Flexible sensors based on assembled carbon nanotubes

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Abstract
Flexible sensors have attracted significant attention as they could be directly attached to/implanted into the body or incorporated into textiles to monitor human activities and give feedbacks for healthcare. A typical fabrication method is the direct use of intrinsically flexible active materials such as carbon nanotubes (CNTs). CNTs are generally assembled into aligned structures to extend their remarkable chemical, mechanical, and electrical properties to macroscopic scale to afford high sensing performances. In this review, we present the recent advance of CNT assemblies as electrodes or functional materials for flexible sensors. The realizations of aligned CNTs are firstly investigated. A variety of flexible sensors based on the aligned CNTs are then carefully explored, with an emphasis on understanding the working mechanism for their high sensing properties. The main attention is later paid to comparing two main categories of flexible sensors with fiber and film shapes. The remaining challenges are finally highlighted to offer some insights for future study.

KEYWORDS
carbon nanotube, flexible, sensor

INTRODUCTION
With the coordinated development of materials, devices, and engineering, flexible electronics have been increasingly popular and important.\textsuperscript{1,2} Compared with the conventional electronics fabricated on rigid and brittle substrates, flexible electronic devices can adapt to arbitrary surfaces and movements of human bodies. Especially, flexible sensors that play an important role in the field of flexible/wearable electronics are booming in the recent decade.\textsuperscript{2} Detections of external signals such as strain, stress, temperature, light, moisture, and chemical/biological species by conformal contact sensors could monitor human activity conditions in real time.\textsuperscript{3} Consequently, flexible sensors afford a broad range of applications including artificial intelligent robotics,\textsuperscript{4} in vitro diagnostics,\textsuperscript{5,6} electronic skin,\textsuperscript{6,7} human-machine interactive systems,\textsuperscript{8} and artificial neurons.\textsuperscript{9} To improve the performances of flexible sensors, a spectrum of nanomaterials such as carbon nanomaterials\textsuperscript{10,11} have been explored and utilized for the design and construction of electrode or functional materials.

Carbon nanomaterials comprising the element carbon are one of the most abundant elements on earth. Carbon nanomaterials have various nanoallotropes with different low-dimensional structures, which are zero-dimensional fullerene/carbon quantum dots, one-dimensional carbon nanotube (CNT), and two-dimensional graphene. The chemical and physical properties vary with the different structures of carbon materials. Among the various carbon nanomaterials, one-dimensional CNTs with sp² bonding and cylinder structure simultaneously show high aspect ratio (> 10⁶),\textsuperscript{12} elastic modulus (~1 TPa), tensile strength (~100 GPa),\textsuperscript{13}...
electrical conductivity (up to 200 MS/m), and thermal conductivity (3500 W/mK). Due to their remarkable properties and advantages, CNTs have been widely explored for flexible sensors. Displacement of CNTs into the matrix accounts for a typical strategy for the construction of flexible sensors. Owing to the strong conjugation within the large-sized conjugated structure, CNTs could generate \( \pi-\pi \) stacking with other conjugated materials and thus enable an easy and compact combination. The performances of these sensors mainly depend on the properties of matrix materials, where CNTs generally act as conductive and reinforced components. With the aim to fully extend the advantages of excellent properties of individual CNTs to macroscopic scale and to expand their application areas, CNTs have been extensively assembled into aligned structures with macroscopic architectures.

Due to the resulting remarkable electrical and mechanical properties, the assembled CNTs are promising for various applications including flexible sensors, actuators, energy conversion and storage devices (such as solar cells, lithium-ion batteries, and supercapacitors), and implantable electronics. Therefore, the aligned CNT for sensing applications forms the central theme of this review.

In this review article, we focus on a variety of sensors from aligned CNT sheet and fiber as electrodes and active materials in comparison to CNT network film. The preparations of aligned CNT sheet and fiber are first compared with CNT network film. Several typical kinds of flexible sensors, such as strain and pressure sensors, electrochemical sensors, solvent/vapor sensors and light sensors, are then carefully investigated with an emphasis on construction strategy to reveal the influence of structure on sensing property. The main challenges and future directions are finally highlighted for flexible sensors based on the CNT materials.

PREPARATION OF ASSEMBLED CNTs

Preparing CNT network film by vacuum filtration, that is, filtering a CNT suspension over a porous filter membrane followed by rinsing to remove the surfactant, represents one of the simplest methods to date. The resultant CNT network film can be transferred onto various substrates or functional materials for the construction of flexible sensors. For example, a transparent CNT film could be transferred onto quartz, a sapphire, or a Mylar substrate by dissolving a filter membrane on them. In addition, there are also other simple methods such as using the CNT suspension or ink through drop-drying, inkjet printing, slow evaporation, and spray deposition. The key of the fabrication lies in the homogenous dispersion of CNTs, which can be enhanced by sonication, surface functionalization of CNTs, or the addition of surfactants. It is obvious that the as-prepared CNT network film is typically non-aligned. The use of CNT network films can contribute to the capability to sense stress and strain in multiple directions due to their isotropic structures.

However, due to the formation of a lot of contact points among CNTs, the electrical conductivities of CNT network films are relatively low (<1 MS/m), and the charges also have to experience long pathways during transport. In addition, the building CNTs tend to separate from each other under drawing, and the tensile strengths of CNT network films are lower than 1 GPa. Both electrical conductivities and tensile strengths are much lower than individual CNTs (electrical conductivity: \( \sim 200 \) MS/m, tensile strength: \( \sim 100 \) GPa), which is unfavorable for high sensing properties. These properties may be further improved by the formation of the aligned structure, which in turn, enhance the performance of sensors based on CNTs.

