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Low-dimensional Carbon Based Sensors and Sensing Network for Wearable Health and Environmental Monitoring

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ABSTRACT

With the advent of the era of big data and Internet of Things, wearable electronics are becoming more imperative than ever before, which prompts the continuous and fruitful research on wearable sensors and sensing network for health and environmental monitoring. This article presents an in-depth overview and review of this fertile area, focusing on sensors and sensing networks for strain, pressure, surface bio-potential, gas and temperature, which are made from low-dimensional carbon nano-materials and their composites. It covers materials, device structures, fabrication, performance and applications. It's evident that the appropriate and deliberate selection of low-dimensional carbon materials, matrix and substrate materials, and their interactions, as well as effective structural designs, are essential for highly sensitive and stable performance. Finally, the current status of industrial application is presented, possible hindrances for the adaptation of the technology are discussed, and future directions of development are indicated.

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Contents

1. Introduction

Both the global aging population and the continuous improvement of human living standard create a growing demand on personalized wearable health and environmental monitoring equipment. Wearable sensor technology, a key member of these wearable monitoring devices, has achieved marked progress in recent years. Among which, wearable low-dimensional carbon based sensors have long been a research focus in both academia and the industrial sector, because of their extraordinary electrical and mechanical properties. However, only a few products based on these sensors are available in the market. In this article, therefore, we will conduct a comprehensive overview and review on the development of wearable low-dimensional carbon based sensors for health and environmental monitoring during the past decade.

The low-dimensional carbon nano-materials normally refer to zero-dimensional (0-D), one-dimensional (1-D) and two dimensional (2-D) carbon allotropes. Their nano-sized assemblies are also included like carbon nano-particles (carbon black) and graphite particles representatively in a wearable setting, they are 0-D carbon black (CB), 1-D carbon nanotubes (CNTs) and 2D graphene. 1-D carbon materials also include carbon nanofibers and carbon nanotubes, which however are seldom used in wearable devices because of doubts on safety. Therefore, this review involves wearable sensors based on these three types of conductive materials.

Among all the requirements for wearable devices, the most basic is arguably safety and non-toxicity. The 0-D CBs are non-toxic and they are widely used in vehicle tires. The 2-D graphene is non-toxic either. Graphene can be simply exfoliated from graphite (1), what we use in pencils for many centuries. However herein, it should be emphasized that CNTs may pose serious hazardous concerns for human health and the environment (2-4). Such toxicity is more worrying in the scenarios of wearable devices, because these devices involve large-area manufacturing technologies and the final products may come into direct contact with the human skin. The future roll-to-roll mass production may speed up the spreading of CNTs to the environment, while direct skin contact means higher risks of toxicity. Therefore, we hope in all seriousness that scientific peers could seriously consider the potential toxicity of CNTs in the research on wearable electronics.

This review is organized as follows. First, wearable strain and pressure sensors, the basic types of mechanical sensors, will be summarized and analyzed in terms of materials, sensing mechanisms, fabrication technologies and their performance. Secondly, wearable bio-potential sensors and environmental sensors, will be fully reviewed including gas and temperature sensors. Thirdly, wearable sensing networks will be studied and promising applications of these sensors and sensing network will be indicated. Finally, conclusions will be drawn along with future outlook.

2. Wearable strain sensors

Strain sensors, or strain gauges, are sensors for deformation measurement. They transduce mechanical deformation into electrical signals. Traditional strain gauges comprise a patterned metal foil on a polymeric backing for easy attachment onto an object (5). Highly precise as they are, the conventional metallic or silicon based strain sensors can hardly be used in wearable electronics, which is primarily due to their insufficient stretchability, conformability, and a limited strain measuring range, normally below 5%. In comparison, low-dimensional carbon based strain sensors can be stretchable, confirmable and highly sensitive, thus more promising for wearable devices. In these devices, carbon materials are usually mixed with elastomers to make conductive composite. The elastomer renders the composite stretchable and conformable, while the conductive carbon components offer the composite piezoresistivity (6) thereby enabling a good strain sensitivity. Significant progress has been made in carbon based strain sensors over the past decade. The detailed review of the progress is arranged in an ascending dimensional order of the carbon materials.

2.1. 0-D carbon black

Carbon blacks, a 0-dimensional carbonaceous material, are made through incomplete combustion of hydrocarbon in a fixed gaseous atmosphere. Carbon blacks have long been used as conductive fillers in pressure sensitive rubbers (PSR) due to their low cost, safety, a high surface-area-to-volume ratio and good electrical conductivity (7). When used in wearable sensors, the primary sensing principle is the piezoresistive effect of CB/polymer composites.

Being the most cost-efficient material in the low-dimensional carbon family, CBs have been effectively employed in the research of wearable strain sensors. According to the substrate material, there are two main categories of sensor: fabric strain sensors which are easy for garment integration, and polydimethylsiloxane (PDMS) based strain sensors ready for skin mounting.

2.1.1. CB composite coating on fabric

Clothing made from fabrics are an ideal platform for wearable sensors as they are soft, deformable, breathable, washable, and durable (8). We wear garments made from fabrics almost 24 hours a day. Next we will illustrate the evolution of CB based fabric strain sensors by introducing four representative sensors.

Early in 2007, Cédric et al. made a fabric strain sensor by printing a conductive polymer composite of CBs and Styrene-Butadiene-Styrene (SBS) co-polymer, onto a Nylon woven fabric using a mask and a blade. The sensor showed a linear resistance change upon strains above 15% and the gauge factor was 80 (9). In the same year, De Rossi (10) at the University of Pisa reported a sensor-printed garment for kinesthetic monitoring (Fig. 1a). A composite of silicone matrix and CB powder was printed onto a Lycra®/cotton fabric. The composite functioned as both strain sensing materials and wire connections. This garment could provide real time feedbacks on limb orientation of the wearer. Although no specifications were provided for the strain sensors, this is a very typical work of early CB based wearable strain sensors. On year later, Tröster's group (11) at ETH Zürich presented a more systematic study on a sensor for measuring strain in textiles. A composite of SEBS-Block copolymer and CB particles was made into a thread through a self-developed wet spinning process. Then the sensor thread was integrated onto a knitted fabric using silicone and connected to silver-coated nylon yarns using conductive epoxy (Fig. 1b). This sensor measures a strain up to 80% with a gauge factor of 20. In addition, it has a fatigue life over 3800 cycles and can be washed for 8 times (Fig. 1c).

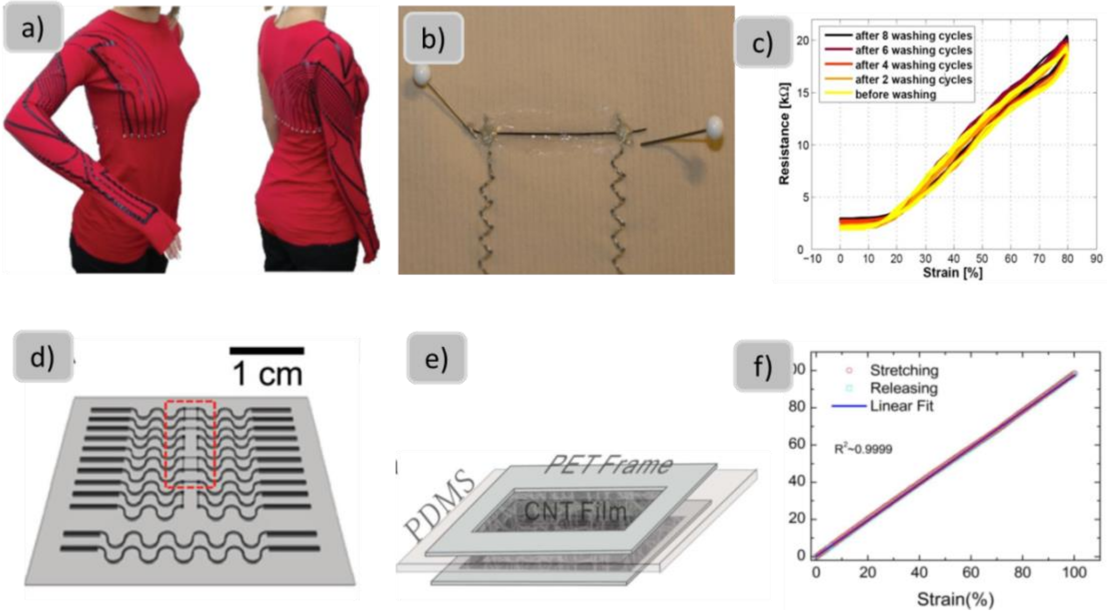

Fig.1. Typical CB and CNT based strain sensors. a) Garment printed with CB strain sensors for kinesthetic monitoring. Reprinted with permission from (10). Copyright (2011), Cambridge University Press. b) A CB-PDMS fabric strain sensor and c) its performance during washing tests. Reprinted with permission from (11), Copyright (2008), Sensors. d) CB-PDMS strain sensors based entirely on elastomers. Reprinted with permission from (5), Copyright (2012), Advanced Functional Material. e) A CNT-PDMS capacitive strain gauge and f) its calibration curves. Reprinted with permission from (12), Copyright (2012), Smart Materials and Structures.

Our group at The Hong Kong Polytechnic University conducted an extensive study on wearable strain sensors and made distinct progress (6, 12-14). We developed a resistive fabric strain sensor with extraordinary fatigue resistance of over 100 000 tensile cycles at 50% strain. The sensor has a tunable gauge factor from 1 to 100. Moreover, it can be machine washed for over 35 times without noticeable degradation in performance (12). To date, this kind of sensor has been produced in bulk volume and used in a range of wearable products by a company in Hong Kong (15). The sensors were fabricated by screen printing a composite of CB and silicone elastomer onto a fabric and connecting conductive yarns. They can be used to accurately monitor human respiration ,and limb muscle contraction (14). Accuracy in upper limb muscle contraction was as high as 93.8%. More wearable health-care products are being explored by the company at present. On the other hand, soft pressure sensors have also been developed from this fabric strain sensor as illustrated in section 3.1 of this review.

2.1.2. CB composite on PDMS

All the above strain sensors use fabrics as the backing material. The other main category adopts PDMS as the substrate. These sensors aim to realize skin-mountable devices for health monitoring and motion detection, etc.

