

Article

Review of Flexible Piezoresistive Strain Sensors in Civil Structural Health Monitoring

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Abstract: Owing to the outstanding sensing properties, especially high sensitivity and large stretchability, flexible piezoresistive strain sensors are advantageous for achieving intelligent sensing and have become a popular topic in the field of civil structural health monitoring (SHM). To explore advanced flexible strain sensors for civil SHM, this paper summarizes the recent research progress, achievements and challenges in flexible piezoresistive strain sensors. First, four common piezoresistive mechanisms are introduced theoretically. Sensor materials, including conductive materials, flexible substrates and electrodes, are explained in detail. Second, essential sensing parameters are interpreted and then followed by specific explanations of improvement strategies for the sensor performance in terms of each parameter. Third, applications of flexible piezoresistive strain sensors in the deformation measurement and damage detection of steel structures, concrete structures and fiber-reinforced composite structures are presented. Existing challenges and prospects in the practical application and large-scale production of flexible strain sensors are also reported. Last but not least, strategies for the selection of piezoresistive sensors for civil SHM are explained.

Keywords: flexible strain sensor; piezoresistive mechanism; nanoscale composites; civil structural health monitoring; intelligent sensing



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1. Introduction

With the worldwide development of infrastructure construction, enormous numbers of buildings, bridges, tunnels, railways and other infrastructure have been constructed during the past half century. Given that much infrastructure has been operated for decades, degradation and damage of structures are inevitable because of environmental erosion, fatigue load and natural disasters [1,2]. For instance, in 1995, the collapse of the Mitutoyo Department Store in Seoul, Korea, caused by functional modifications and design flaws, killed 502 people. In 2021, the collapse of a 12-story residential building in Miami, USA, killed 97 people [3]. Consequently, demands for structural health monitoring (SHM) and safety assessment of civil structures have been growing in the past decade [4,5].

Structural strain is a direct reflection of structural deformation under external force or environmental change. Thus, accurate strain measurement is one of the most important things in SHM to analyze structural performance and detect potential damage [6,7]. In practice, structural strain is usually measured using metallic wire resistance sensors, which depend on the variation in the sensor resistance under deformation. For instance, the relative change in resistance $\Delta R/R$ of a metal wire is given by $(1 + 2\nu)A/(\rho l)$, where ν , ρ , l and A are the Poisson's ratio, electrical resistivity, wire length and cross-sectional area of the metal wire, respectively [8]. The gauge factor (GF) of a metal foil strain sensor is around 2 because the Poisson's ratio is generally within -1 – 0.5 . This sensitivity level is insufficient for a low-strain measurement, making the signal vulnerable to external noises. Meanwhile, metal wire sensors usually sustain a maximum effective strain range lower than 2% strain,

which is incapable of addressing large structural deformation. Aside from the low sensitivity and small working range, conventional metal sensors also encounter short fatigue life and environmental problems [9]. In the past few years, as novel alternatives, nanomaterial-based flexible strain sensors have displayed much higher sensitivity and larger range because of their ultrasensitive conductive material and elastic substrate. One benefit of the flexible sensor is the capability to measure both small and large strains [10]. Therefore, the flexible strain sensors show promising applicability in various fields, including SHM [11], human health care [12,13] and human–machine interfaces [14,15].

The construction industry is one of the industries with the largest carbon dioxide (CO₂) emissions in the world. According to the global status report for buildings and construction publicized by the United Nations Environment Program, in 2021, the construction and operation of buildings were responsible for 37% of global energy-related CO₂ emissions and 36% of global final energy consumption [16]. Under this circumstance, the development of smart SHM based on flexible strain sensors will assist managers to make maintenance decisions, which is effective for ensuring the safety of civil structures and extending the service life of structures [17]. Thus, SHM is helpful in reducing CO₂ emissions indirectly. Moreover, SHM is of great significance to ensure the safety of people's lives and property and to achieve a sustainable society (e.g., requirements of energy consumption) and economy (e.g., investment in building energy efficiency) [18].

In accordance with the sensing principle of different conductive materials, flexible strain sensors are mainly divided into three categories, namely, piezoelectric-, capacitive- and piezoresistive-based strain sensors. Flexible piezoelectric sensors measure strain on the basis of the relative potential difference between the top and bottom electrodes of the piezoelectric material under deformation. The electric potential changes immediately when the strain changes, leading to high sensitivity and fast response, which are valued in dynamic strain monitoring [19]. However, piezoelectric sensors cannot measure static or pseudo-static strain because the potential difference in the material will soon return to zero when the deformation process stops. Flexible capacitive sensors measure strain by translating the external deformation into capacitance change of the conductive material. Benefits of capacitive sensors include high sensitivity to tiny deformation, high linearity and low hysteresis [20]. Nonetheless, the fabrication process for capacitive sensors is more difficult than that for other kinds of sensors. Compared with piezoelectric and capacitive sensors, piezoresistive sensors present easier fabrication, high sensitivity and long durability. As a result, piezoresistive-based flexible strain sensors are the most widely used flexible sensors [21].

Piezoresistive strain sensors are composites that usually comprise two-phase materials, namely, the conductive phase and the substrate phase. The conductive phase provides the sensing properties of the sensors, and the substrate phase provides the shape and structural integrity of the sensors [22]. The constituents of these two-phase materials cooperate with and complement each other to achieve the functions of flexible strain sensors. One of the most important things in designing a flexible piezoresistive sensor is the choice of suitable conductive and substrate materials that meet both the sensing and mechanical requirements for practical application. Researchers tend to adjust the concentration of the conductive and substrate materials to improve the functional properties of the piezoresistive strain sensor or add extra phase materials to the composite to endow extra functionality [23]. Nevertheless, existing composite materials still encounter challenges in practical application, such as low sensitivity for tiny strain, degradation of sensing performance caused by material fatigue and plastic deformation of the polymer substrate during large cyclic deformation [24].

In practice, civil structures often suffer time-variant environments and different service conditions, such as seasonal temperature changes, construction loads, fatigue loads, typhoons and earthquakes [25–27]. Under these conditions, the effectiveness and stability of strain sensors in the long term are difficult to guarantee [28]. Therefore, sensors installed on structures are required to have integrated performance of high sensitivity, large stretchability and durability [29]. Specifically, measurement of the full strain range from the elastic

stage to plastic buckling and then to crack propagation is very difficult for traditional strain sensors. Therefore, novel piezoresistive flexible strain sensors with improved structural design and fabrication technology are promising for realizing accurate monitoring of the complete deformation process, which is significant to intelligent civil SHM [30].

To explore advanced methodologies for monitoring the full process of structural deformation from small strain to crack propagation, this study focuses on the state-of-the-art research progress of piezoresistive-based flexible strain sensors in civil SHM. The keywords in the Abstract are used for searching and retrieving related papers before the writing of this review, namely the piezoresistive mechanism, flexible strain sensor and civil structural health monitoring. The methodology of the selection of appropriate papers for this review includes three criteria. The first criterion is that the selected papers should be highly related to the topic and keywords of this review. The second criterion is that highly cited papers and papers from high-authority journals are preferable for selection. The third criterion is the publication time of the paper. Papers published in the recent five years are more valuable because of the rapid growth and improvement of piezoresistive technologies. Aiming at a clear and comprehensive overview, the mechanisms, materials, performance improvements and applications of piezoresistive strain sensors are all considered, which can help researchers find the proper ways to achieve accurate and reliable monitoring of structural deformation. First, the basic piezoresistive sensing principles are explained, followed by an elaboration on the sensing materials, substrates and electrodes. Second, improvement approaches to sensor performance are introduced in detail by optimizing the sensing parameters of sensitivity, stretchability, linearity, hysteresis and durability. Third, applications of flexible piezoresistive strain sensors in terms of SHM are presented. The existing challenges and potential prospects are indicated. The strategies for selecting proper piezoresistive strain sensors for civil SHM are also presented.

2. Piezoresistive Mechanisms

The resistance variation of a sensor caused by deformation is known as piezoresistivity. In other words, piezoresistive strain sensors can transform strain changes into resistance changes of the sensor material. Considering that the piezoresistive materials used for SHM are almost composites that contain carbonaceous or metallic nanomaterials as conductive fillers and polymers as substrates, this review mainly focuses on the piezoresistive principles of those composites. The primary piezoresistive principles are mainly divided into four categories, namely, the interface contact effect, quantum tunneling effect, percolation network and crack propagation.

2.1. Interface Contact Effect

At the beginning stage of strain, the interface contact effect dominates amongst all principles. Given that the material surfaces are rough and uneven on the micro- or nanoscale, the interface contact effect occurs at the contact points between the conductive materials. The current is transmitted through these contact points [31] (Figure 1a). The number and size of contact points change with the deformation process, leading to the change in the total contact area; thus, the resistance varies. Through adjusting the number and size of contact points, high sensitivity can be obtained if the relative change rate of the contact area is large [32]. Flexible piezoresistive sensors based on the interface contact mechanism are divided into two categories. One category is adjusting the interface contact resistance between the electrode and the active layer. For example, Gao et al. used silver nanowire-coated cotton paper as conductive fiber and nanocellulose paper as the bottom substrate of a printed interdigital electrode [33]. The compression deformation of the cotton paper will cause more conductive fibers to contact the interdigital electrode to form more conductive paths; hence, the contact resistance of the sensor is reduced (Figure 1b). The other category is regulating the contact resistance between the conductive particles inside the active layer. For example, Doshi et al. fabricated a flexible sensor by electrodepositing polyethyleneimine-functionalized carbon nanotubes (CNTs) on nonconductive fibers as

coatings [34]. The compression deformation of the nanocomposite leads to the formation of fiber electrical contact in the active layer, which changes the resistance (Figure 1c).

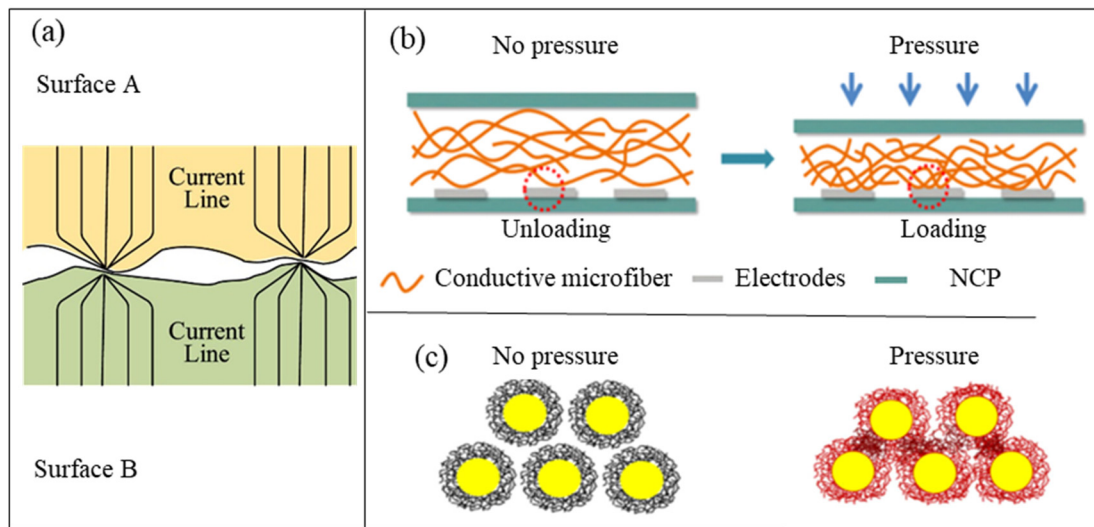


Figure 1. (a) conductive paths via the contact points [31] (Copyright 2017, Elsevier Ltd.); (b) variation of the contact resistance under pressure [33] (Copyright 2019, American Chemical Society); (c) interface effect of CN-coated fibers under external pressure [34] (Copyright 2018, American Chemical Society).

