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Recent developments in bio-monitoring via advanced polymer nanocomposite-based wearable strain sensors

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ABSTRACT

Recent years, an explosive growth of wearable technology has been witnessed. A highly stretchable and sensitive wearable strain sensor which can monitor motions is in great demand in various fields such as healthcare, robotic systems, prosthetics, visual realities, professional sports, entertainments, *etc*. An ideal strain sensor should be highly stretchable, sensitive, and robust enough for long-term use without degradation in performance. This review focuses on recent advances in polymer nanocomposite based wearable strain sensors. With the merits of highly stretchable polymeric matrix and excellent electrical conductivity of nanomaterials, polymer nanocomposite based strain sensors are successfully developed with superior performance. Unlike conventional strain gauge, new sensing mechanisms include disconnection, crack propagation, and tunneling effects leading to drastically resistance change play an important role. A rational choice of materials selection and structure design are required to achieve high sensitivity and stretchability. Lastly, prospects and challenges are discussed for future polymer nanocomposite based wearable strain sensor and their potential applications.

1. Introduction

With the recent advances in modern wearable bioelectronic devices and nanotechnology, great effort have been made in polymer nanocomposite based wearable strain sensors for healthcare and medical diagnosis, robotic systems, prosthetics, visual realities, professional sports, entertainments, among others (Rogers et al., 2010). Wearable devices can be attached onto clothes, garments or directly worn on the human skin by adhesive tapes or elastic straps for monitoring physical, biochemical signals and motions (Park et al., 2015b; Sazonov and Neuman, 2014). Mechanical flexibility is extremely important for wearable devices to minimize the discomfort of worn or attached electronics, in short, it should behave like the human skin (Sazonov and Neuman, 2014; Someya et al., 2016). Due to the soft, complaint nature of human tissues and the natural bending or rotational motion associated by joints, both the structure and materials should be soft and mechanically robust enough to bend, stretch, press, and twist in response to motions of wearer (Lu et al., 2016; Pantelopoulos and Bourbakis, 2010; Rogers et al., 2010; Salvatore and Tröster, 2015). Strain sensing is one of the most important applications of wearable sensor devices. Though many different types of flexible strain sensors have been developed or reported, including fiber Bragg grating (FBG), raman shift, liquid metals, triboelectricity and piezoelectricity based strain sensors (Chossat et al., 2013; Gullapalli et al., 2010). However, the complexity of fabrication, poor dynamic performance, low resolution and requirement of advanced measurement devices have constrained their applications. In contrast, resistive-type and capacitive-type strain sensors are more practical and popular due to their low cost, ease of fabrication and integration, easy read-out unit, high flexibility and high strechability (Cai et al., 2013; Larimi et al., 2018). They respond to the mechanical deformations by the change of resistance or capacitance. Conventional strain sensors are inexpensive; however, they typically have very limited stretchability (< 5%) and bulky components (Amjadi et al., 2014; Monty et al., 2013; Wujcik et al., 2013).

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Table 1
Mechanical properties of some common materials (Someya et al., 2016).

Material	Young's Modulus	Elongation at Break
Silicon	130 GPa	1%
Bone	~ 20 GPa	1%
Plastics	1 GPa	5%
Elastomer	0.01–10 MPa	50–4000%
Gel	1–1000 kPa	10–2000%

The demand for high performance wearable strain sensor has dramatically increased with the advances of science and technology. Developing novel stretchable sensor which could sustain a high strain with the preservation of conductive pathways is a key point (Zhao et al., 2017a). Conventional semiconductors including silicon, and metal oxide films possess an intrinsic brittle and rigid nature, which limits their applications in wearable devices requiring intimate integration with curvilinear surfaces (clothes, human skin, etc.) and large deformation (bending, stretching, twisting, etc.) (Blasdel et al., 2015; Yao and Zhu, 2015). Improved conformability and bendability can be achieved by using ultrathin films of silicon or metal oxide semiconductors. However, reducing the thickness does not change the Young's modulus and stretchability (low elongation at break) of the silicon film which make it impractical for strain sensors (Table 1). To address this challenge, there are generally two main strategies in designing stretchable strain sensors: "structures that stretch" and "materials that stretch" (Khang et al., 2006). A polymer nanocomposite based strain sensor typically has two main components, one is the conductive network (act as the sensing element) which can provide conductive pathways to produce signal, and the other component is an elastomeric polymer to provide flexibility, stretchability and protection for conductive network (Zhao et al., 2017a). As for the operation of stretchable strain sensors, it should be easy to establish as they are no-invasive wearables at current stage of applications. Due to the fact that signals are produced by the physical strain changes of underlying subjects such as skin, clothing, etc. the strain sensor does not require intimate direct contact with skin as long as the adhesion is strong enough to continuously experience the same degrees of strain for accurate monitoring. The encapsulation of strain sensor using an appropriate biocompatible, stretchable barrier polymer (such as silicone based elastomers: poly(dimethylsiloxane) (PDMS), Ecoflex, etc.) or adhere strain sensor onto skin tight clothes can help prevent the direct interaction with body fluids and external environment which may interfere the receiving resistive or capacitive signals. The majority of reported elastomeric polymers (See Section 4.1 for details) as matrix have a wide useful working conditions as they are chemically inert, not light sensitive, waterproof in temperature ranges between − 45 to 200 °C. Note that some types of conductive fillers might limit the useful working conditions.

This review focuses on the background and recent advances of polymer nanocomposite based wearable strain sensors in four sections: First, strain sensing mechanisms are introduced. Second, performance metrics such as stretchability, gauge factor (GF), linearity, hysteresis and features including self-healing, transparancy, *etc*. of recent reported strain sensors are discussed. Third, the choice of materials selection including elastomeric substrate and conductive nanomaterials are reviewed. Finally, we point out some challenges that need to be addressed to make current stretchable strain sensors more practical in future applications.

2. Sensing mechanisms

Because polymer nanocomposite based strain sensors have multiphase, designed structures and conductive networks constructed by nanomaterials, their sensing mechanisms are different from conventional strain gauges which solely dependent on geometric and intrinsic piezoresistive effects. For stretchable resistive-type strain sensors, the change of resistance are mainly due to disconnection of conductive fillers, crack propagation at large strain and tunneling effect between closely positioned conductive fillers. Strain is defined as the ratio of the change in length of a material to the original length, as shown in Fig. 1. Strain can be positive when material is elongated by tensile stress, while it can also be negative when compressed. There are four types of strains: axial, bending, shear, and torsional, in which axial and bending strains are most common.

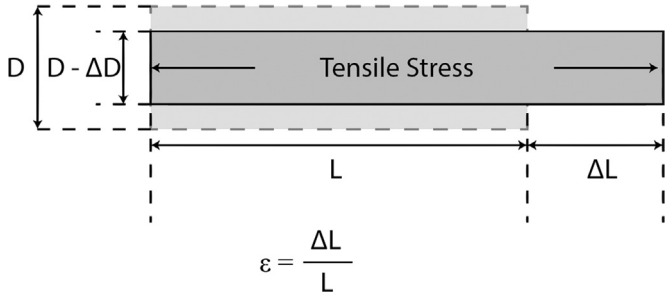


Fig. 1. Illustration of strain in materials.