In 2012, the Rogers' research group proposed a highly sensitive strain sensor based entirely on elastomers (5). A composite of CBs and PDMS was stencil printed as strain sensing element and another composite of CNT/PDMS was deployed as interconnects with a serpentine design. The sensor measures strains below 20% with a gauge factor of 29 (Fig. 1d). However, no fatigue information was reported. A similar sensor demonstrated a fatigue resistance of 30 cycles in 2014 (16)

Apart from fabric and PDMS substrates, textile yarns are also good platforms for wearable strain sensors. In 2016, a polyurethane (PU) yarn based strain sensor was reported (16) with good reproducibility over 10 000 cycles at merely 1% strain and claimed excellent washing and corrosion resistance. Unfortunately, the wash resistance in this article is not daily laundry using a washing machine but being submerged in acid and alkaline solutions only. Therefore, although a gauge factor of 39 was realized, it is uncertain that such a sensor can be applied into wearable electronics.

2.2. 1-D CNTs

Carbon nanotubes are one-dimensional carbonaceous materials. They are seamless cylinders of one or more layers of graphene, with an exceedingly large length-to-diameter ratio of over 10^6 (17). By virtue of their superior current carrying capability, high Young's modulus over 1 TPa, and fibril nanostructures, CNTs are very competitive candidates for stretchable devices especially when transparency is required (18). Therefore ever since 2006, the global CNT production has surged over 10 fold, and CNT-related journal publications and issued patents have been growing continuously in number (17). Various applications of CNT have been demonstrated including stretchable interconnects and electrodes, organic light-emitting diodes (OLEDs), super capacitors, field effect emitting devices, as well as strain, pressure and environmental sensors (18).

Traditional PSR composite has relatively large resistivity and strain dependence, which hamper their application in wearable sensors. Therefore, recent researchers improved performance of conductive elastomers by replacing CBs with more conductive and structurally advantageous materials including CNTs (18) Wearable strain sensors using CNTs can be both capacitive and resistive.

2.2.1. Capacitive CNT-based strain sensors

Capacitive CNT-based sensors merit excellent linearity and low hysteresis. In 2012, Daniel et al. (19) reported a highly elastic capacitive strain sensor made from percolating CNT networks. SWNTs were vacuum infiltrated and silicone hydrophobic-patterned to produce the percolating networks. The strain sensor could withstand 3 000 cycles of tensile strains at 3%, while it was claimed that the sensor can undergo 100% strain or higher with a gauge factor of 0.99. The linearity of sensor was as high as 0.9978. One year later, Le Cai et al. (18) improved the linearity of capacitive CNT strain sensor to 0.9999 (Fig. 1e and 1f). Moreover, strain measuring range was raised to over 300% and the sensor's fatigue life was dramatically enhanced to 10 000 cycles at 100% strain. Such marked advancement was resulted from the usage of both single and double walled CNTs films as well as a floating catalyst vapor deposition method. Potential applications in wearable devices were demonstrated through a prototypical glove and a respiration monitor.

2.2.2. Resistive CNT-based strain sensors

In recent research, typical resistive CNT-based strain sensors involve PDMS (20, 21), cotton/PU core-spun yarns (22), and very interestingly, chewing gum (23) as their structural materials. Since CNTs are mixed with insulating materials, they need to remain connected to each other to form a conductive network even upon deformation. In PDMS sensors, vertically aligned CNT films can be manually arranged with overlapping boundaries (20), or they were spun into continuous fibers (21) so the stretchability of sensors can be dramatically increased to 280% (20) and 300% (21) respectively. For the core-spun yarn strain sensors (22), PU SWNTs were coated onto cotton/PU filament core-spun yarns in a repeated dip coating approach. The sensor showed excellent performance and durability. Nevertheless, the CNTs were uncovered on the core-spun yarns. Hence further research is needed on effective encapsulation before application, especially in view of the toxicity of SWNTs. The sensor using chewing gum (23) achieved a uniform and stable distribution of CNTs by stretching and folding for around 500 times. Detailed specifications of these CNT-based strain sensors are summarized in Table 1. Note that the strain range here is not stretchability or the highest strain measured, but the strain used for fatigue test if fatigue test was conducted, because sensors without a satisfactory fatigue life are only potentially hopeful but barely applicable at present.

Table 1 Summary of performance of CB and CNT based strain sensors

Type of sensor	Materials	Strain range (%)*	Gauge factor	Fatigue life	Washability	Ref.
Resistive	CB-SBS-Nylon fabric	15	80	-	-	(9)
Resistive	CB-silicone-Lycra®/cotton fabric	-	-	-	-	(10)
Resistive	CB-SEBS-Block copolymer	0-80	20	3 800	8	(11)
Resistive	CB-silicone elastomer-Fabric	0-50	1-100	> 100 000	>35	(12)
Resistive	CB-PDMS	0-20	29.1	-	-	(5)
Resistive	CB-PDMS	10	1.8	30	-	(16)
Resistive	CB-cellulose-PU yarn	1	39	>10 000	-	(24)
Capacitive	CNT-PDMS	100	0.97	10 000	-	(18)
Capacitive	SWNT-silicone elastomer	100	0.99	3 000	-	(19)
Resistive	Aligned SWNTs-PDMS	150	0.82	10 000	-	(20)
Resistive	Aligned CNTs-EcoFlex	300	0.24	10 000	-	(21)
Resistive	PU-cotton-SWNTs	40	0.82	300 000	-	(22)
Resistive	Chewing gum-CNTs-PDMS	200	12-25	1 000	-	(23)

2.3. 2-D graphene

Graphene is a two-dimensional allotrope of carbon. Specifically, it is a single-layer of carbon atoms packed in an sp^2-bonded hexagonal lattice (25) and it can simply described as a mono-layer of graphitic film in graphite (26). Graphene has outstanding electrical, mechanical, thermal, and optical properties, which make it a promising candidate for flexible/wearable electronics (26, 27). Therefore, a great number of studies have been conducted to promote its application, and considerable progress has been reported (26). The application in wearable strain sensors mainly takes advantage of the excellent conductivity of graphene. Similar to the categories in

CB strain sensors, strain sensors based on graphene use silicone, PET, PU, rubber and other materials as matrix or substrates. Next, these sensors will be discussed accordingly.

2.3.1. Graphene with silicone

The intrinsic piezoresistivity of single graphene is rather limited because the hexagonal mesh of graphene can withstand strains only below 6% (28). Therefore, in this category of wearable strain sensors, the piezoresistivity also originates from the change in conductive networks, including breaking of contacts, contact area and change of spacing upon stretching (29). In 2011, Xue wen et al. (30) made a strain sensor based on a single layer of graphene on PDMS substrate. The sensor could measure strains between 2.4% and 4.5%. Yi et al. improved the strain measuring range to 20% by transferring a PMMA film coated with graphene to a pre-strained PDMS membrane (27). To further enlarge the strain measuring range, nanocellulose fibrils were added into the graphene layer radically enhancing the stretchability from 6% to 100% (29) (showed in Fig. 2b). The consequent strain sensors showed a gauge factor of 7.1 at 100% strain. In 2016, CNTs were also used to reinforce the sensing network of graphene (31). Apart from vacuum infiltration method, assembled graphene films have been rapidly scalable-made at liquid/air interface by Marangoni effect (32). The resultant strain sensor, however, worked at strains below 2% only.

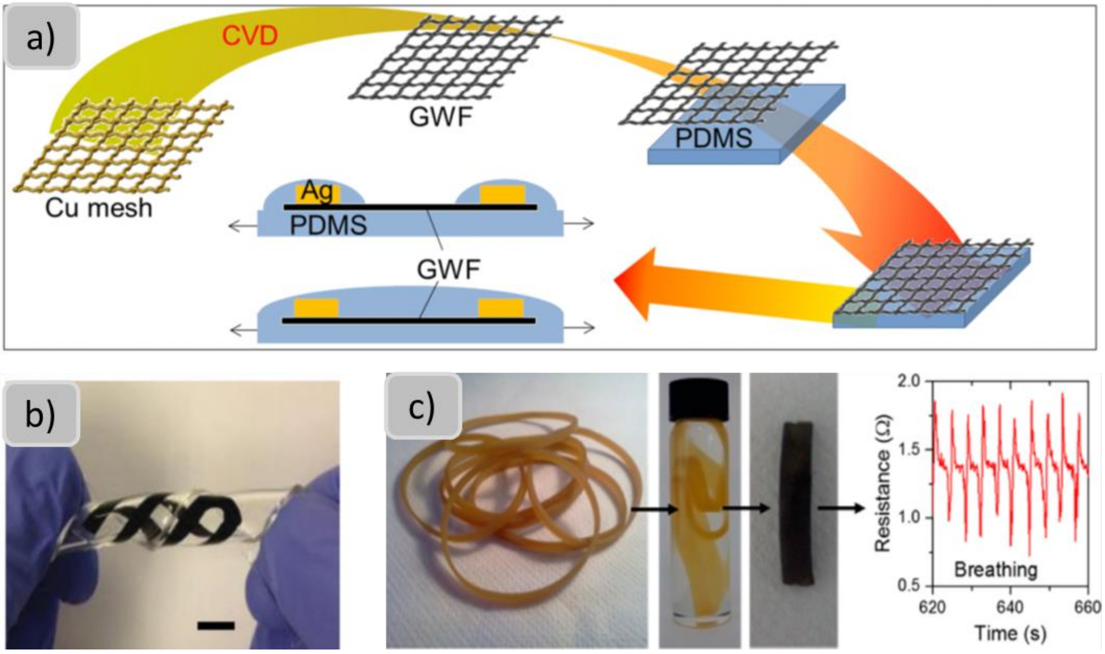

Fig.2. Typical graphene based strain sensors. a) Schematic of the graphene woven fabric sensor production. Reprinted with permission from (33). Copyright (2012), Scientific Reports. b) A graphene-nanocellulose strain sensing being twisted. Reprinted with permission from (29). Copyright (2014), Advanced Materials. c) Graphene-rubber strain sensor, fabrication process and breathing signals monitored. Reprinted with permission from (34). Copyright (2014), American Chemical Society

In 2016, graphene aerogel (GA)/PDMS (GA) nanocomposites were explored for wearable strain sensor applications (35). Strain sensor based on this type of nanocomposite showed a gauge factor of up to ~61.3 and such piezoresistivity remained virtually stable even after 10 000 cycles of tensile strains at 10%. Besides, the gauge factor can be readily adjusted by changing the concentration of precursor and the freezing temperature in the fabrication process of GA. Such favorable performance benefitted from the three dimensional macroporous cellular structure of the composite. Fabrication of the sensors was both cost-effective and time-effective. First, graphene oxide (GO) and vitamin C were used to prepare the GA. Then the aerogel was thermally treated in a furnace to reduce the GO into graphene. Second, vacuum-assisted infiltration method was adopted to prepare the GA/PDMS nanocomposite. Finally, copper wires were connected to both ends of the composites using conductive silver-paste adhesive for sensor characterization. Among others, this work is promising for wearable applications.