2.2. Tunneling Effect

As the strain increases, parts of the contacting conductive fillers begin to separate, and, hence, the tunneling effect appears. With this mechanism, the electrons between the conductive fillers can pass through the so-called energy barrier between adjacent but uncontacted conductive particles, resulting in tunneling currents [35,36].

Hu proposed an improved 3D statistical resistance network model, which can evaluate the tunneling effect between randomly distributed CNTs in a polymer matrix and predict the change in the CNT network [35]. In terms of a small strain level, the change in resistivity of a CNT nanocomposite sensor is mainly due to the tunneling effect between CNTs, leading to its nonlinear piezoresistive characteristics. Scanning electron microscopy was used to realize random distribution and a possible tunnel path between adjacent CNTs (Figure 2a). This physical diagram is simplified into a model for evaluating resistance (Figure 2b). The tunneling resistance between two adjacent CNTs is estimated as:

$$R_{tunnel} = \frac{V}{AJ} = \frac{(h^2d)}{(Ae^2\sqrt{2m\lambda})} \exp\left(\frac{4\pi d}{h}\sqrt{2m\lambda}\right) \quad (1)$$

where J is the tunneling current density, V is the potential difference, e is the quantum of electricity, m is the mass of electrons, h is the Planck constant, d is the distance between CNTs, λ is the energy barrier height and A is the cross-sectional area of the tunnel (the cross-sectional area of CNTs is used as an approximate value).

Yang et al. introduced the quantum tunneling effect to establish a representative volume element model to predict the conductivity, percolation value and mechanical properties of graphene rubber composites accurately [37]. In this model, graphene was randomly and uniformly distributed and large deformation characteristics of the rubber matrix were considered. Additionally, current density calculation results with and without considering tunneling effect were compared for further illustration of the influence of the tunneling effect. As shown in Figure 3, the current density of the rubber matrix without tunneling effect was the smallest, resulting in an insulated state of the graphene rubber composite (Figure 3a). By contrast, the tunnel current density of the composites increased

significantly by about 106 times when the tunneling effect was considered (Figure 3b). Therefore, the tunneling effect is beneficial to format conductive paths in composites and is very important for accurately predicting the conductivity and seepage behavior of conductive polymer composites (CPCs).

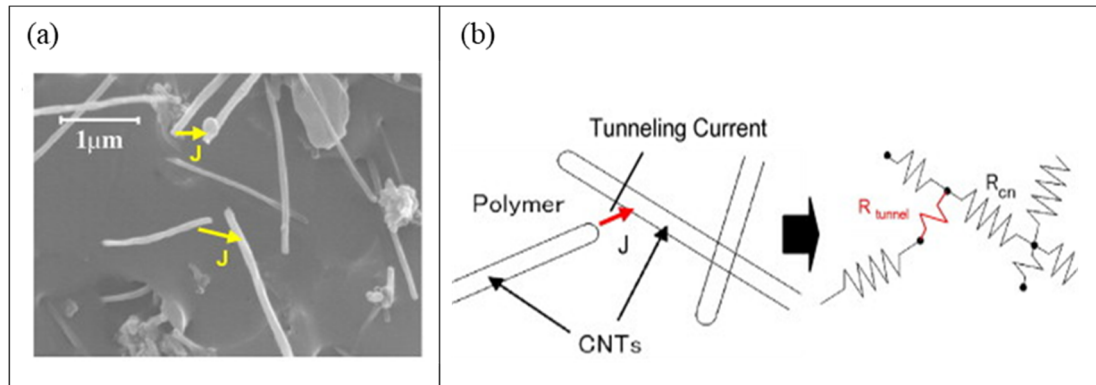


Figure 2. (a) SEM image of possible tunneling effect between CNTs in nanocomposites; (b) tunnel resistance modeling of resistance network [35] (Copyright 2008, Elsevier Ltd.).

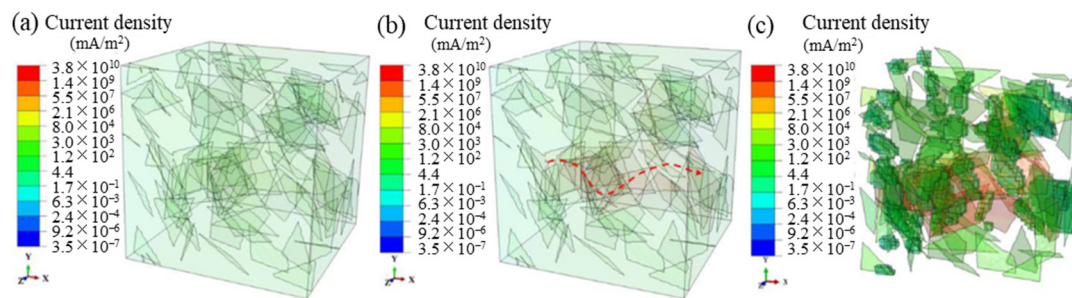


Figure 3. Current density distribution of graphene rubber composite: (a) ignoring tunnel effect; (b) considering tunnel effect; (c) conductive paths formed in graphene rubber composites [37] (Copyright 2020, Elsevier Ltd.).

2.3. Percolation Network

As the distance between adjacent conductive fillers increases with strain, the tunneling effect decreases gradually. In this situation, electrons can just pass through the contacting and overlapping nanomaterials. The conductive pathways form a percolation network. Piezoresistive films are usually constructed as a double-layer structure that is composed of a conductive layer and an elastomer substrate layer. Nevertheless, the conductive layer will crack or even fall off from the substrate layer in the cyclic test of long-term stability [38]. To improve the long-term stability, percolation network methodologies based on CPCs with good performance have been developed [39–41].

The resistivity of conductive composites depends on the content of conductive filler per unit volume in the polymer matrix. When the conductive filler has very low content, the average distance between conductive particles is large. In this case, very few conductive paths are formed; thus, the resistivity is too high for an insulator. With the increase in filler content, the average distance between conductive particles decreases, whilst the number of conductive paths increases to form a percolation network, leading to decreased resistivity of the composite. When the filler content is large enough, the conductive particles are connected to or overlap each other to form a conductive network throughout, transforming the CPC film from an insulator into a conductor. The critical volume ratio of the conductive filler at the transforming point is regarded as the percolation threshold. Figure 4a,b illustrate the development of a percolation network as strain increases in graphene flake- and silver nanowire-based CPC films, respectively.

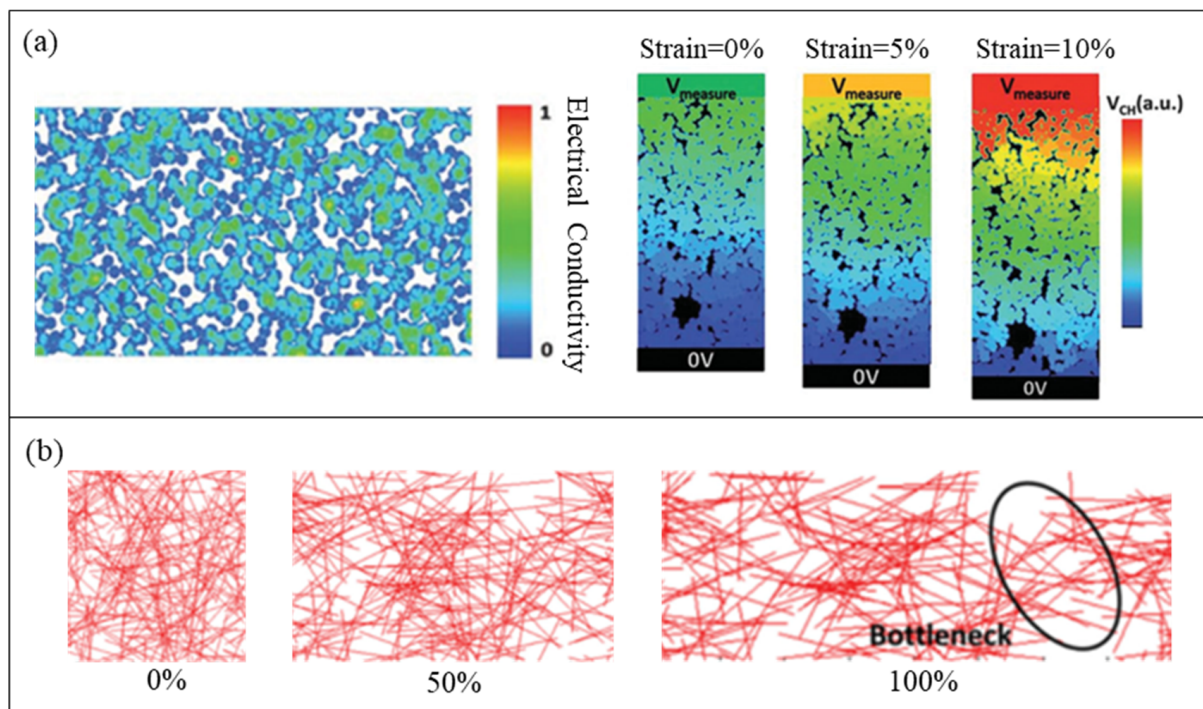


Figure 4. (a) percolation network of the randomly assigned graphene flakes [23] (Copyright 2019, Elsevier Ltd.); (b) percolation network of silver nanowires with low filler density [42] (Copyright 2016, WILEY-VCH).

2.4. Crack Propagation

Crack-based flexible strain sensors rely on the reconstructible disconnection and reconnection of cracks [43–45]. For strain sensors with thin nanomaterial films coated on flexible substrates, nano/microcracks generate at the stress-concentrated areas to release stress [23]. The quick departure of adjacent crack junctions caused by the applied strain considerably confines the conduction of electrons through the film, inducing a substantial change in the sensor resistivity [42,46]. Therefore, the crack propagation principle is advantageous to develop ultra-high-sensitive and flexible strain sensors [47–49]. Moreover, the sensing performance and working range are significantly influenced by parameters such as the length, width, depth and density of cracks [44,50].

