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Flexible devices: from materials, architectures to applications*

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Abstract: Flexible devices, such as flexible electronic devices and flexible energy storage devices, have attracted a significant amount of attention in recent years for their potential applications in modern human lives. The development of flexible devices is moving forward rapidly, as the innovation of methods and manufacturing processes has greatly encouraged the research of flexible devices. This review focuses on advanced materials, architecture designs and abundant applications of flexible devices, and discusses the problems and challenges in current situations of flexible devices. We summarize the discovery of novel materials and the design of new architectures for improving the performance of flexible devices. Finally, we introduce the applications of flexible devices as key components in real life.

Key words: flexible devices; flexible architectures; nanomaterials; stretchability

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1. Introduction

With the arrival of the mobile Internet era, portable electronic devices, as one of the most important information processing devices, have become an indispensable part of human life. Wearable electronic devices are a type of portable electronic device integrated with clothes or directly touching the skin. Through the integrated design, processing information will be more convenient and lifestyles will be changed greatly. The research of wearable electronic devices to satisfy the growing demands generated by vast amounts of consumers has become an irresistible trend. Traditional electronics are based on the integrated circuits which are mostly manufactured in rigid and planar semiconductor wafers, which cannot fit irregular, soft or moving objects such as wrinkled clothes and human skin^[1–3]. The dilemma of the traditional electronics results in the birth of flexible electronic technology directly. The recent dramatic progress in this research field has opened a new prospect for future electronics. Flexible electronic technology offers a wide-variety of applications in cutting-edge technological areas, such as E-skin in robotics and prostheses^[4–7], biosensors^[8–15] and biomedical instruments in the medical area^[16–18]. On top of these, it also plays a vital role in flexible batteries^[19–23], transistors^[24–28], electrodes^[29–30], display devices^[31] and conformable RFID tags^[32]; all of these have generated immense interest for the scientific and industrial world. There have been many flexible electronic research advancements, and some related products are also designed and manufactured.

Flexible electronics entail the ability to undergo large mechanical deformation, such as bending, stretching, twisting and folding, even to deform into more complex shapes

while keeping the high performance of the devices. These ideal properties demand that the materials applied to flexible electronics embrace great flexibility and other excellent physical and chemical characteristics. There is no doubt that finding and synthesizing new materials has made a great difference in putting forward the development of flexible electronic technology. Numerous materials can be used in manufacturing flexible electronics, such as graphene^[15, 33], carbon nanotubes (CNTs)^[9, 34–36], liquid metals^[37] and conductive polymers^[38–40].

Aside from finding new materials to fabricate flexible electronic devices, designing specific flexible and stretchable architectures also has significant meaning for flexible electronic devices, especially for some rigid inorganic materials. Through flexible architecture, the stretchability and bendability of the devices will be improved greatly. This article serves as a review which mainly focuses on the unique flexible architectures and materials that are used to manufacture flexible electronics, which have abundant applications in various fields.

2. Materials

Silicon materials, though widely used in the traditional electronic industry, have limited applications in manufacturing flexible electronic devices due to the intrinsic inflexibility of silicon wafers and the complicated fabricating processes of its microstructure. Hence, searching for new and highly flexible materials as substitutes for traditional silicon material is of great importance. Numerous materials can be used to fabricate flexible electronic devices, such as graph-

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ene^[33, 41], carbon nanotubes^[34–36], GaAs^[42], liquid metals^[37], conductive polymers^[38–40] and so on. These materials can be divided into two classes: inorganic materials such as the traditional silicon and inorganic nanomaterials, and organic materials, which includes conductive polymers and electroluminescence organic molecules. Traditional inorganic materials have excellent physical and chemical properties such as high electric and thermal conductivity, however, most of traditional inorganic materials also have obvious disadvantages such as fragile, terrible bendability and stretchability. To solve those problems, scientists designed a series of flexible architectures mentioned above to overcome the disadvantages, however, these methods just solve the problem partially. In this context, some fresh inorganic materials especially nanomaterials are attracting lots of attention. The low dimensional nanomaterials not only possess outstanding physical and chemical properties containing the merits of bulk inorganic materials, but they also have excellent intrinsic flexibility, so they are very suitable to manufacture the flexible electronic devices. Contrary to the inorganic materials, organic materials have outstanding intrinsic flexibility making them more suitable for fabricating flexible electronic devices, especially the large scale of display devices. Nevertheless, organic materials also have some deadly weaknesses, which affect their development heavily. Compared with inorganic semiconductors, the conductive mobility of organic semiconducting materials is several orders of magnitude lower. The conductivity of inorganic metals is also several orders of magnitude higher than organic conductive polymers. Besides, the conversion efficiency of optoelectronic converter devices based on organic semiconductors is far less than that of inorganic semiconductor devices. The limited physical and chemical properties of organic semiconductors restrict the development of organic flexible electronic technology^[43]. But considering the industrial applications, the manufacturing process of organic materials is simpler than inorganic materials. Moreover, the manufacturing process of inorganic materials usually needs some corrosive chemicals, which are inconvenient and dangerous^[43]. For most of the inorganic materials, the key to breaking through the bottleneck is the materials' lack of intrinsic flexibility; however, for organic materials, regulating the characteristics of the materials themselves is the key to gaining high performance of devices. We introduce the current process of flexible devices using low dimensional inorganic nanomaterials and organic materials as follows.

2.1. Inorganic nanomaterials

2.1.1. Two-dimensional (2D) materials

When the lateral and vertical scale of materials decrease to the nanometer range, the stretchability and anti-bending ability will be improved greatly. Two-dimensional materials (2D materials) are a class of materials which consist of individual freestanding layer materials with a thickness of a few atoms or even single-atom. 2D materials have a lot of excellent electric, optical, thermal and mechanical properties. The materials' flexibility can be greatly enhanced with the decrease of thickness. Hence, 2D materials are a type of material which

is really suitable for manufacturing flexible electronic devices^[44].

Graphene is a typical 2D material that possesses outstanding electrical, chemical and mechanical properties^[45]. These remarkable properties made graphene become a rapidly rising star on the horizon of material science in recent years, and also make it attractive for applications in flexible electronics. As it is known to all, flexible electronics has a wide range of research areas. The applications using graphene in these areas include high-performance electric and optical devices, energy storage devices, and biological sensors. The transparent and flexible electrodes play a vital role in the electronic devices. The common commercial transparent electrode materials are indium tin oxides (ITO), while the rigid properties hinder the fabrication of flexible transparent electrodes. Because of the ultrathin feature, graphene possesses excellent light transmittance and flexibility. Meanwhile, it also has good electric conductivity, making graphene very suitable for manufacturing transparent flexible electrodes. Bae *et al.* reported the roll-to-roll production of 30-inch graphene films for transparent electrodes^[33]. They used the chemical vapor deposition (CVD) method to grow a large scale of graphene with 97.4% light transmittance on the copper foils. A layer-by-layer stacking method was used to stack a four-layer graphene film with about 90% transparency, and its sheet resistance was as low as 30 Ω/\square . The transparent graphene film was fabricated as electrodes and incorporated into fully functional touch-screen panel devices. The graphene-based panel resists up to 6% strain, better than an ITO based touch panel, which can only resist up to 2%–3% strain. The strain range was limited by the printed silver electrode, rather than graphene itself. Lee *et al.* fabricated a kind of flexible resistive random access memory device, which used graphene as the substrate materials and grown NiO_x/GaN microdisk arrays on graphene films^[41]. The device showed excellent performance even after 1000 bending cycles, and the resistive switching characteristics of the device were nearly unchanged.

Due to the zero band gap of graphene, there are a lot of leakage currents phenomenon in the applications using graphene as active materials. So scientists try to use some chemical methods to modify graphene and open the band gap. Graphene Oxide (GO) is manufactured to functionalize graphene using a strong oxidizer such as KMnO_4 to oxidize the graphene. As the defects and oxygen functionalization, the GO is a kind of semiconductor which exhibits a transport gap greater than 0.5 eV at room temperature^[46, 47]. The oxygenated functional groups of the Oxide Graphene are always on the basal plane and edges, this derivatization breaks the π -conjugated network and results in the change of band gap of this type of material^[48]. Because GO embraces excellent physical and chemical characteristics which are similar to graphene and contains the proper band gap at the same time, it has broader applications in flexible electronics. Jeong *et al.* used a room temperature spin-casting method to manufacture a type of flexible GO based memory device on flexible substrates^[49]. When the device was continuously bent 1000 times, the performance of the devices was nearly unchanged. The devices can maintain a large bending radius that can

reach 7 mm, which can be considered as an “extremely flexed state”. Their work also had attractive applications in flexible charge-based memory devices such as dynamic random access memory (DRAM) and flash memory.

Reduced GO (rGO) is a kind of carbon materials which is similar to graphene but with some residual oxygen and structural defects. The conductivity is comparable to the doped conductive polymer^[50] and far higher than the doped silicon materials^[51]. Because of splendid properties of rGO, it has roused lots of scientists’ interests. Liu *et al.* reported a kind of flexible composite anode material for sodium-ion batteries made by red phosphorus nanodots and rGO sheets^[52]. The red phosphorus nanodots were deposited onto the rGO sheets to decrease the sodium ion diffusion length and the sodiation/desodiation stresses, and the rGO network also provided an electron pathway and free space to adapt to the volume variation of phosphorus particles. The authors also measured the mechanical properties of rGO/graphene composites. When the film was bent at the bending radius larger than 3.4 mm, no obvious resistance change was observed (<10%). When the bending force became larger, a 17% resistance increase was observed at the bending radius as small as 2 mm. Finally, when the test was finished, the films were free-standing and the resistance rarely changed. Experimental results proved that rGO based composite material is a kind of outstanding flexible material.