In order to arrange graphene effectively, copper meshes in a plain woven structure were used as sacrificial supporting structure (33, 36), where graphene first grew on the surface of copper meshes by chemical vapor deposition at room atmospheric pressure, then the copper meshes were etched away and the residual graphene was coated with or imbedded into PDMS. Such graphene films changed more dramatically in polycrystalline structure because dense cracks could form and propagate upon deformation, giving the strain sensor an extremely high sensitivity at $\sim 10^3$ for 2~6% strain (33) (Fig. 2a) and $\sim 10^4$ under 8% strain (36). Unfortunately, only a 100-cycle test was conducted (33) which apparently is insufficient for real application, although phonation, expression change, blink, as well as breathing and pulse can be detected using these sensors (36).

One more fantastic work of graphene in 2016 involved Silly Putty, a slightly cross-linked silicone, as the matrix (37). The nanocomposites were prepared by transforming nanosheets of graphene from liquidly exfoliated graphite to chloroform and mixing with homemade Silly Putty. The as-made nanocomposites worked as highly sensitive electromechanical sensors with a gauge factor of over 500 and even the superlight footsteps of a small spider could be detected. It is possible to measure pulse and blood pressure using this composite.

2.3.2. Graphene with other materials

Besides PDMS, various other materials have been explored to make wearable graphene based strain sensors, including PET (38), 3M elastic adhesive tape (39), PU (40), as well as very interestingly, natural rubber elastic bands (34), and human hair (41). The summary of these sensors is listed in Table 2.

Sensors using graphene on PET film could withstand only 150 repetitive strains at 7.5% (38). Strain sensors with fish-scale like graphene layer on an elastic tape could further sense strains up to 82% and its stability reached over 5 000 cycles (39). Such improvement benefits from the fabrication process. The adhesive tape with the first layer of reduced graphene oxide (rGO) was pre-stretched to 50% for the addition of the 2^{nd} rGO layer. The tape with bilayer rGO was subsequently stretched to 100% to generate fish-scale-like cracks for a high sensitivity and stability. Aside from the pre-stretch and bilayer method, Ag nanoparticles (AgNPs) have been introduced to form synergetic conductive network (40) for highly stretchable graphene/AgNPs/PU strain sensors. During elongation, graphene was believed to fulfill the cracks of AgNPs thus an ultrahigh strain at 1000% could be detected and the sensor could work at 50% for over 1 000 cycles.

It is worth noting that, in 2014, Conor et al. (34) reported an impressive high-strain high-rate sensor based on graphene-rubbers composites, in which rubbers are only store-bought elastic bands that we use in everyday life (Fig. 2c). The fabrication process was simple. Natural rubber bands swelled due to soaking in toluene, opening up many tiny pores for graphene to go inside. Strikingly, this simple sensor worked at strains exceeding 800% with a gauge factor of 35. Moreover, dynamic response of the sensor was excellent at even 160 Hz. Strains above 6% could be detected even if strain rates were over 6 000%/s at 60 Hz. Such extraordinary performance is ascribed to both the mechanical properties of natural rubber and the dense network of graphene infused inside. This type of sensor is highly promising for motion detection as well as breathing and pulse rate monitoring.

Table 2 Summary of performance of graphene based strain sensors

Type of sensor	Materials	Strain range (%)	Detection limit (%)	Gauge factor	Fatigue life	Ref.
Resistive	Graphene ripples-PDMS	20	0.3	-2	-	(27)
Resistive	Graphene-nanpcellulose-PDMS	100	0.4	7.1	-	(28)
Resistive	Graphene monolayer on PDMS	5	0	151	-	(30)
Resistive	Graphene-CNTs-PDMS	20	1	0.36	-	(31)
Resistive	Graphene-PDMS	2	0	1037	-	(32)
Resistive	Graphene aerogel-PDMS	10	0	61.3	10 000	(35)
Resistive	Graphene woven fabrics-PDMS	>6	2	1000	100	(33)
Resistive	Graphene woven fabrics-PDMS	8	0.2	10^4-0.07	-	(36)
Resistive	Graphene-Silly Putty	10	0	>500	-	(37)
Resistive	Graphene on PET	7.5	0	9.49	150	(38)
Resistive	Fish scale-like rGO/tape film	82	0.1	16.2-150	5 000	(39)
Resistive	Graphene-AgNPs-PDMS	50	0.5	7	1 000	(40)
Resistive	Graphene-rubber	800	0	35	500-1000	(34)
Resistive	Graphene on human hairs	5	0	4.46	400	(41)

3. Wearable pressure sensors

A pressure sensor is a device that transduces mechanical forces into electrical signals. Wearable pressure sensors are essential elements for force and pressure detection in both health and environmental monitoring, as well as in human-machine interfaces. In the past decade, a large number of wearable pressure sensors based on CB, CNTs, and graphene have been studied and the summary is listed in Table 3.

3.1. CB pressure sensors

CB pressure sensors can be made typically through intrinsic piezoresistivity of PSR

(42) or CB@PU sponges (43) as well as using CB based fabric strain sensors (12, 13).

CB/PDMS nanocomposites, due to their piezoresistivity (see section 2.1), can be used as pressure sensors for a pressure up to 1 MPa (42). Unfortunately, the limited flexibility and conformability impede their application in wearable devices.

In 2016, Wu et al. (43) presented soft pressure sensors made from microcrack designed CB@PU sponges (Fig. 3a). CBs assembled onto PU sponges in a water-assisted layer-by-layer fashion. The sensor comprising the CB@PU sponge and conductive silver pastes was reported to be both sensitive for tiny (91 Pa) and large motion monitoring (16.4 kPa), owing to its two sensing mechanisms, i.e. microcrack junction mechanism and compressive contacting of sponge bones respectively. In addition, the sensor showed fast response of below 20 ms and good reproducibility of over 50 000 cycles (at 40% strain compression). However, the sensor needs to be well-encapsulated before application and it's relatively thick and bulky in size.

In the past few years, our group developed soft pressure sensors for both low-medium pressure (12) and large pressure measurement (13), both of which were based on CB fabric strain sensors as described in section 2.1.1.

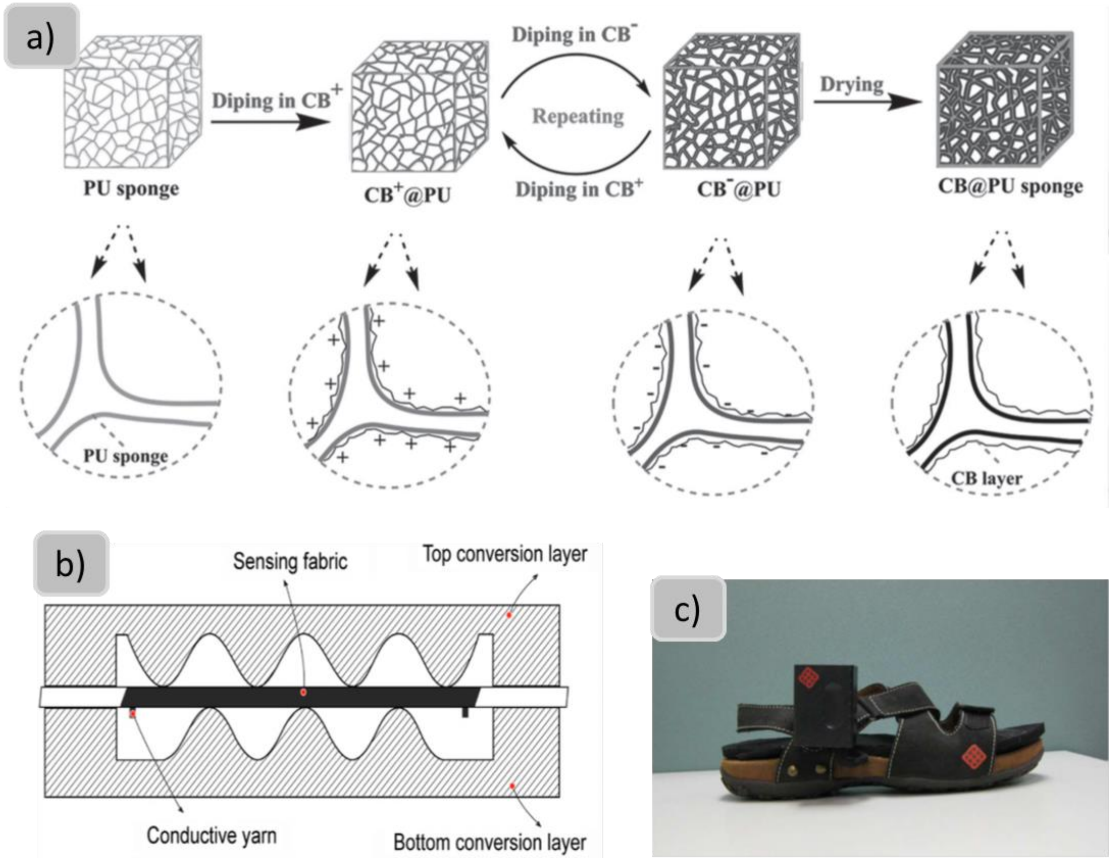

Fig. 3. Typical CB based pressure sensors. a) Fabrication process of CB-PU sponge pressure sensors. Reprinted with permission from (43). Copyright (2016), Advanced Functional Material. b) Schematic of a fabric pressure sensor with tooth-structured layers. Reprinted with permission from (12) . Copyright (2011), Smart Materials And Structures. c) A picture of i-Shoe, which is equipped with sensors in b) to monitor distribution of foot pressure in most daily activities. Reprinted with permission from (44). Copyright (2011), IEEE Transactions On Information Technology In Biomedicine.

The first type of pressure sensor developed was published in 2011 (12). It was made by sandwiching a CB fabric strain sensor between two tooth-structured PDMS layers as illustrated in Fig. 3b. The highly durable CB strain sensor and the simple structure impart the pressure sensors with robustness, flexibility and excellent fatigue resistance. The pressure sensors were stable even after 100 000 compressive cycles. They could measure pressure from 0 to 2 MPa with a sensitivity of 2.98 MPa^{-1}, covering the required monitoring range of human foot pressure. They were therefore adopted in i-Shoe (44) (Fig. 3c), a foot pressure mapping system that records spatial and temporal plantar pressure distributions in most daily activities.