2.1. Geometric effect

Geometric effect is the main mechanism for capacitive-type strain gauges. This effect also plays a role in highly stretchable resistive-type strain sensors but is believed not as crucial as other mechanisms. When the materials are stretched, they tend to contract in transverse directions to the stretched direction. Similarly, when they are compressed, they tend to expand in transverse directions following Poisson's ratio of ν. For highly stretchable elastomers, the Poisson's ratio value is nearly 0.5. From Ohm's Law, the resistance of a conductor is given by $R = \rho L/A$, where ρ is the electrical resistivity, L is length and A is the cross section area of the conductor. When the conductor is at a stretched state, L increases while A decreases, which makes an increment in measured resistance (Hempel et al., 2012). However, for resistive-type strain sensor, the large degree of deformation takes place at elastomer matrix phase, while conductive fillers slip out each other or conductive network sustains a structure deformation. The geometric change of conductive materials is extremely limited. For capacitive-type strain sensor, it can be treated as a simple parallel-plate capacitor where the overlapped length is l_o, width is w_o, and thickness of dielectric layer is d_o (Yao and Zhu, 2014). The initial capacitance is given by Eq. (1):

$$C_0 = \varepsilon_0 \varepsilon_r \frac{l_0 w_0}{d_0} \tag{1}$$

Where ε_o and ε_r are the electric and dielectric constant of dielectric layer, respectively. At strain ε, the length l are stretched to $(1 + \varepsilon)\ l_o$, while width and thickness are contracted to $(1- \nu_{\text{electrode}}\ \varepsilon)\ w_o$ and $(1- \nu_{\text{dielectric}}\ \varepsilon)\ d_o$, respectively, the capacitance will be:

$$C = \varepsilon_0 \varepsilon_r \frac{(1+\varepsilon) l_0 (1- \nu_{electrode}\varepsilon) w_0}{(1- \nu_{dielectric}\varepsilon) d_0} \tag{2}$$

For most stretchable strain sensors, the Poisson's ratios of electrode and dielectric layers are close to 0.5. The capacitance at stretched state is close to $C = (1 + \varepsilon)C_0$, giving capacitive-type strain sensor an excellent linearity (Yao and Zhu, 2014). However, some studies reported that when strain is too high, the linear relationship will not be valid anymore (Amjadi et al., 2016).

2.2. Piezoresistive effect

The piezoresistive effect is a change in electrical resistivity of materials when a mechanical strain is applied. For metals and

semiconductors, the change of resistance at strain ε can be written as:

$$\frac{\Delta R}{R} = (1 + 2\nu)\varepsilon + \frac{\Delta\rho}{\rho} \tag{3}$$

Where $(1 + 2\nu)\varepsilon$ is the geometric effect, and $\frac{\Delta\rho}{\rho}$ is the fractional change in resistivity (intrinsic piezoresistive effect) (Barlian et al., 2009). Piezoresistive effect for metal is as small as around 0.3. However, for semiconductors such as silicon and germanium under certain directions, the change in resistivity can be 50–100 times larger than the geometric effect due to the change of bandgap and inter-atomic spacings. Bulk semiconductor strain sensor can have a very high sensitivity, but they are not suitable for wearable applications due to limited stretchability and flexibility. Stretchable polymer nanocomposite based strain sensors combine the merits of elastomeric materials and conductive nanomaterials. The piezoresistive effect is low for composite strain sensors as the large mismatch of elasticity between conductive network and elastomeric polymer leading to a relatively low adhesion.

2.3. Disconnection mechanism

Conductive nanomaterials form a conductive network inside elastomer matrix which produces signal relating to the strain. Electrons can pass through the connection of overlapped nanomaterials within the percolation network. When the strain sensor is stretched, especially at a large strain, the overlapped nanomaterials tend to disconnect as they have a much smaller elongation at break and a higher Young's modulus compared with elastomeric matrix. The disconnection of conductive nanomaterials blocks the pathway of electrical connection, consequently decreases the conductivity of the strain sensor. As the strain increases, the effect of disconnection mechanism increases as more overlapped nanomaterials slip until a critical strain is reached where resistance goes infinity. The performance of strain sensor greatly relies on disconnection and reconnection process. Permanent disconnection of overlapped nanomaterials causes the irreversible degradation of strain sensing accuracy.

2.4. Crack propagation

Fatigue crack propagation is the most common type of structural failure caused by static or cyclic loadings, especially for polymers (Ramsteiner and Armbrust, 2001). Cracks are initiated at points of high stress and defects. During the subsequent stretching, the initiated cracks will grow and propagate till at a point the material suffers fracture. Within the critical strain and attributed to viscoelastic polymer matrix, the electrical conductivity is limited by crack propagation in nanomaterials and strain sensor is able to resume its initial conductivity by closing the cracks when applied strain is removed. This crack propagation mechanism is more dominant for 2D nanomaterials such as graphene or polymer thin films than 1D nanowires. Kang et al. reported an ultrasensitive graphene-on-polymer strain sensors with a gauge factor of 6×10^6 at 8% strain (Li et al., 2012). A graphene woven fabrics (GWFs) prepared from chemical vapor deposition (CVD) was placed on top of elastomeric poly(dimethylsiloxane) (PDMS). By applying strain around 1–8%, a significant amount of cracks in GWFs were initiated, propagated, and broke causing a significant resistance change. The current pathway through a fractured GWF is shown in Fig. 2.

2.5. Tunneling effect

For resistive-type polymer nanocomposite strain sensors, disconnection mechanism typically has a dominant effect. However, when the separation distance between conductive nanomaterials and polymer matrix is sufficiently small, electrons can pass through forming a complete pathway through tunneling effect. John Simmons had

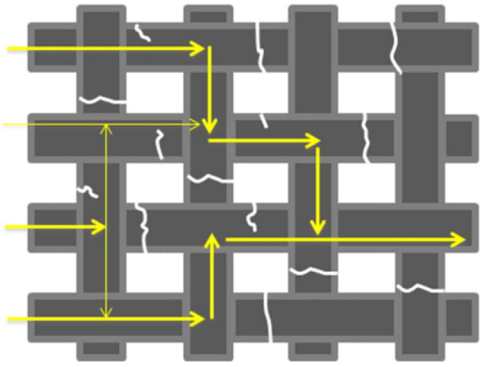


Fig. 2. Current pathway within fractured GWF under stretched state. Reproduced with permission (Li et al., 2012), Springer Nature.

formulated an equation (Eq. (4)) to calculate the electric tunneling resistance:

$$R_{tunnel} = \frac{V}{AJ} = \frac{h^2 d}{Ae^2\sqrt{2m\lambda}} exp\left(\frac{4\pi d}{h}\sqrt{2m\lambda}\right) \tag{4}$$

where J is tunneling current density, V is electrical potential, e is the quantum of electricity, m the mass of electron, h Plank' constant, d the distance between conductor and elastomer, λ the height of energy barrier of elastomer, and A for cross-section area of the conductor. Hu et al. has conducted a comprehensive study on the tunneling effect on a carbon nanotubes (CNTs)/polymer nanocomposite strain sensor (Hu et al., 2008). For a small CNT volume fraction, the piezoresistivity is weak while tunneling effect is considered to be the principal mechanism of the sensor under small strain observed both from experiments and numerical simulation.