Aside from the graphene materials family, there are numerous other 2D materials which can be used in flexible electronic devices. Transition metal dichalcogenides are a class of 2D materials which have high carrier mobility, photoconductivity, thickness-dependent electronic band structure, and environmental sensitivity. Hence, they have better prospects for applying in electronics compared to other materials. Except transition metal dichalcogenides, 2D boron nitride is also an important dielectric material which can be used to fabricate field-effect transistors. Lee *et al.* fabricated vertical heterostructures using hexagonal boron nitride and graphene, and then, flexible and transparent MoS₂ field-effect transistors were manufactured on the heterostructures^[25]. The electrical measurement was performed when the devices were under uniaxial strain by bending. The statistics indicated that the performance of the flexible device remained unchanged with applied uniaxial strain up to 1.5%. Georgiou *et al.* designed a type of vertical field-effect transistor based on graphene-WS₂ heterostructures. These devices can operate on transparent and flexible substrates. The experimental statistics indicated that the devices were insensitive to bending and the devices still showed excellent performance when the devices withstood 4% strain^[53].

2.1.2. One dimensional (1D) materials

Beyond 2D materials, other nanomaterials also play significant roles in the flexible electronics area. One dimensional materials (1D materials), including nanotubes, nanowires and nanoribbons have a lot of different electrical, thermal and mechanical properties from bulk materials.

Carbon nanotubes (CNTs) is a promising material for flexible electronics for their significant mechanical flexibility, conductivity and intrinsic carrier mobility, which can be used to

serve as the channel materials in field-effect transistors, and also can be fabricated as films for transparent electrodes^[54]. Besides, CNTs is also a highly suitable candidate for other flexible electronic applications. Yakobson *et al.* set up a model molecular dynamics method (MD), to analyze the deformation of CNTs when CNTs were stretched by applied strain; he predicted the theoretically maximal strain of the CNTs can reach to 15%, indicating the high flexibility of CNTs^[55]. The theoretical prediction was then identified by experimental results^[56, 57]. Actually, many CNTs materials possess numerous defects, so pure CNT yarn in previous reports can bear only 7–8% strain as the fractures will appear when the strain increases to the maximal limit^[58, 59]. In this case, some approaches were put forward to strengthen the flexibility and stretchability of the CNT yarns. One approach adopted some polymer materials as support media to fabricate carbon-polymer composite ribbons, and the other strengthened the stretchability of original CNTs yarns by designing some flexible structure. For example, Shang *et al.* reported a yarn-derived spring-like CNT rope. In this case, the CNTs yarns could elongate several times of its original length and the maximal strain was 285%^[60]. Through these processes, the flexibility of CNTs materials will be greatly improved and make them more suitable to be applied to flexible electronics. Xia *et al.* designed a type of all-carbon-nanotube transistor on flexible and transparent substrates indicating extreme stretchability. The transistor exhibited high mechanical stability when the length change was less than 5% (22.5% strain was applied). They also found that the on–off ratio increased with the increase of stretch strain, while mobility initially increased and then decreased with the increase of stretch strain^[34]. Xu *et al.* manufactured a kind of transistor using CNTs, ion gel and buckled metal composite films. The FETs maintained high performance when a stretch change up to 50% was applied. The intrinsic flexibility of ion gel allowed it to maintain a high-quality interface with the CNTs during stretching, so the CNTs films can endure large stretching strains^[36]. Chortos *et al.* fabricated the transistors employing polymer-sorted CNTs as the semiconductor, unsorted CNTs as the conductor, and a tough thermoplastic polyurethane (TPU) as the dielectric and substrate, respectively. The devices can maintain constant performance when stretched 1000 cycles, whose stretchable range reached to 100% strain at each cycle^[35].

Except for CNTs, there are some other materials which also play important roles in flexible electronics. Silicon is a traditional material which is mostly used in the semiconductor industry. To solve the problem that silicon cannot be bendable and stretched, silicon nanoribbon material was invented to improve the flexible ability of silicon materials. Rogers’ group has done lots of meaningful work in this area. They fabricated high quality buckled silicon nanoribbons, and used them in manufacturing the transistors. Their devices exhibited excellent performance under 1.4% strain^[61]. Besides, III–V compounds were also investigated, as these materials have high saturated drift velocity, high electron mobility, wide direct band gap, and a wide range of working temperatures. For example, vertical aligned GaAs or InP nanowires can be grown with pre-depositing metal catalyst on GaAs or InP wafers via the VLS process. After being transferred to a plastic substrate, some simple two-terminal diode devices can be directly fabricated;

the GaAs nanowires with a width of ~ 400 nm could withstand the strain of $\sim 1.3\%$ [42].

Compared with metal materials, the conductivities of CNTs and conducting polymer materials are poor, which results in their limited applications and performance. In this case, metallic nanowires (NWs) have roused immense interest of researchers. To the best of our knowledge, most metallic nanowires research is focusing on seeking the substitutes of indium tin oxide (ITO)[62, 63], while only a few groups are studying the flexible electronic applications of metallic nanowires. Lee *et al.* reported that a new type of highly stretchable and highly conducting metal electrode was fabricated using long Ag NWs. The long Ag NWs could retain high performance when it was stretched. The resistance of the nanowires hardly changed and kept on the low sheet under the 460% strain[64].

Various 1D materials play significant roles in the flexible electronics field with different electric properties and mechanic features making them suitable for different applications.

2.2. Organic materials

2.2.1. Polymers

Varieties of polymers play a very important role in the field of flexible electronics, as the majority of flexible electronics need flexible, stretchable and bendable substrates. Due to polymer materials' splendid intrinsic stretchability, they are the most ideal substrate material[65]. Polydimethylsiloxane (PDMS) is the most common substrate material used in flexible electronic devices. Aside from PDMS, there are numerous other polymer materials which can be used in different aspects of flexible electronics. For instance, conductive polymers can be used for interconnects, and semiconductor polymers can be fabricated as organic transistors. Intrinsic stretchability is the most important characteristic of most polymers. Semiconductor polymers have a cost advantage over traditional inorganic semiconductors such as silicon which require complicated process technology and expensive equipment[66]. Elastic conductive polymers possess excellent stretchability, more than traditional metal wires, and their electric properties maintained their original level when accommodating the large strain. Aside from these, flexible electronic devices that use polymer materials are easy to fabricate through the printing technology. Usually, carbon nanotubes (CNTs) will be added into polymer materials to prepare composite materials whose conductivity and stretchability are strengthened.

In 1977, Shirakawa *et al.* found the conductive polyacetylene, the first conductive polymer, and opened a door for the research of conductive polymers[67]. Nevertheless, organic conductors with pure ingredients are still in needed. Conjugated polymers containing a special band structure, is attracting a great deal of attention from researchers. Through doping or modifying functional groups in the side chains to control the whole band structure, the conductivity of polymer can be greatly increased.

Poly (3,4-ethylenedioxythiophene): polystyrene sulfonate (PEDOT:PSS) is a kind of water solution conductive poly-

mer, which has great electric conductivity and is most widely used in the conductive polymer materials area. In recent years, scientists carry out a lot of works in strengthening the conductivity and stability of PEDOT:PSS. Some volatile solvents, such as ethylene glycol and dimethyl sulfoxide have been used to increase the conductivity of conductive polymer materials[68, 69]. Vosgueritchian *et al.* reported a method which used a small fraction of a nonionic fluorosurfactant polymer (Zonyl) (0.1 wt%) with DMSO that has a synergistic effect to enhance the conductivity of PEDOT:PSS[70]. The acid treatment can even increase the conductivity of PEDOT:PSS materials to the same order of magnitude of indium tin oxide (ITOs). Because PEDOT:PSS materials are easily fractured when they are bent, Jin *et al.* mixed an excess amount of a non-volatile surfactant plasticizer, Triton X-100 ($C_{14}H_{22}O(C_2H_4O)_n$) ($n = 9-10$) with PEDOT:PSS, to modify the nanostructure and viscoelastic property of PEDOT:PSS. Through such a process, the mechanical property of these materials was greatly improved[39]. PEDOT:PSS is the representative of conductive polymer materials, increasing the conductivity and stability which is the key point in the researching progress.

Semiconducting polymers materials are other important candidates used in the flexible electronics field. They are intrinsically stretchable and can be manufactured using standard process methods. These properties make them very suitable for fabricating the film based field-effect transistors which can be applied to wearable electronic devices. Similar to conductive polymers, semiconducting polymers are also not stable when they are stretched or bent. The chains of normal polymers are easy to break when the polymers undergo large applied strains. So enhancing the mechanical stability of semiconducting polymers and even achieving the self-healing property have always draw the attention of researchers. Lots of semiconducting polymers are conjugated polymers. Usually, the conjugated polymers which contain modified side-chains and segmented backbones are infused with more flexible molecular building blocks to enhance the stretchability greatly. Jin *et al.* reported a kind of self-healing film field-effect transistor using non-covalent crosslinking to enhance the stability of the polymer and achieve the target of self-healing. When the devices were stretched, the non-covalent crosslinking will be broken and undergo the energy dissipation, while the high charge transport abilities will be kept. The transistor can recover the high performance even after a hundred cycles at the applied strain of 100%. Chortos *et al.* reported a stretchable organic transistor which can withstand $>250\%$ strain[26].