The second type of pressure sensor was reported in 2014 (13) (Fig. 4a). The sensor was fabricated by mounting a ring-shape CB fabric strain sensor onto a PDMS cylinder. It could measure pressures up to 8 MPa with a sensitivity of 1 MPa^{-1}. Besides, the sensor was suitable for dynamic impact tests. To examine its applicability, smart clothing integrated with such soft pressure sensors (Fig. 4b and c) was made and examined in frontal sled crash tests with three-point belted Hybrid III, a regulated anthropometric test device in auto industry. Two rounds of sled crash tests at 40 km/h and 30 km/h were carried out. Results showed that pressures on the two ends of the shoulder belt and lap belt were significantly larger than those in the central part, with peak values around 6 MPa for the shoulder belt and 5 MPa for the lap belt. Such measured pressure distribution basically agreed with the results of numerical simulation. For both impact velocities, the peak pressure on dummy's torso was ~20% larger than that on abdomen. The above findings with quantitative pressure values from an in-situ pressure evaluation system provide useful information for the study of occupant injury in vehicle crash. The smart clothing could also help in improving vehicle crashworthiness design and in training sportsmen to avoid impact injury.

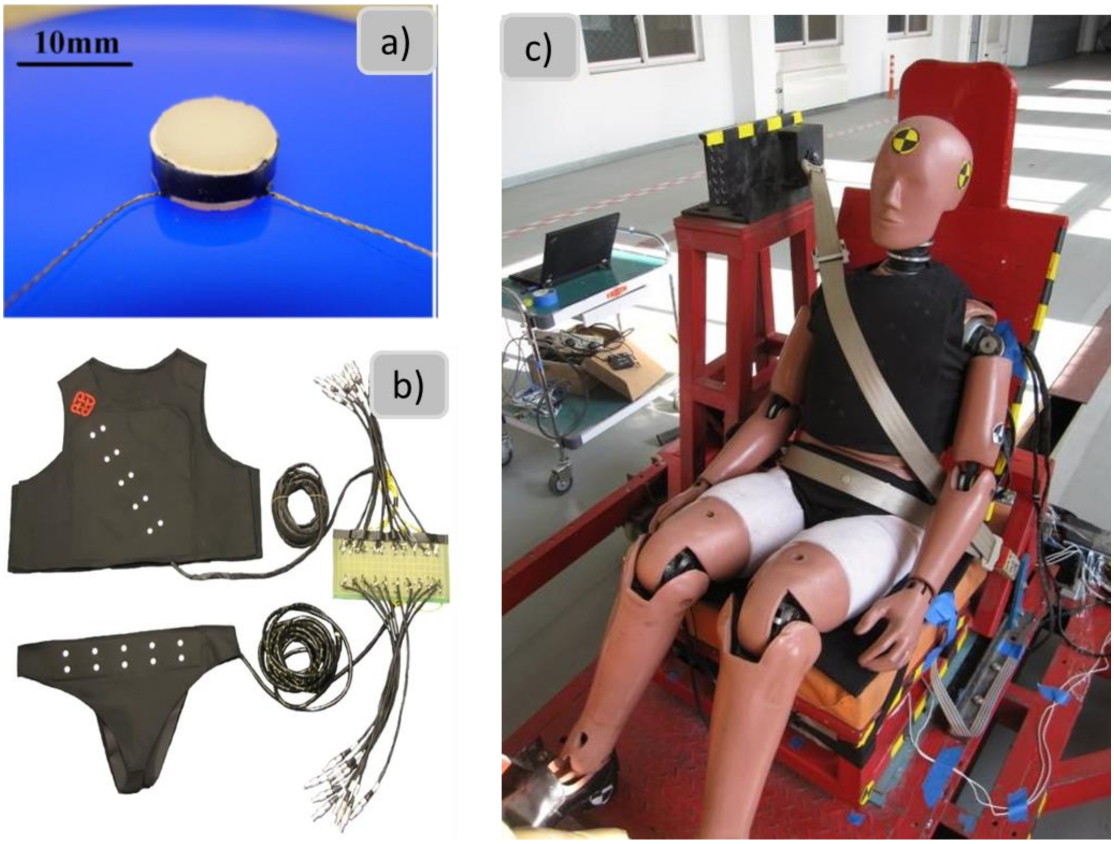

Fig. 4. CB based pressure sensors for large pressure measurement. a) A sample of fabric pressure sensor for large pressure measurement. Reprinted with permission from (13). Copyright (2014), Smart Materials And Structures. b) Prototype of smart clothing for sled crash test. The clothing was designed for dummy wearing. And c) Installation of dummy's apparel on the sled.

3.2. CNT pressure sensors

Compared with CB based pressure sensors, more sensitive pressure sensors have been made using CNTs, and some are exceedingly sensitive. Typical approaches include spraying CNTs onto PDMS films for capacitive pressure sensors (45, 46) or combining CNT films with replicated micro (47, 48) or porous structures for resistive pressure sensors.

In 2011, Bao's group (45) reported a skin-like pressure and strain sensor based on transparent CNTs films (Fig. 5a). CNTs were spray-coated according to pattern design onto PDMS substrates. After stretch and release treatment in one direction, two PDMS films were laminated together using silicone elastomer, with CNTs films facing each other and CNTs stripes crossed. Further stretch and release stabilized the morphology of CNTs film. The sensor arrays could detect pressures between 50 kPa and 1 MPa with a transparency above 68%. For such sensors, delamination of the CNTs layer under repetitive loadings may be a big concern because the CNTs are deposited to PDMS without robust bonding. Subsequently, in 2013, Wu et al. (46) furthered Bao's work and improved the mechanical bonding between CNT film and PDMS.

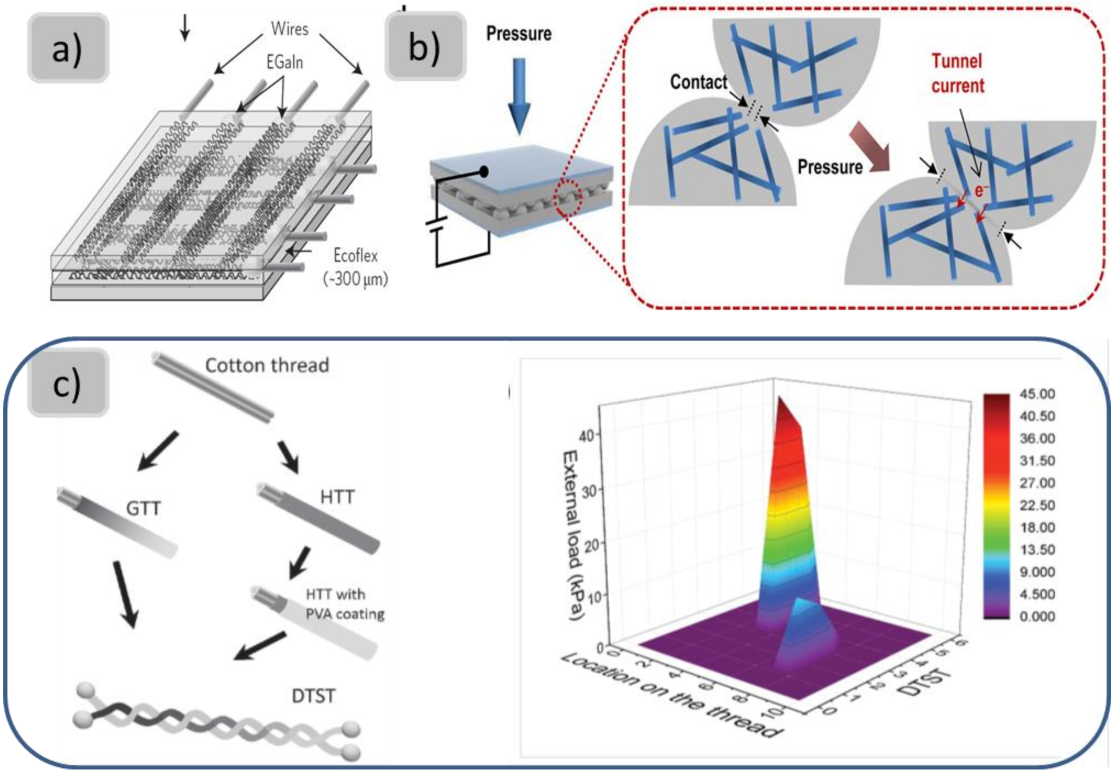

Fig.5 Typical CNT and graphene based pressure sensors. a) Transparent CNT pressure sensor arrays. Reprinted with permission from (45). Copyright (2011), Nature Nanotechnology. b) CNT based pressure sensors with dome-shape structures and its working mechanism. Structure and working principle of contact mode CNT pressure sensors. Reprinted with permission from (47). Copyright (2014), American Chemical Society. c) Double-twisted conductive smart threads. Left is schematic of fabrication process. Right is a 3D profile of calculated amplitude and location of applied forces. Reprinted with permission from (49). Copyright (2016), Advanced Functional Materials.

Resistive CNT pressure sensors can also obtain high sensitivity by using replicated microstructures. In 2014, Jonghwa et al. (47) reported such a pressure sensor. A composite of MWNTs and PDMS was silicon molded into films with dome features (Fig. 5b). Two films were placed in a pair with domes facing each other. When pressure applied, the contact area of the two composite films will increase significantly resulting in a decrease in contact resistance. The sensors could detect pressures as low as 0.2 Pa within 0.04 s. Apart from using silicon mold, in the same year, Xuewen et al. (48) made highly sensitive pressure sensors using silk fabric as a mold. With a similar working principle as Jonghwa's sensors, this silk-molded sensor could detect pressure low to 0.6 Pa and it demonstrated a fatigue life over 67 500 cyclic compressions.

Highly sensitive pressure sensors have also been fabricated by introducing tiny pores into CNT/PDMS composites (50). Reverse micelles were used to produce pores. The incorporation of pores significantly improved the pressure sensitivity to around five folds. This method shows possible routes to improve sensitivity of traditional PSR. However, the incorporation of CNTs comes with potential hazards (see section 1). Another form of pores can be realized by using porous sponges as the structural materials (51). Similar to Xiaodong's research in section 3.1, sponges were used to make piezoresistive pressure sensors along with CNTs and AgNPs. The simple "dip and dry" method of composite fabrication was both facile and scalable. The sensors could detect pressures below 61.81 kPa with a fatigue resistance above 2 000 cycles.

One distinctive work on CNT pressure sensors was published in 2016 by Yanlong et al. (49) (Fig. 5c). This work realized multidimensional pressure sensing through thin threads only. Each thread comprised two cotton yarns coated with SWNTs differently.