3. Performance and features of strain sensors

To design a high-performance strain sensor, performance metrics include but not limited in sensitivity (i.e. Gauge Factor, GF), linearity, stretchability, response speed, durability, hysteresis, fabrication cost, simplicity, safety, ease of packaging and some preferable features such as self-healing, self-cleaning, self-powering, transparency, printability, *etc*.

3.1. Gauge factor (Sensitivity)

The simplest performance metric is the gauge factor (GF) which describes the slope of the relative change of electrical signal (resistance or capacitance) vs. applied strain, seen below in Eq. (5):

$$GF = \frac{\Delta R}{R_0\varepsilon}, \quad GF = \frac{\Delta C}{C_0\varepsilon} \tag{5}$$

Where ΔR or ΔC is change of resistance or capacitance, R_0 or C_0 is the initial resistance or capacitance at strain $\varepsilon = 0\%$, and ε is the applied strain. GF represents the sensitivity of a strain sensor. Conventional metallic foil and semiconductor based strain gauge possess a relatively high GF (2–5 for metal foil and 100–1000 for semiconductors) while the limitation in stretchability make them impractical for wearable devices. With the recent developments of nanotechnology and microelectronics, highly stretchable strain sensors with superior GF values have been continuously reported (Frutiger et al., 2015; Larimi et al., 2018; Slobodian et al., 2018; Yan et al., 2014). For capacitive-type based strain sensors, the main sensing mechanism is geometric effect. The limitation of geometric change have limit the GF value of capacitive-type strain sensor within 1. For resistive-type based strain sensors, the main mechanisms affect GF include disconnections between conductive blocks, crack propagation in the thin films, and tunneling effect. Considerable structure change leads to a significant change of resistance

Table 2
Summary of some recent reported polymer nanocomposite based strain sensors.

Material	Sensing Mechanisms	Stretchability	Gauge Factor
Ag NP-SBS (Park et al., 2012)	Resistive	200%	25
Ag NW-PDMS (Kim et al., 2015)	Resistive	35%	20
Ag-PU fiber (Ma et al., 2015)	Resistive	150%	211
Crumpled graphene-PDMS (Yan et al., 2014)	Resistive	100%	7.1
PEDOT:PSS-PU (Seyedin et al., 2014)	Resistive	350%	6
Ag NP/CNT-PDMS (Takei et al., 2014)	Resistive	2.4%	95
PEDOT: PSS-polyvinyl pyrrolidone (PVP) (Sun et al., 2013)	Resistive	4%	360
Graphene-Plastic (Hempel et al., 2012)	Resistive	2%	150
Ag NW-Hydrogel (Lee et al., 2012)	Resistive	460%	1.52
Pt-PUA (Kang et al., 2014)	Resistive	2%	2000
Graphene Mesh-PDMS (Li et al., 2012)	Resistive	8%	6×10^6
AuNWs-Latex Rubber (Gong et al., 2015)	Resistive	350%	9.9
Ag NW-Ecoflex (Yao and Zhu, 2014)	Capacitive	50%	0.7
CNT-silicone (Cohen et al., 2012)	Capacitive	100%	0.99
Ionic fluid-silicone (Frutiger et al., 2015)	Capacitive	700%	0.35

which in turn results in a high GF value. Amjadi et al. designed a strain sensor with a sandwiched structure of PDMS/Silver Nanowires (AgNWs)/PDMS (Amjadi et al., 2014). The elastomer nanocomposite based strain sensor showed a tunable GF within the range of 2–14 and a maximum strain of 70%. The AgNWs layer endorsed the electrical conductivity to the sensor and the GF was controlled by the number density of AgNW percolation network. Lower number density gives a rising amount of disconnections between AgNWs which considerably increases the resistance of the device giving a high GF value. Cai et al. designed a capacitive-type strain sensor based on CNT film on PDMS film. The sensor is found to detect strains up to 300% with excellent durability (Cai et al., 2013). The gauge factor is very close to 1 which is the theoretical limitation value for capacitive-type strain sensor.

3.2. Stretchability

As mentioned in introduction section, the stretchability of polymer nanocomposite based strain sensor derived from elastomeric matrix made of polymeric materials and stretchable structure of conductive network. Table 2 shows a summary of the materials selection, gauge factor and stretchability of some recent reported polymer nanocomposite based strain sensors. 1D nanomaterials such as CNTs and AgNWs are considered preferable than other nanomaterials such as carbon blacks (CBs), graphene, and silver nanoparticles (AgNPs) because of their high aspect ratios, and capability to sustain a percolation network at large strain. The selections of elastomeric polymer matrix and conductive nanomaterials will be discussed in detail in Section 4: Materials Selection for Stretchable Strain Sensors.

For conductive nanomaterials including carbon nanomaterials, metallic nanomaterials or conductive polymers, none of them is intrinsically elastic enough to bear a strain over 5%. So a designed conductive network which can retain mearsurable electrical conductivity at a high strain while effectively transfer the tensile stress to elastomeric matrix is needed. There are mainly two effective approaches in fabricating a stretchable strain sensor: 1. Simultaneous blending of conductive materials and stretchable matrix. 2. Construct a stretchable conductive network (such as buckling, spring, coiled, and open mesh structures, etc.) embedded inside an elastic matrix. The first approach is both simple and scalable while typically produces uniform nanocomposites. However, there are issues need to be addressed including: a homogeneous dispersion should be achieved for good linearity of signal, an insulating polymeric matrix reduces the conductivity of the device, a high percolation threshold needs a relative high loading of conductive fillers, *etc*. The second approach can provide high conductivity, especially at large strain compared with the first method. But the fabrication procedure is generally more complex, the stretch direction is often limited, and mechanical property can be low due to the poor adhesion between conductive layer and polymer layer. Seyedin et al. successfully fabricated a strain responsive polyurethane (PU)/poly (3,4-ethylenedioxythiophene): polystyrene sulfonate (PEDOT: PSS) elastomeric composite fibers by wet-spinning techniques (Seyedin et al., 2014). The conductive PEDOT: PSS was homogenously dispersed in PU matrix. The composite fiber shows an increased Young's modulus and yield stress but a decreased elongation at break due to the presence of conductive polymer. The electrical conductivity of $\sim 25\ S\ cm^{-1}$ can be achived at 25% loading. A model simulates the reversible and irreversible deformations of PEDOT: PSS conductive network within PU was proposed. Lipomi et al. (2011) designed a strain sensor composed of CNTs/PDMS. CNTs were spray-coated onto the pre-stretched substrate and forming a spring-like structure after releasing the strain. The nanotube film can accommodate strains up to 150% with an excellent conductivity of $2200\ S\ cm^{-1}$.

