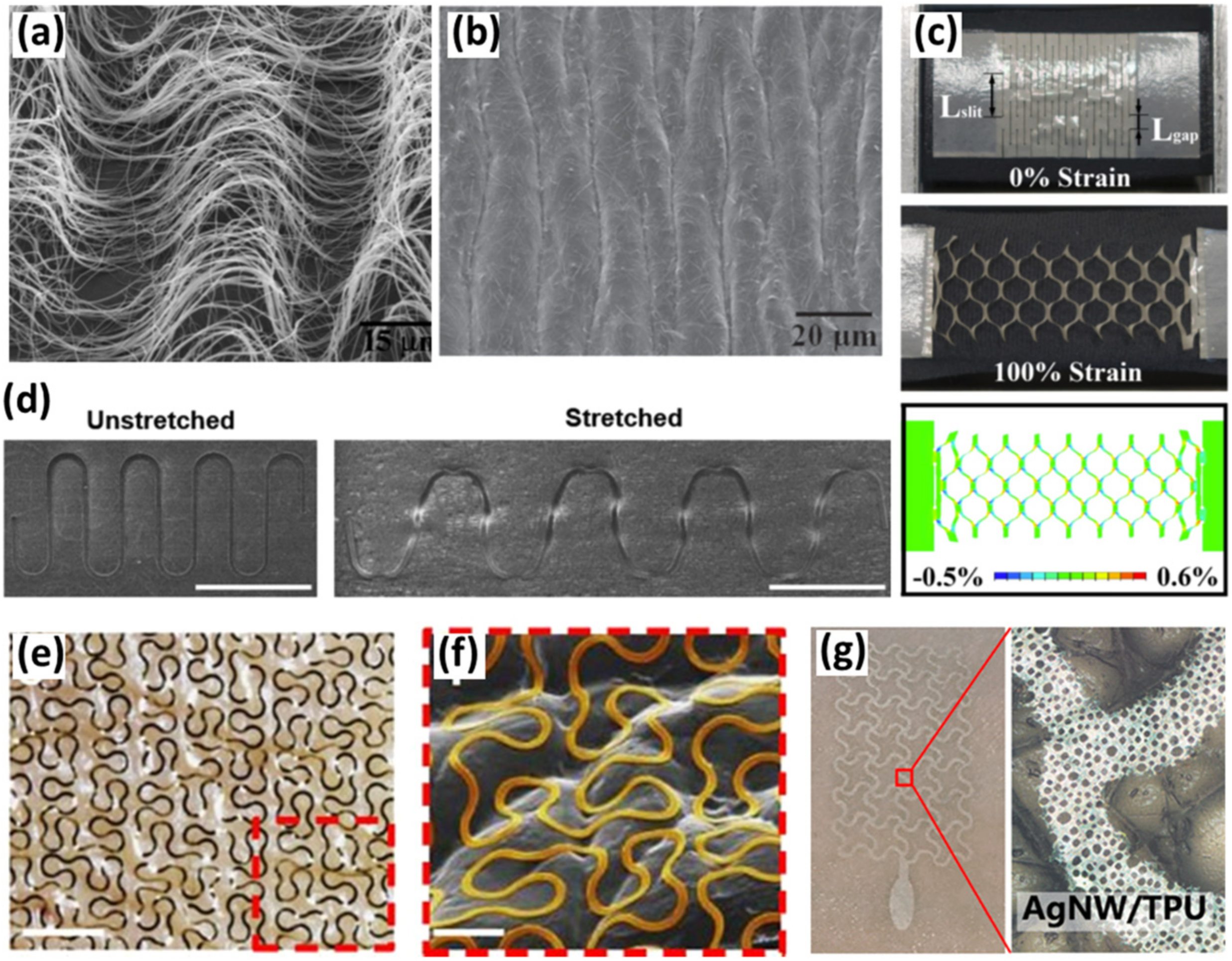

Fig. 1 (a) SEM images of buckled carbon nanotube (CNT) ribbon fabricated by prestrain-release-buckling strategy. (b) SEM image of the silver nanowire/Polydimethylsiloxane (AgNW/PDMS) buckling surface fabricated by the stretch-release-buckling strategy. (c) Optical images of a kirigami structured AgNW/Polyimide (AgNW/PI) temperature sensor at tensile strains of 0% and 100% (top two) and at 100% strain from finite element analysis (bottom). (d) SEM images of the serpentines on an Ecoflex substrate at 0 (left) and 100% (right) strain. The scale bar represents 500 μm. (e) Optical image of a fractal patterned metal film on skin replica. (f) SEM image of the magnified fractal pattern in (e), showing the conformal contact of the pattern on the substrate. Scale bars in (e) and (f) are 2 mm and 500 μm, respectively. (g) Optical images of a porous AgNW/thermoplastic polyurethane (TPU) electrode on an artificial skin. Reproduced from (a) Xu, F., Wang, X., Zhu, Y., Zhu, Y., 2012. Wavy ribbons of carbon nanotubes for stretchable conductors. Advanced Functional Materials 22, 1279-1283. (b) Xu, F., Zhu, Y. 2012. Highly conductive and stretchable silver nanowire conductors. Advanced Materials 24, 5117–5122. (c) Cui, Z., Poblete, F.R., Zhu, Y. 2019. Tailoring the temperature coefficient of resistance of silver nanowire nanocomposites and their application as stretchable temperature sensors. ACS Applied Materials Interfaces 11, 17836–17842. (d) Pan, T., Pharr, M., Ma, Y., *et al.*, 2017. Experimental and theoretical studies of serpentine interconnects on ultrathin elastomers for stretchable electronics. Advanced Functional Materials 27, 1702589. (e–f) Fan, J.A., Yeo, W.H., Su, Y., *et al.*, 2014. Fractal design concepts for stretchable electronics. Nature Communications 5, 3266. (g) Zhou, W., Yao, S., Wang, H., *et al.*, 2020. Gas-permeable, ultrathin, stretchable epidermal electronics with porous electrodes. ACS Nano 14, 5798–5805.

much attention due to the easy processing for both the functional nanomaterial layer and the substrate layer. With the proper geometrical design of the kirigami pattern, the stretchability can be increased by over 100% (Cui *et al.*, 2019). Porous structures generally have better stretchability than the solid counterpart, which has been used as the matrix material for nanomaterials to accommodate large deformation of the wearable devices (Zhou *et al.*, 2020; Qin *et al.*, 2015).

Metal Nanomaterials

Conductors are the most important building block in wearable electronics. The electrodes of resistor- and capacitor-based sensors, the source, drain, and gate electrodes for a field-effect transistor (FET), the Joule-heating-based heaters and related thermal actuators, the electrodes of electrophysiology sensors such as electrocardiography (ECG), electromyography (EMG), and electroencephalography (EEG), and the interconnects typically need flexible, stretchable, and highly conductive materials. Metal is the most commonly used conductive material. Typical metals possess electrical conductivity on the order of $\sim 10^7$ S/m, where silver (Ag), copper (Cu), and gold (Au) are the top three most conductive metals. Cu is mostly used since Cu is cheaper (1% the cost of Ag) and more abundant (1000 times than Ag) (Hu *et al.*, 2011). Ag is used for devices with requirements of higher conductivity and better anti-oxidation. Au, although with even better resistance to oxidation and better biocompatibility than Ag and Cu, is less used in wearable devices due to the high cost. Apart from these highly conductive metals, other metal materials such as cobalt (Co) (Gao *et al.*, 2017; Yang *et al.*, 2018b), nickel (Ni) (Shi *et al.*, 2017), iron (Fe) (Wu *et al.*, 2020a; Zhou *et al.*, 2018b), and zinc (Zn) (Wang and Song, 2006; Khan *et al.*, 2012) are also widely used in wearable devices as sensing materials, catalysts, and electrodes. However, metals are typically not flexible or stretchable, with limited failure strain $< 1\%$, which hinders their applications in wearable devices. Metal thin films fabricated using conventional microfabrication processes, i.e., deposition and patterning using photolithography, can exhibit reasonable flexibility; flexibility increases with decreasing film thickness. However, the stretchability of such thin films is limited, showing weak dependence on the film thickness, which impedes their applications that require high stretchability, conformability, wearability, and durability, such as continuous healthcare monitoring. With the development of advanced fabrication procedures and nanomaterials, metal nanomaterials have been used to fulfill the increasing need for flexible and stretchable wearable devices.

Using nanostructured metal materials allows tuning of the structure, size, and surface status via crystal facet engineering, which makes the nanomaterials exhibit unique electrical and mechanical properties due to size effects. These properties provide engineering possibilities beyond the realm of their bulk counterparts. Compared with the top-down fabrication for metal thin films, metal nanomaterials can be fabricated by both bottom-up and top-down approaches (De Oliveira et al., 2020; Franke et al., 2006; Jamkhande et al., 2019; Ye et al., 2014). Metal nanomaterials can exist in various forms. The most significant forms are 0D structured metal NPs (**Fig. 2**(a and b)) and 1D structured metal NWs and NRs (**Fig. 2**(c and d)). Metal nanomaterials are widely used as conductors in a wearable device, because they offer high electrical conductivity of metals while being compatible with facile and scalable manufacturing processes including solution-based (bottom-up) synthesis and printing. Their physical, chemical, and optoelectronic properties, processing procedures, biocompatibility, and cost should be considered when being used as the components for wearable devices.

NPs are the most widely used nanostructured metals due to their relatively easy synthesis, while NWs/NRs are more effective in forming conductive pathways compared to NPs according to the percolation theory (Lee *et al.*, 2012b; Liang *et al.*, 2014). 2D structured metals such as nanoflakes and nanosheets are also widely used due to their large surface area. These metal nanomaterials have shown superior material properties in terms of mechanical, electrical, optoelectronic properties compared with their bulk counterparts (Wu *et al.*, 2005; Zhu and Li, 2010; Richter *et al.*, 2009; Zhu *et al.*, 2009; Xu *et al.*, 2010; Zhu *et al.*, 2012; Zhu, 2017), making them highly promising to meet the demands of soft wearable electronics. Facile solution-based fabrication processes for wearable devices have emerged in the past decade, which makes scalable manufacturing technologies such as coating and printing on soft substrates with low working temperatures including plastic film and elastomers feasible. In this section, metal nanomaterials with 0D, 1D, and 2D nanostructures used as conducting materials in soft wearable electronics will be discussed.

Metal Nanoparticles

By the top-down approach, NPs can be produced by breaking bulk metals using milling, grinding, etching, and pyrolysis (Teow *et al.*, 2011; Kamyshny and Magdassi, 2014; Huang and Zhu, 2019). By the bottom-up approach, NPs are formed from ionic precursors by reaction with proper reducing agents in solution, protected by a capping layer, using a wet chemistry method. The bottom-up approach offers more precise control of the dimension and surface status of NPs, making it the dominant approach for fabricating NPs. For example, the particle size, dimension uniformity, and dispersion stability of the NPs can be controlled by adjusting the experimental parameters during the synthesis. When using $AgNO_3$ or CH_3COOAg as the Ag precursor, AgNPs with different mean particle sizes of 20 nm and 10 nm were synthesized (Ahn *et al.*, 2009; Magdassi *et al.*, 2010).

Attributed to the surface properties and the easy surface modification of NPs, superior performance can be achieved by NP-based materials with proper surface modification. One of the most significant applications of metal NPs is used as the sensing materials in sensors. As shown in **Fig. 3**(a), a self-healing wearable sensor array to detect volatile organic compounds (VOCs) was fabricated by integrating five types of differently functionalized AuNPs into the array, which exhibited selective detection of different types of VOCs related to human health, enabled by the specific bonding between the right pairs of VOCs and functionalized AuNPs (Jin *et al.*, 2016). To thoroughly evaluate human health, a AuNP-based resistive pressure sensor was also developed based on this array to monitor heart and breathing rates. Li *et al.* (2021) recently developed a plant wearable sensor patch that can sense VOCs emitted by plants, allowing noninvasive and early diagnosis of plant diseases such as late blight caused by Phytophthora infestans. AuNPs modified by different ligands were mixed with reduced graphene oxides (rGO), where AuNPs were used as the sensing materials while rGO was used to enhance the conductivity, as shown in **Fig. 3**(b). With AgNW electrodes and a kirigami-inspired stretchable substrate, the sensor array with 8 sensors modified by different ligands was mounted on live tomato plants (as shown in **Fig. 3**(c and d)) to profile key plant volatiles at low-ppm concentrations with fast response (< 20 s). The multiplexed sensor array allowed accurate detection and classification of 13 individual plant volatiles with $> 97\%$ classification accuracy.