According to recent studies, microcracks were generated in thin CNT-, silver nanoparticle-, gold nanowire-, graphene- and other conductive-filler-based films coated on flexible substrates [51–53]. Figure 5 depicts the process of microcrack generation and development in a graphene-polydimethylsiloxane (PDMS) composite film under cyclic stretching and releasing strain. It is shown that the density and size of microcracks increased with the stretching and then recovered to their initial states after the release of the thin films. In addition, microcrack edges were reconnected upon the release of the applied strain, ensuring complete recovery of the electrical resistance [54].

In summary, the four categories of piezoresistive mechanisms explained above are common in flexible piezoresistive strain sensors. Different mechanisms predominate the electrical conductivity of strain sensors at various strain levels or concentrations of conductive fillers, and they work together to form a complete conductivity system during the overall strain range of flexible sensors. In addition, the above mechanisms can be regarded as extrinsic conductive mechanisms because the conductivity of the composite is attributed to the additional carbonaceous or noncarbonaceous conductive fillers rather than the polymer substrates [22]. The conductivity can be tuned by changing the content of the fillers, leading to different piezoresistive mechanisms.

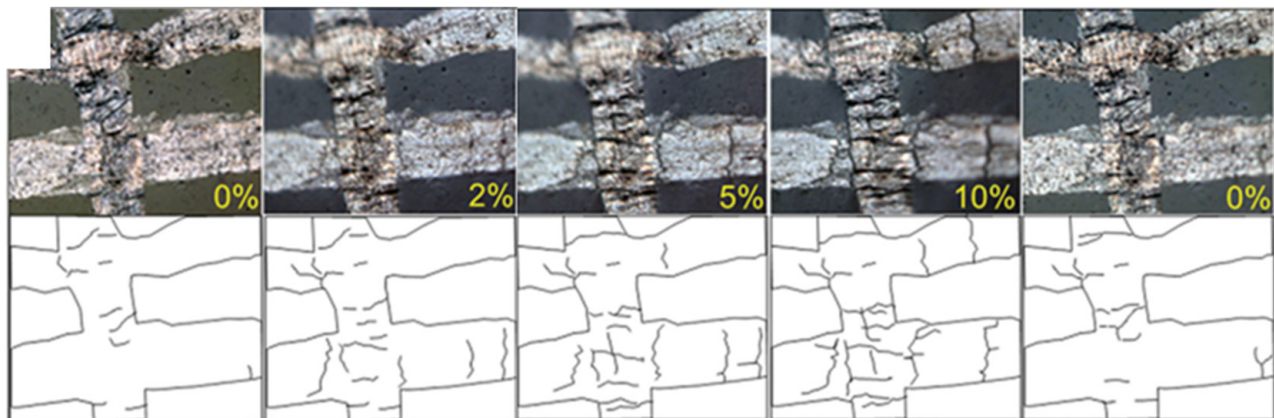


Figure 5. Crack propagation under SEM view: microcrack propagation on the graphene–PDMS composite film under stretching [54] (Copyright 2012, the authors).

3. Elaboration of Sensor Materials

The selection of material type is crucial and challenging in the preparation of flexible piezoresistive strain sensors because the material characters determine the working performance of the sensors. In general, a flexible piezoresistive strain sensor is composed of three parts: conductive layer, flexible substrate and electrode. When external strain is applied, the substrate begins to stretch, inducing resistivity change in the conductive layer and hence resistance change in the sensor. The resistance change is then transformed into an identifiable electrical signal through the electrode. Table 1 gives the symbols and abbreviations of the materials mentioned in this paper. This section presents specific introductions to the characteristics of the sensor materials as follows.

Table 1. Symbols and abbreviations of the materials.

Abbreviations	Material	Abbreviations	Material
AFM	Atomic Force Microscope	PDMS	Polydimethylsiloxane
AgNPs	Silver Nanoparticles	PEDOT: PSS	Polyethylenedioxythiophene: Polystyrene Sulfonate
AgNWs	Silver Nanowires	PET	<i>Polyethylene terephthalate</i>
AuNWs	Gold Nanowires	PI	Polyimide
CB	Carbon Black	PMVS	Polymethylvinylsiloxane
CNTs	Carbon Nanotubes	POE	Polyolefin Elastomer
CPCs	Conductive Polymer Composites	PP	Polypropylene
CSF	Carbonized Silk Fabric	PPy	Polypyrrole
CuNWs	Copper Nanowires	PU	Polyurethane
DI	Deionized Water	PVA	Polyvinyl Alcohol
EG	Ethylene Glycol	PVC	Polyvinyl Chloride
FBGs	Fiber Bragg Gratings	PVDF	Polyvinylidene Fluoride
FDM	Fused Deposition Modelling	rGO	Reduced Graphene Oxide
FGS	Fragmentized Graphene Sponges	SBCs	Styrenic Block Copolymers
FSG	Fish-Scale-Like Graphene	SBR	Styrene-Butadiene Rubber
GWF	Graphene Woven Fabric	SBS	Polystyrene-Block-Butadiene-Block-Styrene
IL	Ionic Liquid	SEBS	Styrene-Ethylene/Butylene-Styrene Triblock Copolymer
ILBW	Ionic-Liquid-Based Wavy	SEM	Scanning Electron Microscopy
MWCNTs	Multi-Walled Carbon Nanotubes	SPX	Spandex
LPG	Laser Patterned Graphene	SWCNTs	Single-Walled Carbon Nanotubes
NaCl	Sodium Chloride	TPU	Thermoplastic Polyurethane
NR	Natural Rubber	TPV	Thermoplastic Vulcanizate
OBC	Olefin Block Copolymer	WEP	Waterborne Epoxy
PANI	Polyaniline		

3.1. Conductive Materials

The conductive layer is the core part of a flexible piezoresistive sensor, which not only needs to provide conductivity for the sensor but also has the elasticity to adapt to applied external force. The conductive layer usually refers to the conductive polymer composite formed by mixing conductive fillers and insulative elastomer or conductive nanomaterials coated on insulative elastomer substrates. Materials of the conductive layer are divided into three categories: metallic materials, carbon-type nanomaterials and hybrid materials.

In accordance with the dimensions of the conductive fillers, conductive materials are divided into 0D/1D/2D/3D materials. 0D materials mainly include nanometallic particles and CB; 1D materials mainly contain metal nanowires and CNTs; 2D materials refer to graphene flakes; and 3D materials refer to conductive polymers [46]. Amongst these materials, 1D materials have a high aspect ratio, which is helpful for forming conductive paths. Therefore, most flexible conductive composites have been prepared using 1D metal nanowires or CNTs.

3.1.1. Metallic Materials

Metallic nanomaterials, including 0D nanoparticles [55–57] and 1D metal Cu/Au/Ag nanowires [58–60], have good electrical properties and mechanical flexibility. The distance between metallic nanoparticles is usually small; thus, most electrons are transmitted via the tunneling effect. Because the quantum tunneling effect is extremely sensitive to the distance change between particles, a small strain on the metallic nanoparticle array will cause destruction and regeneration of the conductive network, resulting in a large change in resistance. Therefore, metallic nanoparticles can be used to fabricate highly sensitive piezoresistive strain sensors with assisted microcracks.

AgNW-based conductive inks have been developed and applied to print large-area stretchable strain sensors [61], but the weak connectivity between AgNWs will lead to defects, such as a small sensing range, low sensitivity and nonlinear mechanical deformation. In addition, silver is relatively expensive, which significantly increases the manufacturing cost [43]. As an alternative, CuNWs have similar conductivity to AgNWs but their cost is only one-third that of AgNWs. Nevertheless, CuNWs easily oxidize and require further methods to reduce or prevent oxidation [62].

3.1.2. Carbon-Type Materials

Commonly used carbon-type nanomaterials, including CB, CNTs and graphene, usually have excellent electrical properties and chemical stability [63,64].

CB is an amorphous 0D conductive particle. The conductivity of CB is influenced by the particle size, structure and roughness of CB (Figure 6a). Compared with CNTs and graphene, CB is much cheaper. So far, CB-based strain sensors have been manufactured by screen printing a CB-silicon elastomer composite on a fabric to produce a series of wearable flexible sensors [65]. Nonetheless, CB is easy to agglomerate and difficult to disperse because of its tiny particle size. In this case, a high content is required to obtain good conductivity, but this leads to reduction in the stretchability. In addition, the conductive path amongst CB particles is easily damaged and reconstructed when the flexible sensor is deformed, resulting in unstable sensing performance [66].

CNTs are 1D conductive nanofibers with a low percolation threshold and excellent mechanical properties (Figure 6b). They are divided into single-walled CNTs (SWCNTs) and multi-walled CNTs (MWCNTs) [67,68]. The cost of MWCNTs is just one-tenth of that of SWCNTs. Moreover, the purity of MWCNTs in solutions can reach 99%—higher than the 90% purity of SWCNTs. Therefore, MWCNTs have much lower cost and higher purity than SWCNTs. Strain sensors based on MWCNTs have excellent sensing characteristics and a large working range. The curved structure of CNTs allows easy winding around each other in the substrate to form many contact points, which is helpful to improve the piezoresistive property of the composites [67,68]. Although CNTs have high stretchability, CNT-filled elastic polymer composites usually have relatively low GF under a low strain level and

nonlinear GF under a large strain level. In addition, CNT-based strain sensors often exhibit resistance hysteresis under cyclic loading owing to the irreversible change and degradation of the conductive network. Accordingly, structural stability is very important in realizing high performance, and many strategies have been employed to resist the buckling and binding of CNT networks during cyclic deformation [69].

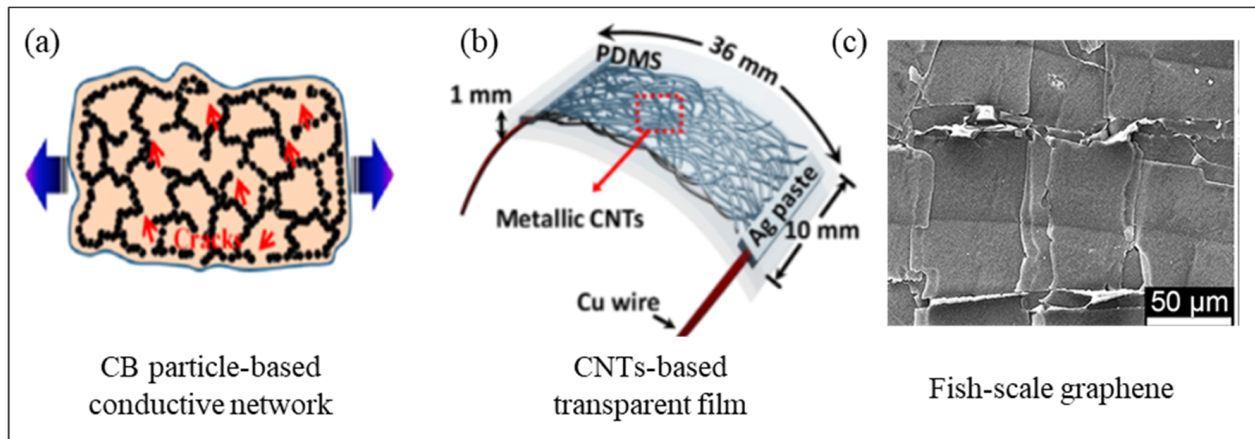


Figure 6. Carbon-type conductive materials.