One yarn for detection of pressure amplitude was homogeneously coated with SWNTs and the other for identification of force location was coated in a gradient manner. The two yarns were twisted together and then encapsulated with thin PDMS films. After complex calibration, such thread systems were able to detect pressures below 50 kPa as well as its position. More importantly, dynamic fatigue life reached over 10^4 cycles, indicating superior durability. This special design of two different yarns is referential and enlightening to peer-researchers working on wearable pressure sensor technologies.

3.3. Graphene pressure sensors

As stated previously, intrinsic piezoresistivity of single graphene is quite limited because graphene itself is barely deformable. Therefore, similar to those in section 2.3.1, the graphene based wearable pressure sensors also realize high sensitivity from the change in conductive networks, such as the size of contact area (52-55), breaking and joining of contacts, and change of spacing (56). Among others, nanosuspension of GO has been utilized for microfluidic tactile sensors (57).

The pressure sensing mechanism of contact area change has been widely used by researchers, either using one layer of graphene or two layers. In 2014, one graphene film was assembled onto a silicon mould patterned PDMS layer (52). Then the assembled layer was integrated with an indium tin oxide (ITO)/PET layer to form a pressure sensor capable of detecting a pressure as low as 1.5 Pa. In comparison, two-layer-graphene mechanisms were more popular. In 2014, two isolated and patterned single graphene films were used to detect pressures from 1 to 14 kPa (53).

Then in 2015, a double-layer graphene structure (54) could detect pressures either below 250 Pa or above 1 000 Pa (and below 10 kPa). It was claimed that low pressures were measured through change in contact area and larger pressures through electromechanical effect of graphene itself. No fatigue life was provided for these two sensors. In 2016, Tran et al. (55) reported a new strategy to fabricate graphene from graphite through liquid-phase exfoliation with sonication, where GO was used as a dispersant. When such graphene layers were coated onto PDMS, the sensors could detect pressures below 7.3 kPa even after 1 000 cycles of compression.

An impressively sensitive and durable soft pressure sensor was reported in 2013 using graphene-PU sponge with a fractured design (56). After GO was coated onto PU sponge, graphene was reduced and the whole structure was hydrothermally treated. Then the sponge was compressed at 95% strain for 2 hours to fracture the microfibers of sponge. This fracturing treatment induced more broken branches and made the whole structure stable, thereby improving the pressure sensitivity by two orders of magnitude for pressures below 2 kPa and one order of magnitude between 2 kPa and 10 kPa, when compared with sponge before treatment. The sensor also displayed a fatigue life over 10 000 cycles. Such pressures sensors are low-cost and easily scalable, but further techniques need to be developed for reliable elastic wire connection and encapsulation.

Table 3 Summary of performance of low-dimensional carbon based pressure sensors

Type of sensor	Materials	Pressure range (kPa)	Sensitivity	Fatigue life	Ref.
Resistive	CB fabric strain sensor-PDMS	0-2 000	2.98 MPa^{-1}	100 000	(12)
Resistive	CB fabric strain sensor-PDMS	8 000	>1 MPa^{-1}	-	(13)

Resistive	CB-PDMS with Cu polyimide films	25-1 000	>1.7 MPa^{-1}	-	(42)
Resistive	CB-PU sponge	0.091-16.4	0.023-0.068 kPa^{-1}	50 000	(43)
Capacitive	SWNTs-PDMS-EcoFlex	50-1 000	0.23 MPa^{-1}	-	(45)
Capacitive	SWNTs-PDMS-EcoFlex	10-200	0.59 kPa^{-1}	-	(46)
Resistive	MWNTs-PDMS	0.0002-80	-15.1 kPa^{-1}	-	(47)
Resistive	SWNT ultrathin film-PDMS	0.0006-1.2	1.8 kPa^{-1}	-	(48)
Resistive	MWNTs-PDMS	0.25-100	2.5 kPa^{-1}	-	(50)
Resistive	CNTs-AgNPs-Sponge	2.24-61.81	2.12-9.08 kPa^{-1}	2 000	(51)
Resistive	SWNTs-Cotton thread-PDMS	50	0.1 1.56 kPa^{-1}	10 000	(49)
Resistive	rGO-PDMS-ITO/PET	0-0.1	-5.5 Pa^{-1}	-	(52)
Resistive	Graphene-PDMS-PET	1-14	3.9* 10^{-8} kPa^{-1}	120	(53)
Resistive	Graphene-PDMS	<0.25/(1-8)	-0.24 / 0.039 kPa^{-1}	-	(54)
Resistive	Graphene-PDMS	0-10	-0.268 kPa^{-1}	1 000	(55)
Resistive	rGO-PU sponge	0-2/2-10	0.26/0.03 kPa^{-1}	10 000	(56)
Resistive	GO nanosuspension-EcoFlex	0.07-0.25 N	0.0338 kPa^{-1}	100	(57)

4. Wearable bio-potential sensors

The electrocardiography (ECG), electroencephalography (EEG) and electromyography (EMG) are widely accepted methods for surface bio-potential monitoring. While the most used Ag/AgCl wet electrodes are not suitable for continuous monitoring because the use of conductive gel on the skin is inconvenient and can lead to irritation and allergic reactions (58-60). Therefore, researchers have been investigating alternative electrodes which are dry and even do not need skin preparation (61, 62). Carbon based dry electrodes for long-term electrophysiological signals monitoring have received much interest recently due to the high surface area, superior conductivity and excellent flexibility of carbon materials.

The carbon based bio-potential sensors work by converting the ionic electric current within the human body into electric current in the sensors. For example, two or more wearable ECG electrodes are placed on the body to acquire the changes in differential voltage between them, in which the variation comes from electrical fluctuations generated by the heart. Despite of the unmatched wearing comfort of capacitive electrodes, the carbon-based dry electrodes are rarely capacitive ones. Instead, most come into direct contact with the skin or even penetrate the skin, of which two the working mechanisms are slightly different. For electrodes placed only on the skin surface, the electrode-skin surface can be simply modeled by a parallel connected capacitor C and resistor R of the stratum corneum (an insulating layer formed by dead cells), series connected to another parallel RC of the epidermis. While for the penetrating counterparts which step over stratum corneum, the first parallel of C and R no longer exist.

Next, different kinds of carbon based electrodes including their fabrication processes and performances will be introduced, followed by issues and challenges.

4.1. CNT based dry electrode

Ruffini et al. (63, 64) employed a penetrating ECG electrode based on MWCNT forest. A large number of CNTs formed a brush-like structure which guaranteed a stable electrical contact interface with low impedance between electrode and skin. The developed electrode minimized the possible infection risk by barely penetrating the outer layer of the skin and avoiding nerve cells. A comparable spectral densities were achieved from both CNT based electrodes and wet electrodes. Subjects did not report any side effects after 6 months human trials which confirmed the application of the CNT electrode for long-term bio-potential recording (64).

CNT/PDMS composites, which combine the excellent electrical properties and elastic properties, are often used to fabricate flexible and conductive dry electrodes (65-68). However the large surface areas of CNT lead to aggregation with each other, making their uniform dispersion into polymer challenging. In order to achieve a good dispersion of CNT into PDMS, Lee et al. (69) developed a novel wetting and flow stress dispersion method. Even though the homogeneous dispersed flexible CNT/PDMS composite material in the electrode enabled the close contact of the electrode to the ear, the contact impedance was still about four-times larger than that of Ag/AgCl electrode. The same group later fabricated a self-adhesive ECG electrode through dispersing CNTs into an adhesive PDMS (aPDMS) adopting flow stress dispersion method (Fig. 6a) (67). ECG electrodes could embed on other electronic components through soldering (Fig. 6 c-f). Skin compatibility of CNT/aPDMS electrode was tested by attaching it to the skin for a week without side effects (Fig. 6g). The developed CNT/aPDMS based electrode demonstrated an adhesive force (1.1 N/cm^2) large for skin attachment. Although the robust monitoring of bio-potential signal could be achieved, the poor reusability of this electrode limited its commercial application in human health monitoring. Similarly, Liu and co-workers (65) fabricated polymer electrodes which consist of a PDMS matrix and CNTs. An even CNTs-PDMS mixture was obtained by using a cost-effective replica technology. A systematic study was performed to investigate the effects of ultrasonication time, and effects of CNT concentration by evaluating impedance and ECG measurements. The signal amplitude obtained with the wet electrodes and CNTs–PDMS electrodes (ultrasonication time: 12 h and CNT content: 5 wt%) showed no significant difference.

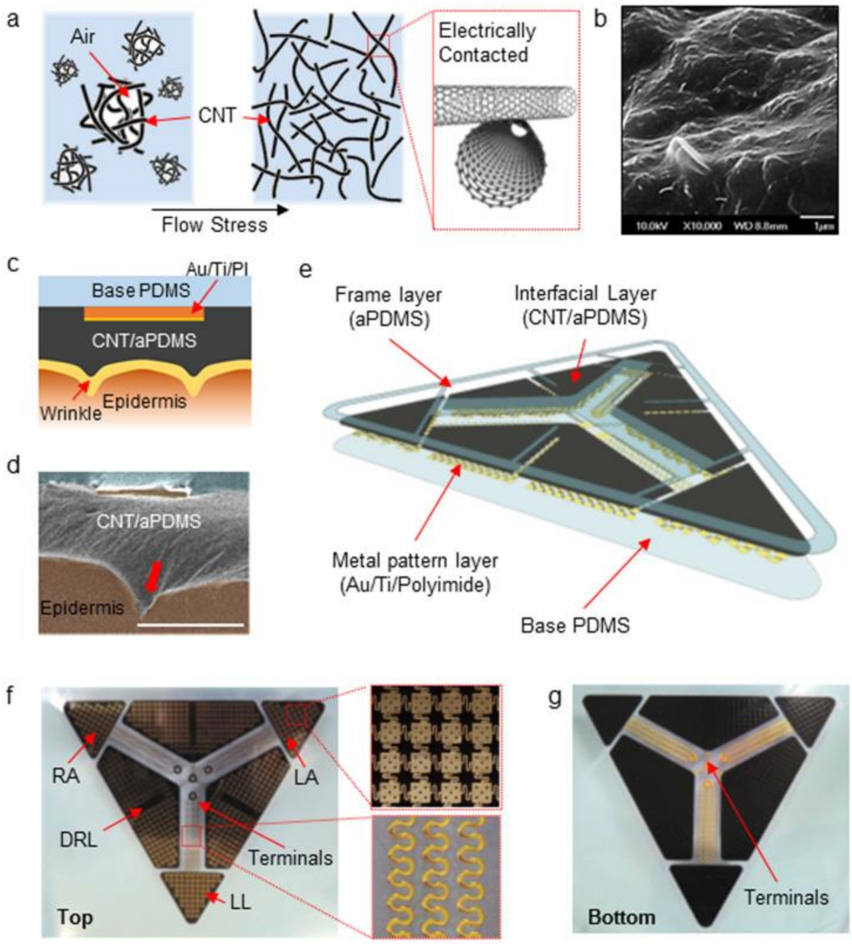

Fig. 6. Dry electrode based on CNT/aPDMS. a) Mixing aggregated CNTs with aPDMS using flow stress. b) A SEM image of CNT/aPDMS. c) Robust contact between CNT/aPDMS and skin d) SEM image of a CNT/aPDMS layer attached to epidermis. e) Structure of an ECG electrode. f) Top view of the CNT/aPDMS based electrode. g) Photograph of skin after one-week continuous attachment of electrode. Reprinted with permission from (67). Copyright (2014), Scientific Reports.