CNT sheet

Compared with CNT network films, the aligned CNT sheet generally exhibits better electrical conductivity (1.3–3.2 MS/m). The aligned structure of CNTs may be induced by applying external forces, such as electric and magnetic fields and mechanical shearing during the formation process of CNT network films. The above-mentioned solution-processing methods operated at room temperature can be easily integrated with other preparation techniques such as jet printing. However, the preparation of uniform CNT dispersion normally requires the addition of surfactants and pre-treatments such as strong acid or ultrasound. These treatments greatly damage and shorten used CNTs and thus degrade both their electrical and mechanical properties that are dependent on their lengths. In addition, the undesirable residual organics also reduce their electrical and mechanical properties.

Apart from the solution-processing method, the CNT sheet can be directly drawn from a spinnable CNT array grown on a silicon substrate with pre-deposited catalysts. The synthesis of CNT film is reproducible and scalable. However, based on our own experience, it is pretty challenging to synthesize such spinnable CNT arrays that are required to have suitable packing density, thickness, alignment, and clean surface. Although the fundamental mechanism of drawing continuous film remains not very clear yet, the interactions among CNT bundles at ends are believed to play a key role in the formation of continuous CNT sheets. It is still difficult to determine the critical values for such interactions. If they are too low, the neighboring CNTs could not be drawn out to form the desiring continuous structure. If they are too high, the whole CNT array would be peeled off from the substrate as a whole.

CNT sheets can also be obtained from CNT aerogel during a floating catalyst chemical vapor deposition process. Carrying reactant gas such as hydrogen and catalyst like ferrocene and thiophene, \( n \)-hexane or ethanol as carbon source was injected into reacting furnace heated to the pyrolysis temperature of carbon source. Compared with the direct spinning method from the CNT array, the floating chemical vapor deposition process merits a much lower cost because it can be easily scaled up for industrial production.

CNT fiber

The preparation strategies of CNT fibers mainly include wet spinning from CNT solution, dry spinning from CNT aerogel synthesized by floating catalyst chemical vapor deposition, and dry drawing/spinning from spinnable CNT array grown on substrates with pre-deposited
FIGURE 1 Various applications of flexible sensors based on carbon nanotube (CNT) assemblies. Inset figures show specific examples. “Motion detection.” Reproduced with permission: Copyright 2018, Wiley-VCH. [30] “Arm movement detection.” Reproduced with permission: Copyright 2016, American Chemical Society. [31] “Hands movement detection.” Reproduced with permission: Copyright 2013, Springer Nature. [32] “Torsion sensor.” Reproduced with permission: Copyright 2016, American Chemical Society. [33] “Human-machine interaction.” Reproduced with permission: Copyright 2017, Royal Society of Chemistry. [34] “Wearable ultraviolet (UV) detector.” Reproduced with permission: Copyright 2018, Wiley-VCH. [35] “Remote electric switch.” Reproduced with permission: Copyright 2012, Wiley-VCH. [36] “Smart windows.” Reproduced with permission: Copyright 2015, Wiley-VCH. [37] “Neural probe.” Reproduced with permission: Copyright 2020, Royal Society of Chemistry. [38] “Wireless health detection.” Reproduced with permission: Copyright 2018, Wiley-VCH.

catalysts (Figure 3). [52] Each strategy shows its advantages and disadvantages, depending on the requirements of specific applications. [53]

The preparation of CNT fibers started from the solution-spinning method in 2000. [51] Figure 3A shows a typical process that started by dispersing CNTs in aqueous solutions with surfactants such as sodium dodecyl sulfate. The CNT-containing solution was then injected through a syringe into polyvinyl alcohol solution. After washing and drying the as-prepared gel fiber to remove surfactants and polymers, a CNT fiber was obtained with electrical conductivity of 10 S/cm and tensile strength of 300 MPa. [51] Compared with the other methods, a variety of CNTs with different sizes may be made into fibers by such a solution method, and it is easier to scale up with lower cost through the well-established chemical fiber technology. However, it is more difficult to disperse relatively longer CNTs in aqueous solutions, and the use of shorter CNTs produces poorer electrical conductivities and tensile strengths. Compared with short CNTs (~0.5 μm), longer CNTs (3–7 μm) showed much higher electrical conductivities (enhanced from 0.5 to about 8.5 MS/m) and tensile strengths (enhanced from 0.11 to 2.4 GPa). [54, 55]

CNT fibers had also been easily produced from CNT aerogels synthesized by floating catalyst chemical vapor depo-
sition (Figure 3B). [50, 56] The shrinking of CNT aerogels by water effectively decreased the distances among CNTs to form the fiber shape, and the drawing force induced the alignment of them along the drawing direction. Compared with the solution method, it is also easy to scale up but with a much stricter synthesis that required much higher cost, and multi-walled CNTs have been mainly produced on the basis of current technologies. The electrical conductivities (~2 MS/m) and tensile strengths (3–5 GPa) of these fibers are relatively higher as compact CNT assemblies had been designed to build the fiber. [57, 58] With further iodine doping, the conductivity could reach 6.5 MS/m. [59]

A CNT sheet from a spinnable CNT array can be twisted into a continuous fiber (Figure 3C). [52, 60] As the building CNTs showed typical lengths of hundreds of micrometers and were highly aligned in the array, the resulting fiber showed electrical conductivities of around 1 MS/m [61] and tensile strengths of up to 3.3 GPa. [60, 62, 63] However, it is very challenging to synthesize spinnable CNT arrays, and the synthesis is also a time-consuming process. In addition, the size of the CNT array is very limited, generally ranging from millimeters to several centimeters, which directly determines the lengths of resulting CNT fibers. In other words, it is difficult
to scale up this synthetic process, and the cost is much higher than the previous two methods.