Graphene is a hexagonal 2D carbon nanomaterial with a honeycomb lattice composed of hybrid orbital carbon atoms (Figure 6c). It has a high transmittance of more than 90%, a large specific surface area ($2630 \text{ m}^2/\text{g}$) for a large sensing area per unit volume under strain and outstanding mechanical and thermal properties [70]. In particular, graphene has the advantages of high ductility, high recovery, high elastic stiffness (340 N/m) and good carrier mobility at room temperature, making it very suitable for flexible strain sensors with good sensing performance and stability [71,72].

3.1.3. Hybrid Materials

According to the explanations above, each type of conductive material has its own pros and cons in application. To improve the sensing and mechanical properties of single-material sensors, numerous studies have focused on the combination of nanomaterials with different dimensional sizes and properties. One value of hybrid materials is the ability to compensate for the disadvantage of a single material by combining the merits of other materials, as shown in Figure 7. For example, compared with graphene- or CNT-based strain sensors, strain sensors based on graphene-hybridized CNT films are beneficial for achieving higher linearity and reproducibility under cyclic tensile strains of up to 20%. This was ascribed to the strong interaction and effective load transfer within the hybridized films that successfully resisted the buckling of CNTs. In addition, soft AuNWs and hard AgNWs were combined to make stretchable transparent films [73], which showed a wide measurable strain range of 0.05–70% and a high sensitivity of 236.6 within the 5% strain level. These attributes are difficult to achieve with a single component of either AuNWs or AgNWs.

Another benefit of hybrid conductive fillers is the ability to tune the piezoresistive sensitivity of flexible strain sensors. For example, the incorporation of CB reduced entanglement in the MWCNT network, which promoted strain sensitivity [74]. Similarly, a composite film composed of CB and CNTs exhibited high sensitivity, high durability and electrical stability under fatigue cycles owing to the synergistic effect of the dual conductive network formed by CNTs and CB [75]. Furthermore, combinations of MWCNTs and CB [74,76], CNTs and graphene [77], AgNWs and graphene particles [43] and AgNPs and SWCNTs [47] have been reported.

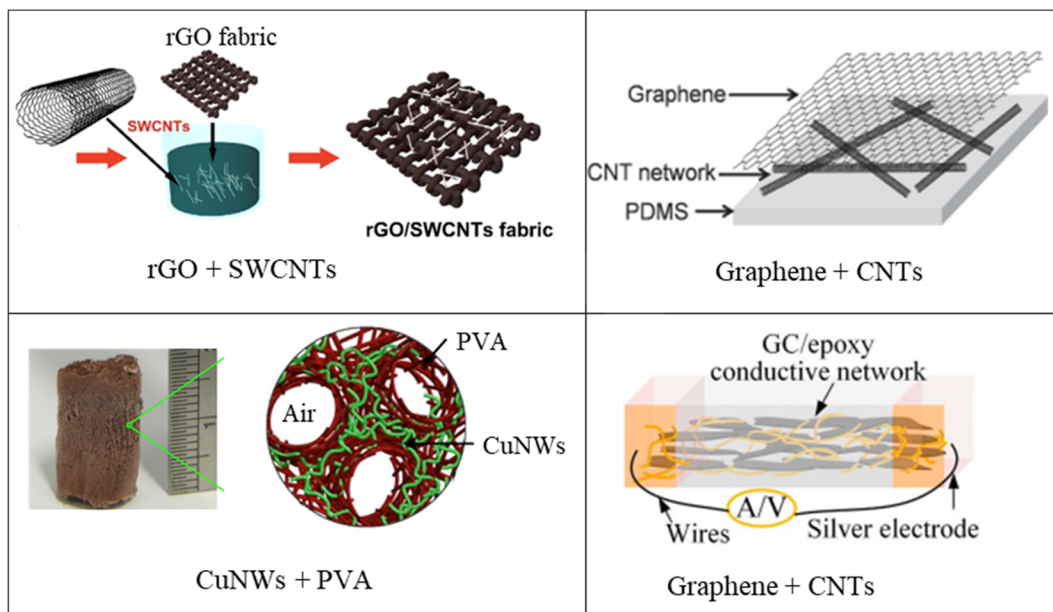


Figure 7. Hybrid conductive materials.

In a word, hybrid fillers show benefits for the improvement of electrical conductivity, sensing properties and stretchable range in flexible piezoresistive sensors because of the various conductive connections and the complementary mechanical properties amongst different materials.

3.2. Flexible Substrates

Flexible substrates provide a large stretchable range for piezoresistive sensors. In the application of SHM, the substrates are pasted to the structural surface by using epoxy or another adhesive. The generally used flexible substrates in SHM include PDMS [74,78,79], Ecoflex [80] and thermoplastic polyurethane (TPU) [81,82].

PDMS has the advantages of low elastic modulus, high elasticity, good formability, transparency, good thermal stability and corrosion resistance; therefore, it is the most widely used substrate for fabricating flexible sensors. Zhang et al. prepared a strain sensor based on CNT arrays encapsulated by PDMS [74]. In their study, the flexibility of PDMS was consistent with that of CNTs; hence, the sensor still maintained reliability and stability after cyclic bending it 8000 times. Lee et al. produced a metallic CNTs/PDMS composite film, which had 92% high transparency in the visible light range and all-round outstanding performance in sensitivity, hysteresis, linearity and drift [79]. Nevertheless, PDMS is sensitive to temperature change and is prone to ageing in the long term.

Ecoflex has excellent mechanical flexibility, water resistance and long-term stability. As an example, Amjadi et al. employed Ecoflex as a substrate and CNTs as conductive filler to fabricate an ultrasoft strain sensor. Given the strong interface between the CNTs and the Ecoflex, the composite sensor has high reliability, high linearity and an ultra-large strain range [80].

TPU has higher toughness, higher tear strength and stronger resistance to mechanical damage than PDMS. Moreover, TPU-based substrates still maintain the original function after more than 1000 large-tensile cycles. As a result, TPU has gradually become utilized as a common matrix material for flexible sensors in recent years [81,82].

High-performance polymers, such as polyimide (PI) [83], polyurethane [84], polyvinyl alcohol [85] and polyethylene terephthalate [86,87], have also been employed as substrates in different application scenarios.

Table 2 shows the materials of the above piezoresistive sensors and their corresponding sensing properties. Specifically, the maximum detectable strain range, the GF within the

working range, the linearity and the cyclic times for long-term durability are illustrated. It is found that the properties of different piezoresistive sensors are quite different from each other, even though they have the same conductive filler. Another phenomenon is that a flexible sensor can hardly achieve co-existence of high GF, large detectable range and good linearity. A sensor with a relatively large range and high GF always has a nonlinear strain-resistance relation, while a sensor that possesses a large range and good linearity regularly meets a low GF. The main reason for this phenomenon is probably the mutual conflict between the concentration of conductive filler and the ratio of resistance variation. Therefore, the sensing properties should be improved to meet the demands of practical application, as presented in the following Section 4.

Table 2. Conductive materials and flexible substrates (ϵ' is strain).

No.	Materials Filler + Substrate	Maximum Detectable Range	Gauge Factor/GF (Strain Range)	Linearity (Strain Range)	Cyclic Times for Durability
[58]	CuNWs + PDMS	7.5%		Nonlinear	
[65]	CB + Knitted Fabric	60%	1~100	Nonlinear	100,000 ($\epsilon < 60\%$)
[67]	CNT + TPU	400%	4~69	Nonlinear	1000
[68]	MWCNT + SBC	45%	$5.39 \sim 8.58 \times 10^7$	Nonlinear	600
[69]	CNT/Graphene + PDMS	10%	0.4	Linear ($\epsilon < 10\%$)	3000 ($\epsilon < 10\%$)
[73]	AuNWs/AgNWs + PDMS	70%	236 ($\epsilon < 5\%$)	Nonlinear	1000 ($\epsilon < 10\%$)
[74]	MWCNT + PU	200%	$5 \sim 1.4 \times 10^5$	Nonlinear	
[75]	CNT/CB + PMVS	60%	1~192	Nonlinear	10,000 ($\epsilon < 30\%$)
[76]	CNT/CB + PVDF	10%	0.6	Nonlinear	
[77]	CNT/Graphene + TPU	30%	152	Nonlinear	
[79]	CNT + PDMS	30%	3	Linear ($\epsilon < 30\%$)	600
[80]	CNT + Ecoflex	500%	2.4	Linear ($\epsilon < 100\%$)	2000 ($\epsilon < 300\%$)
[82]	SWCNT + TPU	25%	0.2	Linear ($\epsilon < 25\%$)	1000 ($\epsilon < 20\%$)
[85]	Graphene + PVA	55%	271 ($\epsilon < 35\%$)	Nonlinear	10,000 ($\epsilon < 55\%$)
[87]	CrNP + PET	3%	20 ($\epsilon < 0.36\%$)	Nonlinear	

3.3. Electrodes

Electrodes serve as the connections between a sensor and a signal receiver instrument. Electrical signals generated from the conductive layer are transmitted to the receiver through electrodes; thus, electrodes should have good conductivity and stability during the overall deformation process of the strain sensor. Stretchable electrodes based on metallic nanoparticles or nanowires, ionic hydrogels and conductive polymers have been widely studied [88–90].

For instance, polyacrylamide–sodium alginate hydrogel was used to fabricate a scalable, transparent (90% transmittance) and ultrathin (380 μm) electrode that can collect energy from human motion [88]. An aluminum electrode was used to link polymer-based aerogel to the signal receiver [89]. A filament serpentine gold electrode, which has stronger adhesion than ionic hydrogels, was used on epidermal electronic systems [90]. Generally, metallic electrodes are the most commonly used electrode materials in SHM.

4. Optimization of Sensor Performance

To evaluate the working performance of the flexible strain sensor, several parameters are indispensable, including sensitivity, stretchability, linearity, hysteresis and dynamic durability. As mentioned in Section 3, the selection of the appropriate material type is es-

essential to the working performances of strain sensors; nevertheless, there are still problems beyond that. For instance, the incomplete stress transfer between the conductive layer and the substrate layer will reduce the sensing accuracy because of the elastic mismatch and weak interfacial adhesion between them [91]. Moreover, the prevalent resistance hysteresis between the loading and unloading processes is ascribed to the internal friction between the material layers and the inherent viscoelastic character of the polymer substrates [91,92]. Additionally, the destruction and reconstruction of the conductive networks are unstable during deformation process, leading to unstable sensitivity and low linearity. Besides, the sensing performance usually shows a degradation trend during cyclic stretching/releasing loadings. This is attributed to plastic breaks of the polymer substrate and the buckling of the conductive layer during large cyclic deformation [76]. As a result, improved structural design and fabrication technology are requested to deal with the above problems for better sensing performance.