3.3. Linearity

Linearity is another performance parameter for strain sensor. A linear fit of gauge factor or change of resistance over applied strain can provide the strain sensor with high accuracy and simple calibration. Unfortunately, most reported resistive-type strain sensor exhibit a poor linearity, especially at large strain. The nonhomogeneous morphology upon stretching is believed to be the main reason of nonlinearity. Calibration curve is required to correlate the signal to strain value and periodic recalibration is recommended. Yu et al. (2017) developed a wearable strain sensor composed of carbonized nano-sponge encapsulated by silicone rubber. Their sensor showed three different gauge factors (GF of 4.3, 18.4 and 8.4) for strain ranges of 0–10%, 10–25%, 25–40%, respectively.

Capacitive-type stretchable strain sensor showed an excellent linearity as discussed in Section 2.1: Geometric Effect. But the linearity is limited due to the variation of Poisson's ratio at large strain. Yao and Zhu (2014) fabricated a highly stretchable pressure and strain sensor based on AgNW/PDMS/Ecoflex films. The AgNW was patterned on PDMS by screen printing, and ecoflex film as dielectric material was orthogonally placed between two AgNW/PDMS films. The sensor showed a linear response to strains up to 50% with a GF of 0.7 (Fig. 3). In addition, the sensor showed a fast response time of 40 ms and good pressure mapping function.

3.4. Hysteresis

Hysteresis is an important metric for wearable strain sensors especially when monitoring dynamic motions such as walking, running, speaking, heartbeat, breathing, *etc*. For capacitive-type strain sensors, the hysteresis is usually negligible while resistive-type strain sensors

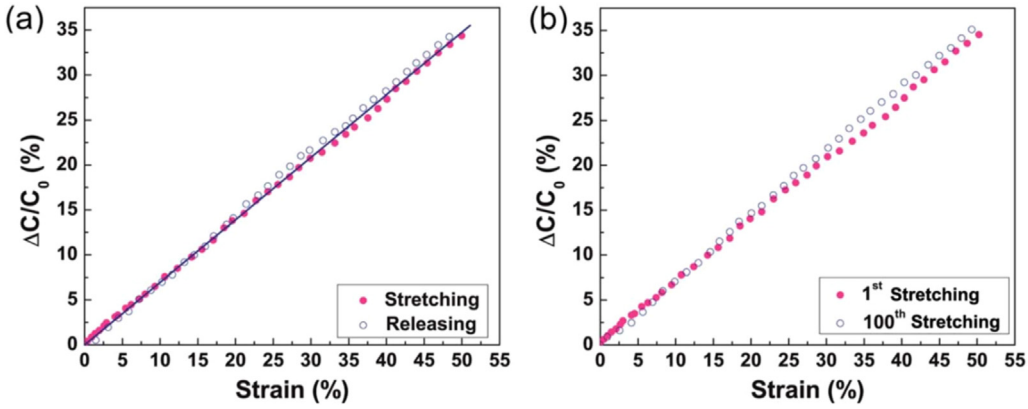
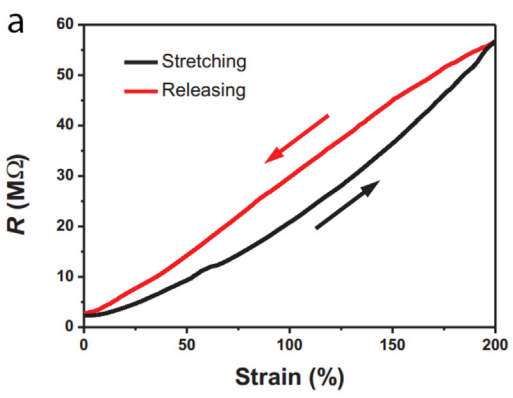


Fig. 3. Strain sensing performance: a) Linear fit of strain sensing capability of the capacitive sensor up to 50%. b) Relative capacitance change for 1st and 100th stretching of the capacitive sensor. Reproduced with permission (Yao and Zhu, 2014), Royal Society of Chemistry.



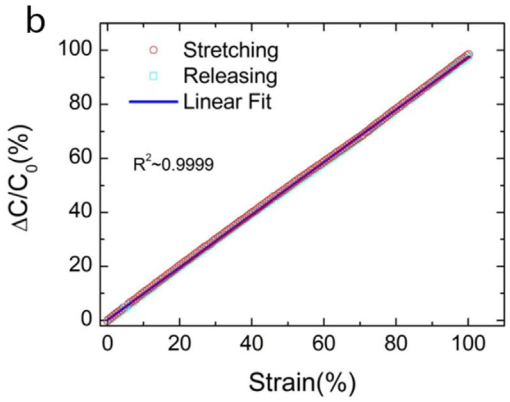


Fig. 4. Hysteresis: a) AuNWs-latex rubber sensor: Resistance change as a function of strain at 3 mm S^{-1} exhibiting hysteresis of resistance change. Reproduced with permission (Gong et al., 2015), John Wiley & Sons. b) CNT-PDMS sensor: Relative capacitance change as a function of strain with negligible hysteresis. Reproduced with permission (Cai et al., 2013), Springer Nature.

typically suffer large hysteresis (Fig. 4) (Gong et al., 2015; Yao and Zhu, 2014). Capacitive-type strain sensor relies on the geometry of overlapped dielectric layer between two electrodes. Though polymeric elastomer is known for elastic hysteresis caused by energy dissipation due to material internal friction. However, because capacitance is directly related to the strain, not stress, based on Eq. (2), the effect of stress-strain loop caused hysteresis is negligible. For resistive-type strain sensor, because of their unique conductive mechanisms, the stretchable conductive network needs time to slide back to its original position. Higher frequency is likely to cause incomplete deformation leading to an amplitude decay. The stronger binding with the matrix can help reduce the effect of hysteresis (Gong et al., 2015).

3.5. Self-healing

Self-healing materials have attracted tremendous attention in various fields due to their great potential for structural restoration and function recovery after mechanical damage (Kuang et al., 2018; Liu et al., 2018; Wang et al., 2018a; Yang et al., 2018b). When materials are used for a long time, they are often damaged caused by mechanical stress and/or harsh environment (e.g. temperature, pH, light, etc.) which are responsible for the loss of stability and shortening cycle life. Self-healing materials can prevent or reduce damage and thereby extend the durability and stability. Self-healing materials are usually categorized into two distinct systems: intrinsic and extrinsic self-healing materials (Liu et al., 2017). Intrinsic self-healing materials can heal the damage through inherent reversible bond without additional self-healing agents. Intrinsic self-healing processes can be accomplished through dynamic physical or chemical interactions including dynamic covalent bond (Lai et al., 2016), polymer chains entanglement, hydrogen bond (Tee et al., 2012) and non-covalent interactions such as ionic bond (Xu et al., 2016), metal coordination (Li et al., 2016a), *etc*. On the contrary, extrinsic self-healing materials rely on external healing agents to repair damages. Extrinsic self-healing can self-heal their damage by taking advantage of healing agents encapsulated in vehicles such as microcapsules/microvascular (Patrick et al., 2014; Zhu et al., 2015), and guest/host structures (Highley et al., 2015). When there is a crack or damage, incorporated healing agents are released and diffused to the damaged areas to repair defects.