Metal NPs are widely used as electrodes for wearable devices due to their low cost and high conductivity. In these applications, metal NPs are typically required to have stable mechanical and electrical properties under repeated loading-unloading deformation

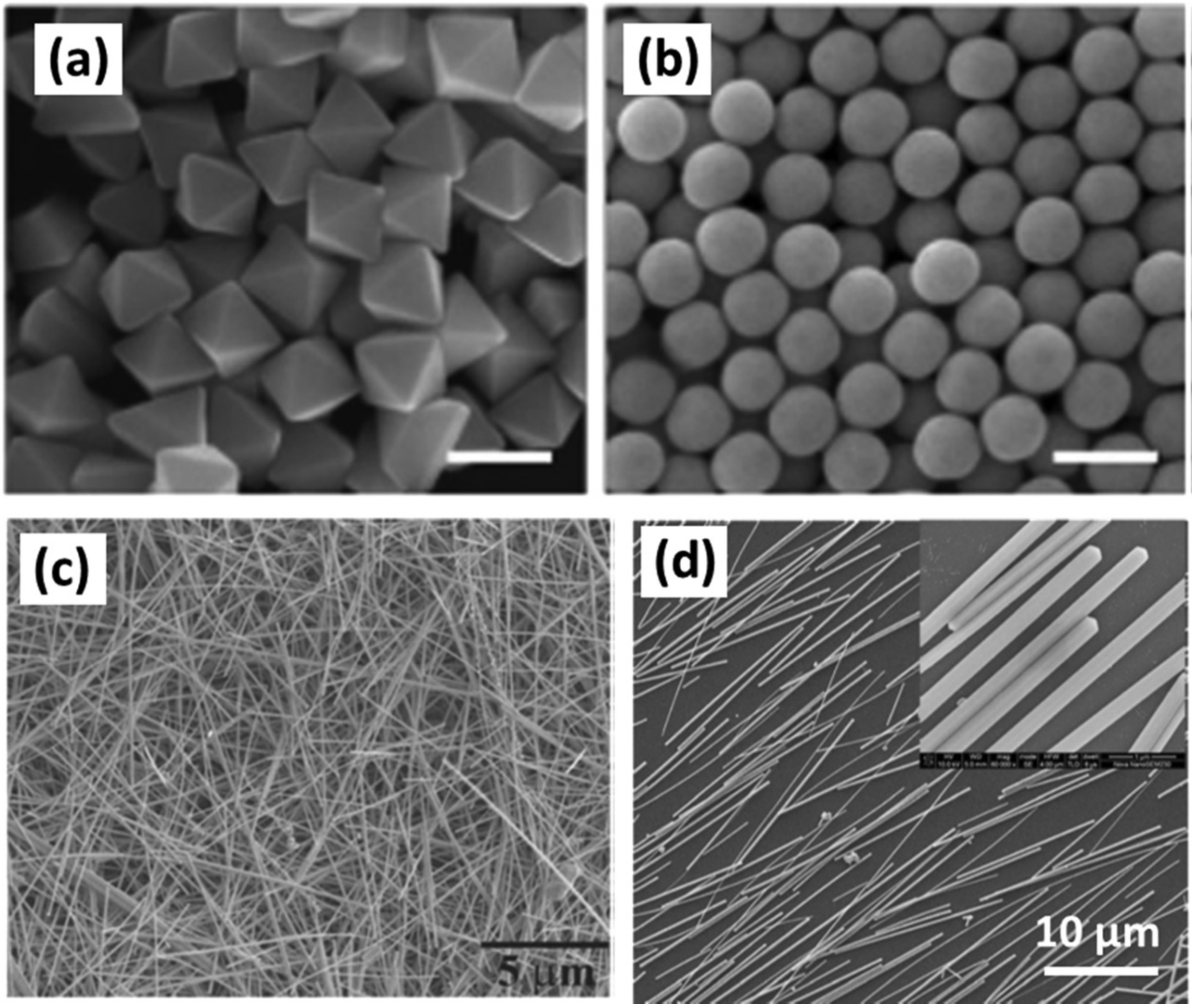

Fig. 2 SEM images of Au (a) octahedra NPs and (b) spherical NPs. (c) SEM image of a random AgNW network (Xu and Zhu, 2012). (d) SEM image of aligned AgNWs (Kang *et al.*, 2018). Reproduced from (a–b) Montes-Garcia, V., Squillaci, M.A., Diez-Castellnou, M., *et al.*, 2021. Chemical sensing with Au and Ag nanoparticles. Chemical Society Reviews 50, 1269–1304. (c) Xu, F., Zhu, Y. 2012. Highly conductive and stretchable silver nanowire conductors. Advanced Materials 24, 5117–5122. (d) Kang, L., Chen, H., Yang, Z.-J., *et al.*, 2018. Seesaw-like polarized transmission behavior of silver nanowire arrays aligned by off-center spin-coating. Journal of Applied Physics 123, 205110.

such as bending, compression, stretching, and twisting. For wearable electrophysiological monitor devices such as ECG (Casson *et al.*, 2017), EMG (Marco, 2015), and EEG (Marinou *et al.*, 2020) sensors, it is desirable to have highly conductive electrodes. The electrodes used in these devices adhere to the surface of human skin and collect the biopotential signals of the human body. Due to the close contact between the electrode and the human skin, the electrodes need to be biocompatible, chemically resistive, and conformal with human skin. Khan *et al.* (2016) developed a wearable sensor system with printed AuNP electrodes for ECG measurement, as shown in **Fig. 3**(b–d). While the power module, data acquisition module, and Bluetooth communication module were rigid and mounted onto a flexible polyimide (PI) substrate, the AuNP electrodes ensured high-quality contact with the skin.

Similar to the electrodes for electrophysiology sensors, the source/drain electrodes for transistors, the electrodes for gas sensors, and the electrodes for electrochemical sensors have the same requirements and metal NPs have been considered as a highly promising material to enable better flexibility, compared to conventional metal films. In addition, NPs usually show higher gauge factors when used in strain sensors compared to NWs or thin film due to the relatively large separation between neighboring NPs under deformation (Segev-Bar and Haick, 2013; Lee *et al.*, 2014; Liu *et al.*, 2017). However, metal NPs possess several drawbacks for wearable devices. First, the manufacturing of metal NP-based devices usually includes a sintering process to enhance the conductivity, which sacrifices the flexibility and is not compatible with some low-temperature components. Second, NPs show low electrical stability during mechanical deformation due to the high percolation threshold needed to maintain the conductive path, hence the device could easily fail under mechanical deformation. Third, metal NPs, like all other metal-based materials have a relatively high density (compared to carbon-based materials), which makes the device heavier, not ideal for wearable applications.

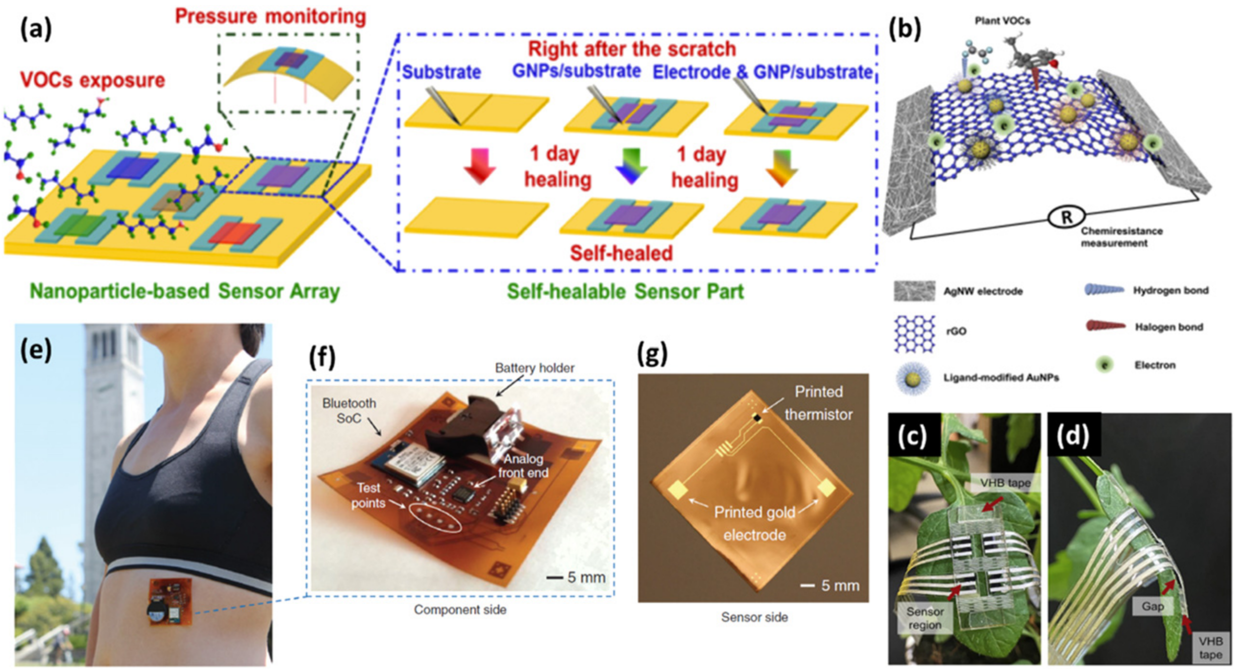

Fig. 3 (a) Schematics of a gas sensor array consisting of 5 self-healable sensors on a polymer substrate with a molecularly functionalized AgNP film as the sensing material. (b) Schematic diagram of the wearable sensor with rGO/AuNP sensing materials and soft AgNW electrodes. (c) Top view and (d) side view of the wearable sensor patch on a tomato leaf. (e) An ECG measurement system mounted on a person's lower left rib cage. (f) The component side and (g) the sensor side of the patch shown in (e). Reproduced from (a) Jin, H., Huynh, T.P., Haick, H., 2016. Self-healable sensors based nanoparticles for detecting physiological markers via skin and breath: Toward disease prevention via wearable devices. Nano Letters 16, 4194–4202. (c–d) Li, Z., Liu, Y., Hossain, O., *et al.*, 2021. Real-time monitoring of plant stresses via chemiresistive profiling of leaf volatiles by a wearable sensor. Matter 4, 2553–2570. (f–g) Khan, Y., Garg, M., Gui, Q., *et al.*, 2016. Flexible hybrid electronics: Direct interfacing of soft and hard electronics for wearable health monitoring. Advanced Functional Materials 26, 8764–8775.

Metal Nanowires

Metal NWs such as CuNWs, AgNWs, and AuNWs have emerged in recent years to address the challenges encountered by NPs, e.g., low mechanical stretchability and low electrical stability when deformed. According to the percolation theory, the electric conductivity σ is given by

$$\sigma \propto \left(N - \frac{5.71}{\overline{L}^2}\right)^t \tag{1}$$

where N is the density of the NW network, $\overline{L}$ is the average length of the NWs, t is the universal conductivity exponent, and the term $\frac{5.71}{\overline{L}^2}$ is the threshold density to keep a conductive network, indicating that the longer the NWs, the lower the density needed for keeping conductivity. Considering that the NWs have a much higher aspect ratio than NPs, the loading density of NWs needed for achieving the same conductivity is much lower. Furthermore, the network structure can maintain the conducting threshold even under large mechanical deformation such as stretching, which makes NWs more desirable for wearable applications where large strain due to body motion is expected.

The most widely used synthesis process for metal NWs involves solution-phase reduction, including the polyol method (Korte *et al.*, 2008; Sun *et al.*, 2003) and the hydrothermal reaction method (Wang *et al.*, 2005; Bari *et al.*, 2016), both of which have facile synthesis and high yield. In a typical solution-phase reduction synthesis, precursors, reduction agents, and capping agents are needed. And similar to the synthesis of NPs, modification of the dimension and surface status can be achieved by varying the reactors' species and synthesis parameters. Typically, longer NWs are conducive to the mechanical and electrical performance of wearable devices, but they are more difficult to be dispersed uniformly. In the mechanical aspect, longer NWs provide higher intrinsic stretchability; in the electrical aspect, longer NWs make it easier to retain the electrical contact between NWs when the percolation network deforms (Lee *et al.*, 2016). Multistep growth and using lower temperature with longer reaction time have been proved to be effective in increasing the length of AgNWs (Lee *et al.*, 2012a; Jiu *et al.*, 2014). To mitigate the agglomeration issue caused by long NWs, the filtering method (Beheshti *et al.*, 2015) and ultrasonication method (Chen *et al.*, 2004) have been reported.