4.1. Improvement of Sensitivity

The sensitivity of a sensor is also called the gauge factor (GF), which reflects the amplification factor of the conductive materials for resistivity response to external deformation. It is regarded as the most important parameter of sensor performance, especially for subtle strains or cracks. In terms of a piezoresistive strain sensor, the GF is defined as the ratio of the relative resistance variation to external strain [92]. Note that the GF is usually a linear regression constant within a linear strain range of the sensor [93]. The GF value is influenced by many factors, such as the type, content and dispersion of the conductive fillers, and the interaction between fillers and substrates. In general, fillers with a content near the electrical percolation threshold show the largest GF value, while this GF hardly reappears because of the unstable conductive network. In this case, the sensitivity is mainly determined by the ratio of tunnel resistance to total resistance [94].

Therefore, the key point of sensitivity improvement is to regulate the morphology of the conductive network. An effective way to tune the conductive network is to combine different types of fillers. For example, the sensitivity of a CNTs-CB hybrid filler is regulated by changing the mass ratio of CNTs to CB. The CNTs act as conductive pathways in the conductive network, and the CB particles are located between the adjacent CNTs. When the CNT has a low content, the connections between CB particles and between CB particles and CNTs were weakened as the sensor stretched. In this case, the conductive structure became more susceptible to tensile strain; therefore, the lower mass ratio led to a higher GF value at a low strain level. Additionally, strong interfacial binding between conductive fillers and elastic substrates is also helpful to realize higher GF. For example, Lin et al. developed tunable sensitivity composites using CNT and thermoplastic elastomer [74], and Kang et al. combined reduced graphene oxide with polyvinyl fibers [95].

On the other hand, high sensitivity can also be achieved through crack-assisted conductive structures and micro- or nanostructures [96,97]. Inspired by the crack-shaped organs of spiders, a microcrack-assisted piezoresistive strain sensor using AgNWs and patterned PDMS substrates was fabricated, with many microcracks inside the composite. Attributed to its ultrasensitive microstructure, the sensor showed a remarkable GF value of 1.5×10^5 within a 60% strain range [49]. Besides, a crack-assisted strain sensor composed of AgNWs and graphene was developed, which showed a GF = 20 within a 0.3% strain, GF = 10^3 within a 0.3~0.5% strain, and GF = 4×10^3 between a 0.5~1% strain [43]. According to recently studies, microcracks are generally generated by the pre-stretching or bending, thermal solvent evaporation, and cooling process [43,98]. Moreover, other microstructures, such as interlocked structure, whisker structures and cotton fabric molded structures, have also been applied for higher sensitivity [99]. In addition to high sensitivity, the microstructure-assisted strain sensors also have the benefits of fast response and high stability [100,101].

4.2. Improvement of Stretchability

As for civil SHM, the stretchability of traditional strain sensors that is below the 2% range is sufficient for regular deformation monitoring but insufficient for detection of structural cracks and damages [29]. Hence, a wider working range is necessary for large-strain and damage tracking. Basically, stretchability of the flexible sensor depends on not only the elasticity of the polymer matrix, but also the stability of the conductive network during stretching [78].

There are two categories of approach to improve the stretchability. The first category is adopting stretchable conductive materials, as well as elastic substrates. Commonly used stretchable conductive fillers such as 1D CNTs, 1D AgNWs and 2D graphene have high-aspect ratios that can form many twists in the network [102,103]. These nanomaterials have been either mixed into an elastomeric polymer matrix or integrated in the middle layer/on the top layer of substrates [104,105]. In addition, a strong binding between the conductive materials and the polymer substrates is valuable for achieving high stretchability [80]. In a word, approaches of the first category are based on exploiting the intrinsic properties of sensor materials; therefore, they are helpful for simple fabrication and cost-effective production [106]. The second category is the special structural design of the flexible sensor, including out-of-plane designs such as 3D wavy, wrinkled and coiled shapes [107,108], as well as in-plane designs such as buckled, serpentine pattern, and mesh-like geometries [109,110]. Note that these designs help sensors to withstand much larger strain and there is no need to be concerned about sensors breaking during stretching [111]. Nevertheless, fabrications of these sensors require more complicated multi-processes and higher costs than the approaches of the first category [112].

Furthermore, there exists a common problem in pursuing high sensitivity and high stretchability at the same time [113], which is the balance between these two parameters. Specifically, a wide sensing range requires that the active material can keep its structural connections at a high strain level, whereas detection of subtle strain demands the resistivity of the conductive fillers to change significantly under a low strain level [91]. In this case, proper design and control of the connection types of sensing materials are vital to guarantee both high sensitivity and high stretchability [114]. For example, Amjadi et al. developed an ultrasensitive and highly stretchable strain sensor based on thin graphite films with parallel microcracks coated on top of Ecoflex, showing a high GF value of 5.2×10^2 at a large strain of 50% [50]. Furthermore, a combination of microcracks and scale-like structures was utilized to develop a high-performance strain sensor that possesses high sensitivity, high stretchability (GF value of 10^7 at 120% strain), fast response and remarkable stability.

4.3. Improvement of Linearity

The linearity directly reflects the sensing stability of a strain sensor, which is evaluated by the coefficient of determination (R^2) of the linear regression between the electrical signal and the applied strain. A larger R^2 indicates a stronger linear behavior of the sensor [115], while nonlinear behaviors are more common in practice [116,117]. In fact, the linearity relies on the degree of conductivity of the sensor. In other words, good linearity comes from high filler content, while low filler content contributes to nonlinearity [118]. This is because the dominant conductive mechanism changes gradually from the contact effect to the tunneling effect and then to the percolation effect as the filler content decreases.

The resistance change induced by the contact effect is the dominant mechanism at the initial strain stage, leading to a linear response [119]. As for a filler content close to percolation threshold, there are not enough conductive particles to form reversible conductive paths so conductivity is mainly achieved by the tunneling effect, resulting in a nonlinear strain response under stretching [120,121]. For instance, a strain sensor based on an Ecoflex/CNT/Ecoflex sandwich-like structure was produced with a high-dense network. The strain sensor possessed marvelous stretchability of over 500% and high linearity ($R^2 > 0.95$). However, the corresponding GF is low because of the stable

conductive network formed by the high-dense filler. Other reasons for the excellent linearity of this sensor are the strong binding between the CNTs and Ecoflex substrates [80].

In addition, the piezoresistive linearity is also influenced by the substrate property. Yang et al. recently developed a highly sensitive strain sensor with graphene-woven fabrics (GWFs) on a PDMS substrate. By tuning the Young's modulus of the PDMS substrate from hundreds of kPa to a few MPa, the R^2 value was improved from 0.91 to 0.98 [122]. Note that some studies achieved high sensitivity and linearity simultaneously by integrating hybrid sensing materials on polymer composites [123]. Benchirouf et al. reported that an rGO/MWCNT nanocomposite film on a flexible substrate exhibited a GF of 8.5 with a high R^2 of 0.98. Lately, Xin et al. developed a crack-assisted strain sensor with high sensitivity, high stretchability ($GF > 4.2 \times 10^4$ at 150% strain) and high linearity ($R^2 = 0.98$ within 15% strain, and $R^2 = 0.96$ from 22% to 150% strain), which were ascribed to the dense and well-controlled network cracks [124].

In summary, the piezoresistive linearity is relevant to the change in the dominant conductivity mechanism corresponding to specific strain domains [125,126], so that high dense, stable and robust networks will lead to a higher linearity [127]. On the other hand, the incompatibility of high sensitivity, high stretchability and satisfactory linearity was still a great challenge in developing high-performance strain sensors [42,118].

4.4. Improvement of Hysteresis and Durability

Hysteresis and dynamic durability are key parameters to indicate sensing performances under applied dynamic forces. Hysteresis is defined as the inconsistent or non-monotonic resistivity variations during a loading–unloading cycle [128]. Dynamic durability reveals the endurance of strain sensors to long-term cyclic loadings, including stable and reproducible resistive responses and mechanical integrity. In general, hysteretic behavior and response delay exist in all polymer-based strain sensors. They are regularly attributed to reasons that include the intrinsic viscoelastic nature of polymers, the viscoelastic characteristics of conductive fillers, poor interfacial adhesion, and the imperfect recovery of conduction paths because of friction between the sensing element and the elastomeric matrix [92,118].

First, a large interfacial area of conductive fillers and polymer raises the time delay [129]; thus, strong binding between the elastomeric fillers and the polymer matrix is helpful to eliminate the hysteresis effect [42,80]. Second, ultra-soft substrate provides a lower force for quick formation of the conductive network [97]. Third, polymers with smaller viscoelastic properties are recommended to ensure accurate strain measurement [92]. Besides, woven-based piezoresistive fiber/yarn/textiles also provide solutions to lower hysteresis because of the high stretchability and fast response of the woven structure for overcoming the viscoelastic delay of polymer substrates [108,109]. Generally, the piezoresistive fibers/yarns/textiles are fabricated by integrating conductive materials on the surface of the fiber/yarns through coating, ink writing and wrapping processes [130,131].

In fact, the hysteretic behavior depends on the integrity and robustness of the conductive network at different strain levels [93,118]. For instance, the “shoulder peak” hysteretic behavior for CNT/TPU composites at a 30% strain is much more obvious than that at a 5% strain [77]. This is because the weak destruction of the CNT network is easy to recover under small strain, while the destruction and establishment of the conductive network under large strain is more complicated. Similarly, Duan et al. presented that the evenly distributed and densely packed CNT conductive network in polymer suffered almost no evident changes, resulting in stable dynamic durability under a wide strain range within 10% to 50% [68]. Additionally, the response time of the crack-based strain sensors is faster because of their quick and easy reconnection–disconnection transformation [129,132].

In terms of long-term application, there is usually a declining trend of the sensing performance with the increasing number of cyclic stretching/releasing [133,134], owing to the unequal deconstruction and reconstruction of the conductive pathways during cyclic loadings. Another reason for the degradation is the irreversible plastic deformation of

substrates under large strain, and the fracture of conductive fillers. Additionally, long-term exposure to a harsh working environment will also lead to the performance decline and service life reduction in a flexible sensor [42,118]. In this case, growing efforts toward self-healing technologies have been made to improve the sensor's durability. At present, the self-healing of a polymer structure is achieved via passive extrinsic methodologies and active intrinsic methodologies [135]. Extrinsic self-healing is being performed via the leakage of a pre-embedded healing agent or catalyst from cracked microcapsules or microvascular systems. Extrinsic self-healing is usually restricted to only happening once and is incapable of multiple healings. Conversely, intrinsic self-healing is offered by reversible covalent bonding and supramolecular interactions, which are capable of multiple healings. Once the healing cycle proceeds, the sensor material will recover partial mechanical properties with or without external trigger intervention. Self-healing material will significantly extend the service life of a flexible strain sensor and reduce the maintenance cost of civil SHM [136].