2.2.2. Other organic materials

Aside from polymer materials, there are numerous other organic materials such as small organic molecules, organometallic complexes and so on. Organometallic complexes are a class of materials which contain lots of interesting electrical, optical and magnetic properties. The outstanding optoelectronic characteristics make these materials very suitable for making some luminance applications such as OLED which is a very promising flexible electronic display device. Partially

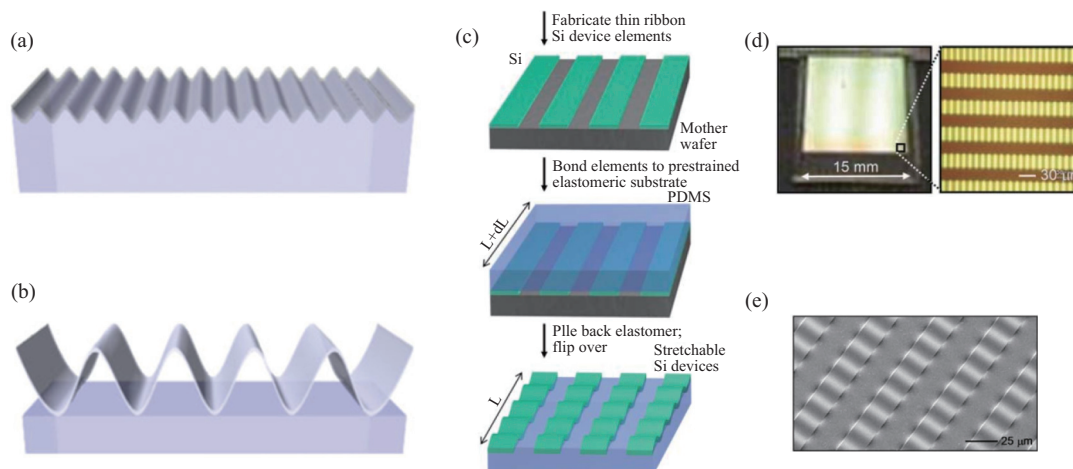


Fig. 1. (Color online) (a, b) Schematic illustration of two different contact structures^[74]. Reprinted with permission, Copyright 2008 John Wiley and Sons. (c) Two-step manufacturing processes of the buckled silicon nanoribbon devices. (d) Optical images of a large-scale aligned array of silicon nanoribbon wave (left) and single-crystal Si ribbons on PDMS (right). (e) Angled-view scanning electron microscope image of four wavy Si nanoribbons in (d)^[75]. Reprinted with permission, Copyright 2005 American Association for the Advancement of Science.

OLED products with excellent performance have even achieved commercial targets at current industrial situations.

3. Flexible architectures

3.1. Flexible planar structures

Traditional silicon integrated circuits with basic units as silicon transistors can handle the task of addressing sensors, enabling signal readout and amplification to achieve great success in the present situation^[71]. However, the rigid and brittle features of silicon hinder its further applications, such as flexible electronic devices. Traditional devices are usually manufactured on rigid silicon wafers by micro-fabrication technology. Due to the drawback of brittleness of silicon, integrated circuits fabricated by this method cannot be incorporated into clothing or attached directly to the human body. To achieve flexible electronic circuitry, the principal block for silicon integrated circuits that silicon materials lack of flexible and bendable properties must be removed. Therefore, a series of effective methods were put forward to tackle that problem. The typical approach focuses on decreasing the thickness of the silicon wafers. As the bending stiffness is in inverse ratio to the thickness of the bulk material, the bending stiffness of silicon wafers becomes lower when the thickness becomes thinner^[72]. Besides, the induced peak strains will decrease linearly with the thickness of materials for a constant bending radius^[72]. Hence, decreasing the thickness of silicon wafers will improve their bendability and stretchability effectively. However, this method just reduces the stiffness of inorganic materials partially, more effective approaches need to be proposed to enable thin brittle inorganic layer materials to withstand a large bending force or applied strains. Therefore, scientists have designed numerous interesting stretchable architectures to enable the devices to embrace excellent flexibility and stretchability.

Recent studies proposed a buckling strategy which means making rigid semiconductor nanowire, nanoribbon or nanosheet materials wrinkled and configured into ‘wavy’

shapes to accommodate large applied strains^[73]. Usually, the wavy structure will be bonded to an elastomeric membrane such as PDMS or plastics polymer to support the active channel and strengthen the flexibility simultaneously. As shown in Figs. 1(a) and 1(b), there are two kinds of manufacturing methods, one is bonding the wavy structure with elastomeric membrane everywhere. Another is bonding the wavy structure with the elastomeric membrane only at the positions of the troughs. The second method was used more often^[74]. Khang *et al.* fabricated stretchable silicon devices using the buckling strategy, as schematically illustrated in Fig. 1(c), a two-step method was adopted to manufacture the wave structure^[75]. First of all, silicon nanoribbons were fabricated on a mother wafer using conventional lithographic processing. Then the pre-strained PDMS was attached to silicon nanoribbons, and the shrink of PDMS would enable the buckling of nanoribbons. Fig. 1(d) shows the optical images of a large-scale aligned array of wavy single-crystal Si nanoribbons on PDMS, and Fig. 1(e) illustrates the corresponding angled-view scanning electron microscope image of four wavy Si nanoribbons in Fig. 1(d). The amplitude of the wavy-shape silicon nanoribbons and peak strain would decrease to resist the applied strains when the devices were stretched. Otherwise, if the applied strain increases continually, the wavy structure would recover to its original plane structure and then elongate until retaining its limits. The compression strain limit and the stretched strain limit for 0.9% pre-strained devices can reach -27% and 2.9% , respectively. Though the pre-strain can be even heavy and the structure will have a larger strain limit, the PDMS membrane endured strain will not increase as it will fracture before the silicon wavy structure gets damaged.

Another architecture, unlike the full-ranged wave, is an open mass geometry in which a mesh structure similar to a fishing net was used^[73]. The typical mesh structure unit is formed by two groups of vertical lines that form a net containing a lot of rectangular meshes. The meshes of the net will become smaller and the shape of the meshes will turn into a long rhombus rather than a square, when the architecture is

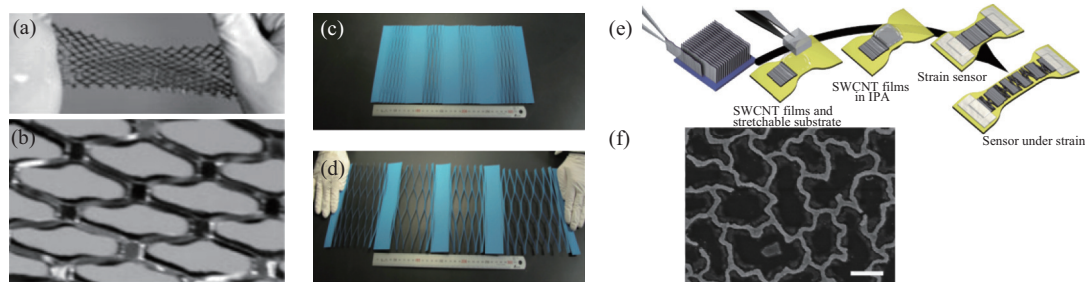


Fig. 2. (Color online) (a, b) Optical images of a net-like organic transistors based device, the device can extend by 25% when it is stretched^[76]. Reprinted with permission, Copyright 2005 National Academy of Sciences. (c, d) Optical images of the stretch process of mesh structure formed by parallel lines and linked points^[77]. (e) Schematic illustration of strain sensor fabricated by SWCNTs^[77]. Reprinted with permission, Copyright 2011 Nature Publishing Group. (f) SEM image of irregular Au nanomesh structure which acts as highly stretchable and transparent electrodes^[78]. Reprinted with permission, Copyright 2014 Nature Publishing Group.

stretched along the lateral direction. Otherwise, the blank area will disappear and the ‘X’ structure will become two parallel lines when the mesh structure shrinks. The applied force in plane was released by meshes of mesh structure rather than the material itself. Someya *et al.* designed a type of pressure and thermal sensor using the open mass geometries, as shown in Figs. 2(a) and 2(b)^[76]. A plastic film, together with organic transistors and pressure-sensitive rubber, formed the net-shaped structure, which enabled the device to extend to 25% when the devices were stretched.

The mesh structure can be hidden, in some other electronic devices, where only parallel lines were revealed and linked at some special points. When the structure was stretched along the lateral direction by applied force, the meshes revealed and endured the strain, as shown in Figs. 2(c) and 2(d)^[77]. Yamada *et al.* demonstrated a carbon nanotube based stretchable sensor for human-motion detection^[77]. They used water-assisted chemical vapor deposition to grow vertically aligned SWCNTs films. Then, the carbon nanotube films were supported on PDMS and the array of the SWCNTs kept perpendicular to the strain axis. The structure image was illustrated in Fig. 2(e). When the sensor was stretched, partial points of SWCNTs films were still connected and most of SWCNTs would be separated, forming the meshes structure. Usually, the stretchability of a device is evaluated by its maximal endurance, which limited the strain of the structure. However, the electrical performance of the devices should also be considered. For example, some flexible electronic devices use metal foils, the metal foils will become narrower and longer which will result in the increase of the resistance and destroy the electrical conductance of performance devices. The electrical response of the strain sensor remained nearly unchanged under 10 000 cycles at 100 and 150% strains. The performance of the devices was limited by the substrate until the substrate ruptured, rather than limited by the stretchable architecture. Apart from some standardized mesh structure, some irregular mesh structures also show outstanding stretchability. Guo *et al.* fabricated a kind of highly stretchable and transparent nanomesh electrode and Fig. 2(f) illustrates the mesh structure^[78]. The especially irregular Au nanomesh structure enabled the electrical resistance of the device to increase modestly when a one-time strain of 160% was applied. In contrast, other materials such as car-

bon nanotube films and graphene sheets, which lack the special architecture, the electrical resistance would increase steeply under strain <100%. The special nanomesh structure improved the electrical character of devices well.

Although two strategies mentioned above can improve the stretchability of the devices, the two architectures merely bear the applied strain from one direction. When the devices are stretched from several directions or bent, they will lose their effectiveness and be damaged irreversibly. In this case, researchers came up with a more effective architecture which utilizes the inspiration of the above two strategies and combines advantages of the two architectures. Fig. 3(a) illustrates a new architecture named ‘islands and bridges’ structure. The whole rigid silicon wafers are divided into numerous small pieces and linked with some stretchable conductive wires, just like linking the islands with bridges. The active channel parts of the devices are located in the silicon islands. The buckling strategy is the most common method used in constructing stretchable interconnects, and the whole array of bridges and islands formed the mesh structure^[79]. Usually, the whole structure will be bonded with an elastomeric membrane to support it and help gain better flexibility. When the structure is stretched, the stretchable conductive wires will bear all the deformation and the active device islands experience nearly no deformation. So the applied strains have little influence on the electronic characteristics of the devices. Based on the islands and bridges structure, the flexible electronic devices can not only be stretched, bent and twisted, but also integrated into a variety of curved surfaces.