Besides dispersing CNTs into polymer matrix, printing the CNTs on surface of flexible and lightweight cotton fabric (70) and patterning synthesis of vertical aligned CNTs on the circular stainless steel substrate (71) were also reported to fabricate CNT based electrodes.

4.2. Graphene based dry electrode

Yapici et al.(72) fabricated a graphene-clad textile electrode through dipping textile with hydrophilic surface property into diluted GO suspension, and then chemically converting the GO into rGO. The signals recorded from the electrodes revealed good correlation both in time and frequency domain to those from wet electrodes. The graphene-clad textile electrode demonstrated excellent washable and durable characteristics, superior comfort and flexibility, which makes it suitable for health monitoring applications. Furthermore, Celik et al. (73) proposed a graphene based electrode by coating a graphene onto the target electrodes. The experimental results clearly showed that the signal-to-noise ratio improved significantly and the signal shape was much better when compared with traditional electrodes.

To overcome the poor electrical percolation of CNT, recently Kim et al. (74) fabricated a hybrid carbon based electrode by incorporating CNT and graphene into an elastomeric matrix simultaneously. The body-attachable carbon based electrodes can be used for surface bio-potential monitoring under different conditions even underwater and movements which is due to its excellent stretchable and water-proof characteristics.

Dry electrodes based on carbon nanomaterials have been fabricated to overcome the drawbacks of commercial wet ones and to meet the needs of long-term monitoring, such as ECG, EEG and EMG etc. The advantages of carbon based electrodes include higher flexibility or confirmability, less irritation and allergic reactions, as well as good wearing comfort and robustness. Furthermore, they are more likely to be machine washed especially when printed onto fabrics. Although carbon based electrodes have the above merits, the noise induced by movement and pressure variation or often called motion artifacts are still poorly understood (75) , which remains a problem for the dry electrodes, thus wet ones still dominate the markets. In addition, the electrical conductivity of many carbon-based electrodes is much lower than their metallic counterparts. More studies should be conducted on low-cost fabrication methods for carbon based bio-potential electrode and novel designs to obtain low contact impedance and stable electrode–skin interface. Besides, the garments or belts carrying the carbon-based electrodes need to be carefully designed to minimize the effect of human body movement on bio-potential monitoring.

5. Wearable environmental sensors

The world-wide air pollution in recent years has seriously threatened human health and environment, which results in booming research on gas sensors (76, 77). Wearable and portable gas sensors prove to be effective devices for health and environment monitoring, because air quality which changes with time and place as well exhaled as gas in a certain people can help diagnose disease and monitor physical signals (76, 78). To achieve highly sensitive wearable gas sensors, a large number of published studies explored low-dimensional carbon materials. Such materials have a high large surface-to-volume ratio which is advantageous for adsorption of gas molecules. The performance of wearable gas sensors based on carbon materials is listed in Table 4. In addition to gas detection, light monitoring is another critical aspect of environmental condition. By taking advantage of the graphene, light sensors have shown promising properties (79-81), which will be introduced in section 5.3.

Table 4 Summary of the performance of gas sensors

Materials	Gas	$Conc_{min}$.(ppm)	Response	Response time	Recovery time ;.	Ref.
MWCNT	Benzene	0.05	~2.5%	1200s	-	(82)
MWCNT	NH_3	0.01	~9%	7s	15s	(83)
CNT	NO_2	0.25	~80%	20min	~7min	(84)

	Cl_2	0.5	~90%	~20min	~10min	(84)
MWCNT	Dimethyl methylphosphonate	10	~0.2%	-	-	(85)
MWCNT	CO	50	~29%	-	-	(86)
RGO	NO_2	0.5	~74.6%	12s	20s	(87)
RGO	NO_2	0.06	~55%	5min	5min	(88)
	NH_3	0.01	~9%	5min	5min	(88)
Carbon	NO_2	1.0	0.5%	10min	20min	(89)
Graphene	NO_2	0.2	~8%	~2min	~2min	(90)
RGO	NH_3	5	~5.5%	~3min	-	(91)

Response=$\frac{R_g-R_a}{R_a}$; R_a, sensor resistance in the air before test; R_g, sensor resistance after exposure to a test gas.

5.1. CNT based gas sensors

CNTs are ideal candidates for gas sensors because their intrinsic electronic properties make them very sensitive to the local gas molecules. When exposed to gas molecules, the electrical conductivity of CNTs will increase or decrease dramatically. Many studies have proven that the surface functionalization of CNTs could promote the charge transfer between a specific chemical species and the decorated carbon materials, which leads to better sensing performance and selectivity (92-94).

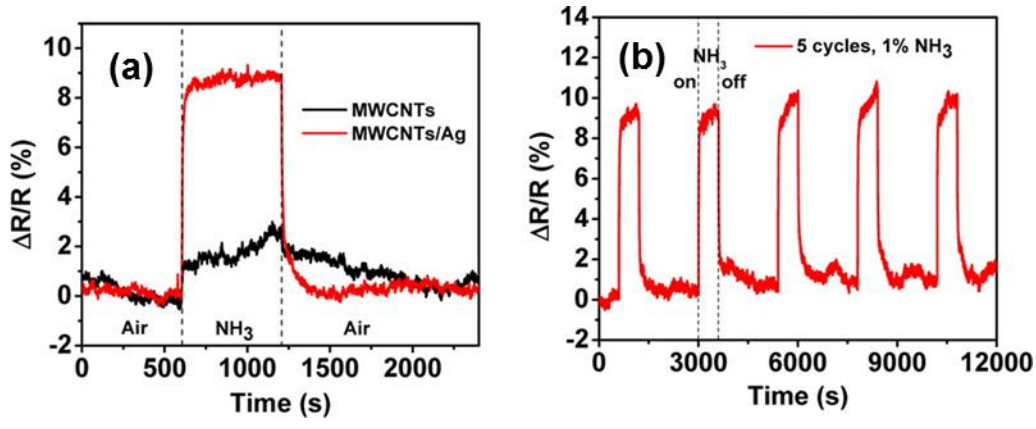

Fig. 7. a) Sensing response with time before and after Ag NCs decorating MWCNTs at RT. b) Sensing behavior of five cycles of the Ag-NC-MWCNT sensor under 1% NH3 contribution appears quite repeatable. Reprinted with permission from (83). Copyright (2012), American Chemical Society.

The electronic characteristics of CNTs remain almost unchanged when exposed to benzene due to poor interactions between CNTs and benzene molecule (95). Through decorating CNTs with rhodium (Rh) or platinum (Pt) nanoparticles, Leghrib et al. (82) developed hybrid materials which can be used to selectively detect benzene with a detection limit below 50 ppb at room temperature (RT). In addition to Rh and Pt, silver (Ag) nanocrystals (NCs) have also been used to fabricate functional CNTs for gas sensing (83, 96). A fast and selective gas sensor for NH_3 detection at RT using Ag NCs multi-walled CNTs (MWCNTs) has been studied by Cui et al. (83). This study showed that the hybrid Ag-NCs-MWCNT sensor could efficiently improve the sensing performance of MWCNT sensor (Fig. 7a) and indicated a good stability (Fig. 7b).

Although metal decorated CNT sensors have demonstrated superior performance, the incorporation of these sensors into wearable devices remains challenging thus hindering their real application. On the other hand, Srikanth et al. (84) developed wearable gas sensors made of CNT bundles on cellulosics (paper and cloth), which can be used to detect aggressive oxidizing vapors (e.g. NO_2 and Cl_2) at 250 and 500 ppb, respectively, at RT. It was the first time that cellulosics was used as sensing substrates for reversible gas detection without thermal or photoirradiation methods. Furthermore, a high-performance wearable gas sensor has been developed by Kittipong et al. (85) based on vacuum-assisted spray layer-by-layer assembly technique. The technique can create highly porous networks of conformal MWCNT multilayers on individual porous electrospun fiber substrates. This work demonstrated that ultrasensitive sensing platform for real-time gas detection could be achieved using engineered textiles.

5.2. Graphene based gas sensors

Graphene and reduced grapheme oxide (RGO) (97) based materials have been widely explored for gas detection due to their large specific surface (2630 m^2 g^{-1}) (98) and high sensitivity of resistance upon gas molecule adsorption. It has been shown that graphene based gas sensors had high sensitivity when detecting different kinds of gases including NO_2, NH_3 and CO under low concentrations (99-101). The poor performance (poor selectivity, slow response and recovery characteristic) of gas sensor based on intrinsic graphene results in a number of studies of functionalized graphene (87, 102, 103).

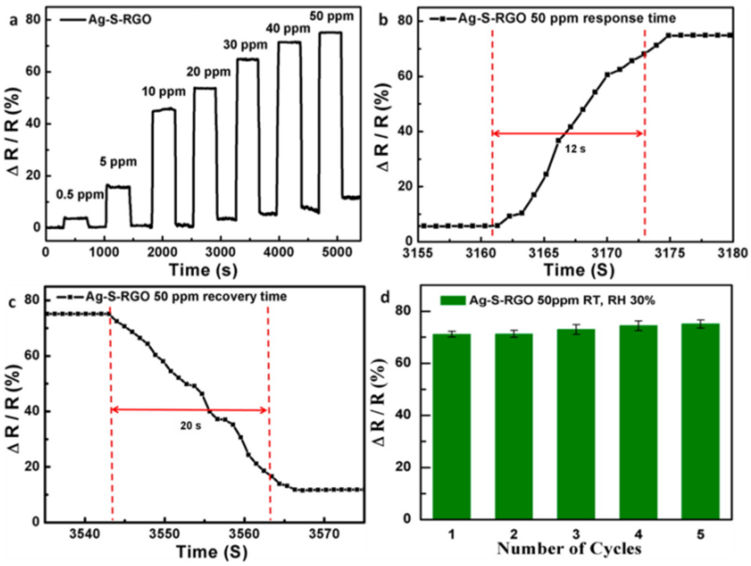

Fig. 8 a) Dynamic response of Ag-S-RGO sensor to different concentrations of NO_2 gas. b, c) Response−recovery characteristics of the Ag-S-RGO sensor. d) Five sensing cycles of the Ag-S-RGO sensor for detecting of 50 ppm of NO_2, indicating a good stability. Reprinted with permission from (87) . Copyright (2014), American Chemical Society.