Table 3 shows some typical flexible strain sensors with improved sensing properties. Compared with the property values in Table 2, the most remarkable improvement of the flexible sensors shown in Table 3 is the more accurate linearity. Most of the sensors showed linear or multi-stage linear characters. For instance, the flexible sensor proposed in [91] showed multi-stage linearity within the strain ranges of $\varepsilon < 2\%$, $2\% < \varepsilon < 15\%$ and $\varepsilon > 15\%$. Owing to the improved linearity, the mathematical sensing model became more precise, namely with fixed GF values and clear working ranges. The improved linearity and precise GF are mainly attributed to the implementation of braid micro-structures or micro-cracks, the reinforced bonding between the conductive filler and substrate, and the integration of hybrid materials. In addition, a few sensors showed the compatibility of multiple properties. Noting that the sensor described in [105] achieved all-round performances of large range (200% strain), relatively good sensitivity (GF = 10.5), good linearity and outstanding durability (180,000 cycles under 30% strain), this all-round sensor is very promising not only in the civil SHM but also other fields.

Table 3. Flexible strain sensor with improved sensing performance (' ε ' is strain).

Ref.	Materials Filler + Substrate	Maximum Detectable Range	Gauge Factor (Sensing Range)	Linearity (Strain Range)	Cyclic Times for Durability
[91]	SWCNT+ PDMS	150%	161 ($\varepsilon < 2\%$), 9.8 ($2\% < \varepsilon < 15\%$), 0.58 ($\varepsilon > 15\%$)	Multi-stage linear	100,000 ($\varepsilon < 60\%$)
[94]	CNT + PI	1%	40	Near linear ($\varepsilon < 0.8\%$)	500 ($\varepsilon < 0.8\%$)
[95]	Graphene + PVA	1%	2.3 ($\varepsilon < 1\%$)	Linear	1800
[96]	CNT + PDMS	120%	27.8 ($\varepsilon < 40\%$), 1084 ($40\% < \varepsilon < 90\%$)	Multi-stage linear	
[97]	Pt + PDMS	4%	0.75 ($\varepsilon < 4\%$) 3.4 ($\varepsilon < 2.5\%$)	Linear	
[99]	Graphene + PDMS	7.5%	100 ($2.5\% < \varepsilon < 5\%$), 400 ($5\% < \varepsilon < 7.5\%$)	Multi-stage linear	
[101]	Graphene + PDMS	3%	223 ($\varepsilon < 3\%$)	Linear	1000
[105]	MWCNTs + PDMS	200%	10.5	Linear ($\varepsilon < 100\%$)	180,000 ($\varepsilon < 30\%$)
[110]	CNTs + PDMS	11%	1 ($\varepsilon < 30\%$), 2 ($30\% < \varepsilon < 50\%$)	Multi-stage linear	1000 ($\varepsilon < 20\%$)
[114]	Graphene + Ecoflex	100%	457 ($\varepsilon < 35\%$), 268 ($35\% < \varepsilon < 100\%$)	Multi-stage linear	180
[123]	GO/MWCNTs + Kapton HN	0.6%	8.5	Near linear ($\varepsilon < 0.5\%$)	
[124]	CNT +PDMS	150%	2.2×10^3 ($\varepsilon < 15\%$), 4.2×10^4 ($22\% < \varepsilon < 150\%$)	Multi-stage linear	5000

Table 3. Cont.

Ref.	Materials Filler + Substrate	Maximum Detectable Range	Gauge Factor (Sensing Range)	Linearity (Strain Range)	Cyclic Times for Durability
[125]	MWCNTs + Ecoflex	300%	1.5	Near linear ($\epsilon < 300\%$)	
[127]	CNT sheets + Rubber	600%	0.5 ($\epsilon < 200\%$), 0.14 ($200\% < \epsilon < 600\%$)	Near linear ($\epsilon < 1\%$)	5000 ($\epsilon < 100\%$)
[130]	CNT + SPX	80%	0.2~0.3	Nonlinear	270,000 ($\epsilon < 40\%$)
[132]	CB + paper	0.22%	647 ($\epsilon < 0.14\%$), 33 ($\epsilon < 0.22\%$)	Multi-stage linear	70,000

Above all, the sensing performances of the flexible sensors were effectively improved through different methods, which inspired researchers to develop more advanced flexible strain sensors. Nevertheless, the compatibility of high sensitivity, high stretchability, low hysteresis and long-term stability is still a common challenge, especially for civil SHM, in which not only subtle strain but also large strain on the structural surface need to be captured.

5. Applications and Prospects in Civil SHM

In the past decades, cement-based embedded piezoresistive sensors have been applied in concrete structures because of their high sensitivity and good linearity. Nevertheless, sensing performances of the cement-based strain sensor are influenced by temperature and humidity. Its poor plasticity also placed restrictions in large deformation monitoring [137,138]. In contrast, conductive polymer-based flexible strain sensors are capable of overcoming the above shortcomings because of their high sensitivity being compatible with large stretchability, which is helpful in monitoring not only strain but also cracks or damage in such infrastructure as bridges, high-rise buildings and other civil structures. With the development of conductive materials and fabrication technologies, flexible piezoresistive sensors in the SHM of concrete structures, steel structures and fiber-reinforced composite structures have been proposed.

5.1. Steel Structures

Steel structures usually suffer yield or bulking damage from large deformations; hence, the target of strain monitoring of steel structures focuses on sensing large strain. Terada et al. used carbon particles as conductive filler and epoxy resin as substrate to prepare a thin film for monitoring the integrity of steel structures. Results showed that the thin film was able to accurately measure the strain of the steel structure with a gauge factor of 25 and a sensing range of 0~0.6% strain, whereas the sensitivity of the sensor decreased significantly when the tensile strain exceeded 0.6% [137]. Compared with the traditional metallic foil strain sensor, this film strain sensor has a much higher sensitivity ($25 \gg 2$) but a smaller sensing range ($0.6\% < 2\%$).

Ku-Herrera et al. made a MWCNTs/vinyl resin composite strain sensor, which showed a relatively large detectable range of 20% strain. The sensor also has a linear relationship between deformation and electrical resistance within 1% tensile strain, whereas it has a nonlinear relationship in compressive strain [138]. The gauge factor of this sensor is about 2.6 ($\epsilon < 1\%$), which is similar to the metallic foil strain sensor. Besides, the durability of the sensor is unknown because the cyclic times of stretching and releasing in their experiment was just 10 times.

Lundberg et al. developed a strain sensor with CNTs as filler and epoxy resin as substrate for monitoring the deformation of steel structures. This sensor performed well within a 0.12% compression strain and 0.14% tension strain under low loading frequency and less cyclic loading times [139]. Nevertheless, the corresponding sensitivity (GF) and the sensor performance for a strain range larger than 0.14% are not given.

The sensing properties of the above piezoresistive sensors are also shown in the first three rows in Table 4. It is indicated that the above sensors for the SHM of steel structures are still not absolutely advantageous over the traditional foil sensor, since they have either higher sensitivity but smaller sensing range, or a larger strain range but unknown durability. More work is required on the compatibility of multiple properties.

5.2. Concrete Structures

Different from steel structures, concrete structures crack under large tensile and compressive deformation. Researchers developed piezoresistive strain sensors using carbonaceous fillers or metallic fillers. In terms of carbonaceous fillers, Ji et al. prepared CB/epoxy composite film sensors to monitor the overall stress and deformation process of concrete columns and reinforced concrete beams [140]. In this case, this film sensor showed a sensing strain range of 1.2% and a GF higher than 11, leading to the accurate measurement of strain and the preliminary location of the crack in the monitoring area [141]. In addition, Zhang et al. designed a composite polymer film by blending solutions of MWCNTs and PDMS, and then investigated the sensing properties of the polymer film for the deformation and crack monitoring of concrete structures. This polymer film showed a large sensing range of 50%, but a nonlinear strain-resistance relationship with various GFs from 1.5 to 6 [142]. Wu et al. fabricated a flexible sensor using graphene oxide as conductive filler, cellulose nanofiber as dispersant, and waterborne epoxy as polymer matrix. This sensor showed large stretchability of a 55% strain, a GF of about 34, and good linearity and repeatability within a 4% strain range. Full-scale tests demonstrated the sensor's capability to monitor the strain and detect cracks on a concrete surface (Figure 8) [143,144].

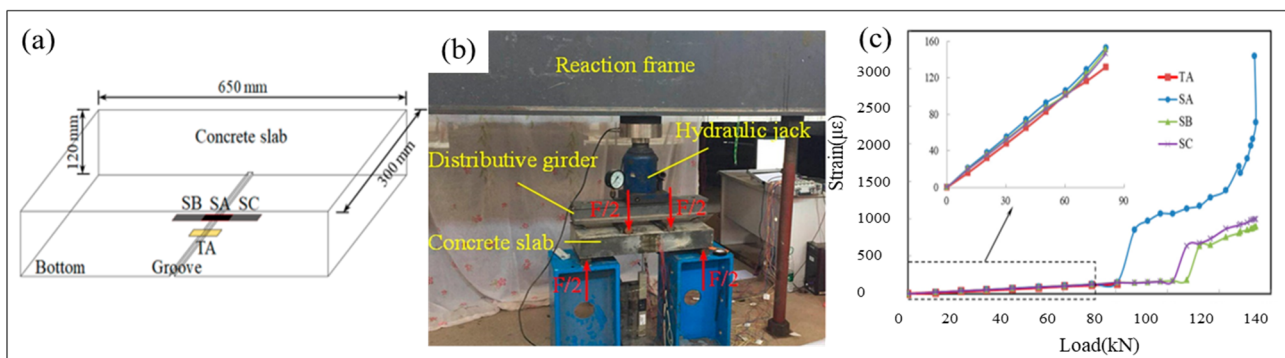


Figure 8. Deformation and crack monitoring [143] (a) the positions of the film sensor and resistance strain gauge; (b) four-point loading chart; (c) strains of composite film sensors (SA, SB, and SC) and traditional resistance strain gauge (TA) during loading. (open access, MDPI).

On the other hand, in terms of metallic fillers, Zhang et al. proposed a crack-width monitoring approach by screen printing CuNWs with different ultimate strains on an epoxy-resin structural surface. They established a multilayered stress-transfer model to quantify the proportional relationship between the crack width and the ultimate strain of the CuNWs, and then redesigned a smart film to assess cracks on a practical large-scale bridge [145]. Besides, Ryu et al. prepared a AuNPs/PDMS composite by an in situ reduction method, and then studied its optical properties under tensile strain. This composite showed a linearity between the light absorption rate and the tensile strain within a proper strain range [146]. Moreover, Hallaji et al. applied conductive silver glue to fabricate a multi-electrode sensing skin, the resistivity of which was monitored by electrical resistance tomography [147]. By using a differential imaging scheme, it is worth noting that this skin was able to locate the through crack of a concrete beam and track the process of crack propagation, as shown in Figure 9. Nevertheless, the sensitivity of the skin is relatively low because of the very tiny variation of the resistance of the conductive silver glue, and the quantitative relationship between resistance variation and crack length/width

was not provided in their work. Additionally, the resolution and accuracy of crack sensing is highly related to the quantity of the electrodes, whereas it is impractical to set many electrodes on a sensing film in field monitoring.