As discussed above, compared with the conventional networked structure, the aligned structure offers CNT materials with both enhanced electrical conductivity and tensile strength. The aligned CNT fiber had been well explored and further reviewed also over many commercialized materials like polymer fibers and metal wires in electrical and mechanical properties (Figure 4). Flexible fiber sensors from aligned CNT fibers share many advantages including light weight, high surface-to-volume ratios, breathability, permeability, comfortability, and suitability for integration into smart textiles that are booming in recent years.

STRAIN AND PRESSURE SENSORS

The response to mechanical stimuli such as strain and stress has been widely studied for a variety of applications. For instance, strain and pressure sensors are extensively explored for wearables to detect various motions on human skins. The performances of flexible strain sensors are estimated from several aspects including sensitivity, stretchability, linearity, strain/pressure range, stability, and hysteresis. Different parameters are suggested for different application scenarios, for example, high stretchability that is endurable of large strain is required for the detection of large-scale motions like joint movements and bending movements of hands, arms, and legs, while high sensitivity to a small strain is significant in detection of small-scale motions such as subtle movements in the chest and neck during breathing, swallowing, and speaking.

Based on different working mechanisms, it is effective to design a spectrum of flexible strain and pressure sensors such as resistive and capacitive sensors. Accordingly, they convert external deformations into the corresponding change in resistance and capacitance, respectively.

Resistive sensors

The strain or pressure resistive sensor relies on the change of resistance, that is, mechanical stimulus induces a change of its resistance. The resistive sensors generally share a single electrode structure compared with the capacitive ones comprising several electrodes. Therefore, they show the advantages of simple fabrications and measurements and broad application areas.

For strain sensors, sensitivity is a very important parameter. Sensitivity can be quantified by gauge factor (GF) and defined as:

\[ GF = \frac{\Delta R / R_0}{\Delta \varepsilon} \]

which is the relative change in resistance (\(\Delta R / R_0\)) per unit of applied strain, in which \(\Delta R\) and \(R_0\) are the change of...
resistance and the initial resistance, respectively; $\Delta L$ and $L_0$ are the change of length and the initial length, respectively; $\varepsilon$ refers to strain.

According to the above formula, the sensitivity is highly dependent on the resistance behavior. Therefore, the resistance response of the CNT assemblies upon mechanical deformation is key and should be carefully investigated. The electrical resistance of CNT assembly depends on the combination of conductive network pathways and contact resistances among neighboring CNTs. For the CNT sheet, here the contact resistance can be divided into two parts, that is, contact resistances between two neighboring aligned CNTs layers in the thickness direction and among paralleled CNTs in the width direction.

As individual CNTs are rigid, the resulting CNT assemblies typically show low elongations upon breakage (failure strain of less than 5%).\textsuperscript{67} For instance, aligned CNT fibers themselves can act as fiber strain sensors because their electrical conductivities demonstrated a linear relationship upon the increasing strain.\textsuperscript{68,69} However, it can only be used to detect subtle deformations (<10% strain) with low sensitivity, poor restorability, and low GF value (GF < 2).\textsuperscript{70} Therefore, to further enhance the flexibility and stretchability of strain sensors, effective strategies were proposed in the fabrication of stretchable sensors by preparing stretchable conductors with varied electrical conductance under large strains.\textsuperscript{71} Generally, there are two typical methods to enhance the strain range, that is, design of stretchable structure based on pure conductive materials and preparation of intrinsically stretchable conductive materials.

The assembled CNTs can enhance the strain range by the design of specific architectures that offer them stretchable, for example, linear, zigzag, wavy, serpentine and Greek-key patterns,\textsuperscript{72} helical micro-structures by overtwisting CNT fibers,\textsuperscript{18,73} and restorable entanglements by twisting CNT fibers.\textsuperscript{73} However, in the case of CNT fibers with stretchable architectures, they present poor sensitivities with small GF values ranged from 0.03 to 1.13 due to the low relative displacement among neighbouring CNTs, which results in an unobvious change in resistance of the CNT fiber during stretching. For this strategy, the slippage of CNTs in the fiber was limited during stretching.
Stretchable sensors can also be achieved by incorporating CNTs with intrinsically elastic materials. The flexibility and low Young’s moduli of the incorporated elastic materials could effectively provide stretchability for the resulting sensors. To date, various elastic materials have been designed into stretchable sensors such as silicone rubber,[75] epoxy,[76] poly (methyl methacrylate) (PMMA),[9] and polycarbonate (PC)-urethane resin.[31] It is noteworthy that the strain sensors based on aligned CNT sheets normally delivered higher strain endurances compared with those from CNT network films, as shown in Table 1.[64,65,77] CNT networks are more vulnerable to stretching deformations because they generally suffer from more easily breaking into fragments than aligned CNT assemblies. When stretched, the networked CNTs tend to rearrange along the load direction, and slippage simultaneously occurs among them with poor mechanical properties. Therefore, the strain sensors based on aligned CNTs exhibited higher strain endurances compared with those based on networked CNTs.[64,65,77] A sensor with strains up to 200% and short sensing delays of less than 15 ms could be achieved from aligned CNT sheets spun from the CNT array.[31] During the stretching process, gaps were formed in the CNT sheet to favor the elongation of the integrated devices (Figure 5A). The CNT bundles in the gap could form a porous structure and maintain electrical pathways for conduction. Note that the highly aligned CNT sheets prepared from CNT arrays showed more suspended CNT bundles bridging the gaps compared with those synthesized by the other methods, preventing their rupturing. For instance, a strain sensor from aligned CNT sheet on polydimethylsiloxane (PDMS) matrix showed only 50% strain, where the aligned CNT sheet was prepared onto a glass slide by a sliding coating method.[177] When CNT sheets were attached to the longitudinal pre-stretched Ecoflex substrate, the resulting strain sensor exhibited ultra-high tolerance to strains along the CNT longitudinal axis, even for strains greater than 900%.[78]