Metal NWs enabled flexible/stretchable wearable devices have attracted extensive attention. The Zhu group developed highly conductive and stretchable AgNW conductors using a simple fabrication method (Xu and Zhu, 2012). AgNWs were embedded below the surface of a PDMS matrix. Upon stretching and releasing, wrinkling occurred in the AgNW layer, which led to a nearly constant resistance upon further stretching. In parallel, the Ko group developed stretchable AgNW conductors using ultralong

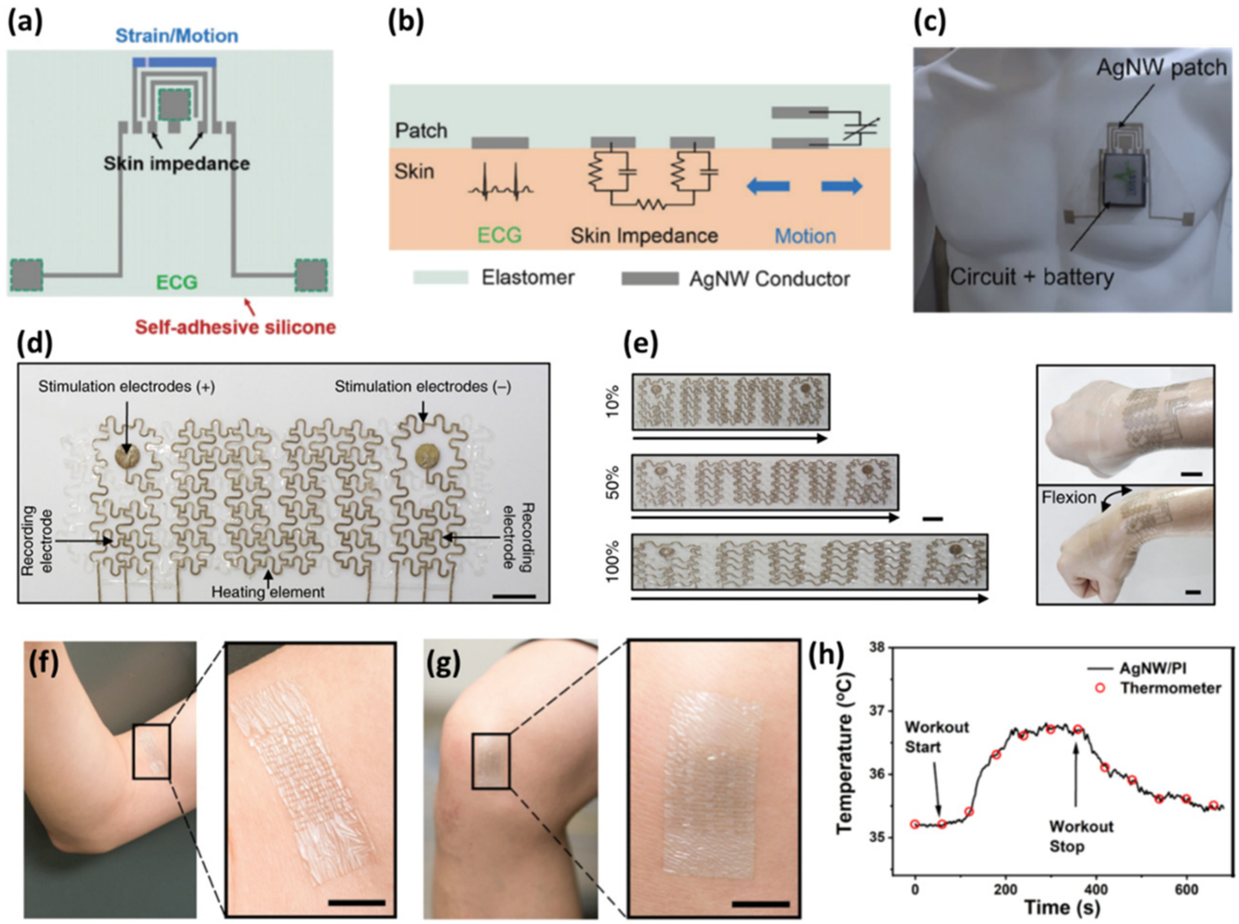

Fig. 4 (a) Layout of a multimodal wearable sensor including three ECG electrodes (boxed), a strain sensor, and an impedance- hydration sensor. (b) Schematic illustration of the sensing concepts of the multimodal sensor. (c) A chest patch with the multimodal sensor and the circuit board and battery enclosed in a 3D printed box, self-adhering to the chest of a mannequin. (d) Optical image of a multifunctional wearable electronic patch consisting of ECG, EMG sensors, and a heater. Scale bar, 1 cm. (e) Optical images of the wearable patch being stretched to 10%, 50%, and 100% (left, scale bar, 1 cm) and adhered on the wrist (right, scale bar, 2 cm). AgNW/PI temperature sensor is attached to the skin near (f) biceps and (g) patella. (h) Temperatures recorded by the wearable temperature sensor and an IR thermometer during biceps workout. Reproduced from (c) Yao, S., Myers, A., Malhotra, A., *et al.*, 2017. A wearable hydration sensor with conformal nanowire electrodes. Advanced Healthcare Materials 6, 1601159. (e) Choi, S., Han, S.I., Jung, D., *et al.*, 2018. Highly conductive, stretchable and biocompatible Ag-Au core-sheath nanowire composite for wearable and implantable bioelectronics. Nature Nanotechnology 13, 1048–1056. (h) Cui, Z., Poblete, F.R., Zhu, Y. 2019. Tailoring the temperature coefficient of resistance of silver nanowire nanocomposites and their application as stretchable temperature sensors. ACS Applied Materials Interfaces 11, 17836–17842.

AgNWs (Lee *et al.*, 2012b). The Zhu group further developed an all-AgNW enabled multifunctional sensor that can be used for strain, pressure, and touch sensing, and multimodal sensor system in the forms of wearable chest patch and wristband (**Fig. 4**(a–c)) (Yao *et al.*, 2017). The sensor system consists of three ECG electrodes, a hydration sensor, and a capacitive strain, all fabricated by AgNWs, to monitor the ECG signals, skin hydration, and body motions, respectively. The Kim group fabricated a multimodal wearable implantable healthcare system using Ag-Au core-sheath NWs as the conductive electrodes for ECG and EMG sensors (**Fig. 4**(d and e)) (Choi *et al.*, 2018). The Ag-Au core-sheath NWs were dispersed in a poly(styrene-butadiene-styrene) (SBS) matrix to provide stretchability as high as 840%. The high conductivity of AgNWs combined with inert Au shell improves biocompatibility and oxidation resistance of the fabricated electrodes.

Metals typically possess large positive temperature coefficients of resistance (TCR), which make them excellent candidates as temperature sensors; a temperature sensor based on this mechanism is categorized as a resistance temperature detector (RTD). The resistance of the sensor should show a linear response to the temperature change, with a constant TCR defined by

$$TCR = \frac{1}{R(T_0)} \frac{R(T) - R(T_0)}{T - T_0} \tag{2}$$

where $R(T_0)$ is the resistance at T_0 (usually room temperature) and $R(T)$ is the resistance at T. A larger TCR represents a higher sensitivity of the temperature sensor. A wearable temperature sensor is of great importance in real-time monitoring of human body temperature, enabling remote diagnosis, therapy, and treatment of patients. NW-based temperature sensors have shown promise in these scenarios. For example, Cui *et al.* (2019) reported a breathable, uniaxially stretchable, and strain insensitive temperature sensor based on a composite material consisting of a AgNW network and a PI matrix, with the TCR of 3.32×10^{-3}/°C. To achieve large stretchability for wearable applications and true temperature sensing that is insensitive to strain, a kirigami-inspired structure was adopted, as shown in **Fig. 4**(f and g). As a result, the temperature sensor can be used under a large strain (up to 100%) without interference from strain. The temperature sensor was used to monitor the skin temperature at the biceps and knees when workout, as shown in **Fig. 4**(h), which was validated with a commercial IR thermometer. Recently the same group extended to develop a biaxially stretchable temperature sensor that can be mounted on joints such as knees for thermal therapy of arthritis (Wu *et al.*, 2021). Taking advantage of the self-sensing capability of the heater, feedback control was realized to stabilize the heater performance under varying environments.

Metal Compound Nanomaterials

Metal compound nanomaterials including metal oxides/nitrides/sulfides/carbides have attracted considerable interest as the functional components in soft wearable devices due to their low cost, large specific area, ease of manufacturing, and tunable bandgap (Comini, 2016; Carlos *et al.*, 2020; Kim *et al.*, 2016; Sim *et al.*, 2019; Yoon *et al.*, 2021; Wang *et al.*, 2020a). They are widely used in many applications including gas sensors, temperature sensors, motion detection sensors, electrochemical energy storage devices, and displays. There are two integration approaches that can be used to combine nanomaterials with proper substrates for device fabrication. One is directly growing nanostructures on a substrate by including the substrate during the synthesis procedure. When fabricating metal compounds nanomaterials for flexible and stretchable devices, this approach is typically not applicable due to the high temperature of conventional synthesis procedures that may damage the soft substrates. Recently, low-temperature manufacturing processes such as thermal solvent methods and electrodeposition has made it feasible to synthesize metal compound nanomaterials directly on flexible and stretchable substrates (Zhou *et al.*, 2018b). The other is to print/coat metal compound nanomaterials on soft substrates or mix them with the soft matrices (Cai *et al.*, 2018). For example, the synthesized metal compound nanomaterials such as NPs and NWs can be drop-casted on flexible substrates (He *et al.*, 2021; Comini, 2013; Zheng *et al.*, 2015; Kumar *et al.*, 2020). Various types of printing methods have been reported to print metal compound nanomaterials on substrates used for wearable devices (Cai *et al.*, 2018; Sundriyal and Bhattacharya, 2020; Carlos *et al.*, 2020; Kim *et al.*, 2016). In this part, soft wearable devices that contain metal compound nanomaterials will be discussed. The commonly used nanostructures of these materials include NPs, NWs, and nanosheets.

Metal Compound Nanoparticles

For metal compound nanomaterials, NPs have become the most common form for applications. Typical synthesis methods of metal compound NPs include wet chemical methods and sintering-based methods (Das *et al.*, 2012; Nikam *et al.*, 2018; Oskam, 2006). The synthesized NPs can be in form of dispersion solution or powder for further use. Unlike metal NPs, metal compound NPs usually do not need an extra annealing process to enhance the conductivity after coating or patterning, since metal compounds are mainly used as functional materials rather than conducing materials; hence, they are compatible with most flexible and stretchable substrates and device fabrication processes.

Metal compound NPs have been used in wearable optical devices. The Someya group developed a self-powered wearable system consisting of an ultra-flexible organic photovoltaic (OPV) power source and an organic electrochemical transistor (OECT) based ECG sensor (**Fig. 5**(a and b)) (Park *et al.*, 2018). ZnO NPs, used as the electron-transporting layer for OPV, were spin-coated and patterned into 1D nanograting structures using a PDMS mold. The thickness of the whole device was only a few micrometers, allowing for intimate contact with human skin or biological tissues. The OPV showed a high power-conversion efficiency of 10.49% and reliable performance under cyclic compression, with weak dependence on the angle of light illumination. By integrating the OPV with the OECT based ECG sensor, where the potential difference between a gel electrode on the chest and the channel of the OECT on the fingertip served as the input gate bias, self-powered ECG sensing was achieved.

Metal compound NPs typically have low conductivity. Although super high conductivity is not required, higher conductivity is still preferred to enhance the electron transfer in optical devices, sensors, and so on. The composition of NPs and other functional materials with higher conductivity is a general strategy to broaden the applications of metal compound NPs Chen *et al.* (2021a). fabricated a fully integrated wireless system with a wearable NO_2 gas sensor based on ZnS NPs/nitrogen-doped reduced graphene oxide (ZnS NP/N-rGO) flake to enable real-time NO_2 monitoring, as shown in **Fig. 5**(c). The device exhibited a wide response range (2.2–10 ppm), fast recovery (724 s), ultra-low theoretical limit of detection (69 ppb), and low power consumption (0.52 μW). The excellent mechanical stability of the device was proved by over 1000 bending tests.