The simplest stretchable interconnect medium is an arch structure. Heung *et al.* invented a hemispherical electronic eye camera using this strategy, as shown in Fig. 3(b)^[80]. Fig. 3(c) illustrates the transfer process of islands and bridges array from PDMS to a hemispherical glass lens substrate^[80]. Compressible metal interconnects which look like arch structure clearly can be viewed. The silicon device islands are photo-detector and p-n diode. This design can tolerate compression and stretch with large levels of strain (50% or more). Based on the simplest arch structure interconnects, scientists changed the shapes and structures of interconnects and optimized their flexibility and stretchability.

Although some unique structures are not complicated, they are effective. The easiest approach proposed is using

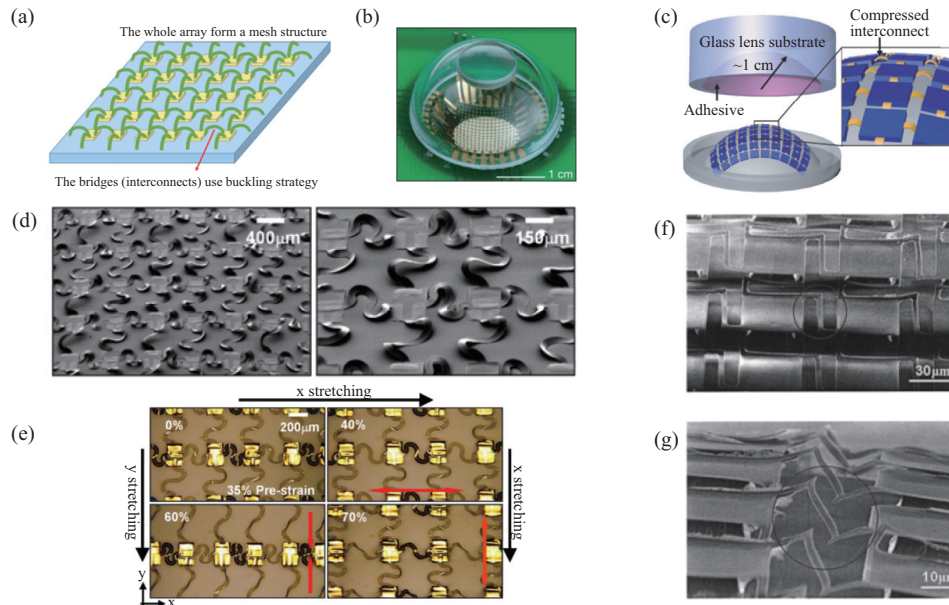


Fig. 3. (Color online) (a) Schematic illustration of the basic islands and bridges structure. (b) Optical images of hemispherical electronic eye camera with islands and bridges structure^[80]. (c) Schematic description of the transfer process of islands and bridges array from PDMS to hemispherical glass lens substrate^[80]. Reprinted with permission, Copyright 2008 Nature Publishing Group. (d) SEM images of the self-similar serpentine structure. (e) The stretched process of the self-similar serpentine structure^[81]. Reprinted with permission, Copyright 2008 National Academy of Sciences. (f) SEM image of a type of S-shaped microfabricated suspensions. (g) Magnified SEM image of the shape-changed process of the structure with S-shaped microfabricated suspensions under pressure^[82]. Reprinted with permission, Copyright 2004 AIP Publishing LLC.

two or more arch ligaments to connect the active device islands^[79]. The second approach is making interconnects themselves have flexibility and stretchability. There are several interesting works to increase the flexibility and stretchability of interconnects. Kim *et al.* designed a type of self-similar serpentine structure presented in Fig. 3(d)^[81]. The structure can increase the quantity of arches and transform the non-plane structure to a plane structure, which used a simple technology process such as printing. The additional radian can enable more potential energy to achieve better stretchability. Fig. 3(e) demonstrates the changing process when the structure was stretched. This approach can be used in manufacturing transistors, the basic active unit of integrated circuits. In addition, Hung *et al.* designed a type of S-shaped micro-fabricated suspension made by Au and Cr, to provide a good electrical conductivity of the structure, as illustrated in Fig. 3(f)^[82]. The structure showed low spring constant; the S-shaped microfabricated suspensions would deform but remain connected and their resistance was nearly unchanged even under 50 KPa pump pressure. Fig. 3(g) shows the shape-change of the structure when the pressure was applied. The structure designed by Huang *et al.* is more creative. Deep reactive ion etching was used to construct a stretchable and two-dimensional wired network in a monolithic silicon substrate, which can cover and interconnect the large planar or curved surfaces to realize outstanding, large-area, monolithic silicon electronics in a cost-effective manner. The key point of the structure was the spiral ribbons which connected the silicon device islands^[83]. When the structure was stretched, the spirals unroll, and the whole architecture would expand to resist the induced strain. The stretching range would attain the maximal limit when the

spirals turn into a straight line. So the stretched limit is decided by the diameter of the innermost spiral winding (D) and the thickness of the spiral ribbons (t), $\epsilon_{\max} = t/D$. The maximum strain of the structure reported by this group can reach to about 3.1%. Although this statistic is not very outstanding, the design of the architecture is unique and attractive.

Among those interconnect-designed structures, self-similar serpentine structures attract plenty of interest. Some groups have designed several different curve-shape interconnects and compared them through Finite element modeling (FEM) or experimental tests^[84–85]. Gonzalez *et al.* used copper tracks to connect the silicon active device islands. They have designed several types of curved copper tracks, just as shown in Fig. 4(a)^[84]. They also explored the best shape of copper tracks through the aforementioned methods, as presented in Figs. 4(b) and 4(c). They found that a horse shoe like shape is the optimal structure to acquire the best performance, and Fig. 4(d) shows this shape^[85].

Xu *et al.* used a self-similar serpentine interconnected structure to manufacture a type of stretchable Lithium battery^[86]. Figs. 5(a) and 5(b) show the flexible structure of the batteries, the fine structure is illustrated in Fig. 5(b). They improved the original self-similar structure and proposed a kind of hierarchical buckling strategy. As illustrated in Fig. 5(c)^[87] and 5(d), two straight copper lines were used to connect three columns of copper serpentine wires and form a two level self-similar structure. The black line in Fig. 5(d) indicates the first level structure which has a ‘short’ wavelength serpentine, and the yellow line shows the second level structure which has a ‘long’ wavelength serpentine. Owing to the hierarchical buckling strategy, the flexibility and stretchability of

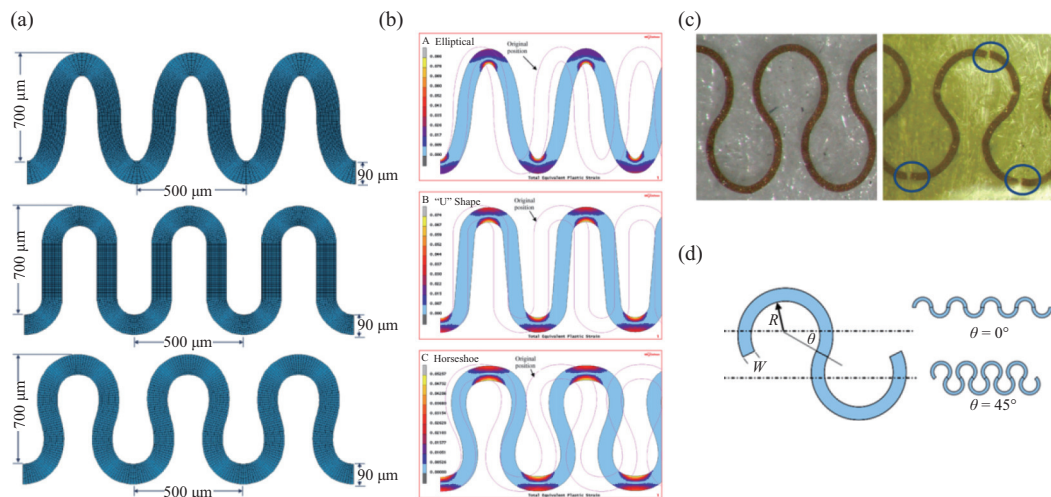


Fig. 4. (Color online) (a) Schematic illustration of different curve-shaped interconnects. (b) The deformation and pressure circumstance when the interconnected tracks are under same pressure. (c) Optical image of the fracture of the copper tracks, the position of the fracture agrees well with the prediction^[84]. Reprinted with permission, Copyright 2008 Elsevier. (d) Calculated radius and angles of the most optimal horseshoe-like structure^[85]. Reprinted with permission, Copyright 2009 Emerald.

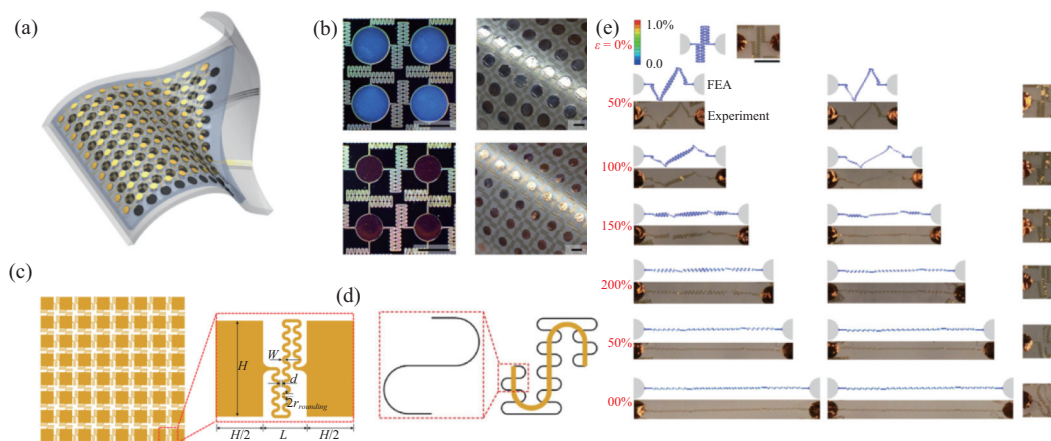


Fig. 5. (Color online) (a) Stretchable lithium batteries with self-similar serpentine interconnects structure. (b) Optical microscope images of the microstructure of the batteries^[86]. (c, Reprinted with permission^[87], Copyright 2013 Elsevier, d^[86]) Large area and magnified scheme of the mechanical model of self-similar serpentine. (e) The specific process of experimental and computational studies (FEA) of buckling physics interconnects with self-similar serpentine layouts^[86]. Reprinted with permission, Copyright 2013 Nature Publishing Group.

the self-similar structure have increased greatly. Finite elements analysis (FEA) predicted that the stretchability of the structure was 321% and made a good agreement with the experimental observations. Fig. 5(e) demonstrates the specific process of experimental and computational studies (FEA) of buckling physics in interconnects with self-similar serpentine layouts. At the same time, resistance interconnected parts also changed little when accommodating large strains. Zhang *et al.* analyzed the stretchability and durability of a high order self-similar serpentine interconnected structure by theoretical calculations, which was mentioned in the discussion of Xu's group works^[87]. They also analyzed the multilevel hierarchical structures and set up recursive formulae at different orders of self-similarity. Their analytic results agreed well with FEA, and their research findings indicated that the higher order self-similar interconnect structures can supply large stretchability to the system. The research findings and analyt-

ic models will provide lots of inspirations to the flexible electronics which require large areal coverage of active devices, such as flexible solar batteries systems and electronic eyeball cameras.