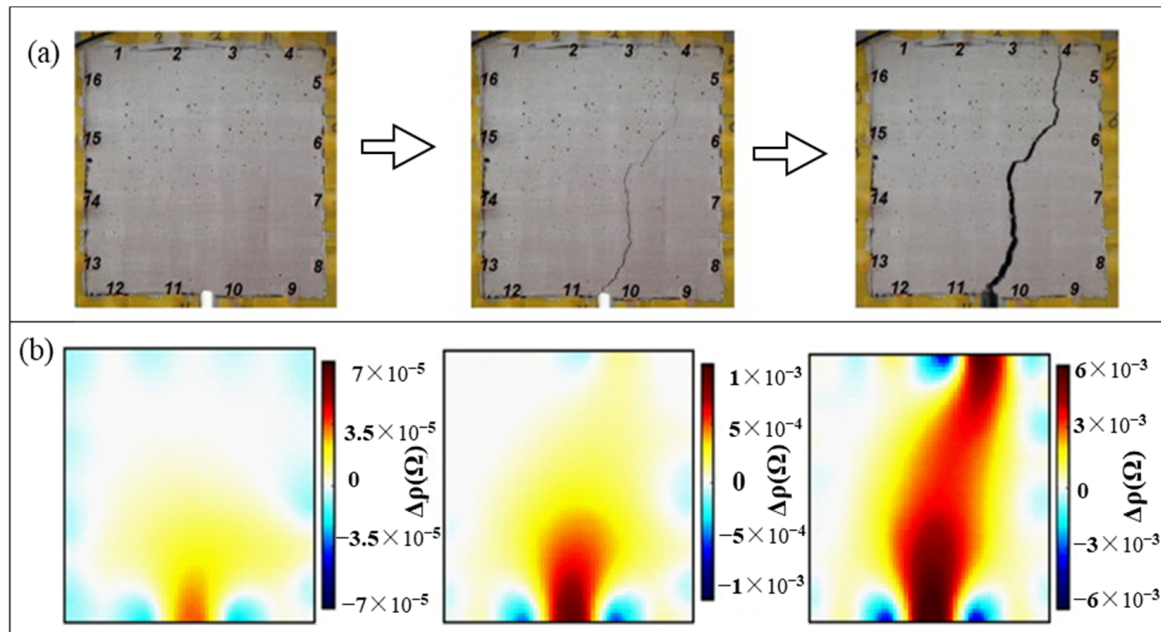


Figure 9. (a) crack expansion process of concrete square slab, (b) corresponding electrical resistance tomography of sensing skin under for crack detection [147] (Copyright 2014, Elsevier Ltd.).

5.3. Fiber Reinforced Composite Structures

With the increasing demand for fiber-reinforced polymer (FRP) materials in structural strengthen and renovation [148], the safety and reliability of the FRP-related materials become crucial [149]. Traditional approaches for monitoring the FRP materials were based on fiber Bragg gratings (FBGs), as well as ultrasonic sensors, which require complicated analysis processes and expensive resolving devices [150]. Moreover, an FBG sensor can only detect the strain or crack of a single point at the sensor area; thus, it is difficult to monitor the deformation and damage at different locations on FRP composites. In addition, FBGs are vulnerable to water and corrosive chemicals, so they need protective sleeves and cannot be compatible with FRP material because their diameters are very different [150].

Compared with FBG-based methods, the electrical resistivity effect of the FRP materials themselves were noticed by scientists and developed for self-sensing composites [151,152]. Nevertheless, resistivity parameters of the rough FRP materials without other additives are hard to control, which brings negative impacts to the sensing performance. To enhance the sensing ability, carbon-based fillers were considered to improve the sensitivity and stability of the detection performance of the FRP materials [153–155]. In general, CNTs or graphene are bonded to the surface of the FRP through coating and chemical vapor deposition. Due to the tiny size of CNTs, conductive networks are formed in the interfaces of carbon fibers. Therefore, local area monitoring is achieved by regulating the deposition of CNTs to the damage-prone area of the FRP composite structure [156,157].

As an example, Salvado et al. preliminarily investigated the properties of several continuous carbon-fiber epoxy-resin composites for strengthening concrete structures [158]. They improved the sensing ability of small-range strain by setting various geometric shapes and arrangement modes of the carbon fibers. As shown in Figure 10, composite materials were made on site for structural reinforcement in civil engineering. Results showed that the composite cannot only sense a small deformation of less than 1% strain, but also increase the strength and bearing capacity of the original concrete structure.

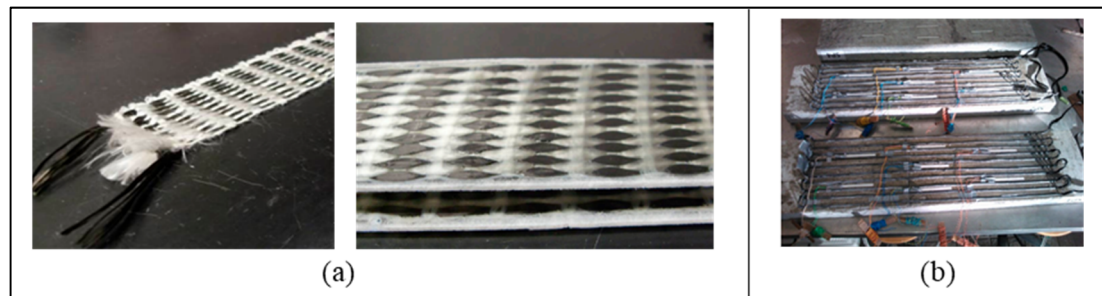


Figure 10. (a) textile-pattern strain sensor; (b) concrete specimens reinforced with carbon fiber epoxy resin composites [158] (open access, MDPI).

In addition, FRP materials are usually classified as carbon-FRP (CFRP) and glass-FRP (GFRP). Nag-Chowdhury et al. fabricated a nano-composite quantum resistance sensor, which can be embedded and distributed in GFRP without affecting its mechanical properties. Fatigue tests illustrated that the closing/reopening and expansion of cracks along with the increase in cyclic loading led to damage accumulation of the GFRP material [150]. This sensor was able to track the strain and damage of the GFRP structure in both the elastic and plastic stages until it failed. On the other hand, electrical resistance measurements were applied for the fracture detection and failure prediction of cylindrical-type CFRP composites (Figure 11a). Results showed that longitudinal cracks of the CFRP cylinder were easily detected according to the signal jump of the resistivity-change ratio [159]. Moreover, the resistance-variation graph can be used to distinguish different fracture modes of the CFRP cylinder (Figure 11b).

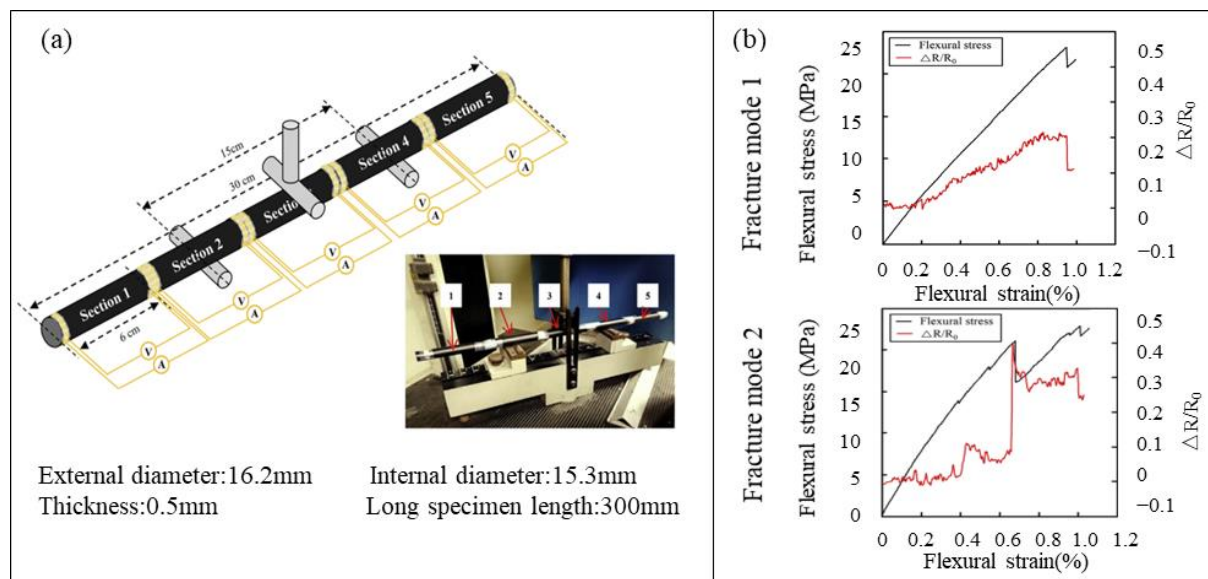


Figure 11. (a) Long CFRP cylinder specimen during flexural test; (b) resistance variation graph under different fracture mode of CFRP cylinders [159] (Copyright 2016, Elsevier Ltd.).

In addition, CNT-epoxy coatings were developed for large-area damage detection in GFRP-based structures [53]. A multichannel electrode network was integrated on the coating to build a two-dimensional map for multi-damage detection of a GFRP structure (Figure 12a). Figure 12c illustrates the location and quantification of a 23 mm long crack according to the normalized resistance variation of the sensing network. As indicated from Figure 12d–e, the CNT-epoxy coating showed a varied GF from 3 to 12 within a 0–2.5% strain range, which is higher than the tradition foil sensor. The excellent sensitivity was attributed to the susceptible destruction and reconstruction processes of the conductive

networks in the ultrathin coating. In addition, the multichannel electrode network detected the damage length with a sensitivity of 0.007%/mm (Figure 12f).