To create self-healable strain sensors, self-healing properties and sensing properties should be incorporated into one device. Based on the intrinsic self-healing concept, Kuang et al. utilized a shape memory polymer, and developed a novel 3D printable semi-interpenetrating polymer network (semi-IPN) elastomer ink using photocurable resin (aliphatic urethane diacrylate) and thermoplastic polymer (polycaprolactam, PCL) (Kuang et al., 2018). After 3D printing and UV irradiation, semi-IPN polymeric structure showed excellent flexibility and strechability up to 600% (Kuang et al., 2018). When the film was scratched by a sharp blade, scratch could be healed by heating the film at 80 °C for 20 min in an oven. This self-healing behavior originated from entanglement of polymer chains and hydrogen bonding between PCL and urethane (Kuang et al., 2018).

The application of external temperature to initiate the healing process may restrict the wide application of the film, which motivate researchers to develop room temperature self-healable composites. Wang *et al.* developed a highly stretchable and self-healable ternary polymer (polyaniline/polyacrylic acid/phytic acid, PANI/PAA/PA) hybrid via a facile in-situ polymerization process, which can heal the cracks at ambient temperature (Wang et al., 2018a). The fabricated ternary system exhibits high stretchability (500%) and electrical conductivity (0.12 S cm^{-1}). The polymer composites showed excellent self-healing properties at ambient temperature (60% Relative Humidity &

25 °C) with mild pressure to come in contact and 99% healing efficiency of mechanical and electrical properties through hydrogen bonding and electrostatic interactions after 24 h healing period (Wang et al., 2018a).

Similarly, Liu et al. (2018) also synthesized an ionic cross-linked double network (DN) hydrogel using polyethylene glycol (PEG)/poly (acrylamide-co-acrylic acid) (PAMAA) with Fe^{3+} infusion into the network with self-healing ability at room temperature. The resulting DN hydrogel can self-heal the impair because of reversible hydrogen bonds and coordination bond, exhibiting impressive self-healing capability and high mechanical strength. The self-healing properties of the DN hydrogel monitored using two pieces, one piece dyed with rhodamine B, kept in contact for 12 h at room temperature. The healed hydrogel showed high stretchability (15 times higher) compared to pristine hydrogel indicates excellent healing efficiency. The incorporation of Fe^{3+} ions also contributed increasing healing efficiency with the increase of the ions content (Liu et al., 2018).

3.6. Transparency

Aesthetics also play a key role in the acceptance of wearable devices. For most applicable scenarios of strain sensor, they can be seen by others which means transparency is an attractive feature. Transparent thin film wearables can be accepted by more consumers for catching less attention. There are many transparent polymeric elastomers but most conductive fillers are opaque especially carbon nanomaterials. Design an ultrathin film with low loading of transparent nanomaterials, such as doped graphene, AgNWs and conductive polymers, can help increase the transparency of stretchable conductive film (Zhao et al., 2017a). Lee et al. (2014) fabricated a CNTs/AgNWs/Ecoflex hybrid nanocomposite with high flexibility, stretchability and transparency. The nanocomposites combine the enhanced mechanical compliance, conductivity and optical transparency of small CNTs (d ~ 1.2 nm), and the highly conductive AgNWs backbone to provide an efficient multi-scale electron transport path. The hybrid nanocomposite possesses a transmittance over 90% and stretchability over 460% (Fig. 5).

3.7. Other features

Some other favorable features include but not limited to powering, self-cleaning, printability, *etc*. There have been very limited literatures reporting stretchable strain sensors possessing these features. Wearable devices require long power life with limited power supply while not sacrificing performance. Most current wearable strain sensor uses data acquisition devices connected by wires. Integration with power system can free up wires making devices truly wearable. Self-cleaning surface can keep wearable device clean despite its surroundings. It is especially important when used in sports or gaming situations where significant amount of sweat is produced and could affect the accuracy of strain sensor. Self-cleaning surface with superhydrophobicity can help sweat slip out and keep device surface clean and dry. However, combining high stretchability and superhydrophobicity is challenging. Cost is always a major consideration to commercialize wearable devices and make them available for most consumers. To achieve scalable production, the design of fully printed electronics is desirable which help automate the fabrication process thus decreasing the cost and manufacturing the standardized products.

4. Materials selection for stretchable strain sensors

The recent advancement of materials science and nanotechnology have greatly contributed to develop next-generation high performance wearable sensors that possess high flexibility, stretchability, and sensitivity with wide detecting range. To obtain these desired properties, it is important to select suitable functional materials and rationally design their architectures. So far, various conductive nanomaterials have been utilized for polymer nanocomposite based strain sensors, including carbon blacks (CB) (Ke et al., 2017; Shintake et al., 2018; Wang et al., 2018b; Wu et al., 2016; Zheng et al., 2015; Zhu et al., 2018), CNTs (Akhmadishina et al., 2013; Christ et al., 2017; Lin et al., 2017; Wang et al., 2014a; Zaporotskova et al., 2016; Zhang and Zhang, 2009), graphene (Li et al., 2015; Liu et al., 2015; Wang et al., 2015; Yang et al., 2017; Zhao et al., 2017b), AgNWs (Hu et al., 2010; Sinha et al., 2016; Wei et al., 2016; Zou et al., 2018), Cu nanowires (Cu NWs) (Hu et al., 2014; Jason et al., 2016; Jung et al., 2016), Au nanowires (Au NWs) (Wu et al., 2017), and conductive polymers such as polypyrrole (PPy) (Li et al., 2014; Yang et al., 2016), polyaniline (PANI) (Lin et al., 2017; Wujcik et al., 2014), and PEDOT:PSS (Choong et al., 2014; Lee et al., 2016; Zhan et al., 2017). These conductive materials provide electrical conductivity that is the basis of sensing mechanism, but often show unsatisfactory mechanical properties such as poor flexibility and stretchability (Bae et al., 2013; Park et al., 2015a; Wang et al., 2014b). For example, graphene can endure strain only up to about 7% that is far below the criteria for stretchable strain sensors. Therefore, it is desirable to create hybrid composite materials having both high sensitivity and good stretchability/flexibility (Amjadi et al., 2016). These two properties can be attained by fabricating composites that consists of electrically conductive fillers and elastomeric matrix, in which conductive fillers provide sensing mechanism and elastomeric matrix endows desired mechanical properties. This part will introduce a couple of key stretchable/flexible matrix materials and electrically conductive materials considering their sensitivity, flexibility and stretchability.