The examples mentioned above supply multitudinous beneficial inspirations to the process of flexible devices. The interconnected structure can be various if the spring constant can satisfy the requirement of the flexible structure. For instance, the spring structure and other complicated flexible structures also need to be concerned. Shang *et al.* reported a yarn-derived spring-like CNT rope which can be greatly elongated when it was stretched and endured tensile strains up to 285%^[60], and the structure is suitable to manufacture the flexible interconnects section. Of course, simpler architectures with high flexibility and stretchability will decrease the cost greatly, as they are easy to be processed and will help spread the word to the industrial world.

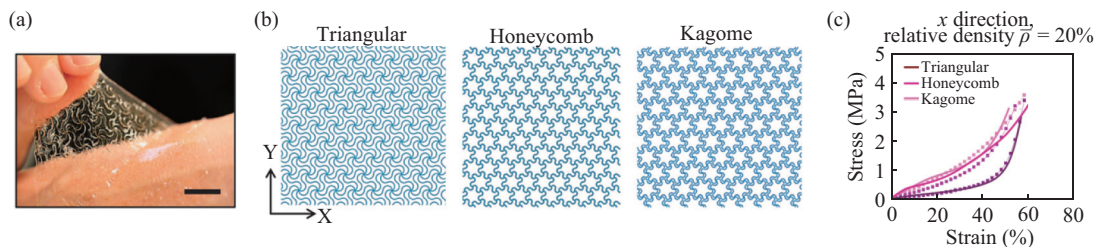


Fig. 6. (Color online) (a) A kind of mesh structure which mimics body tissue. (b) The concrete structure in (a), including triangular, honeycomb and kagome mesh structures. (c) The comparisons of three different architectures' stretchability^[88]. Reprinted with permission, Copyright 2015 Nature Publishing Group.

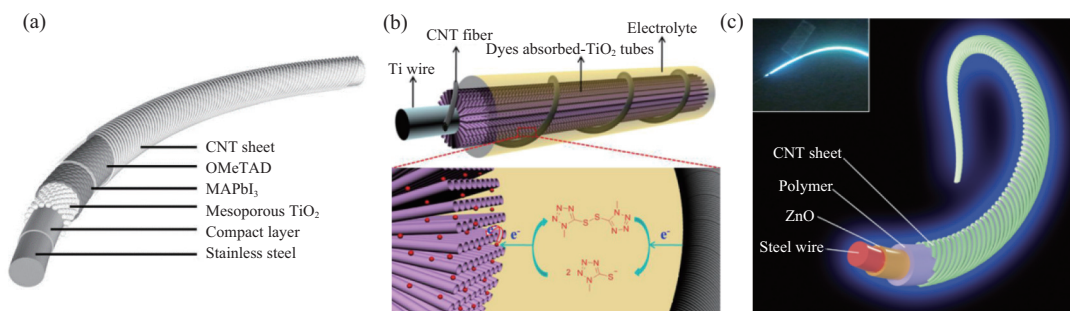


Fig. 7. (Color online) (a) The structure of a fiber-shaped solar cell^[89]. Reprinted with permission, Copyright 2014 John Wiley and Sons. (b) Schematic illustration of a photovoltaic wire^[90]. Reprinted with permission, Copyright 2013 American Chemical Society. (c) The structure of a kind of flexible fiber-shaped PLEC. Inset: photo of a fiber-shaped PLEC biased at 10 V^[92]. Reprinted with permission, Copyright 2015 Nature Publishing Group.

As is presented in Fig. 6(a), Jang *et al.* designed several mesh structures which mimic body tissue and are applied to artificial skin^[88]. Many attractive and irradiative structures in nature, such as cytoskeletons, deserve to be explored and studied. Figs. 6(b) and 6(c) illustrate three architectures with different shapes and compare their ability to withstand strains. The experimental statistics indicated that the maximal sustaining strain of the architecture is about 60%.

3.2. Flexible fiber-shaped structures

Apart from the aforementioned well-studied planar structures, researchers are also fabricating flexible or even stretchable electronics with fiber-shaped structures, such as energy harvesting and light emitting devices.

For energy harvesting, there have been many novel and advancing ideas and device structures to date. Among them, Qiu *et al.* fabricated novel fiber-shaped solar cells via a simple dip-coating process^[89]. As shown in Fig. 7(a), stainless steel with a smooth outer surface was used as the core axis and then a compact TiO₂ layer, a mesoporous TiO₂ layer, and a perovskite CH₃NH₃PbI₃ layer were deposited on the stainless steel in a layer-by-layer pattern, covered by a transparent carbon nanotube (CNT) sheet dry-drawn from a spinnable CNT array working as a cathode. With such device configuration like the one they proposed, the annealing temperature and the mesoporous layer thickness were investigated for optimized device performance and an energy conversion efficiency of 3.3% was achieved, which was not sensitive to the angle of incident light. What is more, such devices could endure harsh bending, and no obvious physical damage or de-

crease in energy conversion efficiencies was observed after bending for 50 cycles. Around the same time, Pan *et al.* fabricated an organic thiolate/disulfide redox couple for novel dye-sensitized photovoltaic wires^[90]. Fig. 7(b) illustrates the device's configuration: perpendicularly aligned dyes-absorbed TiO₂ nanotubes were grown by electrochemical anodization on a Ti wire, and aligned CNT fibers with high flexibility, tensile strength, and electrical conductivity were used as a counter electrode. The thiolate/disulfide redox couple was chosen for its weak corrosion of the current collector and negligible absorption in the visible region, and the CNT fiber was ideal for the counter electrode due to its higher catalytic activity toward organic redox couples compared with traditional Pt counter electrodes. Both of these advantages contributed to a much higher energy conversion efficiency of 7.33% than those counterparts with generally used redox couple and Pt counter electrode. By investigating device performances varying CNT fibers diameters, the optimal diameter for the CNT fiber was around 60 μm as a result of two competing factors, which are that larger CNT fibers have more catalytic sites while larger CNT fibers are less flexible to be woven into uniform twisted structures. Under bending, the *J-V* curves of a typical photovoltaic wire showed no obvious difference compared to those not bent. Besides, key photovoltaic parameters, such as the energy conversion efficiency, maintained fairly well after 100 bent cycles, which indicates a novel direction of highly efficient flexible optoelectronics.

In addition to those energy harvesting applications, fiber structures are also widely utilized in other device fabrication scenarios. For potential applications in sensing, Peng *et al.* fabricated carbon nanotube (CNT)/polydiacetylene (PDA) nano-

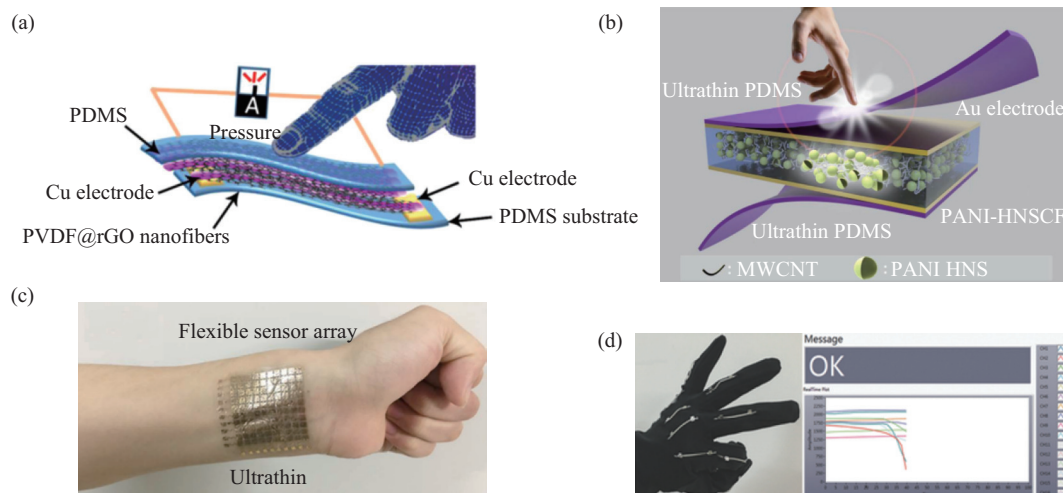


Fig. 8. (Color online) (a) The pressure sensor with PVDF@rGO nanofiber framework film^[95]. Reprinted with permission, Copyright 2016 Elsevier. (b) The structure of a kind of e-skin based on polyaniline hollow nanospheres composite films (PANI-HNSCF)^[96]. (c) The flexible sensor array of the e-skin in (b)^[96]. Reprinted with permission, Copyright 2017 Elsevier. (d) The electronic data glove and the condition when it recognizes the gesture of “OK”^[99]. Reprinted with permission, Copyright 2016 John Wiley and Sons.

composite fibers which rapidly and reversibly responded to an electrical current, with the consequential color change observable to the naked eye^[91]. Pure carbon nanotube fibers were first spun from chemical-vapor-deposition-synthesized nanotube arrays, and composite CNT/PDA fibers were synthesized by directly coating polydiacetylenic precursors onto CNTs, followed by some topochemical polymerization treatments. They further demonstrated a chromatic transition from blue to red in response to an electric current, which was also reversible for certain values of current. Due to the high mechanical strength of nanotube fibers, PDA was presented with mechanochromatism at negligible elongation, which had never been achieved in former research. Apart from electric current, the CNT/PDA fibers also responded to other environmental stimuli, including mechanical abrasion, temperature variation, and chemical and organic vapour, which leads to promising applications in sensors, actuators, and other electronic devices.