Pathak *et al.* (2020) used OH- functionalized multi-walled carbon nanotubes (MWCNTs) (OH-MWCNT) and ZnO NPs composite to fabricate flexible, wearable, visible-blind UV sensors, as shown in **Fig. 5**(d and e). Both the electrode and the sensing material were screen printed on a flexible poly(ethylene terephthalate) (PET) substrate. Pure ZnO-based UV sensors showed low sensitivity. The use of the nanocomposite sensing material enhanced UV sensing performance in terms of sensitivity as well as mechanical stability. Javed *et al.* (2016) applied a facile hydrothermal method to synthesize MnS NPs on a conductive, flexible carbon textile (CT) substrate which enables the wearability of the device. A stable MnS-CT composite was formed due to the strong

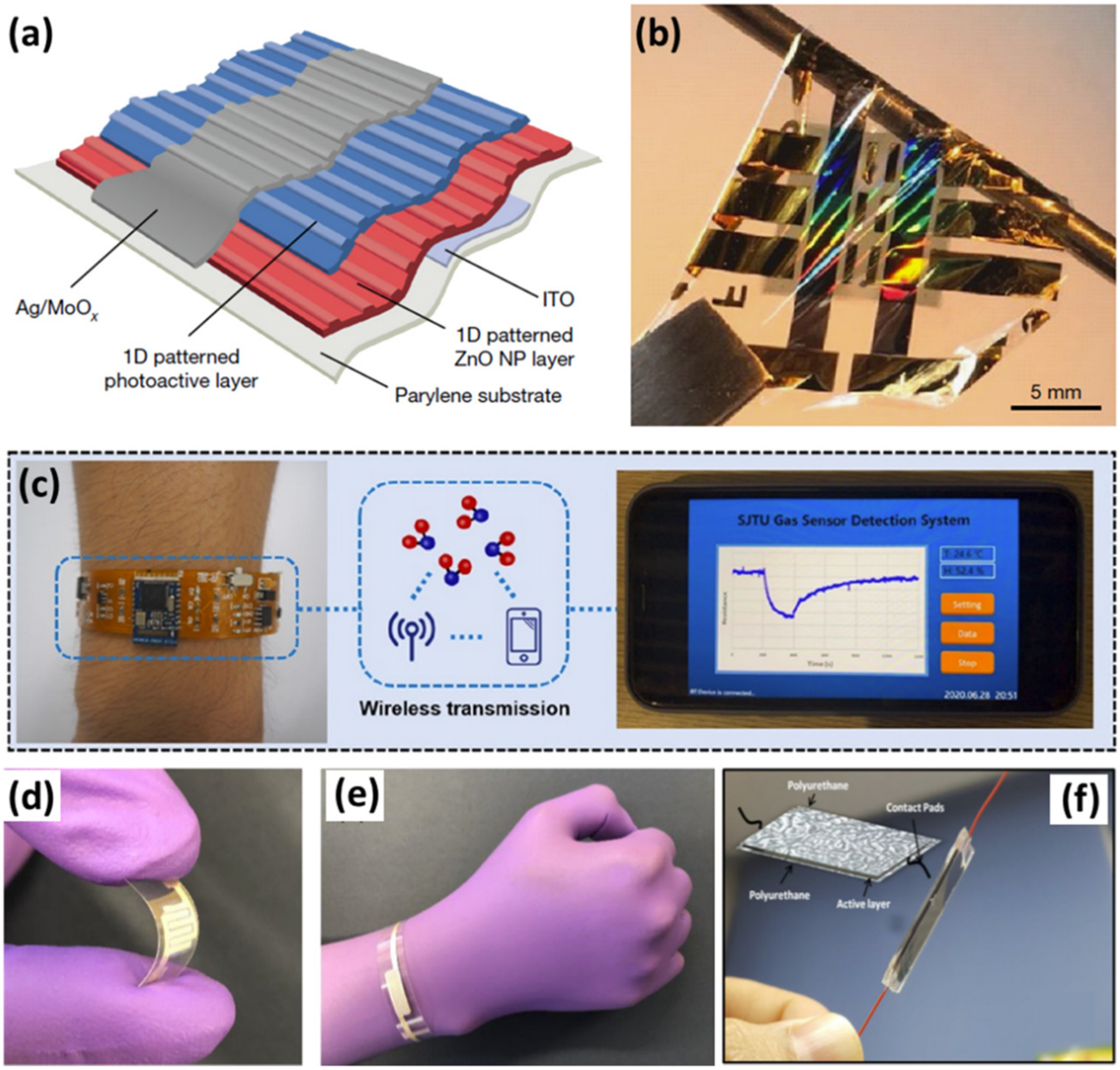

Fig. 5 (a) Structure of the OPV device. (b) Photograph of the OPV device wrapped over a spatula rod and pulled by tweezers. (c) Optical image of the watch-type gas sensor enabled by ZnS NPs/nitrogen-doped reduced graphene oxide (ZnS NP/N-rGO) with a flexible PCB. Continuous detection of the target gas response curve is displayed on a smartphone. (d,e) Optical images of the ZnO NP enabled UV sensor (Pathak *et al.*, 2020). (f) The fabricated strain sensor based on graphene and magnetic iron oxide nano-composite. Reproduced from (b) Park, S., Heo, S.W., Lee, W., *et al.*, 2018. Self-powered ultra-flexible electronics via nano-grating-patterned organic photovoltaics. Nature 561, 516–521. (c) Chen, X., Wang, T., Han, Y., *et al.*, 2021a. Wearable NO_2 sensing and wireless application based on ZnS nanoparticles/nitrogen-doped reduced graphene oxide. Sensors and Actuators B: Chemical 345, 130423. (d–e) Pathak, P., Park, S., Cho, H.J., 2020. A carbon nanotube-metal oxide hybrid material for visible-blind flexible UV-sensor. Micromachines 11, 368. (f) Hassan, G., Khan, M.U., Bae, J., Shuja, A. 2020. Inkjet printed self-healable strain sensor based on graphene and magnetic iron oxide nano-composite on engineered polyurethane substrate. Scientific Reports 10, 18234.

bond between MnS and the CT substrate. The MnS-CT electrodes were used to fabricate all-solid-state supercapacitors, which exhibited outstanding electrochemical performance. A high energy density of 52.03 Wh kg^{-1} at a power density of 307.5 Wh kg^{-1} was achieved.

By adding a small ratio of functional metal compound NPs, the original materials can be modified to own specific functions that may be beneficial for wearable devices. For example, Hassan *et al.* (2020) fabricated a resistive strain sensor based on graphene and magnetic iron oxide NPs on a polyurethane (PU) substrate, as shown in Fig. 5(f). Attributed to the magnetic force response of the iron oxide NPs, the graphene flakes could reconnect due to the magnetic force after the sensor was cut. Along with the engineered self-healable PU substrate, the strain sensor with this nanocomposite active layer could recover most of its stretchability and strain sensitivity without further treatment.

Metal Compound Nanowires

Attributed to the large specific area and capability of redox reactions, metal compound NWs, especially metal oxide NWs, are excellent candidate materials for wearable chemical sensors (Wang *et al.*, 2020b). For integrating the metal compound NWs or NRs with flexible and stretchable substrates, direct growth of NWs on the substrates is the most reliable approach that can result in high mechanical strength at the interface. Ahn *et al.* (2010) reported the integration of ZnO NRs by thermolysis-assisted chemical solution on a PI substrate, as shown in **Fig. 6**(a). The fabricated device was used as a flexible ethanol sensor, which exhibited excellent sensing performances with a sensitivity of 3.11–100 ppm ethanol and a response/recovery time of 3–5 min at an operating temperature of 300°C, comparable to the performance of ZnO NR sensors fabricated on a hard SiO_2 substrate. A caveat is that the high recovery temperature would impede the application of this sensor in a wearable system.

Metal oxide NWs are promising inorganic electrochromic materials that can be used for wearable smart windows. Wang *et al.* (2017) fabricated aligned $W_{18}O_{49}$ NW/Ag NW thin films on a PET substrate using a modified Langmuir-Blodgett technique. The schematic illustration of the flexible electrochromic film is shown in **Fig. 6**(b). The fabricated flexible transparent electrode had tunable conductivity (7–40 Ω/sq) and transmittance (58%–86% at 550 nm) for electrochromic devices as potential wearable displays. By adjusting the layers of loaded $W_{18}O_{49}$, the device with patterned $W_{18}O_{49}$ NWs can show different transmittance under a constant voltage, as shown in **Fig. 6**(c and d). When integrated with $LiClO_4$ based solid electrolyte, an electrochromic eyeglass was assembled, as shown in **Fig. 6**(e–h). Furthermore, a large area electrochromic device with a dimension of 18 cm × 15 cm can also be fabricated.

Metal Compound Nanosheets

After the discovery of graphene, 2D nanomaterials have received extensive attention and enabled a broad spectrum of applications. 2D nanomaterials have many extraordinary physicochemical properties that are different from those of their bulk counterparts, attributed to their unique structural characteristics. For example, ultrathin nanosheet structures enable quantum size effects, leading to the increased bandgap of TiO_2 nanosheets (Sakai *et al.*, 2004). Typical metal compound 2D materials include transition metal oxides (TMOs: ZnO, WO_3, MnO_2, TiO_2, Co_3O_4, etc.), transition metal carbides (MXenes: $Ti_3C_2T_x$ (T: surface functional groups), Ti_2CT_x, etc.), layered metal hydroxides (LMHs: Ni-LMH, NiFe-LMH, MgFe-LMH, etc.), transition metal chalcogenides (TMCs: MoS_2, ReS_2, WSe_2, $MoTe_2$, etc.), and perovskites ($BaTiO_3$, $CH_3NH_3PbI_3$, etc.).

One of the most significant applications of these 2D metal compound nanomaterials in wearable systems is the energy storage devices (Xue *et al.*, 2017; Delbari *et al.*, 2021; Liu *et al.*, 2018). TMO nanomaterials are the most widely used ones as electrodes in electrochemical energy storage devices due to their rich redox capabilities and versatile composition. The well-known Fe_2O_3, MnO_2,

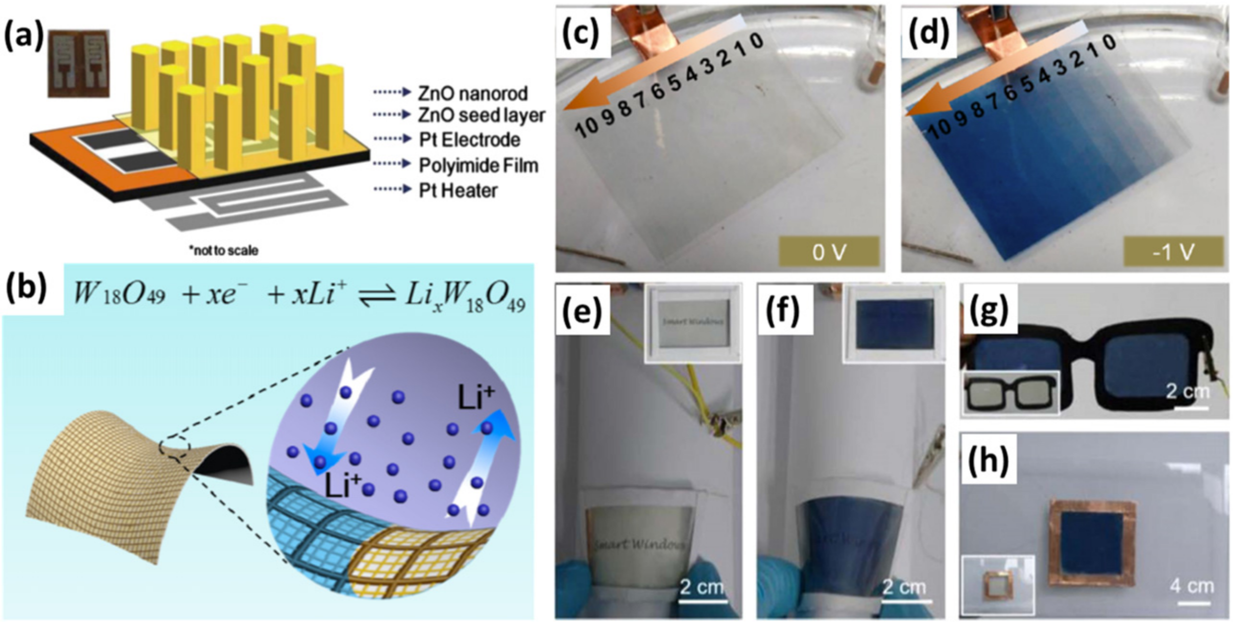

Fig. 6 (a) Schematic showing the integration of ZnO NRs onto a flexible substrate as a sensor. (b) Schematic illustration of the $W_{18}O_{49}$ NW/Ag NW enabled flexible electrochromic film. (c,d) Optical images of the $W_{18}O_{49}$ NW/Ag NW thin films electrochromic device before and after applying an external voltage. (e,f) Photographs of the bleached and colored states of solid electrochromic devices under bending. The insets show the devices without bending. Photographs of (g) the electrochromic glasses model and (h) the electrochromic window model. Reproduced from (a) Ahn, H., Park, J.H., Kim, S.B., *et al.*, 2010. Vertically aligned ZnO nanorod sensor on flexible substrate for ethanol gas monitoring. Electrochemical and Solid State Letters 13, J125-J128. (g) Wang, J.L., Lu, Y.R., Li, H.H., Liu, J.W., Yu, S.H., 2017. Large area co-assembly of nanowires for flexible transparent smart windows. Journal of the American Chemical Society 139, 9921–9926..