Huang et al. (87) presented a fully printed Ag−S-RGO gas sensor through decorating Ag nanoparticles on sulfonated RGO (S-RGO). The Ag-S-RGO sensor has a much shorter response and recovery time (12 and 20 s, as shown in Fig. 8b and c, respectively) when compared with other sensors deriving from graphene. It also demonstrated good dynamic response and stability as illustrated in Fig. 8a and 8d respectively. This work shows that chemical modification of RGO is a promising way to make excellent gas sensors. On the other hand, Wang et al. (89) synthesized an gas sensor based on C-rich carbon nitride which prepared through thermal treating urea-derived porous graphene-C_3N_4 and glucose-derived carbon. The developed porous carbon nitride sensor showed a good sensitivity toward NO_2 and high selectivity to NO2 among other gases at RT. The same group later presented an uncomplicated method of fabricating of holey RGO based gas sensor (88). The developed sensor showed a sensitive detection of NO_2 in ppb levels and a reversible sensing to NH_3 monitoring, while UV irradiation need to be applied to speed up the desorption process. They found that functional decoration and structural modification could be jointly used to improve the sensing performance of RGO.

To address the problem of external power supplies, Yun et al. (90) demonstrated a gas sensor powered by a micro-supercapacitor (MSC) array, which were on the same deformable substrate. The patterned graphene NO_2 sensor developed through integration with MSC could work for over 50 min without using an external power supply. Furthermore, a dual-mode detection sensor combing electrical and visual detection was studied by Duy et al. (91). The sensor can work at both with and without external power conditions. When this dual-mode gas sensor was exposed to NH_3 gas, simultaneous measurements demonstrated that visual detection mode can work at around 45% RH at lower NH_3 concentrations with a faster response time (about 15s) and electrical detection mode can be used at below 80% RH with a slow response time (~3 min) (91).

Specifically, low-dimensional carbon based gas sensors have been widely studied in the development of novel devices as well as in embedding them into smart wearable platforms. However, the energy efficiency for real-time monitoring and the mechanical flexibility which guarantees a successful integration onto skin, textile and other substrates, are still insufficient to obtain wearable platforms. In order to achieve a better gas sensing performance and realize a wide range of real-time human health monitoring, more combination methods, for example integrating power supply or offering alternative method, can be explored in the future. Moreover, future work should focus on detecting and distinguishing multi-component gas mixture at the same time in daily monitoring.

5.3. Light sensors

The band-gap of graphene can by tuned by doping or varying device structures (79, 80). Photon absorption and carrier kinetics of graphene could be improved by integrating photosensitive nanostructures to graphene. Our group synthesized a ZnO nanorod/graphene heterostructure by using in situ growth method (81), and a visible-blind ultraviolet (UV) sensor combing the photosensitivity of ZnO nanorods and conductivity of graphene was developed. The sensor could detect UV only like 357 nm and deeper UV light, and cannot respond to visible light. Future research could focus on developing other photosensitive heterostructures by introducing other 1-D nanomaterials such as PbS, Si and GaN nanowires to graphene.

6. Wearable temperature sensors

Human body temperature is a crucial physiological factor in a human health monitoring system because temperature indicates the health condition of a human. Traditional temperature sensors are based on ceramic (104), Si nanoribbons (105) or serpentine metal (106), while they all suffer from a poor stretchability. Since a good stretchability is needed to integrate temperature sensors into wearable electronics, flexible and stretchable temperature sensors have been developed based on carbon materials such as (graphene (107) or CNTs (108)), organic semiconductors (109) and nanocomposites (110, 111). Among which, carbon based thermistors are the most commonly used temperature sensors for human health monitoring. Their details are presented in Table 5.

Table 5 Summary of the performance of carbon-based temperature sensors

Materials	Working range	Stretchability	Flexibility	Sensitivity [a]	Response time	Ref.
Graphene	30-100°C	50%	Yes	~0.1%/°C*	~20s	(107)
CNT	25-80°C	-	Excellent	~0.075% / °C*	~10s	(108)
CNT	21-80°C	-	Yes	~0.25%/°C*	<~2s	(110)
CNT	20-50°C	-	Excellent	~0.63%/°C*	-	(112)
RGO	30-80°C	-	Excellent	2.48□□10^{-7}/°C**	-	(113)
RGO	25-45°C	0.3%	Excellent	-	-	(114)
RGO	30-80°C	30%	Excellent	1.34%/°C*	-	(115)

[a] Sensitivity: * $=\frac{R-R_0}{R_0}\times\frac{1}{\Delta T}$; ** $=\frac{I_{DS}-I_{DS0}}{I_{DS0}}\times\frac{1}{\Delta T}$; R, sensor resistance at RT; R_0, sensor resistance at set temperature; I_{DS}, normalized current at RT; I_{DS0}, normalized current at set temperature.

Dinh et al. (108) developed a CNT-based thermistor which had a negative temperature coefficient of resistance (NTC), whereby the resistance of CNT yarn decreased with an increase in the temperature. The temperature sensor was integrated with a cellulose paper substrate to fabricate a wearable and portable temperature sensor. The temperature sensor showed almost linear characteristic with resistance change in the temperature range from 25 to 80 °C, and did not show any degradation, even in cyclic tests.

In addition, Harada et al. (110, 112) presented a flexible temperature sensor based on a printed poly (3, 4 - ethylenedioxythiophene) - poly (styrenesulfonate) (PEDOT: PSS) - CNT composite film which works with a temperature range between 21 and 80°C. The devices had a high sensitivity from 0.25 to 0.63% $°C^{-1}$ with a response time lower than 2 s. The results demonstrated that the temperature sensor integrated on a flexible substrate via a printing technique only was promising for applications in human health monitoring. Trung et al. (113, 114) fabricated a flexible temperature sensor integrating R-GO with poly(vinylidenefluoride-co-trifluoroethylene P(VDF-TrFE)) noncomposite as a sensing layer which was coated onto a flexible substrate to fabricate FET. This temperature sensor showed a high sensitivity (about 0.25% per °C) in the temperature range from 30 to 80 °C and it could even detect minute temperature changes as small as 0.1 °C.

In general, different types of temperature sensors based on carbon materials with various detection ranges have been fabricated to monitor human body temperature. However, the sensing ranges of these temperature sensors may be too wide for human body temperature measurement and the suitable ranges are suggested to be lying from 30°C to 50°C. Therefore, high sensitivity and stretchability, stability, precision and resolution are the critical requirements for wearable temperature sensors. Moreover, wireless transmission is a promising way to accelerate the application of temperature sensors in real time human health monitoring.

7. Wearable sensing network

Compared with individual sensor, sensing arrays and networks have enhanced capacity in providing surface or even 3-dimentional sensing functions on the spatial scale, therefore have been increasingly adopted in a number of wearable applications. According to the working principle, carbon based sensing array can be classified into three major types, resistive, capacitive and inductive ones, which correspond to diverse connection, networking, and reading methods. Among which carbon based resistive sensor arrays are the most widely used type in various fields, benefiting from their low cost, superior stability, and less complex networking and reading method.

To form a small scale wearable sensing array, as shown in the first mode in Figure 9 , a straightforward method is using two wires, signal wire and ground wire to connect each individual sensor. Thus $n \times 2$ wires are needed for n sensors. A smart glove system with five graphene based strain sensors attached to the finger joints was developed using 10 electrical wires (29), as shown in Figure 9a. A graphene based five pressure sensor array (Figure 9b) was presented (116). Shu et al formed a carbon black based sensor array using an n+1 wire structure in a wearable foot pressure monitoring system as given in Figure 9d, where n represents the signal wires and 1 refers to the common ground wire, which minimized the array complexity by reducing n-1 ground wires (44). The research group of University of Pisa developed a carbon loaded elastomer (silicone matrix filled with carbon black powder) type sensor array in a sensorized garment for kinesthetic monitoring (Figure 9c), where the same polymer/conductor composite was adopted as material for the connection tracks, avoiding the stiffness of conventional metal wires (10).

To form a large scale wearable resistive sensing array, as illustrated in the second mode in Figure 9, a×b wire structure can be used for an a×b sensor array, where a+b connection wires are necessary. Additional electrical chips are a must for addressing and reading, such as multiplexor and thin-film transistor (TFT). A 20×20 large area tactile sensor array was fabricated using printed carbon nanotube active-matrix backplanes, where the sensing unit was made of a silicone rubber coated with a conductive carbon (117). SWCNT TFT was also used as the row and column selection of a 5×5 stretchable active matrix temperature sensor array (118), as shown in Figure 9g. A 4×4 resistive strain sensor array was invented based on graphene transistors (119), and a 4×4 CNT doped PDMS based capacitive pressure sensor array (Figure 9f) was also fabricated in the a×b sensor array structure (120). In order to minimize redundant chips, a new resistive sensor array readout method was presented for wearable applications, and a 10×10 carbon black based pressure sensor array for sitting monitoring was forwarded (Figure 9e), which achieved a low-complexity and low-crosstalk error performance (121).

To realize a wearable surface mapping function, electrical impedance tomography (EIT) is adopted and it has been widely studied, which is illustrated as the third mode in Figure 9. A pressure mapping imaging device was developed using conductive fabrics (122). A pressure sensitive conductive rubber with conductive carbon filler was made for pressure distribution mapping and tactile distribution sensing in 2-D and 3-D surface mapping mode respectively (123, 124), as seen in Figure 9h and 9i.