Besides, the strain sensitivity of aligned CNT-based sensors is strongly dependent on the deformation direction. Normally, strain sensors can produce high strains and rapid responses to stretching in both parallel and perpendicular directions relative to the CNT-aligned direction. However, the GF value was higher along the parallel direction than that along the perpendicular direction (Figure 5B).[79] For instance, for a strain sensor from an aligned CNT sheet on PDMS substrate, the GF value along the CNT-aligned direction (461 at a strain of 260%) can be hundreds of times higher than that along the perpendicular direction (3.28 at a strain of 400%).[80] When the CNT sheet was stretched along the perpendicular direction, the CNTs tend to slip at the joints, resulting in a rough fracture surface, damage, and rupture of the CNT sheet.[67] While the entanglement and interaction among adjacent CNTs were weak, leading to low variations in resistance along the perpendicular direction. In addition, a much lower initial resistance can be obtained along the CNT-aligned direction. The sensitivity of the strain sensor can be tuned by adjusting the aligned direction of CNTs, for instance, with a certain degree of offset to the pre-stretching direction of the substrate (Figure 5B).[79]

Similarly, the use of elastic polymers as stretchable units also represents an effective route to enhance the strain range of fiber sensors. Polymers, such as polyvinyl alcohol (PVA), can be coated onto the surface of CNT fiber to form a core-sheath structure, increasing the interactions among neighboring CNTs. The GF was slightly increased (2.36) compared with the pure one (1.64) but still with the problem of low strains (<15%).[81] This phenomenon came from the low thickness of the polymer layer. With the production
TABLE 1 Comparison of various strain and pressure sensors based on assembled carbon nanotubes (CNTs) (including CNT sheet and fiber) and CNT network film

| Materials       | Structure                        | Response time (ms) | Strain (%) | GF  | Ref. |
|-----------------|----------------------------------|--------------------|------------|-----|------|
| CNT sheet       |                                  |                    |            |     |      |
| CNT/PDMS       | Aligned                          | 14                 | 280        | 0.8 | [64] |
|                 | CNT network                       | –                  | 5          |     |      |
| CNT/Ecoflex     | Gradient                         | 33                 | > 550      | 13.5| [65] |
|                 | Aligned                          | –                  | 270        | 0.85|      |
|                 | CNT network                       | –                  | 40         | 175 |      |
| CNT/PDMS       | Aligned                          | 98                 | 400        | 0.12| [66] |
| CNT/PCU        | Aligned                          | 15                 | 200        | >10 | [31] |
| CNT/PDMS       | Aligned                          | –                  | Parallel: 59 |     | [77] |
|                 |                                  |                    | Vertical: ∼1|     |      |
| CNT/Silicon rubber | Anisotropic, prestretched          | –                  | Parallel: 400 |     | [79] |
|                 |                                  |                    | Vertical: 380 |     |      |
|                 |                                  | –                  | Parallel: 460 |     | [80] |
|                 |                                  |                    | Vertical: 3.28 |    |      |
| CNT/PDMS       | Leaf-templated                    | –                  | 44         | 22.6| [85] |
| CNT/ Ecoeffick | Vertically aligned                | –                  | 145        | 42.3| [90] |
| CNT network     |                                  |                    |            |     |      |
| CNT/Ecoflex     | Sandwiched                        | 1700               | 400        | 70  | [75] |
| CNT/PDMS       | Pattern array                     | 90                 | 80         | 2.26| [84] |
| CNT/ Ecoeffick | Acid-interface engineering        | –                  | >100       | 1665.9| [89] |
| CNT/PDMS       | Shadow-mask                       | –                  | 100        | 0.99| [92] |
| CNT fiber       |                                  |                    |            |     |      |
| CNT             | Aligned                          | –                  | 1          | 0.38| [69] |
| CNT             | Aligned                          | –                  | 14         | 2   | [70] |
| CNT             | Helical fiber                     | –                  | 25         | 0.14| [74] |
| CNT/Ecoflex     | prestretched                      | 105                | 960        | 64  | [78] |
| CNT/PA         | Core-shell                        | 12.2               | 2.36       |     | [81] |
| CNT/Silicon elastomer | Core-shell | 200                | 300        | 1378| [82] |
| CNT/PDMS       | Micro wire                        | –                  | 15         | 1 × 10⁵ | [88] |

Abbreviations: CNT, carbon nanotube; GF, gauge factor; PCU, polycarbonate-urethane; PDMS, polydimethylsiloxane; PVA, polyvinyl alcohol.

of a thicker polymer layer, the performance, especially the stretchability of fiber strain sensors, had been further enhanced.[82] For instance, a coaxial composite fiber with CNTs as the core and a thick thermoplastic elastomer as the sheath was prepared by a wet-spinning method (Figure 5C).[35] The strain had been improved to 100%, and the resistance was linearly increased (GF = 425 at 100% strain) (Figure 5D).

Nano/microstructure engineering that can effectively tune the GF and stretchability of the sensors represents a promising strategy to further improve their performances. A typical paradigm of nano-/microstructure engineering was to design and optimize polymer substrates, such as the formation of waving pattern,[83] micropillar array pattern,[84] leaf molding pattern on polymer layer,[85] or forming 3D porous structures by a porous photosensitive insulation layer between two electrodes.[86] Another way to form the nano-/microstructures in strain sensors is the creation of microcracks in macroscopic CNTs during preparation. The assembled CNTs with cracks can be introduced and controlled by stretching,[87,88] strong acid etching (such as sulfuric acid) on Ecoflex substrates,[89] or embedding the perpendicularly aligned CNTs in silicone elastomer.[90]

Besides the stretching stimulation, a fiber sensor could also be used to detect the rotational torsion.[91] The sensor was fabricated by wrapping aligned CNT sheets onto the surface of a rod with a predetermined and fixed wrapping angle, and the torsion could be measured up to 400 rad/meter. The fiber sensor could be fully integrated into the fabric using an embroidery machine, which will provide flexibility and air breathability in long-term wearable monitoring. The integrated textile sensors provide new opportunities for the development of next-generation wearable electronics.