ZnO, NiCo LDH, CoAl LDH, Co_3O_4, and their composites have been widely studied. Some typical metal oxides used as wearable energy devices are shown in **Fig. 7**(a) (Yoon *et al.*, 2021). These nanomaterials in a 2D sheet morphology are attracting more and more attention due to their excellent mechanical, electrical, and electrochemical properties. The atomic-scale thickness endows flexibility and durability; ultra-large specific surface area and excessive edges with exposed atoms are beneficial to charge storage and transfer, and catalytic processes. What's more, a vast collection of 2D material with various chemical compositions and adjustable electrical and electrochemical properties are available to form composites for tailoring the device performance (Zhou *et al.*, 2021).

Lithium-ion batteries are a prominent type of power source for wearable electronics. Commercial Lithium-ion batteries typically use TMO (e.g., $LiCoO_2$ (LCO), $LiMn_2O_4$(LMO), $LiFePO_4$ (LFP), and $LiNi_xMnyCo_{1-x-y}O_2$ (LNMCO)) electrode materials due to their high electrical conductivity, high mechanical strength, low cost, and high Li^+ diffusion rate (Zhou *et al.*, 2014). Fabricating these well-known materials into 2D form can further enhance the performance. Li *et al.* (2012) developed a flexible lithium-ion battery by using ultralight LFP nanosheets-loaded graphene foam anode (LFP@GF) and $Li_4Ti_5O_{12}$ (LTO) loaded graphene foam cathode (LTO@GF), as shown in **Fig. 7**(b and c). The assembled full battery exhibited ultrathin thickness (<800 mm), high capacity (~133 mA h g^{-1} at 1 C), high rate (~117 mA h g-1 at 10 C, recoverability of ~88%), and excellent cyclic performance (10 C, 100 cycles, retention of ~96%) with excellent flexibility. The high performances make the large-scale application of this flexible battery promising for powering wearable electronics.

Apart from lithium-ion batteries, flexible and stretchable supercapacitors are another option that can power wearable devices due to their low cost, long cycle life, fast charging/discharging, and high efficiency. A fiber-shaped flexible supercapacitor has been developed by Hou *et al.* (2015). 1D fiber-based devices have important advantages such as structural flexibility and feasibility for knitting, making it relatively easy to integrate into wearable systems, especially e-textile systems. The reported fibers were based on 2D nanosheets of titania ($Ti_{0.87}O_2{}^{0.52-}$), as shown in **Fig. 7**(D–f); wet-spinning was used to spin the nanosheet colloidal suspension into fibers. As a result, the unique stacking order endowed the material with an unusual quasi-crystalline feature. The highly ordered stacking structure and strong electrostatic interactions resulted in enhanced mechanical properties, which mitigated the problem of low mechanical strength of metal oxide materials. The optimal fiber exhibited comparable or even better mechanical performance than the graphene analog.

Using solution-based processes is the most viable route to expand the application of 2D materials from the lab scale to the industrial scale. Typical bottom-up strategies to synthesize 2D nanosheets is wet chemical reaction such as materials electrochemical deposition. These methods can achieve anisotropic growth of sheets with an atomic thickness (Xiong *et al.*, 2018). The top-down strategies typically delaminate or exfoliate 2D nanosheets from bulk crystals of layered materials (Li *et al.*, 2011). Bottom-up strategies can directly grow nanosheets on substrates; both bottom-up and top-down strategies can produce nanosheets

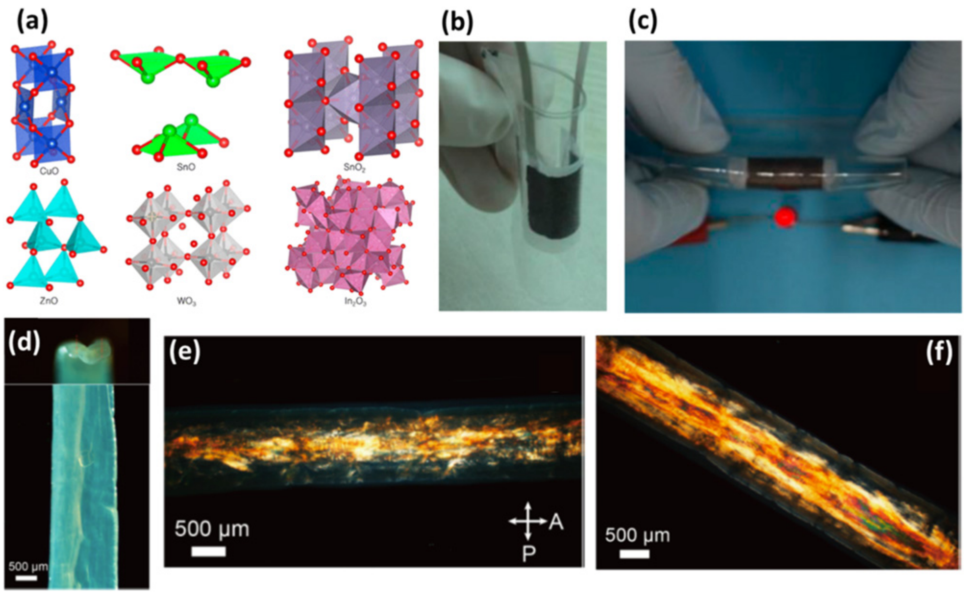

Fig. 7 (a) Typical crystal structures of metal oxide nanomaterials. (b) Photograph of a bent battery, showing its good flexibility. (c) Lighting a red LED device under bending using the flexible lithium-ion battery. (d) Dark field images of the fiber-shaped flexible supercapacitor with a width of ~1300 μm and thickness of ~200 μm. Microscopic image of the ribbon fiber between crossed polarizers at (e) 0° and (f) 45°angle with the polarizer. The directions of the polarizer and the analyzer are indicated by the double arrows, P and A, respectively. Reproduced from (a) Yoon, Y., Truong, P.L., Lee, D., Ko, S.H., 2021. Metal-oxide nanomaterials synthesis and applications in flexible and wearable sensors. ACS Nanoscience Au. (c) Li, N., Chen, Z., Ren, W., Li, F., Cheng, H.M., 2012. Flexible graphene-based lithium ion batteries with ultrafast charge and discharge rates. Proceedings of the National Academy of Sciences of the United States of America 109, 17360–17365. (e–f) Hou, J., Zheng, Y., Su, Y., *et al.*, 2015. Macroscopic and strong ribbons of functionality-rich metal oxides from highly ordered assembly of unilamellar sheets. Journal of the American Chemical Society 137, 13200–13208.

dispersed in solution for coating/printing to fabricate devices. With proper ink formulation using these solutions, the coating/printing process can be compatible with the industrial roll-to-roll manufacturing process.

Carbon Nanomaterials

Carbon nanomaterials have emerged in recent years for wearable devices including carbon nanotubes (CNTs), carbon nanofibers (CNFs), and graphene-based materials (e.g., graphene, graphene oxide (GO), and reduced graphene oxide (rGO)). They possess outstanding merits in electrical conductivity, mechanical robustness, chemical stability, thermal properties, and easy modification, making them promising candidates for the next-generation wearable electronics. In this section, representative carbon materials including CNTs and graphene and their applications in wearable electronics are summarized.

Carbon Nanotubes

CNTs are promising for highly flexible and stretchable devices because of their high electrical conductivity, extremely large aspect ratio, and outstanding flexibility attributed to their unique sp2-bonded honeycomb lattice and 1D structure. CNTs include two types, single-wall carbon nanotubes (SWCNTs) with diameters on the order of a nanometer and MWCNTs consisting of nested SWCNTs weakly bound together by van der Waals interactions (**Fig. 8**(a)) (Reilly, 2007). The typical forms of CNT used as electrodes in wearable sensors include CNT films (Wu *et al.*, 2016; Kim *et al.*, 2006), CNT fibers (Wang *et al.*, 2018b; Senokos *et al.*, 2017), CNT sheets (Xu *et al.*, 2012; Zhu and Xu, 2012; Zhang *et al.*, 2010), vertically aligned CNT array (Zhang *et al.*, 2002; Zhang *et al.*, 2017), and CNTs mixed in the elastomer matrix (Kim *et al.*, 2018; Wang *et al.*, 2018c). Chemical vapor deposition (CVD) is usually used to synthesize CNTs. The produced CNTs can be further processed into CNT films, sheets and fibers by dry or wet approaches.(Lu *et al.*, 2012) Vertically aligned CNT arrays can be directly grown on desired substrates (Lu *et al.*, 2020; Lepró *et al.*, 2010).

CNTs are a promising candidate for strain-insensitive electrodes for wearable devices. Embedding CNTs in a polymer matrix has been developed to fabricate stretchable conductors (Zhang *et al.*, 2010). The Zhu group further developed stretchable conductors based on buckled CNT ribbons (Xu *et al.*, 2012; Zhu and Xu, 2012). The wavy ribbons were fabricated by the prestrain-release-buckling and stretching-release-buckling strategies using transferred CNT ribbons on a PDMS substrate. A representative SEM