To cover more body areas and to guarantee the wearing comfort, wire connected or wireless wearable sensing networks were studied, which is shown as the fourth mode in Figure 9. In a typical wearable sensing network as presented in Figure 9j and 9l, several body-worn sensor nodes are allocated on the human body (125). The placement criteria of sensor nodes for dynamic wearability limit the allocation to large areas that have low movement or flexibility even when the body is in motion. All the placement areas are found to be the most unobtrusive for wearable objects (126, 127). In practice, one-sensor-one-node mode or multiple-sensor-one-node mode can be used, which means the node connects with only one sensor or with multiple sensors. The wearer's physiological and physical information are extracted by sensors and electrodes, processed at the nodes and wirelessly transmitted to the coordinator by Bluetooth, wire connection (Figure 9h), zigbee, Wifi, etc. Fig.9 shows the major four connection and networking modes in this area, where the major challenges can be summarized as follows. On one hand, researchers have tried to increase the accuracy of the wearable sensing array and network, especially to decrease the crosstalk error in the addressing method. On the other hand, the complexity of the wearable array and network should be decreased including the system and connection complexity, as well as fabrication complexity. Some attempts have been conducted, such as reducing the wire number by improving addressing and reading method, and fabricating both sensing and connection components using the same carbon based materials.

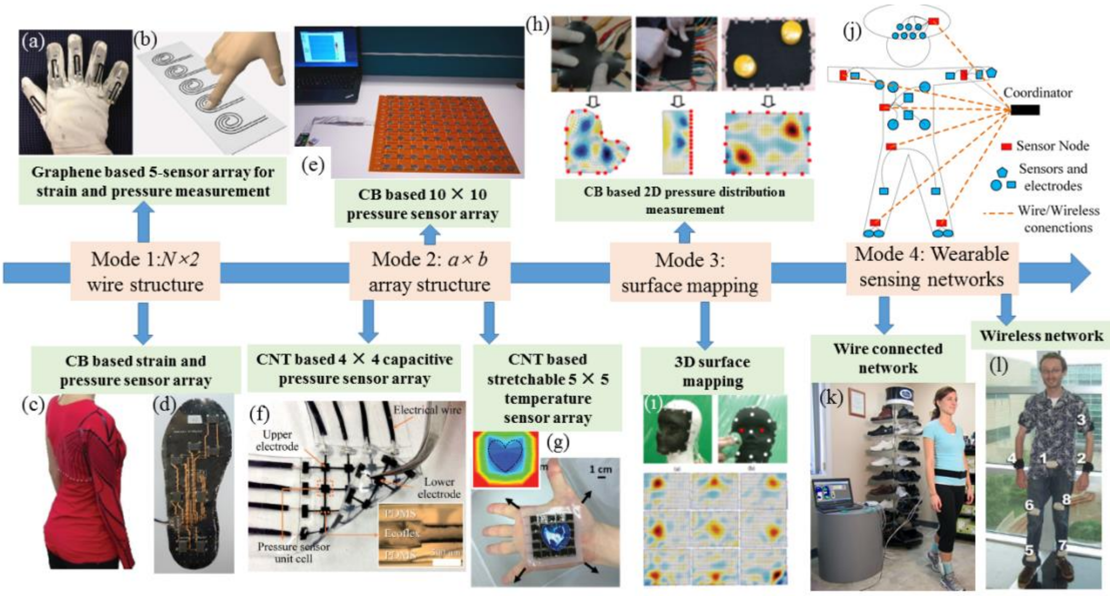

Fig. 9. Wearable carbon based sensing array and network: (a) Graphene based 5-sensor array in a smart glove system. Reprinted with permission from (29). Copyright (2014), Advanced Materials. (b) Graphene based 5-sensor array for pressure measurement. Reprinted with permission from (116). Copyright (2015), Ieee Electron Device Letters. (c) CB based strain sensor array in a sensing garment. Reprinted with permission from (10). Copyright (2007), Mrs Bulletin. (d) CB based pressure sensor array in an intelligent footwear system. Reprinted with permission from (123). Copyright (2007), Humanoids: 2007 7th Ieee-Ras International Conference on Humanoid Robots. (e) CB based 10×10 sensor array for sitting pressure monitoring. Reprinted with permission from (121). Copyright (2015), Ieee Sensors Journal. (f) CNT based 4×4 capacitive pressure sensor array. Reprinted with permission from (120). Copyright (2014), Journal of Materials Chemistry C. (g) CNT based 5×5 temperature sensor array. Reprinted with permission from (118). Copyright (2016), Advanced Materials. (h) CB based 2D pressure distribution measurement and (i) 3D surface sensing. Reprinted with permission from (123). Copyright (2007), Humanoids: 2007 7th Ieee-Ras International Conference on Humanoid Robots. (j) Configuration of a wearable sensor network. (k) Wire connected wearable network. Reprinted with permission from (128). (l) Wireless wearable sensor network. Reprinted with permission from (125), Copyright (2009), Ieee Journal on Selected Areas in Communications.

8. Promising applications of wearable carbon based sensors

As can be seen in the previous sections, the past decade has witnessed considerable progress in a variety of low-dimensional carbon based sensors, such as mechanical, gas, temperature and bio-potential sensors. Potential applications of these wearable sensors involve health and environmental monitoring, voice and motion detection, human-machine interfaces, etc.

8.1. Health monitoring

Benefiting from the advancement in medicine, public health as well as personal and environmental hygiene, human life expectancy has been rising through the past decade. The increasing life expectancy, however, together with the declining birth rate, brings about foreseeable aging population and imposes heavy burdens on these countries. It is therefore of great importance to develop low-cost wearable health monitoring devices for the sake of elderly healthcare (129). Such devices can also be used to monitor physiological signals of patients and people doing sports training in a noninvasive and unobtrusive manner. They may even change the way medical doctors diagnose and treat diseases. The vital physiological signals include heart rate, breathing rate and blood pressure, etc. Most can be detected using low-dimensional carbon based sensors.

The first are two main physiological signs, pulse and breathing rate. Pulse detection has been demonstrated using both wearable strain and pressure sensors, including those based on CB (29), graphene (32, 34, 43), as well as CNTs (48). However, the toxicity of CNT should be paid attention to, since pulse detection involves a direct and sometimes long-time skin contact. Another vital physiological parameter is respiration. Sensors comprising CB (15) , and graphene (34), can be applied for breathing rate monitoring. CNTs based pressure sensors with ultra-high sensitivity (47) can even detect breathing by air flow measurement. Data on continuous monitoring of respiration may provide new and useful information for breathing related diseases, especially at their early onset. Carbon based electrodes have demonstrated their viability for bio-potential (e.g. ECG, EEG and EMG) monitoring due to their excellent flexibility and biocompatibility (68, 74).

The above parameters will ensure an effective monitoring of human health condition thus make it possible to realize remote healthcare for the elderly.

8.2. Environment monitoring

Wearable low-dimensional carbon based sensors also played an important role in environment monitoring. Smart wearable carbon based gas sensors could detect different gases such as NH_3, NO_2 and Benzene which may threaten environment and people health (82-84). The developed portable carbon based gas sensors are believed to have tremendous potential in environment monitoring due to the change of air quality. Wearable low-dimensional carbon based temperature sensors make continuous monitoring of human temperature possible because these sensors have good sensitivity as well as excellent stretchability and flexibility.

8.3. Other applications

Low-dimensional carbon based sensors also enable motion capture and speech recognition. Herein, the motion capture includes finger movement (21, 29, 41), elbow and knee angles (10, 20, 22), body gesture (11), or even facial expressions (22, 24, 51). They may be used to guide and train dancers, sportsmen, actors, or used as control devices for human-machine interfaces. On the other hand, since a number of super-sensitive strain and pressure sensors are being developed, many showed the potential to recognize human speech (20, 24, 34, 39, 40, 47, 48, 51) by attaching the sensors onto throat. Speech signals were captured through the tiny movement or vibration of human throat.

Besides, our group demonstrated other possible applications of CB based wearable sensors. For example, in-vivo muscle girth measurement for studies on human muscles (14), pressure sensors for protective clothing against impact loading for seatbelt pressure recording during car crash (13), as well as i-Shoe (44), a pair of shoes mapping the foot pressure anytime and anywhere. The i-Shoe system has been successfully trial-used for diabetic foot syndrome detection in Hong Kong local hospitals.

9. Conclusions and future outlook

In this article, we conducted a comprehensive review on wearable low-dimensional carbon based sensors for health and environmental monitoring. Carbonaceous materials including CB, CNTs, and graphene are briefly introduced. Special attention has been raised on the toxicity of CNT materials for wearable applications. Then as a major part of the review, wearable sensors, from strain, pressure to gas, temperature sensors, and bio-potential sensors, as well as their sensing network have been carefully studied and discussed. Lots of highly sensitive sensors are being developed. It is evident that the appropriate and deliberate selection of low-dimensional carbon materials, matrix and substrate materials, and their interactions, as well as effective structural designs, are all important for high sensitivity and stable performance of the sensors.

However, despite of the notable progress in the past decade, few products can be found on the wearable market. It is possible that the following problems need to be addressed until successful application of wearable low-dimensional carbon based sensor devices.

(1) A more comprehensive evaluation of the sensors is lacking. Many studies focused on improving the sensitivity or gauge factor of sensors. Others aim at measurement of large strains. But few focused on a balance between the two. Much few conducted reliability test, including fatigue, washability, repeatability, temperature and humidity effect, etc. It should be emphasized that sensors are not sensing and responsive materials only. They must be stable, even after long-term daily use, and under diverse using conditions. On the other hand, softness and conformability need to be given serious consideration in the research, as well as viable and reliable encapsulation, which evidently has not been given sufficient attention in the last decade. Therefore, we recommend a more comprehensive evaluation of the sensors. Indeed, no sensor can be universally applied; nonetheless at least targeted applications should be specified with reasonable and qualitative specifications.

(2) No standard can be found to guide the research on wearable carbon based sensors. Unlike the traditional silicon based strain gauge and pressure sensor industry, that has well-established standards, research on wearable sensor technology now is following its unique path without clearly set targets. Standards in traditional sensor industry are not applicable here, since wearable sensors encounter more complex conditions, including large deformations, repetitive strains, bending, and pressures, the integration with garments, and the need to be machine washed after wearing or a

complete water-resistant encapsulation.

(3) Human and environmental safety needs to be emphasized. Safety is a universal issue in consumer electronics, particularly in healthcare products. This becomes an urgent issue since CNTs have potential hazards (2-4), but they are still being intensively researched for wearable uses. CB and graphene have significantly less hazardous concerns, and the former is more cost-effective. Therefore, more efforts are advised on improving sensitivity of CB based wearable sensors, assuring the safety of CNT based sensors, and realizing cost-effective roll-to-roll production of graphene based sensors.

(4) The last issue is an appropriate selection of the substrate. Wearable sensors are called wearable because consumers will wear them. Except for disposable sensors, textile materials, from fibers, yarns, to fabrics are all excellent substrates to build wearable sensors on, because they are soft, breathable, wearable, washable and durable. Material and electronic scientists are advised to work more closely with fiber scientist and textile engineers to better use textiles as substrate for wearable sensor applications.

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