### Capacitive sensors

Resistive sensors can be easily fabricated, and it is also simple to operate them during use. However, they suffer from poor long-term stabilities, slow responses, and unstable GF values. Compared with resistive sensors, capacitive sensors generally demonstrate superior transient responses. In
capacitive sensors, the resistances of electrodes show no obvious changes when stretched; instead, the electrodes are placed on both sides of an elastomer sheet, whose Poisson deformation is measured by the change of capacitance between the two electrodes.\[92\]

As a typical paradigm, a film capacitive sensor was made from two layers of CNT sheets as two electrodes that sandwiched a dielectric layer (Figure 6A).\[92\] The dielectric layer can be composed of Ecoflex silicone elastomer\[7\] or PDMS.\[26\] The resulting capacitive sensor was transparent and stretchable in the strain range of 0%–50%. The stretchability was further enhanced to over 100%, or even up to 300% by pre-stretching the polymer substrate, followed by attaching CNTs to it.\[26,93\] Alternatively, by pre-stretching the dielectric layer in omnidirections, a strain sensor in response to strain increase in omnidirections was achieved. It enabled reversible inflation/deflation volumetric strains up to 7470%.\[94\]

However, there remain challenges for capacitive sensors. The capacitance changes of capacitive sensors were generally low at the level of pF, which was unfavorable for the accurate detection.\[93\] In addition, the capacitive sensors exhibit low
sensitivities. The sensitivity of the capacitive sensor based on a simple parallel-plate model was close to the theoretical limit for an elastomeric parallel plate capacitor, which predicted a GF of 1 (Figure 6B). \(^{26}\) To be noted, the GF for capacitive sensors is defined as: \[ S = \frac{d (\Delta V/V_s)}{d P_A} \] (2)

where \( V_s \) is the saturated voltage, \( \Delta V \) is the relative change in the voltage, and \( P_A \) is the used pressure.

Similar to resistive sensors, fiber capacitive sensors are promising to sense the motion of torsion, which is difficult for their film counterparts. A single rectangular strip capacitive sensor was fabricated with a silicone rubber dielectric layer between two aligned CNT electrodes (Figure 6C). \(^{32}\) The sandwich-structured fiber generated approximately 26% capacitance change during a giant twist (1700 rad/m or 270 turns/m) (Figure 6D). The sensing performance of the fiber capacitive sensor was better than the fiber resistive one. \(^{91}\)

### Self-powering sensors

For most sensors, such as resistive and capacitive sensors discussed above, an extra power source was required to provide the electrical signal input. Therefore, they generally cannot work independently. In this case, self-powering sensors that combine functions of power supply and sensing into one device are more promising for real-world applications. There are various mechanisms for the construction of self-powering sensors based on CNTs.

The self-powering strain/stress sensors are operated mainly through triboelectricity and piezoelectricity, that is, the coupling effect of triboelectrification and electrostatic induction effect between two different material surfaces, in which the input energy comes from mechanical energy. They could generate electric charges when receiving external pressure and the other stimuli and further transfer these charges to electrical signals. To judge the performance of the self-powering sensor, the sensitivity can be calculated as follows: \(^{95}\)

\[ S = d (\Delta V/V_s) / d P_A \] (2)

where \( V_s \) is the saturated voltage, \( \Delta V \) is the relative change in the voltage, and \( P_A \) is the used pressure.

There are four working modes of sensors for triboelectric generators, including single-electrode, vertical contact-separation, lateral sliding, and free-standing triboelectric-layer modes. \(^{96}\) Among them, single-electrode-based triboelectric devices are most suitable for constructing self-powering and skin-compatible sensors because of their simplicity, broad material choice, and easy access. \(^{97}\) This type of device consists of a frictional layer and an electrode layer, with human skin being another friction layer. Here, CNTs are usually used as flexible electrodes deposited on the substrate with frictional materials on its other side. The frictional materials can be PDMS \(^{98}\) or porous silk, \(^{95}\) with polyvinylidene fluoride nanofiber \(^{99}\) acting as a negative triboelectric layer. The usage of porous substrate contributes to the fabrication of a free-standing sensor with air permeability. For instance, a sensor was prepared by embedding CNTs between electrospun silk nanofiber mats, which showed a high-pressure sensitivity of 0.069 kPa\(^{-1}\) with the pressure ranging from 1 to 14 kPa (Figure 7A). \(^{95}\) The working mechanism is shown in Figure 7B, with the CNTs embedded silk nanofiber mats as one friction layer and skin as the other friction layer. However, the CNTs were spread onto the nanofiber layer with a paintbrush, resulting in a poor interaction with the substrate. In addition, the disadvantage of single-electrode-based triboelectric sensors lied that their performance was sensitive to the environment such as humidity, injury, and contamination.
on the finger, as the contact electrification was a very sensitive interfacial phenomenon. Especially with the increasing humidity, the performance of the sensor obviously decreased, which was caused by the screening effect of water molecules.

To this end, the self-powering sensor based on contact-separation mode was fabricated into a fiber shape with a coaxial structure, and it showed higher stability under various mechanical deformations, especially under bending and stretching. In addition, the performance of self-powering sensors based on contact-separation mode was not affected by the environment. In this mode, an air gap between two electrodes needed to be formed by inserting a spacer or constructing an arch structure. The coaxial fiber triboelectric nanogenerator was fabricated by using aligned CNTs as inner and outer electrodes and designing porous structures in triboelectric polymers of PDMS and PMMA (Figure 7C). The usage of the CNT sheet contributed to a seamless contact with the fiber substrate due to the flexibility and continuously aligned assemblies. An air gap between PMMA and PDMS was generated by dissolving the microsphere particles between these two triboelectric polymer layers for an effective contact and separation of the triboelectric materials. The sensor was flexible and stretchable, and it sensed diverse mechanical stimuli such as pressing, bending, twisting, stretching, and vibrating (Figure 7D). Note that the CNT assemblies usually act as electrodes. While it could also simultaneously act as a negative triboelectric layer when aluminium was used as the top electrode layer.