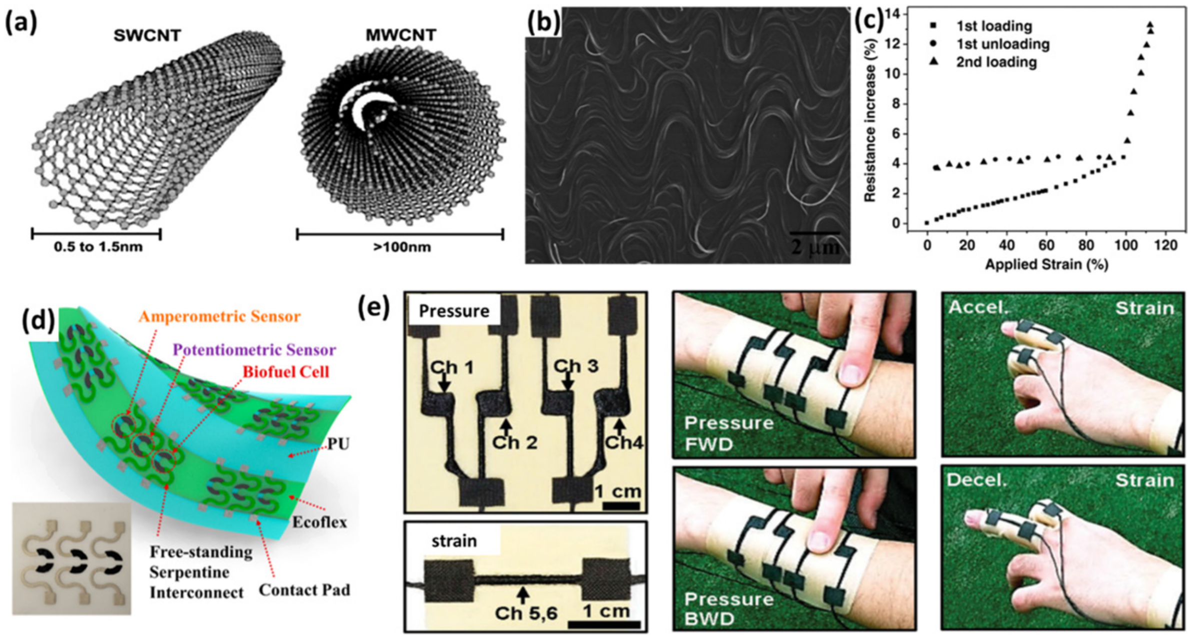

Fig. 8 (a) Conceptual diagram SWCNT and MWCNT. (b) SEM image of the lateral buckling of CNTs on PDMS fabricated by the stretching-release-buckling strategy. (c) Resistance change of a CNT/PDMS film as a function of tensile strain. (d) Schematic for large-scale printed stretchable device arrays along with their various applications (inset shows an image of a printed CNT-based array device). (e) Images of the pressure sensor and strain sensor printed on the commercial elastomeric patch (left), the pressure sensor array (middle), and strain gauge (right). Reproduced from (a) Reilly, R. M. 2007. Carbon nanotubes: Potential benefits and risks of nanotechnology in nuclear medicine. Journal of Nuclear Medicine 48, 1039–1042. (b) Zhu, Y., Xu, F. 2012. Buckling of aligned carbon nanotubes as stretchable conductors: A new manufacturing strategy. Advanced Materials 24, 1073–1077. (c) Xu, F., Wang, X., Zhu, Y., Zhu, Y., 2012. Wavy ribbons of carbon nanotubes for stretchable conductors. Advanced Functional Materials 22, 1279–1283. (d) Bandodkar, A.J., Jeerapan, I., You, J.M., Nunez-Flores, R., Wang, J. 2016b. Highly stretchable fully-printed CNT-based electrochemical sensors and biofuel cells: Combining intrinsic and design-induced stretchability. Nano Letters 16, 721–727. (e) Jung, S., Kim, J.H., Kim, J., *et al.*, 2014. Reverse-micelle-induced porous pressure-sensitive rubber for wearable human-machine interfaces. Advanced Materials 26, 4825–4830.

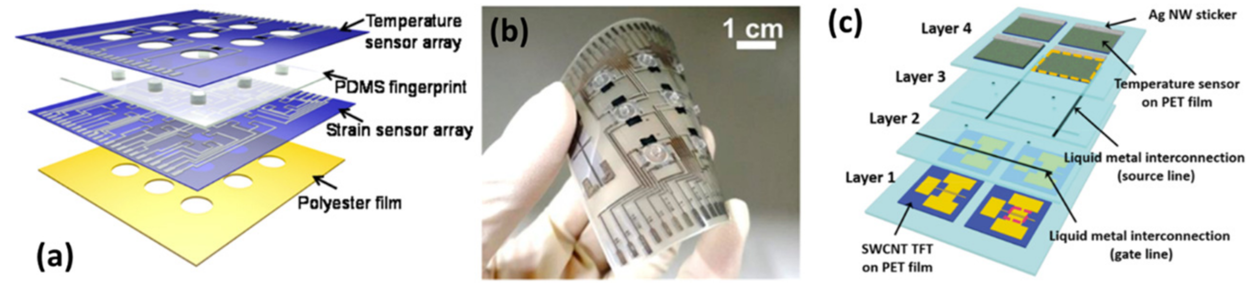

Fig. 9 (a) Schematic for each layer of the temperature sensor array. (b) Picture of a 3 × 3 temperature sensor array. (c) Schematic for assembly of thin-film transistors integrated with stretchable temperature sensors. Reproduced from (b) Harada, S., Kanao, K., Yamamoto, Y., *et al.*, 2014b. Fully printed flexible fingerprint-like three-axis tactile and slip force and temperature sensors for artificial skin. ACS Nano 8, 12851–12857. (c) Hong, S.Y., Lee, Y.H., Park, H., *et al.*, 2016. Stretchable active matrix temperature sensor array of polyaniline nanofibers for electronic skin. Advanced Materials 28, 930–935.

image of CNT ribbons on PDMS substrate fabricated by stretching-release-buckling strategy is shown in **Fig. 8**(b). The conductors can accommodate over 100% strain with little increase in resistance, as shown in **Fig. 8**(c). Another example is printing CNT-based ink on a polymer substrate to act as the electrodes for wearable electrochemical sensors. As shown in **Fig. 8**(d), an electrochemical sensor array consisting of potentiometric ammonium sensors and amperometric glucose sensors was fabricated (Bandodkar *et al.*, 2016b). The CNT-based electrodes were screen printed on an elastomer substrate. The intrinsic stretchability of the CNT networks and the mechanical design strategy using the serpentine structure together contributed to the high stretchability of the sensor array up to 500% with stable electrochemical properties.

CNTs are widely used in wearable strain and pressure sensors with a large linear working range and a large gauge factor. For example, a vertically aligned SWCNT array was synthesized and deposited as a thin film to be used as a wearable strain sensor with high stretchability (Yamada *et al.*, 2011). The island-bridge structure formed when the sensor was stretched; the CNT films fractured into gaps and islands with the CNT bundles bridging the gaps. The resistance between the islands changes due to the reduced conducting paths in the bundles. Benefiting from the unique morphology, the strain sensor showed a working strain range up to 280% with fast response and good cyclic performance. In another example, the CNTs were used as the conductive filler in a porous rubber matrix, and both piezoresistive type strain sensors and pressure sensors were fabricated for multimodal sensing, as illustrated in **Fig. 8**(e) (Jung *et al.*, 2014). The strain and pressure sensors were used to detect the finger bending and the pressure on the finger, respectively, and then the signals were processed by a data acquisition unit to control the movement of a tank-like car remotely via wireless communication.

CNTs have been proved to be an excellent material for thermistors with negative TCR, which can be used for human body temperature detection. CNT/PEDOT:PSS nanocomposites have been used as the ink filler to print wearable flexible temperature sensors with an excellent thermal response. The Takei group developed wearable sensors based on CNT/PEDOT:PSS by different fabrication processes, and these temperature sensors showed tunable sensitivity ranging from 0.25% to 0.63% $^{\circ}C^{-1}$ (Kanao *et al.*, 2015; Harada *et al.*, 2014a; Harada *et al.*, 2014b). A representative wearable temperature system is shown in **Fig. 9**(a and b). The CNT/PEDOT:PSS composite percolation film or 3D matrix showed negative TCR due to the electron hopping at the interface of PEDOT:PSS and CNTs rather than individual nanotubes (Nakata *et al.*, 2017). Apart from being used as the thermal resistive materials in temperature sensors, CNTs can also be used as the channel materials in thin-film transistors. In one example, the CNT transistors were integrated with stretchable temperature sensors using polyaniline nanofibers as the thermoresistive sensing material (**Fig. 9**(c)) (Hong *et al.*, 2016). This kind of transistor-based active matrix can minimize signal crosstalk and thereby offer better spatial resolution and faster response. By interconnecting temperature sensors to the transistor array using liquid metals via holes and microchannels, temperature sensing was achieved with high stretchability (30%) and sensitivity (1.0% $^{\circ}C^{-1}$) (Hong *et al.*, 2016).

Graphene and the Derivatives

Graphene and its derivatives, with an exceptional combination of electronic, optical, and mechanical properties, have shown promising potential for wearable devices. The ultra-thin thickness of graphene makes it conform to a variety of surfaces while keeping much of its transparency and conductivity (Das *et al.*, 2018). There are two typical ways to fabricate graphene and the derivatives, CVD synthesis (bottom-up) (Hwang *et al.*, 2013) and exfoliating graphite (top-down) (Cai *et al.*, 2012) (**Fig. 10**(a and b)). Chemical exfoliation of graphite (e.g., Hummers methods) can produce GO, where solution dispersed graphite is exfoliated by inserting large alkali ions between the graphite layers. The produced GO has abundant defects and side groups (Zhang *et al.*, 2013; Park and Ruoff, 2009). Further reduction of GO can lead to rGO, whose electrical and mechanical properties may depend on the degree of reduction. The chemical exfoliation method owns the merits of scalable production with low cost, benefiting practical applications. GO and rGO synthesized by wet chemical methods can be doped with other elements. Graphene and graphene derivatives can be modified into structures such as ripples (Wang *et al.*, 2011), meshes

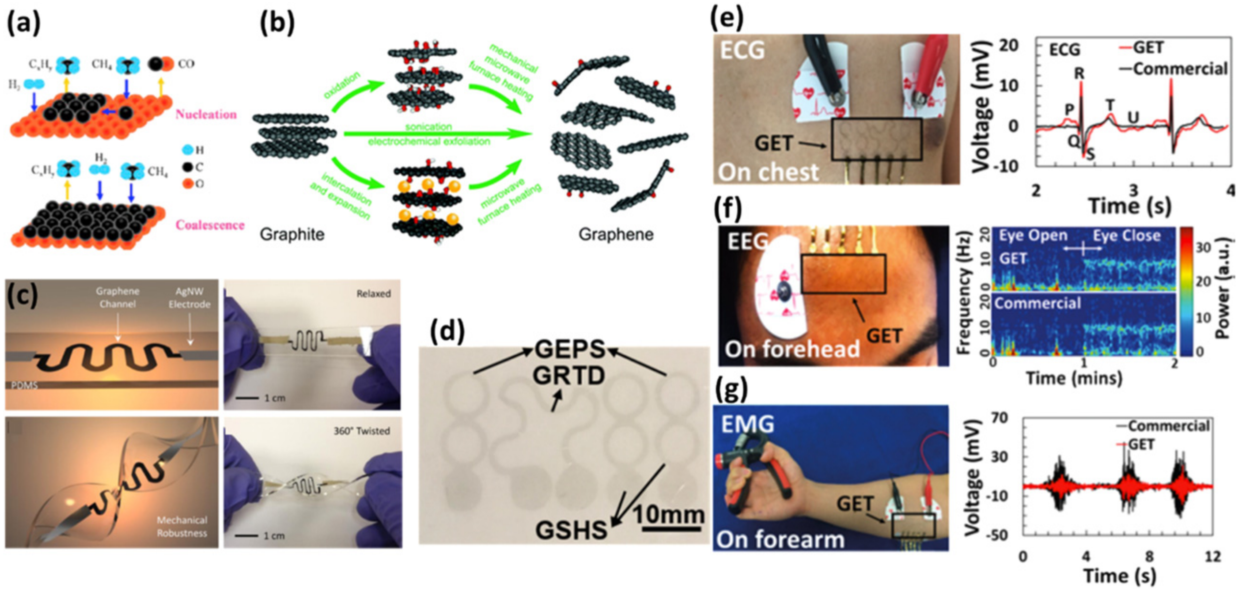

Fig. 10 (a) Mechanism of preparing graphene by CVD. (b) Mechanism of preparing graphene derivatives by chemical exfoliation. (c) Schematic diagrams and images of the stretchable graphene thermistors at relaxed and twisted states. (d) Picture of a graphene-based tattoo sensor array with graphene-based electrophysiological sensors (GEPS), a resistance temperature detector (GRTD), and a skin hydration sensor (GSHS). (e–g) ECG, EEG, and EMG measurement setups for and the corresponding signals from the graphene-based electrophysiological sensors. Reproduced from (a) Hwang, J., Kim, M., Campbell, D., *et al.*, 2013. van der Waals epitaxial growth of graphene on sapphire by chemical vapor deposition without a metal catalyst. ACS Nano 7, 385–395. (b) Hwang, J., Kim, M., Campbell, D., *et al.*, 2013. van der Waals epitaxial growth of graphene on sapphire by chemical vapor deposition without a metal catalyst. ACS Nano 7, 385–395. (c) Yan, C., Wang, J., Lee, P.S., 2015. Stretchable graphene thermistor with tunable thermal index. ACS Nano 9, 2130–2137. (e-g) Kabiri Ameri, S., Ho, R., Jang, H., *et al.*, 2017. Graphene electronic tattoo sensors. ACS Nano 11, 7634–7641.

(Yang *et al.*, 2017; Wang *et al.*, 2014), and crumpled films (Han *et al.*, 2018), which further improves the mechanical robustness and the electrical stability under deformation like bending and stretching. Compared to the CVD-grown graphene, chemically exfoliated graphene derivatives are more suitable for wearable electronics due to the low cost, ease of modification, no need for the transfer process, and compatibility with widely used fabrication processes such as coating and printing.