4.1. Stretchable/flexible elastomeric matrix

Flexible matrices are supporting materials of strain sensors to provide desirable mechanical flexibility and versatility. Ideally, flexible matrix materials should have good mechanical and thermal properties, chemical inertness, low cost, ease processability and good adhesion

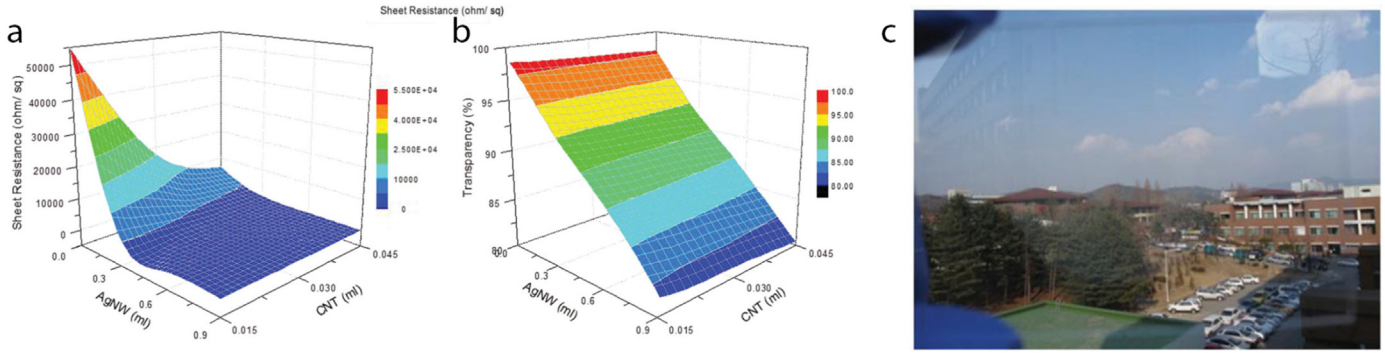


Fig. 5. Surface plot of sheet resistance (a) and transparency (b) for different concentration of CNT/AgNWs concentration. c) High transparency of electrical conductive CNT/AgNWs nanocomposite film. Reproduced with permission (Lee et al., 2014), John Wiley & Sons.

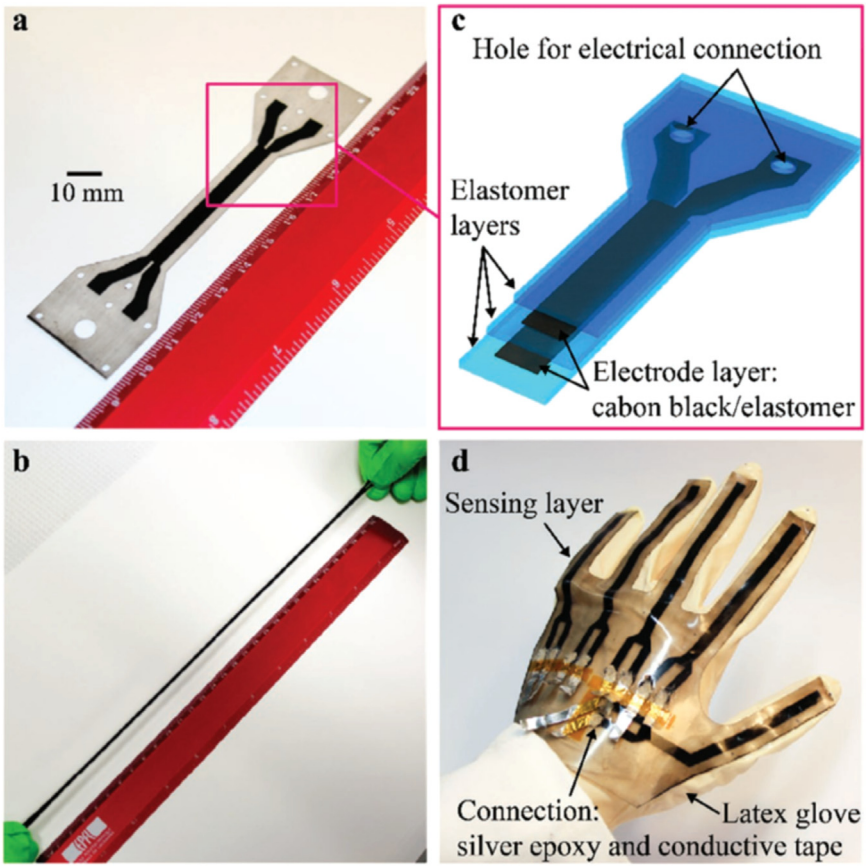


Fig. 6. Illustration of highly stretchable carbon black composite electrodes. a) Fabricated strain sensor. b) Stretchability up to 500%. c) Structure of strain sensors including electrode layers and dielectric layers. d) A smart glove integrated with five strain sensors. Reproduced with permission (Shintake et al., 2018), John Wiley & Sons.

with other functional materials. Most commonly used supporting materials include silicon based elastomers (such as PDMS) and rubbers, PU foam (Chen et al., 2017; Hsu et al., 2017; Park et al., 2016), polyethylene terephthalate (PET) (Hsu et al., 2017), polyimide (PI) (Kaltenbrunner et al., 2013), polyvinylidene fluoride (PVdF) (Zhou et al., 2017b), cotton (Li et al., 2017; Wu et al., 2016), paper (Li et al., 2016b), Ecoflex (Cheng et al., 2017; Lim et al., 2016; Ryu et al., 2015; Zhang et al., 2017), Dragon skin (Cai et al., 2013; Cheng et al., 2017; Frutiger et al., 2015), and Kevlar fiber (Lee et al., 2015a) for flexible strain sensors. Among them, PDMS has been most widely used due to its tunable mechanical properties, facile processability, chemical inertness, low cost, and high tendency to adhere with other functional materials (Larmagnac et al., 2014). The Young's modulus of PDMS matrix is much lower (around 0.4–3.5 MPa) (Amjadi et al., 2014; Fuard et al., 2008) compared with that of other matrices such as PU (15.1–151.4 MPa for 0.16 and 0.32 g cm^{-3} foam, respectively) (Patel et al., 2008), PI (84.1 GPa with 3.25% elongation at break) (Yang et al., 2018a), PVdF (0.84 GPa with 0.86% elongation at break) (Zhang et al., 2018a). This low Young's modulus indicates that PDMS can provide greater flexibility and versatility than other polymers. The Young's modulus of PDMS matrix varies with the thickness of the membrane reported in the literature (Liu et al., 2009). Liu et al. found that the Young's modulus increases with the decrease of membrane thickness. The thinnest membrane (50 μm) showed 1400 KPa Young's modulus with the elongation at break about 125% whereas the thicker membrane (1.8 mm) showed about 600 kPa. Owing to its low Young's modulus and high stretchability, PDMS is a great candidate as matrix in flexible strain sensor fabrication (Liu et al., 2009). Wang et al. incorporated CNT into PDMS substrate using digitally operated printer to fabricate multi-directional CNT/PDMS strain sensors with high stretchability and sensitivity. CNT ink dispersed with a surfactant, Triton X, were deposited on the surface modified PDMS substrates through spray deposition, followed by additional PDMS coating, which gives a sandwich structure, PDMS/CNT/PDMS. The fabricated sensors showed relatively high stretchability (45% strain) and good sensitivity (GF of 35) with excellent durability under cyclic loading-unloading. This sensor also showed a reliable sensing performance having less than 20% deviation, which indicates high accuracy of CNT/PDMS strain sensors to quantify strain-induced motions. Zhang et al. (2018b) demonstrated electroless deposition (ELD) of metals (Cu, Ni, and Ag) on functionalized PDMS substrates the with the aid of polydopamine to increase adhesion and stability for flexible and stretchable sensors. The polydopamine not only serve as the host material for metal ions to enhance the ELD process but also play a vital role in bridging the meal film and the substrate. The fabricated conductive metal-polymer hybrid such as Ag-PDMS, Cu-PDMS and Ni-PDMS showed high electrical conductivity of 1.8×10^7, 1.2×10^7 and 0.7×10^7 S m^{-1}, respectively. The as-prepared Cu-PDMS showed stable metallic conductivity even after 5000 cycles at highest stretch strain (700%).