CNT thin-film transistor-based sensors

Other than above-mentioned transduction mechanisms including resistance variation, capacitance variation, and piezoelectricity/triboelectricity effect, another way for the sensing of pressure is the fabrication of thin-film transistor (TFT) as an electronic switch with CNT films as active materials. By covering with a pressure-sensitive rubber, the CNT TFT could function as a pressure sensor with high response speed. The TFT that responded to input pressure could be integrated into a large area array, and each transistor functions as a pixel for the stressor detection. Typically, semiconducting CNTs with high purity were required for the fabrication of high-performance TFT. Due to the flexibility of CNT network films, large-area flexible pressure sensors based on CNT TFT array had been fabricated. And the resulting TFT with highly purified (<99.99%) semiconducting CNTs showed high accuracy in detecting arbitrary shape on both flat and curved surfaces. A higher purity (99.997%) could further enhance the TFT performance, with higher mobility, lower operation voltage, and higher stability. The advantage of using TFT pressure sensors compared with above-mentioned mechanisms lies that it has a high spatial resolution, which depends on the channel length in the TFT. With further reducing the channel length, the CNT TFT array is expected to show higher density and higher spatial resolution. However, the CNT films for TFT are mainly highly purified CNT network films, instead of aligned structure because of the difficulty in the fabrication of aligned CNT assembly with highly purified semiconducting CNTs. It is expected that the aligned CNT assembly for TFT will further enhance the TFT performance.

ELECTROCHEMICAL SENSORS

Apart from the health-relevant motion of the human body, the chemical constituents from the human body can provide more comprehensive and accurate biochemical information for monitoring health and environmental conditions. Electrochemical sensors, as one of the most important tools for
detecting biochemical signals, are increasingly popular due to their capability to provide useful insights into the health of individuals. Electrochemical sensors may provide real-time, quantized, and diversified health information through measurement of chemical constituents, which are essential in tracking chronic diseases or abnormal/unforeseen situations, such as diabetes and hyperthyroidism.

Electrochemical sensors based on assembled CNTs have great potentials for applications in wearable or implantable biomedical devices due to their high electrical conductivity, high surface area, flexibility, and good chemical stability. Electrochemical sensors usually include two-electrode architecture with working and reference electrodes or three-electrode architecture with an additional counter electrode. Assembled CNTs often serve as working electrodes that determine the performances of electrochemical sensors. Besides acting as flexible electrodes to measure electrochemically active substances involving oxidation-reduction reactions at the electrode,[109] the assembled CNTs also provide a flexible substrate to bond bioreceptor serving as functional units toward electrochemical biosensing. In this case, post-treatments of CNTs are necessary to further enhance their properties for high sensing performances. One typical method is carboxyl functionalization of the CNTs via acid treatment. Carboxyl-functionalized CNT arrays can covalently link glutamate dehydrogenase on the surface for glutamate electrochemical sensor.[110] Another approach is the post-functionalization of CNTs with other materials. For instance, CNT films or fibers were treated with 1-pyrenecarbonylic acid NHS ester or bovine serum albumin to immobilize glucose oxidase enzyme.[111,112] or only incorporated transition metals and their oxides (for example Cu, Cu2O and NiO) nanoparticles as catalysts to fabricate a nonenzymatic electrochemical sensor in order to detect glucose.[113–115]

Compared with the point-of-care flexible electrochemical sensors, implantable electrochemical sensors enable precise measurements of vital activities. A notable advance has been made in developing fiber electrochemical sensors based on CNT fibers, playing an important role in the fabrication of implantable flexible electrochemical sensors with a strong and stable fiber-tissue interfaces. Pure CNT fiber was regarded as an efficient tool for stimulation of neurons and recording single-neuron activity without additional surface treatments.[116] Especially, the tissue contact impedances of CNT fibers were remarkably lower than those of state-of-the-art metal electrodes. Normally, CNT fibers were wounded around the tip of a microneedle for implanting the fiber into the nerve.[117] To further simplify the implantation process, a fiber electrode with alterable elastic moduli was developed (Figure 8A).[27] The electrode based on CNT fiber exhibited as rigid as metal wires to ensure facile and direct implantation and turned as soft as brain tissue after implantation to ensure dynamic and stable interfaces (Figure 8B). It delivered a stable neuron recording for 4 weeks in vivo. In addition to electrical stimulation and recording, CNT fibers had been successfully used as sensors in determining the presence of electroactive analytes, such as dopamine[118] and ascorbic acid.[119] Through injection of the working electrodes into the tissue, the real-time quantification of the target analyte in vivo was established. However, besides the flexible CNT fiber working electrode, the electrochemical measurements still need to use other rigid electrodes such as Ag/AgCl as the reference electrode or platinum wire as the counter electrode, which induce the modulus mismatch of the sensors with soft tissue. Recently, a fully flexible fiber implantable electrochemical sensor based on CNT fibers as core substrate was generated to realize real-time detection of multiple biomarkers simultaneously in vivo (Figures 8C,D).[120] The electrochemical sensor showed a stable fiber-tissue interface and good biointegration after injection into tissue using a syringe, offering a robust tool for long-term sensing applications. It provided the real-time monitoring of H2O2 in solid tumors and Ca2+ and glucose concentrations in blood (Figure 8E).