In the pursuit of smart fabric and light-emitting clothes, Zhang *et al.* sandwiched an electroluminescent polymer layer between a modified metal wire cathode and a conducting aligned carbon nanotube sheet anode using all-solution-based processes, which led to a lightweight, flexible, and wearable fiber-shaped polymer light-emitting electrochemical cell (PLEC) with tunable colors^[92]. Fig. 7(c) presents the device's configuration, where a thin layer of ZnO nanoparticles working as the electron transfer layer was first dip-coated on a stainless steel wire as the cathode. Also using a dip-coating method, the electroluminescent polymer layer, containing a blue light-emitting polymer (PF-B), ethoxylated trimethylpropane triacrylate (ETT-15), and lithium trifluoromethane sulphate (LiTf), was deposited on the modified metal wire. Finally, an aligned CNT sheet was uniformly wrapped around the wire to complete the device fabrication. For flexibility performance, the resistance of the fiber showed a humble increase by less than 6% after 1000 cycles of bending. Due to the unique fiber structure, the brightness of the luminescence was almost independent of the viewing angle, which is import-

ant for practical use of the luminance. More importantly, by simply substituting emissive polymer layers, other colors could also be realized with the same device configurations, which introduces more possibilities as to the applications of the fiber fabricated.

4. Applications

4.1. E-skins

Human skin has remarkable and complicated neuro sensor units and can adapt the body shape change while holding the sense to external environment. E-skin provides great potential applications in substituting human skins, maintaining the sensors' functions while stretching with body^[93]. Also, some of the E-skins exhibit a self-healing characteristic which highly simulates human skin. Li *et al.* reported materials with the characteristics of biological muscles—strong, elastic and capable of self-healing^[94]. They used coordination complexes to link polydimethylsiloxane chains and form a network, which contained the high stretchability and autonomous self-healing ability. The self-healing can take place at low temperature and free from the surface aging and moisture. This material showed great prospect in the application of E-skins. Despite the self-features of E-skins, recognizing various single aspects including pressure, voice, and sight are also very important in human daily life. Compared with sight and voice recognition, pressure recognition has been less investigated as the fabrication of a pressure sensors skin would demand a flexible switching matrix. Lou *et al.* used a type of natural viscoelastic property material P(VDF-TrFe) and conductive reduced GO (rGO) fabricated a kind of composite fibers. Then they manufactured a self-assembled 3D film platform, which can be applied in the highly sensitive piezoresistive pressure sensor with these fibers^[95]. Fig. 8(a) illustrates its structure clearly. The sensor exhibited outstanding performance. It had high sensitivity (15.6 kPa^{-1}), a low working voltage (1 V),

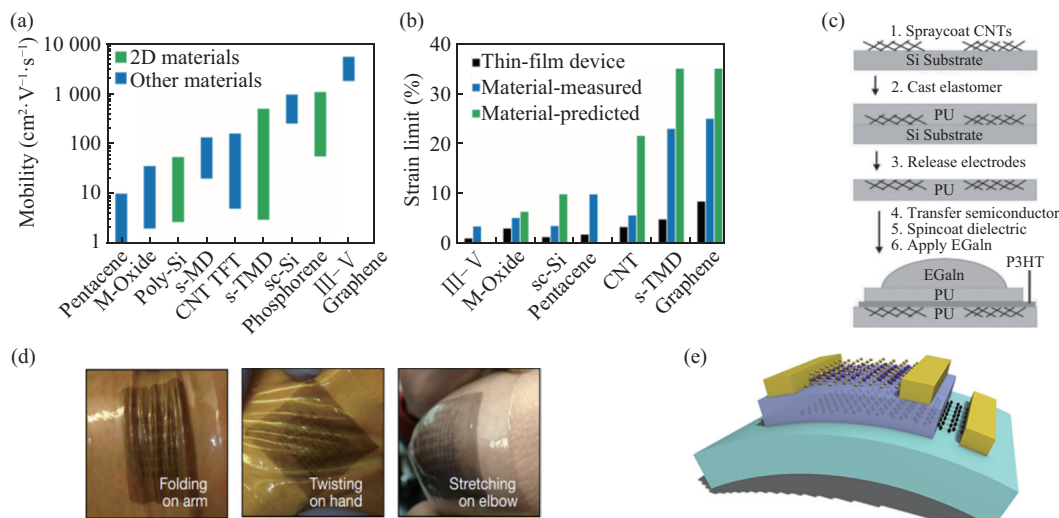


Fig. 9. (Color online) (a, b) The carrier mobility and strain limits comparison of several typical semiconducting materials including organic materials, metal oxide, traditional silicon, and two-dimensional materials^[44]. Reprinted with permission, Copyright 2014 Nature Publishing Group. (c) Schematic illustration of the process of embedding carbon nanotube into elastomer to act as the electrode^[26]. Reprinted with permission, Copyright 2014 John Wiley and Sons. (d) Optical image of skin-inspired stretchable devices using healable semiconducting polymer as active materials^[66]. Reprinted with permission, Copyright 2016 Nature Publishing Group. (e) Scheme of typical all-two-dimensional materials flexible field effect transistor^[104]. Reprinted with permission, Copyright 2013 American Chemical Society.

and detection limit (1.2 Pa). Meanwhile, the sensor can also respond rapidly (5 ms at 50 Hz) and keep stable under 100000 cycles. These experimental statistics indicated that this kind of sensor can be a good choice to be used as highly sensitive electronic skins, which can monitor human physiological signals. Later, Lou *et al.* designed another ultrathin high-performance pressure sensor, which was based on the polyaniline hollow nanospheres composite films (PANI-HNSCF)^[96]. Fig. 8(b) shows the hollow structure of the thin film, which brings it high elasticity and low effective elastic modulus (0.213 MPa). Another ingenious idea is that they mimicked natural skin and designed large ultrathin arrays of pixel high-performance pressure sensors, just as shown in Fig. 8(c)^[96]. Every single unit sensor has excellent sensing performance and can be easily integrated into sensor arrays to improve the whole device's sensing precision. Through amplifying and transforming various external stimuli to independent electrical signals in every single unit, the whole sensor device's sensing precision will be improved effectively. It enables continuous sensing of pressure and temperature with a high pressure sensitivity of 31.6 kPa^{-1} and an accurate temperature resolution of $0.08 \text{ }^\circ\text{C}^{-1}$. Because of the high sensing precision of the device, it will become a more suitable choice to manufacture e-skins. Someya *et al.* integrated organic transistors and rubber pressure sensors providing an ideal way to realize an artificial skin^[97]. The device showed a mobility of $1.4 \text{ cm}^2 \cdot \text{V}^{-1} \cdot \text{s}^{-1}$ and is still electrically functional even wrapped to a 2-mm radius. Despite pressure, flexion-sensitive and tensile-sensitive E-skins were also receiving considerable and rapidly increasing attention as the tensile was well consistent with the finger or joint motions and would be a crucial part for smart and soft wearable devices. Zhu *et al.* fabricated novel multifunctional electronic skins using aligned few-walled carbon nanotube (AFWCNT) polymer composites^[98]. The flexion-sensitive electronic skins showed a piezoresist-

ive function, high precision, linearity, and exhibited no performance degradation even after 15000 bending cycles. Chen *et al.* designed a type of electronic data glove with a highly-stretchable fiber strain sensor^[99]. They used a P(VDF-TrFE) polymer nanofibers mat and a silver nanowires layer to fabricate the fiber strain sensors. The strain sensors can detect high bend and torsion deformation and keep outstanding performance at the same time. They can respond rapidly (20 ms) and keep excellent durability after 10000 strain cycles. These admirable characteristics made it very suitable for monitoring vigorous motions such as figure bending. The electronic data gloves are based on these fiber strain sensors. Fig. 8(d) presents the configuration of the smart data glove, which was integrated with a ten-channel circuit in each joint of the fingers, and the condition that the glove recognize was the gesture of "OK"^[99]. The sensors in the circuits can detect the motions of the fingers to recognize various gestures in real time, which has significant meaning for manufacturing future intelligent wearable electronics, such as human-machine interfacing devices and virtual reality. Aside from this, this smart glove also can be applied in the robots and help them work efficiently.

4.2. Flexible field effect transistors

For flexible field effect transistors, carrier mobility and extension limits are two important parameters to evaluate the performance of flexible field effect transistors. Hone *et al.*^[44] summarized the carrier mobility and strain limits of several typical semiconducting materials including organic materials, metal oxide, traditional silicon and two-dimensional materials with the visualized comparison in Figs. 9(a) and 9(b). The carrier mobility of traditional silicon transistors is about two orders of magnitude lower than new two-dimensional materi-

als including graphene, transition metal dichalcogenides, and phosphorene, but about one order of magnitude higher than organic materials such as pentacene. The comparisons highly encourage researchers to focus on two-dimensional materials for new generation flexible nanotransistors. In the aspect of strain limits, two-dimensional materials also exhibit outstanding flexibility and the strain limits can reach about 35% theoretically. However, a gap still exists between experimentation and theory as the measured strain limit is just the 10% level. Besides carrier mobility and extension limits, optical transparency properties also need to be noticed. For two-dimensional materials, most of them show high optical transparency as the thicknesses is often limited to several nanometers.