Besides PDMS, other silicone-based materials such as Ecoflex and Dragon skin have been employed as elastomeric matrices because of their attractive mechanical properties; Young's moduli of Ecoflex and Dragon skin were 69 kPA and 152 kPA, respectively, which are even lower than PDMS (Amjadi et al., 2015; Cheng et al., 2017). Yu et al. (2017) fabricated highly sensitive strain sensor using Ecoflex as a matrix encapsulating carbon nanosponge. The fabricated composite showed excellent stretchability possessing high fracture strain up to 600% with a reasonable GF ranging from 4 to 19. These sensors also exhibited prompt response ($<$ 100 ms) and excellent stability ($>$ 1000 cycles) with a wide frequency ranges from 0.01 to 1 Hz, indicating its great potential as a skin wearable sensor.

4.2. Electrically conductive materials

Carbon-based nanomaterials such CBs (Ke et al., 2017; Shintake et al., 2017; Wu et al., 2016; Zheng et al., 2015; Zhu et al., 2018), CNTs

(Akhmadishina et al., 2013; Christ et al., 2017; Lin et al., 2017; Wang et al., 2014a; Zaporotskova et al., 2016; Zhang and Zhang, 2009) and graphene (Li et al., 2015; Liu et al., 2015; Wang et al., 2015; Yang et al., 2017) possess high electrical, thermal conductivity, and excellent mechanical strength. When used as conductive fillers, the dimensionality of carbonaceous materials affects electrical and mechanical properties of the composites. The polycrystalline and layered structure of CB empower it as a prominent candidate for conductive network of the composites with high electrical and thermal conductivity. Additionally, CB possesses low density, chemical inertness and low cost, which are also advantageous characteristics as a filler material in elastomeric matrix to fabricate stretchable strain sensors (Ke et al., 2017; Shintake et al., 2017; Wang et al., 2018b; Wu et al., 2016; Zheng et al., 2015; Zhu et al., 2018). With the addition of appropriate amount of CB particles, it is possible to fabricate piezoresistive polymer (silicone, epoxy, PDMS) composites for strain sensing applications (Shang et al., 2016). Shintake et al. fabricated CB-Ecoflex elastomeric composites that have high stretchability up to 500% via film casting and CO_2 laser ablation process (Fig. 6) (Shintake et al., 2018). The fabricated composite allowed the sensors to monitor the strain through either capacitance or resistance. The authors found that the capacitive-type sensor exhibited better performance compared with resistive-type sensor in terms of physical and mechanical compliances except Gauge Factor. The capacitive sensor showed high linearity (R^2 = 0.9995), low hysteresis and high repeatability with consistent strain rates as high as 50% s^{-1}. When the addition of porous CB nanoparticle exceeds the percolation limit, can decrease the resistance performance by 7 orders of magnitude for strain sensors. These sensors can repeatedly be used over 10,000 cycles without mechanical and electrical failures.

CNTs is a one-dimensional sp^2 carbon materials that possess high aspect (length/diameter) ratio, low density. High tensile strength and superior electrical conductivity proved them excellent candidate for sensors as prominent functional materials. CNTs-based polymer hybrids also showed superior sensing performance than the single component-based strain sensors due to their improved mechanical and electrical properties (Wujcik and Monty, 2013). Additionally, these properties of conductive polymer nanocomposites can be tailored depending on aspect ratio and wall thickness of CNT as well as composition of composites.

Recently, CNT-based polymeric strain sensors with high sensitivity and stretchability have been reported. Zheng et al. (2018) fabricated PDMS based CNTs-CB/PDMS elastomeric composites via solution mixing casting method which exhibited excellent stretchability (300%), sensitivity, repeatability and reproducibility in a wide range of strain (up to 200%). The hybrid sensors also exhibited outstanding stability even after 2500 cycles which allows reliable detection of human joint motion such as the motion of finger, wrist, elbow and knee (Zheng et al., 2018).

Christ et al. (2017) successfully 3D printed an elastic strain sensor using thermoplastic polyurethane and multiwalled carbon nanotube (MWCNT). They utilized the 3D printing process using extruded filament of TPU/MWCNT to achieve sensing capability with high mechanical properties. The printed materials loaded with 2% MWCNT showed a highest gauge factor of 176 at 25% applied strain, but the 3% MWCNT loaded sample offered the optimum mechanical and electrical properties up to 100% strain.

Zhou et al. (2017a) developed ultrasensitive strain sensors using single walled carbon nanotube (SWCNT) embedded in PDMS film. The fabricated sensor shows high strain levels with a gauge factor of over 10^7 at 50% strain. This extremely high sensitivity might be ascribed to the low electrical resistance of SWCNT, which maximize a change of resistance in cracked region of the composites. PDMS film plays the key role in providing flexibility and stretchability of the film as CNTs alone do not have satisfied stretchability.

Graphene is an emerging two-dimensional carbon material that can be a prominent candidate for strain sensors because of its attractive electrical, optical and mechanical properties. Chun et al. (2017) proposed graphene (SLG & graphene flake) based sensors on stretchable PDMS substrate which can detect very low strains (around 0.1%) with minimum resistance change. Various human body motions such as stretching, bending and torsion were successfully monitored. The twisted shaped SLG patterned strain sensor exhibited stretchability up to 20% with a gauge factor of 42.2 and bi-directional sensing ability. To further improve sensitivity, Lee et al. (2017) fabricated graphene-based grid patterned sensors on stretchable PDMS polymer substrates via solution dipping layer-by-layer assembly. The layer-by-layer assembly allowed precise thickness control of the film at molecular level to tailor the electrical conductivity and sensitivity of the sensors. They concluded that the mechanical tolerance of the sensors increases with the increase of graphene layer thickness. The 7 bilayer of layer-by-layer coating exhibited electrical failure at 45% strain with a compromised sensitivity, which indicates sensitivity is a function of the graphene layer thickness (Lee et al., 2017). To investigate the subtle and large human body motions, it is necessary to widen the strain range and increase sensitivity limit of the strain sensors. Tao et al. (2017) applied a one-step direct laser patterning method to fabricate Graphene/Ecoflex based strain sensor with high GF at high strain range. They found that the fabricated graphene encapsulated Ecoflex sensor showed an ultra-high GF of 457 at 35% strain and GF of 268 at 100% strain with excellent repeatability which significantly exceeds some reported value (Zhao et al., 2012).