Significant endeavors are underway toward the development of noninvasive electrochemical sensors. The noninvasive electrochemical sensors usually use biofluids, such as sweat, saliva, or tears, that can be readily collected noninvasively and contain a plethora of analytes. For example, the first proof-of-concept demonstration of a wearable platform for real-time health monitoring, to our knowledge, was made by weaving different kinds of sensing fibers based on CNT fibers as the building blocks (Figure 8F,G).[29] The concentrations of glucose, Na+, K+, Ca2+, and pH were tested by the sensing fibers which were constructed by coating active materials onto CNT fibers to form a coaxial structure (Figure 8H). This report opens a new research direction for wearable sensors, which may be commercialized for diversified applications in the near future.

In addition to flexibility, the stretchability of electrochemical sensors is another important issue for the integration with biological tissues because of the soft and stretchable nature of biological tissues. Stretchable electrochemical sensors have been reported for the real-time detection of analytes by depositing CNT assemblies on a stretchable substrate.[121,122] Sensing fibers comprising Pt-containing CNT-sheath and rubber-core can provide amperometric electrochemical sensors for glucose, whose response was unaffected under stretching by 45%.

Despite the above advance, due to the fact that the concentrations of biomarkers in sweat, saliva, and tears are relatively low, the sensitivity and reliability of carbon-based flexible electrochemical sensors needs to be further improved, which may be realized by functionalizing or modifying carbon materials with more active sites. Besides, simultaneous and interference-free monitoring of multiple biomarkers are necessary considering the practical applications of the flexible electrochemical sensors, which request functionalized carbon materials to be highly selective for specific biomarkers.

**GAS/VAPOR/SOLVENT SENSORS**

Determination/detection of the gas/vapor/solvent in the workplace and daily life is important, such as gas sensors in environmental monitoring or detecting air pollutants. The sensing mechanism is the electrical conductance induced by the electron transfer between CNTs and adsorbed gas molecules. Various studies showed that the gas sensors based on aligned CNTs showed faster response and recovery time than CNT networks, including CO2,[123] NO2,[124] and NH3 gases.[125] This may be due to the decreased network density that increases the detection limit of gas sensors.
Besides gas, vapors such as humidity or volatile organic compounds sensing in the environment around the human body or the relative humidity of human skin is also of vital importance in personal healthcare, electronic skin applications, and public safety control and security.\cite{126} In this regard, CNTs can be employed in vapors sensing due to their intrinsically doping effect by the adsorbed vapors molecules via physical van der Waals force that induces electrical changes upon vapors concentration changes.\cite{127} CNTs are usually introduced into polymers to fabricate composites in monitoring various vapors like methanol, chloroform, toluene, and cyclohexane vapors on the basis of change in electrical resistance.\cite{128-131} However, the sensitivities of these sensors are rather limited and calls for further exploration and enhancement. Another way to fabricate flexible CNT-based solvent/vapor sensors is utilizing the assembled
CNTs as a substrate to incorporate functional materials such as polymers. This method combines the advantages of the large specific surface area, high electron mobility, and flexibility of CNT assembly. Typically, poly-ionic liquid and some other polymers have been studied to develop such sensors due to the effective adsorption of solvent/vapor molecules inducing the swelling response. The CNT assembly that serves as a flexible substrate for the functional material such as hydrophilic water-swellable PVA for the flexible humidity sensor could enhance the performance. The resulting humidity sensor exhibited a wide range of detection of the relative humidity from 4% to 95% with a fast response (1.9 s) and short recovery time (1.5 s). The working mechanism of these sensors relies on the changes of electrical property or mechanical movement behavior when the coating polymers contact external solvent/vapor molecules. The CNT assembly provides a network for polymer to swell, thereby inducing an increase in the neighboring nanotube-nanotube distance. Especially, aligned CNT assembly introduces a new structural element to achieve a high level of control to dictate the actuation behavior to solvent/vapor. For instance, a flexible sensor responding to ethanol was made in a bilayer structure with an anisotropic CNT composite layer being deposited on anisotropic substrate (Figure 9A). The composite layer was prepared by spin-coating poly[1-phenyl-2-(p-trimethylsilyl) phenylacetylene] onto aligned CNTs, inducing the orientation of conjugated polymer chains by the aligned CNTs. In this case, the sensor exhibited anisotropic bending and unbending deformations with the controlled direction along the perpendicular direction relative to the CNT length direction (Figure 9B).

Construction of porous structure in the sensor obviously promotes sensitivity. Inspired by the helical structure in plants, we built hierarchically arranged helical fibers through the hierarchical and helical assembly of aligned CNTs (Figure 9C). With further modifications of the CNT surfaces, the CNT fiber could respond to multiple solvents and vapours including water. Rapid response and large actuation stroke (2050 revolutions per meter) with a high strain rate (for example, 340% per second) and high reversibility were achieved. The good performance contributed to the rapid solvent and vapour diffusion through the micrometer-scale and nanoscale gaps. These fiber sensors can be used for smart louvers in response to the change of humidity (Figure 9D).

LIGHT SENSORS

Light sensors play a vital role and have been extensively explored in many fields, such as night vision, information communication systems, and remote control. Because CNTs can absorb light over a wide range of wavelengths and convert the light to local heat, assembled CNTs can be utilized for flexible light sensors based on their unique photothermal and photothermoelectrical effects.