The cracks in the stretch process largely limit the practical application of organic semiconductors. To achieve a transistor with high strain limits and effectively reduce its cracks, Chortos *et al.* reported a stretchable transistor which exhibited transistor properties even under large strains with negligible cracks^[26]. By embedding carbon nanotube into elastomer as an electrode, the semiconductors could be easily embedded between the elastomer layers, which deeply suppressed the formation of cracks (Fig. 9(c)). However, the cycling performance largely depends on the morphology change of organic semiconductors and viscous deformation of elastomers; finding a type of elastomer with minimal viscous deformation can effectively enhance the cycling performance. Also, the cracks can be cured in organic semiconducting materials. Bao *et al.* reported a concept of healable semiconducting polymer to repair the cracks generated in a stretch process using chemical moieties to promote the crosslinking of polymers^[66]. The transistors can recover the high mobility even after a hundred cycles under 100% strain, and the transistors are successfully applied to skin-inspired stretchable devices (Fig. 9(d)).

By fabricating silicon devices on a flexible substrate and reducing the dimension of silicon crystal, flexible silicon field effect transistors can also be achieved. As a typical example, Kim *et al.* reported stretchable and foldable silicon metal-oxide semiconductor integrated circuits by aligning single crystalline silicon nanoribbons on stretchable substrates^[100]. Menard *et al.* adopted a dry transfer printing process to transfer traditional single silicon objects using a rubber stamp^[61]. The microstructure silicon materials' stretchability and flexibility resulted from the stretchable feature of the plastic substrate.

Carbon nanotubes are also promising materials for flexible field effect transistors for their high carrier mobility and large tensile modules^[101]. Xu *et al.* used ion gel as the dielectric layer and semiconducting carbon nanotubes thin film as the channel to fabricate a stretchable carbon nanotube transistor. The stretchable transistors showed a superior on/off ratio of $>10^4$ and carrier mobility of $10 \text{ cm}^2 \cdot \text{V}^{-1} \cdot \text{s}^{-1}$ under low working voltage $<2 \text{ V}$ ^[36]. Chortos *et al.* employed unsorted carbon nanotubes as the gate and the electrode, and semiconductor carbon nanotube as the channel, fabricating a bottom gate and top electrode contact carbon nanotubes transistor^[35]. The resistance of electrodes only increased 60% under strain of 100%, giving a mobility of $0.18 \pm 0.03 \text{ cm}^2 \cdot \text{V}^{-1} \cdot \text{s}^{-1}$. The all-carbon-nanotube flexible field effect transistor using TPU as

dielectrics layers and substrates can present the transistors with more mechanic durability and a strain-independent feature under a limited strain range.

Two-dimensional materials such as graphene, h-BN and transition metal chalcogenides show tunable band gaps under strains and excellent mechanical properties with the thickness limited in a few nanometers^[102]. These two properties indicate that the two-dimensional materials are appropriate candidates for flexible electronic devices. Graphene and transition metal chalcogenides also possess high carrier mobility, which attracts a large amount of attention from researchers and have been studied tremendously in recent years^[103]. Owing to their attractive properties, plenty of high-performance, flexible and wearable devices emerged and are promising in the next generation of flexible transistors. A typical all-two-dimensional materials flexible field effect transistor is shown in Fig. 9(e). Owing to the high conductivity of graphene and insulating BN, graphene and BN are used as the gate electrode and dielectric, respectively^[104]. Two-dimensional MoS_2 was used as the channel, and the devices show a carrier mobility of up to $45 \text{ cm}^2 \cdot \text{V}^{-1} \cdot \text{s}^{-1}$ under the gate voltage below 10 V. As all materials of the heterostructure are flexible and highly transparent, the devices are transferred onto polymer substrate showing constant performance under up to 1.5% strain. As the substrate is flexible plastic, the gate materials used in flexible devices need to be operated at low temperature. Recently, gelation of an ionic liquid which is the so-called ion gel, possessing properties of mechanical flexibility, high ion conductivity and printability has attracted research attention intensively. Pu *et al.* reported a MoS_2 transistor where ion gel was used as the gate dielectrics. The transistors exhibited high mobility ($12.5 \text{ cm}^2 \cdot \text{V}^{-1} \cdot \text{s}^{-1}$) with a low threshold voltage ($<1 \text{ V}$), and a high on/off current ratio (10^5)^[24]. The devices showed almost no performance degradation when the flexible device was bent to a radius of 0.75 mm and this can be attributed to the high flexibility of ion gel. Benefitting from the diversity of two-dimensional materials, they can be assembled as the heterostructure at a vertical dimension, which shows many fantastic electrical and optical properties for flexible transistors. Georgiou *et al.* used two-dimensional WS_2 as an atomically thin barrier between two layers of graphene to fabricate a vertical tunneling transistor^[53]. The transistor showed unprecedented current changing over 1×10^6 with really high ON current, and it can work on transparent and flexible substrates.

Flexible two-dimensional materials transistors show good tendency and potential for the next generation of wearable devices. However, the strain range limits its practical application, and large area atomic two-dimensional thin films still need to be synthesized to create wrinkle easily to increase the stretchable range.

4.3. Flexible lithium-ion batteries

A flexible energy storage device is an indispensable part of portable electronics. For most electronic devices at present, the low energy storage ability of batteries cannot satisfy the continuously growing demand, and has become a serious problem. Meanwhile, due to the batteries taking up a

large space in proportion to the whole device, energy storage problems have been a bottleneck in the development of flexible electronics. For batteries or other energy storage devices used in flexible electronics, they not only need high energy density, but they also demand considerable flexibility, even high stretchability, bendability, and other abilities. Obviously, existing energy storage devices such as Lithium-ion batteries are rigid, which cannot satisfy the need for portable electronics. To solve the problem, a series of new flexible batteries and supercapacitors have been designed.

Lithium-ion batteries have high voltage and large capacity, which are most suitable for portable electronics. In this case, there is no doubt that enabling lithium-ion batteries flexibility has significant meaning. But to achieve the target, some problems must be handled. First of all, new flexible and bendable materials for the current collector must be explored and substituted for traditional metal materials, because traditionally used metal foils are too smooth to combine with other materials, and are hard to recover once been bent. Secondly, the widely used liquid-state electrolytes have two obvious advantages, namely good contact interface and high conductivity. But the disadvantages are also obvious, one of which is that the liquid-state electrolytes will be revealed easily when the batteries are stretched or bent. Moreover, the liquid-state electrolytes are usually poisonous, so they are not suitable for flexible batteries. Thirdly, the manufacture technique of lithium batteries must be improved. Traditional methods are spreading adhesive on the interface between the active materials and current collector to bond them. However, this kind of layer structure is not stable, which is easy to be destroyed when it is stretched or bent, and results in the degradation of the batteries' performance.

At present, using flexible active materials to take the place of copper or tin foils is the most usual method to get flexible lithium-ion batteries. Carbon nanomaterials and polymers are two kinds of materials which have been widely investigated. For carbon nanomaterials, the research mainly focused on CNTs, graphene and GO, etc.

The research on CNTs as electrode materials has received great popularity exclusively, due to their porous networks, high surface specific area, and excellent electric, and mechanical properties^[105]. Intense studies have been carried out to fabricate both flexible anode and cathode materials applying CNTs. Vacuum filtration and chemical vapor deposition (CVD) are the main methods to prepare CNTs for flexible electrodes. For example, Li *et al.* developed a novel procedure to apply the vacuum filtration method to fabricate flexible nanoporous CNTs films, which can be used as a binder-free and current collector-free anode electrode for Li-ion batteries^[106]. In their configuration, the flexible electrodes have much better mechanical strength and flexibility compared to their former counterparts due to the existence of a separator substrate, while the energy density does not see an obvious decrease. However, in this process, the thickness of the active material is dramatically limited ($\sim 1 \mu\text{m}$) and surfactants are strongly involved. The CVD method successfully gets rid of these limits.

As to polymers, due to their excellent mechanical properties and the potential to improve the batteries' mechanical flexibility, flexible solid-state polymer electrolytes are wildly

used as separators and electrolytes. Traditionally, lithium-ion batteries are designed to have liquid-state electrolytes and rigid separators, which prevent a short circuit between cathode and anode. These make the traditional structure fail to meet the requirements of flexible lithium-ion batteries, due to potential electrolyte leakage and short circuits. Lots of research efforts have been made to solve this problem and among many proposals, the idea of solid-state electrolytes is always one of the key focuses. The solid-state battery was first reported in 1969 incorporating LiI as solid-state electrolytes, which demonstrated a poor ionic conductivity at room temperature^[107]. However, such structure inspired people to integrate polymer electrolytes with a separator into a single film, which blocks electron transport but serves as an ion carrier between anode and cathode, and such efforts may potentially meet the requirements of flexible lithium ion batteries. These polymer electrolytes feature high stability against substances within lithium-ion batteries, as well as the inertial nature against corrosion, combustion, and leakage, which make them operate effectively in a wide range of temperatures with negligible self-discharge^[108]. For example, Kil *et al.* developed novel free-standing gel polymer electrolytes using an ultraviolet(UV)-cured trivalent/monovalent acrylate polymer matrix together with a LiPF_6 based liquid electrolyte, where ethoxylated trimethylolpropane triacrylate oligomers introduce strong rigidity and dimensional stability while ethylene glycol methyl ether acrylate oligomers bring mechanical softness and benign contact with electrodes. Another main research focus recently is the development of plastic crystal electrolytes (PCEs), which feature outstanding ionic conductivity at room temperature, inertial nature including thermal stability and nonflammability. Recently, work has been done on the possibility to improve their mechanical properties. For example, Ha *et al.* incorporated the semi-interpenetrating polymer network matrix to fabricate a highly bendable plastic crystal composite electrolyte (S-PCCE), which can be used in a shape conformable all-solid-state lithium-ion battery^[109]. Such obtained S-PCCE showed superior mechanical stability and had great implications on the future of battery manufacture. However, much more research is needed since the majority of solid-state electrolytes still have many disadvantages, such as low room temperature ionic conductivity and a lack of fluidity, etc. Besides, people also pick up thin polymer materials as packing materials for flexible lithium-ion batteries^[110].