Graphene-based thermistor temperature sensors have been extensively studied for healthcare applications due to their higher TCR and robust mechanical properties. As shown in Fig. 10(c), a graphene thermistor temperature sensor was fabricated by embedding graphene in a PDMS matrix (Yan *et al.*, 2015). The thermal index was tunable by simply modifying the applied strain. The unique adjacent crumpled structure led to strain-induced contact change and hence resistance change, leading to large sensitivity. Benefiting from the intrinsic flexibility of graphene and the serpentine design, the sensor reached a stretchability of 50%. However, achieving accurate temperature detection when the sensor was deformed was challenging due to the interference with the strain.

Graphene has also been widely explored to fabricate dry electrodes for biopotential measurement, including ECG, EMG, and EEG. Graphene-based tattoo sensors were developed that can be attached to the chest and forearm to measure ECG, EEG, and EMG signals (Fig. 10(d)). The heart rate was extracted from the ECG signal, the EEG signal was collected when closing and opening eyes, and the EMG signal was measured when the subject squeezed a handgrip, as illustrated in Fig. 10(e–g) (Kabiri Ameri *et al.*, 2017). The ultrathin feature of the graphene electrode facilitated conformal contact of the electrodes on the skin, leading to an comparable impedance between skin and the electrodes to that of commercial pregelled electrodes but at a smaller contact area.

An all-graphene multisensory system was achieved with multiple sensors integrated onto one wearable patch, as shown in Fig. 11(a) (Ho *et al.*, 2016). Capacitive humidity sensor enabled by GO, resistive temperature sensor enabled by rGO, and capacitive pressure sensor using CVD grown graphene as two electrodes were integrated; all the sensors were interconnected by CVD-grown graphene. Another highly integrated wearable system was developed by transfer printing patterned graphene onto the skin (Choi *et al.*, 2015). The resistance of the electrodes can be adjusted by stacking different layers of monolayer graphene. The system included sensing (Fig. 11(b)), display (Fig. 11(c)), and therapy components (Fig. 11(d and e)). To detect human body motions, a graphene strain sensor was included in the platform with a LED light to display the sensing results. Furthermore, the therapy component consisted of a heater and drug-loaded iontophoresis electrodes. Disorder of human motion detected by the strain sensor could trigger the heating or drug delivery functions from the therapy component.

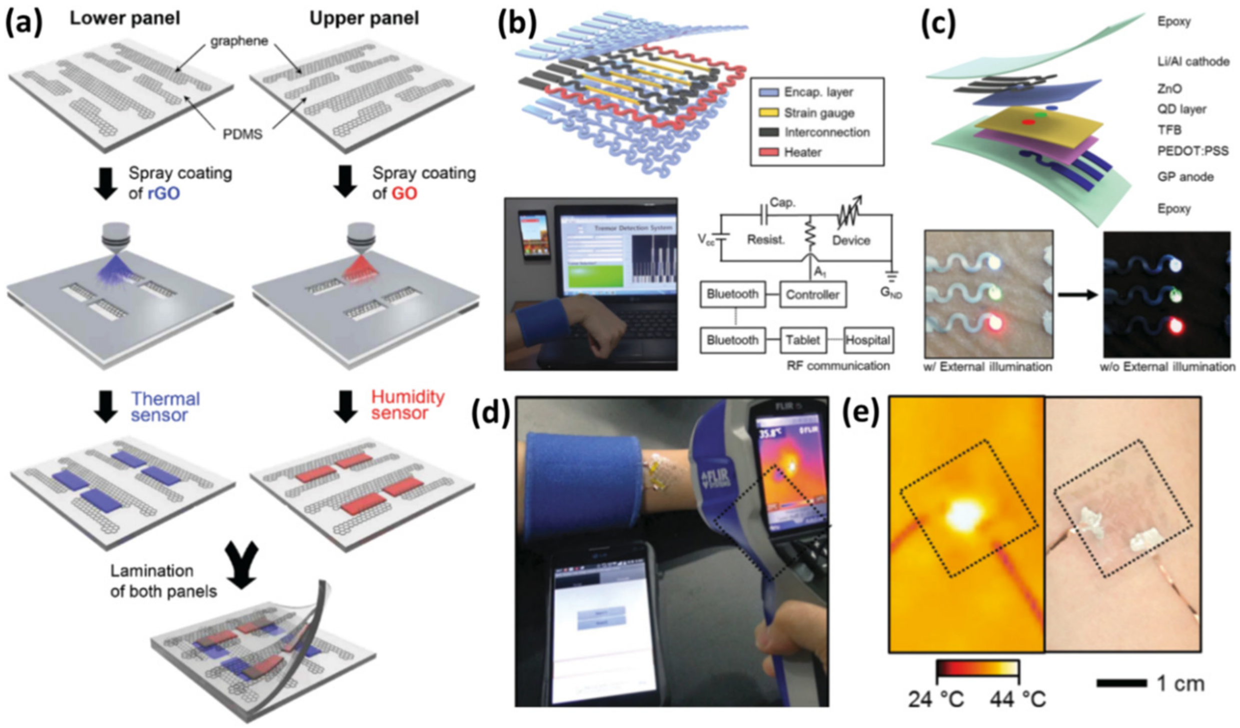

Fig. 11 (a) Fabrication of the stretchable and multimodal all-graphene multisensory system. (b) Exploded illustration of the wearable sensors and heater with a wireless tremor detection system. (c) Exploded illustration of the wearable QLED. The bottom images show the QLED laminated on the human arm with (left) and without (right) the external illumination. (d) Wirelessly controlled thermal actuator using a smartphone application. (e) IR (left) and optical (right) images of the graphene heater laminated on the skin. Reproduced from (a) Ho, D.H., Sun, Q., Kim, S.Y., *et al.*, 2016. Stretchable and multimodal all graphene electronic skin. Advanced Materials 28, 2601-2608. Choi, M.K., Park, I., Kim, D.C., *et al.*, 2015. Thermally controlled, patterned graphene transfer printing for transparent and wearable electronic/optoelectronic system. Advanced Functional Materials 25, 7109–7118.

Hybrid Nanomaterials

As discussed above, advances in material development enable rising numbers on the new nanomaterials with unique properties and merits to be used to construct wearable devices. However, most of the emerging nanomaterials have their drawbacks simultaneously, which are the main reasons that impede the indoctrination and large-scale application of the wearable systems fabricated by nanomaterials. For instance, metal NWs face the problem of performance deterioration during long-term use in ambiance due to the oxidation and sulfuration of the conducting blocks. Carbon-based materials and most of the metal compound materials own limited electrical conductivity, making the signals collection process hard and unreliable. To address these issues, hybrid materials are getting more and more popular and have the potential in offering enhanced mechanical properties, device performance, and system robustness for applications for wearable devices.

Hybridization of two or more conductive nanomaterials is commonly used to improve the electrical properties in two aspects, preventing the conductivity deterioration due to corrosion and enhancing the conductivity. To mitigate the corrosion issue of metal nanomaterials and enhance the mechanical and electrical durability of wearable electronics, coating another protection layer on the surface of the metal nanomaterials has been developed. Core-shell structures are usually formed in these hybrid nanomaterials. One example is to coat a chemical and electrical stable Au thin coating on the surface of AgNW (**Fig. 12**(a and b)) (Kim *et al.*, 2014; Choi *et al.*, 2018). The hybrid NWs showed better anti-oxidation properties than AgNWs, as shown in **Fig. 12**(c). As reported by Choi *et al.* (2018) highly conductive, Au-coated AgNWs were dispersed in elastomer to fabricate a highly stretchable conductor, as shown in **Fig. 12**(d–f). The AgNW core contributes to most of the electric conductivity, while the Au shell enables biocompatibility and oxidation resistance. The hybrid conductor was then patterned into desired electrode structures for monitoring electrophysiological signals including a surface EMG, surface ECG, and intracardiac electrogram.

To enhance the electrical conductivity of carbon nanomaterials and metal compound nanomaterials, hybridization of them with metal NPs and NWs is a promising strategy owing to the outstanding conductivity of metal nanostructures. The hybrid material can also provide tailorable sensing performance by adjusting the hybrid structure, fabrication procedure, and composition ratio. For instance, hybridization of Ag NWs with CNTs, graphene, and metal oxide NWs are commonly used to enhance the electrical conductivity of the transparent conductive electrodes (Hwang *et al.*, 2021; Deng *et al.*, 2015; Pillai *et al.*, 2016; Kim *et al.*, 2013). Hybridizing metal NWs with other conducting materials can significantly enhance electrical conductivity by forming

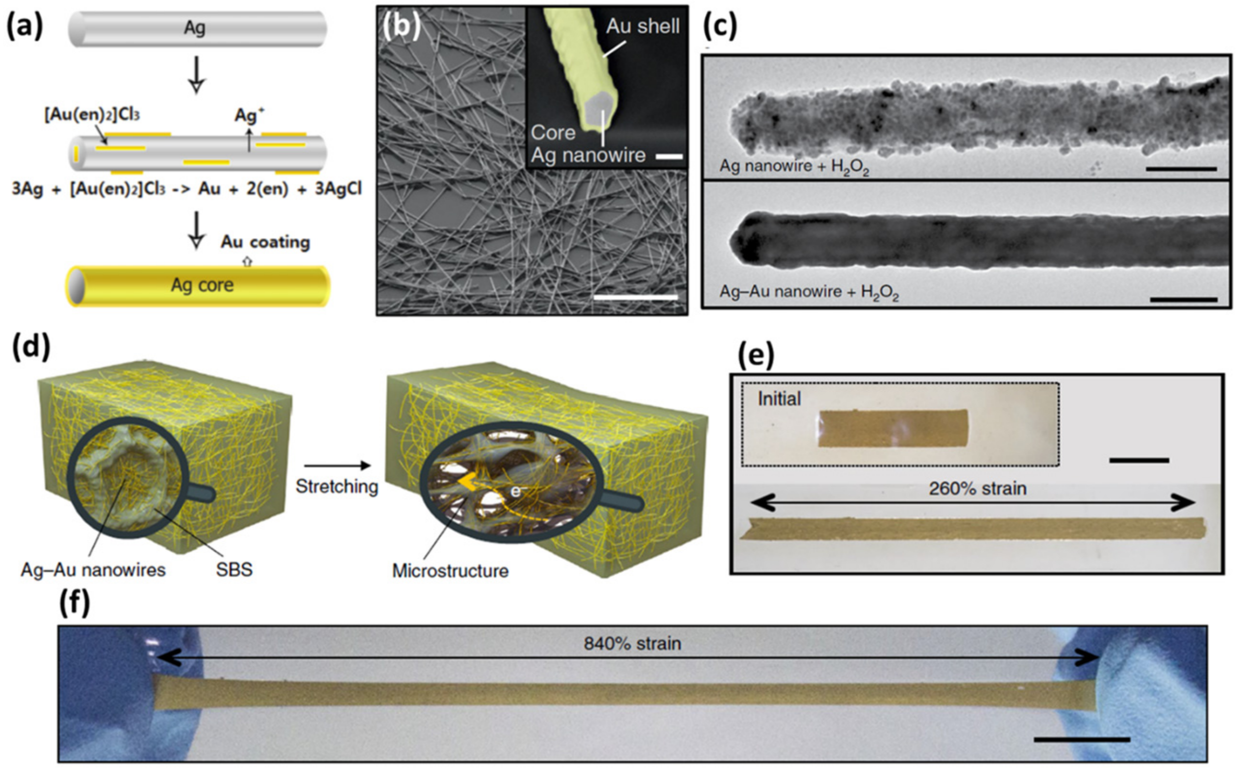

Fig. 12 (a) Schematic diagram and reaction mechanism for the formation of a thin layer of Au on AgNWs via galvanic displacement reaction. (b) SEM images of Ag–Au NWs. Scale bars, 5 μm and 200 nm (inset). (c) TEM image of the AgNW (top) and Ag–Au NW (bottom) treated with 1.5 M H_2O_2. Scale bars, 100 nm (top) and 200 nm (bottom). (d) Schematic illustration of the Ag–Au/elastomer nanocomposite before and after stretching. (e,f) Optical images of the Ag–Au nanocomposite under 260% strain and 840% strain. Reproduced from (a) Kim, T., Canlier, A., Cho, C., *et al.*, 2014. Highly transparent Au-coated Ag nanowire transparent electrode with reduction in haze. ACS Applied Materials and Interfaces 6, 13527–13534. (b) and (d) Choi, S., Han, S.I., Jung, D., *et al.*, 2018. Highly conductive, stretchable and biocompatible Ag-Au core-sheath nanowire composite for wearable and implantable bioelectronics. Nature Nanotechnology 13, 1048–1056.

percolation networks. A FET-based gas sensor with hybrid AgNW-graphene as the electrodes and graphene as the channel was developed (**Fig. 13**(a)) (Park *et al.*, 2016). Metal NWs-graphene hybrid nanostructures exhibited high electrical conductivity, high transparency, and outstanding mechanical robustness. In another example, Lu *et al.* (2014) demonstrated the selective nucleation and growth of AgNPs at the junctions of AgNWs at room temperature and ambient environment to form a hybrid network on a PET substrate, as shown in **Fig. 13**(b and c). As a result, the hybrid network exhibited enhanced electrical conductivity and optical transmittance compared to the AgNW electrodes treated with thermal annealing. As a flexible transparent electrode, the hybrid film achieved an optical transmittance of 89.4% and a sheet resistance of 14.9 ohm/sq.