Yan et al. (2018) fabricated flexible strain sensor using carbon/graphene composites nanofiber yarns (CNY) and thermoplastic polyurethane (TPU) via electrospinning method. They found that the performance of the sensor depends on the structural parameters such as yarn quantity and thickness of substrate. The tradeoff between the TPU film thickness and the sensitivity and stability of the sensors incited the author to find out the optimum structural conditions. The sensor exhibits an average gauge factor above 1700 under 2% strain at an optimum yarn number and substrate thickness are 4 and 129 μm, respectively (Yan et al., 2018).

Besides carbon-based nanomaterials, metal nanostructures such as metal nanoparticles, nanowires can also be utilized as a conductive material for strain sensors. As it is challenging to maintain both high electrical conductivity and flexibility of the device simultaneously, composites approach is similarly preferable. Wei et al. (2017) reported piezoresistive-type hybrid sensors based on biodegradable cotton fiber, AgNWs and rGO. The inherently rough surface of cotton fiber act as the backbone and flexible substrate whereas implanted AgNWs act as conductive bridge toward damaged rGO under tensile loading. Due to the dynamic bridging nature of AgNWs, the fabricated sensor exhibited very low detection limit (0.125 Pa) and high stability (> 10,000 fatigue cycles) (Wei et al., 2017). Lee et al. (2015b) fabricated AgNWs and AgNPs based highly stretchable strain sensor incorporated in a styrene–butadiene–styrene (SBS) elastomeric substrate via spinning method. The fabricated sensor showed high elongation at break (~ 900% strain) and excellent electrical conductivity (2450 S/cm) because of high conductivity of AgNWs and high stretchability of SBS matrix. During tensile stress, the embedded AgNWs come across and bridge the high resistive gap between AgNPs, maintaining the conductivity of the device and stretchability at a high strain. They also found that the conductivity of the device increased with the fiber length at a percolation threshold limit of 20 μm (Lee et al., 2015b).

Conductive polymers such as polypyrrole (PPy) (Li et al., 2014; Yang et al., 2016), polyaniline (PANI) (Lin et al., 2017; Wujcik et al., 2014), and PEDOT:PSS (Choong et al., 2014; Lee et al., 2016; Liu et al., 2011; Zhan et al., 2017) have also attract extensive attention in wearable strain sensor research due to their ease of preparation, inherent flexibility, low Young's modulus, and good adhesion with elastomeric matrix. However, conductive polymer based composites tend to have an inferior conductivity compared with carbon and metal nanofiller based composites. Some researchers have reported strain sensors with

conductive nanofiberous network based on electrospun conductive polymers. Liu et al. fabricated a novel strain sensor based on PEDOT:PSS/Polyvinyl Alcohol (PVA) nanofibers on Kapton substrate (Liu et al., 2011). The conductivity can be tuned by controlling the concentration of the additive dimethylsulfoxide from 4.8×10^{-8} to 1.7×10^{-5} S cm^{-1}. Though the sensor shows excellent stability and high GF of 396, the stretchability is limited at 1.2% due to the nature of nonwoven nanofiberous network. Bin et al. adopted a novel reciprocating-type electrospinning setup to produce PEDOT:PSS/PVP nanofibers with curled architectures (Sun et al., 2013). The GF (which is 360) is close to Liu's report but the stretchability was enhanced to 4%. Seyedin et al. (2014) successfully dispersed PEDOT:PSS homogenously in PU elastomer which greatly enhance the sensor stretchability to over 300%. This study has been discussed in Section 3.2: Stretchability.

5. Conclusions and outlook

Nowadays, wearable device is of great interest due to its easy implementation, easy diagnosis, better user experience and long-term monitoring capabilities. Wearable strain sensors have numerous potential applications such as healthcare and medical diagnosis, robotic systems, prosthetics, visual realities, professional sports, entertainments, among others. The strain limitation in conventional metallic foil or semiconductor strain gauge makes it impractical to be used as wearables. An ideal strain sensor should be highly stretchable, sensitive, and robust enough for long-term use without degradation in performance. During the last decade, owing to the fast advancement of material science, nanotechnology, microelectronics and strong need for wearables, remarkable achievements have been made in this field. This review comprehensively discussed the background, sensing mechanism, performance, features, materials selection, and prospects. A polymer nanocomposite based strain sensor typically has two main components, one is the conductive network which can provide conductive pathways to produce signal, and the other component is an elastomeric polymer to provide flexibility, stretchability and protection for conductive network. A rational choice of materials selection and structure design are required to achieve both high sensitivity and stretchability. In addition to geometric and intrinsic piezoresistive effects, electron conduction mechanisms such as disconnection, crack propagation, and tunneling effects play an important role in strain sensing performance.

Though great advances have been made in wearable strain sensors, there are still many challenges need to be addressed. First of all, material selections are difficult. Conductive nanomaterials including carbon nanomaterials, metal nanowires and conductive polymers all have their disadvantages. Carbon black has limited electrical conductivity, easy to aggregate, and needs high loading to form the percolation network. CNTs typically have a wide range of diameters and poor purity. Graphene based strain sensor has limited stretchability and other issues causing degradation such as irreversible cracks formed during stretching cycles. Metal nanowires such as AgNWs, AuNWs are relatively expensive. And huge Young's modulus difference between metal nanowires and polymeric elastomers can cause weak adhesion. Conductive polymers have lower electrical conductivity and are hard to process. How to minimize above material weaknesses and design a strong and stable conductive network with strong adhesion is a priority. Second, high stretchability, high sensitivity and linearity are hard to achieve simultaneously. High stretchability requires intact morphology under large strain, high sensitivity requires considerable structure change, and linearity requires homogenous morphology during stretching. These requirements are mutually exclusive making strain sensors with high stretchability, high sensitivity and linearity simultaneously difficult. Third, scalability and cost should be considered to commercialize wearable strain sensors. To achieve mass production, the design of fully printed electronics is desirable which help automate the fabrication process thus decreasing the cost and manufacturing the standardized products. Another significant challenge is packaging the wearable strain sensor with power, signal conditioning, communication, and data management units into a complete system. The integration of the whole system with high reliability, robustness, long power life has not been reported yet and needs multidisciplinary teams to address this issue. Finally, some features are attractive and technical issues needs to be tackled when adding into strain sensors. Examples are self-healing, self-powering, self-cleaning, transparency, biodegradability, etc., some have been discussed in Section 3. Further investigation and cooperation of researchers in material science, nanotechnology, microelectronics along with industrial companies are needed to overcome these challenges.

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