Aside from these materials, some composite materials such as fibrin materials or paper also deserve investigation. These materials have extraordinary flexibility and low cost. Although they are dielectric, by composite with some conductive carbon materials, they can become conductive materials. For example, Hu *et al.* reported an integration of all components of a Li-ion battery into a single paper sheet under a simple lamination process^[111]. In such structure, the commercial paper functions as both mechanical support and Li-ion battery membrane due to its flexible texture and porous structure. Such rechargeable energy storage devices are thin, flexible, and lightweight, which present them with great potential in various applications, including RFID tags, functional packaging, and new disposable applications.

On top of materials, the shape of the batteries also needs to be redesigned. Due to the stack layer structure, it is easy to

be damaged when the batteries are bent or stretched. So researchers designed some new structure to satisfy these needs. For instance, the fiber structure is a novel and promising proposal, which has already led to unprecedented device configurations and outstanding performances. Zhang *et al.* proposed a general strategy to fabricate freestanding flexible supercapacitors and lithium-ion batteries using CNT fiber springs as electrodes, and promising electrochemical performance was achieved^[112]. To fabricate CNT fiber springs, they first synthesized spinnable CNT arrays by chemical vapor deposition. Then CNT sheets were peeled from the CNT array and further spun into CNT fibers, which could form a spring-like fiber by over twisting several CNT fibers together. The carefully aligned structure of CNTs in the spring-like fibers presented those fibers with excellent electronic and mechanical properties, including a high electrical conductivity on the level of 10^2 – 10^3 S/cm and an outstanding stretchability of over 300%. For an elastic fiber-shaped supercapacitor, two spring-like fibers coated with PVA/H₃PO₄ gel electrolyte were placed in parallel and the highest specific capacitance achieved was 18.12 F/cm³ with various electrode diameters, which was also maintained over 95% after 1000 charge-discharge cycles. All fabricated supercapacitors showed energy densities up to 0.629 m·Wh·cm⁻³ and power density up to 37.74 m·Wh·cm⁻³. In terms of the flexibility performance, the supercapacitor can be bent in any direction without obvious performance degradation, and the specific capacitance remained at 93.5% after 300 bending cycles. For stretchability, the CV curves remained unchanged and the specific capacitance remained over 90% when the supercapacitor was stretched to 100%, which was much higher than those elastomeric substrate-based stretchable supercapacitors (if the substrate was included). The spring-like fiber can also be used in lithium-ion batteries. Coated with active nanoparticles, the spring-like fiber can serve as electrodes for fiber-shaped lithium-ion batteries. LTO and LMO were introduced to the fiber to fabricate hybrid spring-like fibers, which was achieved by dipping the CNT sheet into LTO and LMO suspension. The capacity remained at 92.1% after running for 100 cycles and also remained under 85% and 100% strain.

Later on, Zhang *et al.* further refined their works to twist three carbon-nanotube-based hybrid fibers for the integration of the lithium-ion battery and the supercapacitor, seeking high energy and power densities^[113]. Within the structure they proposed, they incorporated carbon nanotube (CNT)/ordered mesoporous carbon (OMC), CNT/Li₄Ti₅O₁₂ (LTO) and CNT/LiMn₂O₄ hybrid fibers, which can be woven into various flexible electronics clothes and other device applications.

4.4. Flexible display devices

Compared with a conventional rigid panel display, the next generation of display devices have the advantage of better flexibility. This concept is getting closer to practical usage, due to the recent development of the relevant technologies, involving substrates, barrier layers, electro-optic material, and manufacturing processes. By simply substituting brittle materials and structures for deformable ones, flexible display can be realized based on present liquid crystal displays

(LCD). However, these improvements can hardly make LCD an ideal flexible display with large and highly reversible deformations. So some new technologies are raising up. Flexible organic light-emitting diodes (OLED) are promising devices that offer advantages over the LCDs, such as improved flexibility, lower power consumption, lighter and thinner, and better image quality. Unlike LCD, OLED displays are self-luminous, so structures of OLED devices are quite different from conventional or flexible LCDs. And because of the thin film structures and amorphous organic materials, OLEDs are supposed to be a natural choice for flexible displays. The whole performance of the display depends on several involved techniques, and here we summarize the recent advances in flexible transparent conductive electrodes (TCEs), light-emitting material, and manufacturing processes.

Indium-tin-oxide electrodes are successfully commercialized TCEs, but they are unsuitable for flexible devices because of their brittle properties. Hence, researchers are searching for other alternatives for ITO, some of which include conducting polymers, graphene, carbon nanotubes (CNT), metal networks or thin films, and mostly their composites^[114–115]. For example, some polymers like PEDOT: PSS often serve as a hole-injection layer for graphene, to eliminate the huge hole-injection barrier at the interface^[116]. The high refractive index dielectric layers of the multilayer improve transmittance by reducing the reflection of the metal film caused by destructive interference^[117]. By material engineering, even ITO can be incorporated into a flexible OLED, which shows ultrastability after bending over 2000 times with a critical bending radius of 5 mm by the use of a poly(ethylene-terephthalate)/ITO/Ag/ITO (PET-IAI) structure^[118]. A detailed discussion about TCEs based on different materials can be found in Ref. [119–123].

Ideal light-emitting material should have a high luminescence quantum yield, good carrier mobility, film-forming properties, thermal and oxidative stability, and color purity^[119]. The first generation of OLEDs utilized fluorescent materials, and the second generation of OLEDs was based on phosphorescent materials with much higher efficiency. Today, numerous phosphorescent emitters have been reported and mostly were organometallic complexes containing Pt(II), Ir(III), Os(II), Ru(II), Au(III), etc. The properties (quantum efficiency, emission color, charge injection/transporting property, and stability) of these phosphorescent emitters mostly depend on the organic ligands^[124]. The internal quantum efficiency (IQE) with these materials can reach nearly 100%. Thermally activated delayed fluorescence (TADF) based materials are considered as the third generation, which could perform as well as phosphorescent materials but get rid of the use of heavy metal ions^[125]. The device with light outcoupling enhancement could further improve its external quantum efficiency to over 50%.

The commercialization of the material not only depends on the performance, but also depends on the manufacturing process and the cost. OLED devices can be fabricated from organic materials by both the thermal evaporation and the solution deposition. Compared to thermal evaporation, solution processing is much cheaper without a vacuum system, but suffers from low luminous efficiency. Han *et al.* reported on

highly efficient solution-processed OLEDs that use novel universal electron-transporting host materials based on tetraphenylsilane with pyridine moieties, exhibiting the highest recorded electroluminescent at that time^[126]. On the other hand, OLED materials are quite sensitive to moisture and oxygen species, indicating that an inert flexible protective layer is required to prevent the penetration of water vapors and oxygen. Park *et al.* reported on a flexible encapsulation method called Flex Lami-encapsulation compatible to the roll-to-roll process, in which OLEDs were encapsulated in a laminar structure that consists of a thin metal foil and a rubbery polymer layer^[127]. Wang *et al.* fabricated oxides/polymer composite film as encapsulation that lead to a superior water vapor transmission rate as low as $1.05 \mu\text{g}/(\text{m}^2 \cdot \text{d})$ at 60°C and 100% RH^[128].

4.5. Summary and prospect of applications

Recent advancements in architecture design, and synthesis of advanced materials have enabled multitudinous applications in the field of flexible electronics to become a reality. These applications in medical monitoring, integrated circuit design, energy storage, and flexible display devices will revolutionize the lifestyle of modern society. In the near future, some applications in different flexible electronics fields can be combined together to manufacture some products, which will bring more convenience to our life. For instance, the E-skin monitoring device can be combined with some ultrathin Lithium-ion batteries and flexible display devices to form a system, which can achieve the target of real-time monitoring our physiological parameters, such as pulse, temperature, and respiration. In the meantime, their timely feedback will appear on the display devices of the system to help people adjust their behavior. Lithium-ion batteries will provide great power for the system to ensure its long-time normal operation. This integrated system will help people understand their bodies and prevent some accidents, such as hyperkinesia. On top of this, flexible electronic technology will bring bendable, ultrathin smart phones and other flexible intelligent devices to us. High stretchability and bendability let them have the possibility to integrate with our skin or clothes. Intelligent clothes will start a new offline era of the information society. The intelligent devices of these clothes can receive and send information though WiFi and Bluetooth conveniently, increasing efficiency of handling problems, and helping people save time and energy. Some light emitting devices can decorate these clothes and make them more beautiful. The aforementioned monitoring system also can be integrated into intelligent clothes to achieve multiple different functions. With the progressive efforts of scientists and engineers, these wonderful ideas will become a reality in our lives in the near future. These intelligent systems not only bring convenience to normal peoples' lives, but also bring hope to disabled people, helping them to live better.

5. Conclusion

This article serves as a general and albeit focused review of flexible electronics covering flexible materials, architecture designs, and specific applications. Various novel materi-

als provide numerous opportunities and choices for researchers to further push FED to their limits. Multitudinous nanomaterials contain excellent physical and chemical characteristics, which are very suitable for fabricating all kinds of flexible electronic devices. Organic materials, especially diverse polymer materials, also attract lots of attention because of their intrinsic flexibility and stretchability. However, increasing the conduct mobility of conductive polymers remains a challenging project. Multifarious flexible architectures, ranging from simple mesh structure and buckling strategy to complicated improved islands and bridges structure are also discussed. Flexibility, stretchability, and bendability of devices have all been extremely improved through these ingenious flexible architecture designs. Meanwhile, those designs enable some rigid inorganic materials, regarded before as impossible for manufacturing flexible electronic devices, to be integrated into flexible electronic products. Different and multiple application products of flexible electronics depict a bright future for us. Overall, the rapid rate of advancements suggests that flexible electronics will become a part of our future life, play a vital role in medical, energy, display, and other fields, making our lives more comfortable and convenient.

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