Fabrication

Nanomaterial composites consisted of conducting filler and polymer substrate or matrix dominant the flexible and stretchable conductors, which are one of the most important building blocks in stretchable wearable systems. The goal of combining conducting materials with a polymer matrix is to construct a flexible or even stretchable conductor that can retain the electrical conductivity during stretching with long-term mechanical and electrical durability and stability. There are mainly three approaches to fabricating conductive materials/polymer composites. The first approach is to directly coat or pattern conductive NPs, NWs, CNTs, or graphene onto the surface of the polymer substrates (Ho *et al.*, 2016; Liu *et al.*, 2016; Lipomi *et al.*, 2011). Typical coating techniques include drop-casting, dip coating, spray coating, spin coating, and Mayer rod coating. To pattern the nanomaterial networks on polymer substrate, lithography (photolithography, capillary lithography, nanoimprint lithography), printing (inkjet printing, electrohydrodynamic (EHD) printing, gravure printing, screen printing), laser patterning, mask assisted patterning, and substrate modification patterning have been used (Huang and Zhu, 2021; Huang and Zhu, 2019; Cui *et al.*, 2018). The second is embedding the conducting nanomaterial film or pattern under the surface of the polymer by curing the polymer on the nanomaterial film or pattern and then peeling it off. For example, the conductor constructed by embedding AgNW under the surface of PDMS shows high conductivity and stretchability (Yao *et al.*, 2017; Xu and Zhu, 2012). The third is to disperse the conducting filler materials into the polymer matrix to form 3D body conductivity. An example is to disperse Au-coated AgNWs in SBS elastomer to be used as stretchable conductors (Choi *et al.*, 2018).

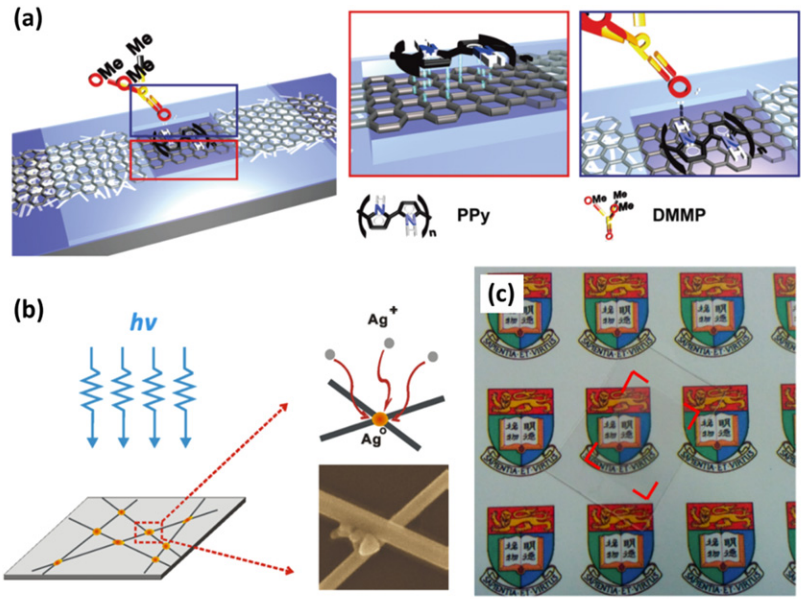

Fig. 13 (a) Schematic illustrations of a FET-based gas sensor using hybrid AgNW-graphene as the electrodes and graphene-based materials as the channel. (b) Schematic illustrations of the selective nucleation and growth of AgNPs at the junctions of AgNWs and the SEM image of the junction area of the fabricated hybrid materials. (c) Photographs of the hybrid network on a PET substrate as a flexible transparent conductive electrode. Reproduced from (a) Park, J., Kim, J., Kim, K., *et al.*, 2016. Wearable, wireless gas sensors using highly stretchable and transparent structures of nanowires and graphene. Nanoscale 8, 10591–10597. (b) Lu, H., Zhang, D., Ren, X., Liu, J., Choy, W.C., 2014. Selective growth and integration of silver nanoparticles on silver nanowires at room conditions for transparent nano-network electrode. ACS Nano 8, 10980–10987.

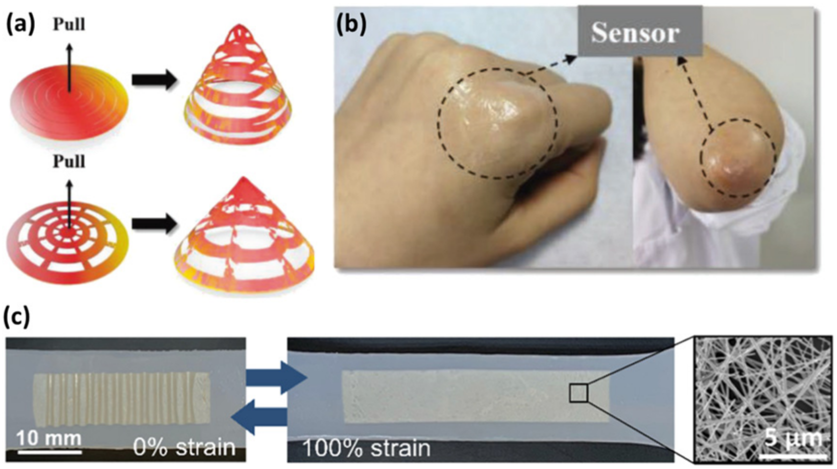

Fig. 14 (a) Schematic of patterning AgNWs/Parylene hybrid electrodes in a 2D plane and then being pulled into a 3D structure. (b) Photos showing AgNWs/Parylene hybrid electrodes enabled sensors attached to the finger joint and elbow. (c) Photographs of the stretchable AgNW/PDMS hybrid conductor stretched from 0% to 100% and the SEM image showing the top surface of the conductor. Reproduced from (b) Yang, C., Zhang, H., Liu, Y., *et al.*, 2018a. Kirigami-inspired deformable 3D structures conformable to curved biological surface. Advanced Science 5, 1801070. (c) Wu, S., Yao, S., Liu, Y., *et al.*, 2020b. Buckle-delamination-enabled stretchable silver nanowire conductors. ACS Applied Materials and Interfaces 12, 41696–41703.

Using these approaches, a rising number of studies on composite design and fabrication are emerging rapidly to fulfill the requirement of conductivity and stretchability of the electrodes and interconnects for wearable electronics. A more advanced approach is to combine the composites with the structural engineering strategy to further increase the stretchability. A kirigami-patterned AgNWs/Parylene hybrid electrodes were developed for wearable stretchable humidity sensor (**Fig. 14**(a and b)) (Yang *et al.*, 2018a). The hybrid showed a sheet resistance of 2.24 Ω sq^{-1} and stability over 60 days. The sensor enabled a conductance change linearly related to the humidity. Wu *et al.* (2020b) reported the combination of using AgNW/PDMS stretchable conductor with a spontaneous buckle-delamination strategy to extend the stretchability of AgNW/PDMS hybrid (**Fig. 14**(c)). By bonding AgNW/PDMS composite film onto a prestrained Eco-flex substrate, uniform buckling structures would form after releasing the applied strain due to the spontaneous delamination. The strategies enabled a stretchability up to 100% while only 3% resistance change was observed. Benefitting from the constant electrical properties, the hybrid structure was integrated with a capacitive touch sensor to detect the pressure of the finger onto other objects.

Compared with 0D nanomaterials and 2D nanomaterials, assembling 1D nanomaterials into aligned structures can endow unique mechanical, electrical, and optical properties attributed to the anisotropic structure. The conventional drop-casting method gives rise to random NW networks. Recently, various assembly strategies have been developed to align 1D nanomaterials on different substrates to be used as functional components in wearable devices. A recent review paper summarizes widely used assembly techniques for oriented NW monolayers or multilayers (Hu *et al.*, 2020). Representative methods include assembly by external fields (Fragouli *et al.*, 2010), shear coating (Park *et al.*, 2015), assembly by template substrates, stretching/releasing of the substrate (Xu *et al.*, 2011), Langmuir–Blodgett technique (Baratto *et al.*, 2020), evaporation induced assembly (Zhang *et al.*, 2009), contact printing (Takahashi *et al.*, 2009), dip-coating (Qi *et al.*, 2018), assembly at liquid–liquid interfaces (He *et al.*, 2007), and layer-by-layer assembly (Wang *et al.*, 2006). The alignment strategy is widely used to assemble metal and metal compound NW transparent electrodes to optimize the electrical and optical properties, because the well manipulated aligned NW networks reduce the ratio of nonconductive portion compared with a random network (Kang *et al.*, 2015; Trotsenko *et al.*, 2015). Apart from the applications of transparent electrodes, the aligned metal compound NWs are also widely used for chemical sensors (Yadav *et al.*, 2020; Zhou *et al.*, 2018a). One of the sensor configurations is using a few NWs to bridge the two electrodes, where the alignment of the metal compound NWs is the prerequisite so that the sensor consists of several individual NWs without NW-NW junctions.

Outlook and Perspectives

Nanomaterials play a significant role in the rise of soft wearable electronics. However, the practice applications of such soft wearable electronics are still in an early stage. Interdisciplinary collaborations are of great importance to translate these research discoveries into industrially amenable products. Below is a list of challenges and future directions for nanomaterial-enabled soft wearable electronics, in our opinion: (1) Scalable synthesis of nanomaterials with high uniformity, low cost, and reproducibility with controlled performance is the prerequisite of using these nanomaterials in commercial products. For instance, CVD-enabled large bench manufacturing of CNTs and graphene is becoming mature. Wet chemical synthesis of metal and metal compounds nanomaterials is attracting more and more attention for fabricating functional materials for wearable electronics. (2) Scalable fabrication methods (e.g., printing) for integrating nanomaterials into wearable devices are in great need. Printing is a promising approach to enabling facile and precise fabrication of nanomaterial-enabled components in wearable devices. Advanced printing methods such as gravure printing, screen printing, and EHD printing have emerged to print nanomaterial patterns with high resolution. Furthermore, printing techniques that can be compatible with industrial roll-to-roll manufacturing have the potential to enable the mass production of these nanomaterial-enabled wearable electronics. (3) More comprehensive wearable systems including sensors, actuators, data acquisition modules, data transmittance modules, displays, and power sources are needed to complete a practically usable system. Multimodal sensing has been employed to fulfill the demand of monitoring multiple signals at the same time. But the interference/crosstalk between sensors is still a challenge that needs to be addressed. For example, how to reduce the strain introduced signal interference for wearable temperature sensors, ECG sensors, and chemical sensors has received extensive studies. (4) New features, such as self-healing, biodegradability, recyclability, and gas-permeability have gained significant interests as they can offer better convenience, user comfort, eco-friendliness, and device robustness. (5) Advanced device integration and packaging techniques, including 3D heterogenous integration, are needed to integrate the diverse sensors, actuators, electronics modules, and other components with advanced polymer materials to ensure the compact size and durability under repeated usage. (6) Standard reliability and stability testing protocols of the soft wearable devices enabled by nanomaterials are highly desirable. (7) Combining the soft wearable sensors with advanced algorithms in data science such as artificial intelligence is critical to analyze the enormous amount of sensor data and extract concise, user-friendly information for the users.

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