A flexible light sensor based on the photothermal effect can be fabricated with a simple bilayer structure containing a CNT network film and another material with a different thermal expansion coefficient, such as PC. Due to the large difference in the coefficients of thermal expansion between PC membrane and CNTs, the structure is strongly curled upon light irradiation. However, this kind of sensor showed specific deformation upon light. The sensor based on aligned CNTs could control the responsive deformation direction. A light sensor comprising aligned CNTs with other functional materials like paraffin wax could swell upon heating. Here, aligned CNT assembly provides the space among aligned CNTs for the functional material to expand perpendicularly in the orientation. The responsive deformation direction could be designed and tunable by varying the aligned direction of CNTs. The integration of the light sensor with the electrostatic effect could further convert the visible light into electricity in an effective manner. Furthermore, with a combination of the thermoelectric effect, a light sensor was built based on p-n junctions formed between two macroscopic films of p- and n-type CNTs. A responsiveness of up to ~3 V/W was observed in these sensors. Other than visible light, terahertz (THz) radiation could also be used by CNT network films due to their high absorptions over the entire THz frequency range and high thermoelectric Seebeck coefficient over 100 µV/K. Furthermore, with the high degree of flexibility, flexible and portable THz imaging systems with CNT network films had been achieved. Basically, they relied on the photothermoelectric effect as the detection mechanism, with CNT network films absorbing the THz radiation and inducing temperature gradient inside the film. Voltage signal was induced due to the carrier diffusing along the temperature gradient. The performance of the detector can be further enhanced by forming p-n junctions with p-type and n-type CNTs, and subsequent chemical doping of the p-n junctions. This THz scanner is expected for non-destructive and non-contact inspections.

In addition to the photothermal properties of CNTs, another role of aligned nanostructure of assembled CNT sheets in light sensors is that they could effectively orient the functional materials along the length direction of the CNTs without using any other aligning layer. For instance, the introduction of aligned CNT assembly could effectively orient crosslinked liquid-crystalline polymers to fabricate a sensor responding to ultraviolet (UV) and visible light or even only UV light. As real-time UV monitoring is important to prevent various skin diseases such as aging and skin cancer, a wearable fiber UV sensor based on p-CuZnS/n-TiO$_2$ heterojunction with titanium wire and CNT fiber as electrodes was successfully developed. The sensor read out ambient UV power density and transmitted data to smartphones via WIFI, presenting a promising wearable health monitor.

A series connection of a commercial light emitting diode (LED) to the gate electrode could control the TFT switch from “on” to “off” states when the LED was illuminated, which generated electrical charge and resulted in biased gate voltage. The integrating of flexible CNT TFT with commercial LED in series connection could be used for the detection of light with different intensities.

OUTLOOK AND PERSPECTIVES

In this review, we discuss the flexible sensors based on assembled CNTs including CNT sheet and CNT fiber. The CNT assembly functions as either electrode or active materials for sensing applications. Specifically different kinds of sensors for various applications have been introduced. However, there remain challenges for the sensors based on assembled CNTs.
For instance, compared with the conventional network structure, the aligned structure offers CNT materials with both enhanced electrical conductivity and tensile strength. However, the aligned CNT sheet shows anisotropic nature, which also limits their use for omni-directional sensors. In addition, most of the strain sensors show two opposite performance trends for different application scenarios, that is, a large deformation range with low sensitivity or a high sensitivity at small deformation range. This is because excessive sensitivity under large deformation may cause excessive signal amplitude, requiring a wider sensing system dynamic range. How to combine the two mutually restricted factors of high sensitivity and large strain/pressure range in the strain and pressure sensors need to be greatly investigated and explored.

Although flexible sensors based on CNT assemblies have shown outstanding sensing performances, these sensors are still in the preliminary stage for real-world applications. There are still some limitations and problems to be solved.
in future research. Firstly, the motion of humans is very complicated, and sensors are therefore required to be conducted with multi-stimulus instead of one stimulus in elaborately designed laboratory conditions. For instance, a strain sensor designed for pulse rate detection needs to simultaneously exclude the signal interference from the bending of the wrist. The signal of sensors needs to be carefully studied toward real-world usage scenarios that are more complex. To analyze the interaction between different stimuli or construction of specific microscale topographies, the sensors should be developed to extract or maximize the signal in a more efficient manner and output the detection data accurately. A deep-learning technique and big-data analysis may help the development of multi-functional sensors. Furthermore, the environment such as temperature and humidity will also affect the sensing performance. Suitable encapsulations make it possible to solve these challenges.

Secondly, the repeatability, stability, and durability of these sensors call for further investigations and optimizations, especially upon realistic usage conditions. For strain sensors, cracks are usually used to achieve high performances. However, the sensors may fracture in some random parts upon applying strain and form new cracks, due to the inhomogeneous feature at the microscale of the assembled carbon nanomaterial. Programming of cracks can be considered to improve the repeatability of devices.

Thirdly, a conformal and stable attachment of sensors on the surface of the target that needs to be detected for the expected signal is a vital factor for accurate recording. So far, many studies usually fastened sensors simply by stabilizing two ends by tapes with weak/no interfacing force between sensor and substrate. The mismatch of mechanical properties between the sensors and the target object usually deteriorates the accuracy and stability of the detected signals, which drastically reduces the sensitivity and shows no guidance in practical usage considering the practicability. Chemical adhesive bonding (e.g., hydrogen bonds) introduced on the surface of sensors is promising for improving adhesion and stability.

Lastly, most application conditions of the sensors are closely related to human body, such as contacting with our skin, and we need to consider the comfortability and biocompatibility. Currently, the majority of sensors are in the form of film or constructed/encapsulated by polymers for enhanced stretchability/adhesion. However, these sensors show poor air permeability and poor excretion of sweat, leading to the poor comfortability or swelling of the sweat ducts of the human body. If excessive sweat is not removed in time, it will lead to skin sensitivity, rash and sweat sore, and gland cystoma. To this end, fiber sensors that can be woven into flexible and breathable textiles provide a promising way to achieve attachment and comfortability at the same time.

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**CONFLICT OF INTEREST**

The authors declare that there is no conflict of interest that could be perceived as prejudicing the impartiality of the research reported.

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Conceptualization, idea, and manuscript preparation, editing and finalization: Sisi He and Longbin Qiu. Conceptualization, idea, and manuscript structure determination, writing, reviewing, editing, and finalization: Huisheng Peng.

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