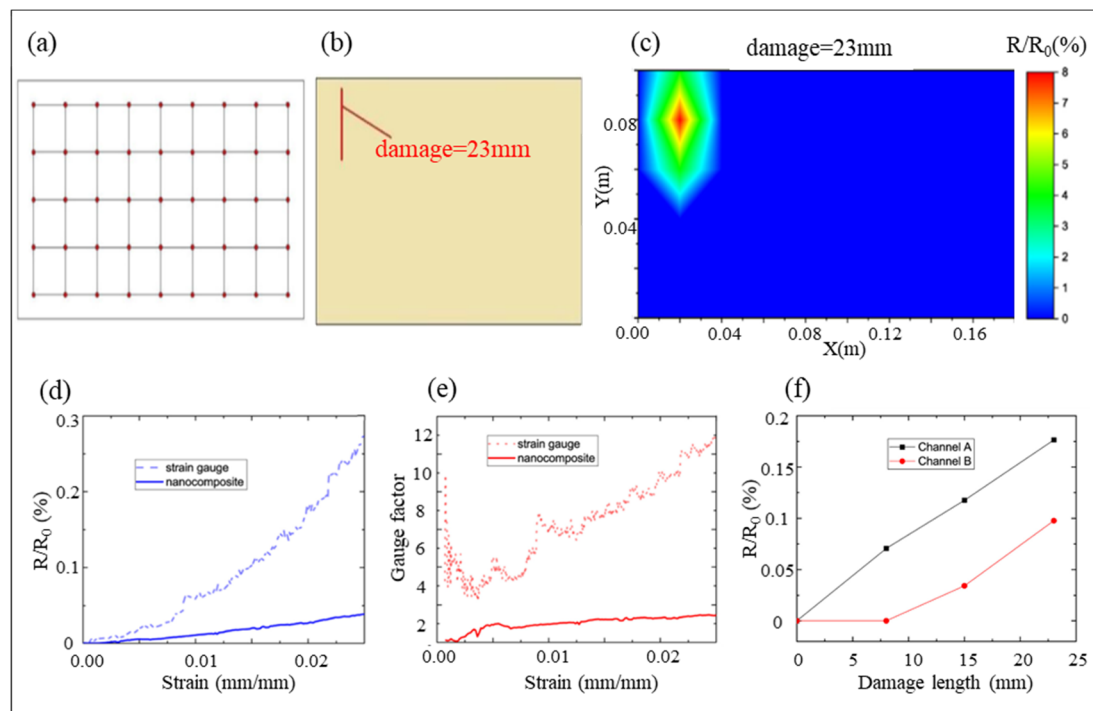


Figure 12. (a) Electrode disposition on the CNT/epoxy coating; (b) damages on the slab; (c) two-dimensional electrical resistance mapping of crack detection; (d) normalized resistance variation under tensile strain; (e) gauge factor variation under tensile strain; (f) normalized resistance variation with damage length [53] (Copyright 2017, Elsevier B.V.).

Table 4. Piezoresistive strain sensors for civil SHM.

Ref.	Materials Filler + Substrate	Maximum Detectable Range	Gauge Factor (Sensing Range)	Linearity (Strain Range)	Cyclic Times for Durability
[137]	Carbon Particle + PI	0.6%	25	Near linear ($\varepsilon < 0.6\%$)	
[138]	MWCNTs + Resins	20%	2.6 ($\varepsilon < 1\%$)	Linear ($\varepsilon < 1\%$)	10 ($\varepsilon < 1\%$)
[139]	MWCNTs + Epoxy	0.14%		Linear ($\varepsilon < 0.14\%$)	1000 ($\varepsilon < 0.14\%$)
[140]	CB + Epoxy	1.2%	11~42 (Depend on filler content)	Nonlinear	
[142]	MWCNTs + PDMS	50%	1.5~6	Nonlinear	
[143]	rGO/Cellulose nanofiber + Epoxy	55%	34~71	Nonlinear	100 ($\varepsilon < 4\%$)
[144]	RGO-CNF + WEP	4%	16~52 (Depend on filler content)	Near linear ($\varepsilon < 4\%$)	4 ($\varepsilon < 4\%$)
[146]	Gold nanoparticles + PDMS	30%	0.04	Near linear	
[151]	MWCNT/FRP + Epoxy	1.23%	5~6	Nonlinear	
[154]	CNTs/Glass fiber + Epoxy	1.2%	2.25	Linear ($0.5\% < \varepsilon < 1\%$)	
[155]	MWCNTs/Glass Fibers + Epoxy	2.5%	3.8 ($\varepsilon < 0.2\%$)	Near linear ($\varepsilon < 0.2\%$)	
[156]	CNT/Glass Fibers + Epoxy	2.2%		Nonlinear	
[157]	Graphene nanoplate + Glass fiber	2%	840~16,400	Nonlinear	
[158]	Carbon fibers + Epoxy	3.5%	10		
[159]	Carbon fibers + Epoxy	1%	0~0.25	Nonlinear	
[53]	CNT + Epoxy resin	2.5%	3~12	Nonlinear	

Last but not least, Table 4 gives a summary of the aforementioned piezoresistive strain sensors used for the SHM of civil structures. As shown in the third column of Table 4, the detectable strain ranges of most of the sensors are below 5%, which is much smaller than the piezoresistive sensors shown in Tables 2 and 3. This is because the elastic and recoverable strain range of civil structures is usually below 1~3%, according to the construction materials. For instance, the FRP-based structure will crack near a 1%

strain (Figure 11b). Hence, the mission of the strain sensor changed to detect cracks or bulking rather than measure strain when the structural strain is beyond the recoverable range. Nonetheless, little consideration has been paid to the long-term durability of the piezoresistive sensors, as indicated from the blanks in the last column of Table 4.

5.4. Challenges and Prospects of Large-Scale Application

Although carbon-type composite materials have shown many advantages for sensing damage in SHM, there are still challenges to realize large-scale application of carbon-based strain sensors. For instance, the high-purity CNTs and their corresponding dispersion have high cost, and the blast risk of CNTs brings safety concerns [67,68]. In addition, a tedious fabrication process hinders the application of graphene-based strain sensors [160]. In fact, most current fabrication techniques of carbon-type strain sensors are still in their prototype stage. Moreover, practical application of the proposed piezoresistive strain sensors requires an overall integrated system, which includes sensors, power supplement, data analysis and processing, wireless data transmission and a remote operation system [93]. Besides, the integrated sensing system should have robustness and high reliability against the performance degradation in long-term monitoring [128]. In terms of the self-healing flexible sensor, there are still challenges for simple, economical and scale-up fabrication strategies. Another important challenge is to keep the strain sensors maintaining good performance even after multiple healings [22].

Furthermore, researchers explored novel ideas of carbon-type sensors using pure macro-graphene, which showed potentials for field application and scale-up production [161]. Wang et al. reported a pure graphene-assembled film (GAF), which can be directly used as a strain sensor without flexible substrates [160]. The GAF significantly simplifies the fabrication process in comparison with most reported strain sensors, and mass production of this material has already been realized. Besides, the resistance of the film remains unchanged with increasing temperatures from 20 °C to 100 °C, indicating a good thermal stability (Figure 13a).

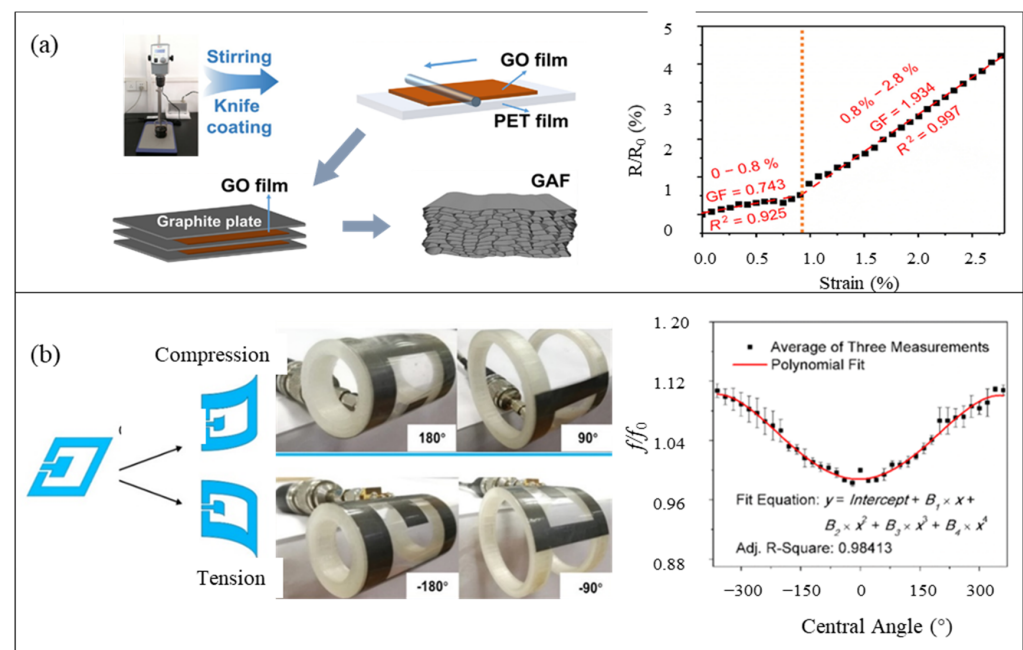


Figure 13. Flexible strain sensor based on pure macro-graphene: (a) fabrication of the GAF [160] (Copyright 2016, Elsevier Ltd.); (b) GAF antenna-based wireless strain sensor for measuring compression and tension [162] (Copyright 2020, American Chemical Society).

On the other hand, the GAF, which was originally used as a wireless antenna with a flexible radio frequency, can also be used as a wireless strain sensor via resolving the

signal variation resulting from the deformation of the GAF antenna [162]. In their work, the high-conductive GAF has advantages of high flexibility and excellent mechanical stability. As a result, a flexible GAF-antenna-based wireless strain sensor was achieved, which has high GF of 35 in measuring tensile bending and compressive bending (Figure 13b). Moreover, a two-dimensional wireless displacement sensor was developed based on a bilayer graphene-based frequency selective surface [163]. A two-dimensional displacement measurement with high accuracy was carried out based on the variation trend of the resonant frequencies of the top and bottom layers of the sensor. One more benefit is that the bilayer graphene material shows outstanding mechanical and chemical stability in complicated environments, indicating long-term-use prospects.

5.5. Strategy for Selection of Piezoresistive Sensor in Civil SHM

Aiming for the requirement of deformation monitoring and damage detection of civil structures, one of the primary targets of this review is to provide appropriate strategies for the selection and improvement of flexible piezoresistive strain sensors. Note that the strain level of civil structures is usually below 1% under normal loading and ambient situations, and large strain or cracks will appear because of long-term plastic deformation and extreme loadings. Therefore, the flexible piezoresistive sensor should have not only enough sensitivity and linearity to measure small strains ($\epsilon < 1\%$) but also a wide strain range and long-term durability to sense large strains and damages. In addition, the selection principle should be comprehensive by considering different aspects such as sensitivity, feasibility and detection performance [22]. To name a few, some strategies are concluded as follows:

First, carbonaceous materials are preferable to be chosen as conductive fillers since they have better piezoresistive properties and lower production costs than metallic materials. Integration of different carbonaceous materials might be able to produce better performance. Second, the sensors proposed in references [138,144] are promising examples for civil SHM because of their balanced performances of relatively high GF, large sensing range and good linearity. The outstanding compatibility of multiple performances is attributed to not only the integration of complementary conductive fillers but also the reinforcement of the substrate. Besides, their feasibility for scale-up fabrication and long-term durability is suggested for further investigation. After that, the sensing performances can be improved via the methods presented in Chapter 4, such as utilizing hybrid reinforced materials or micro-structures. By contrast, a new form of graphene material, namely, the pure GAF, has potential because of its easy fabrication and long-term stability. However, the sensitivity of the pure GAF in a small strain range ($\epsilon < 1\%$) should be further enhanced.

6. Conclusions

This study presented a literature review of flexible piezoresistive strain sensors in several aspects, including typical piezoresistive mechanisms, elaboration of sensor materials, optimization of sensor performances, applications and prospects in civil SHM. The conclusions are summarized as follows:

The four common piezoresistive mechanisms successively predominate the electrical conductivity of the strain sensor within their respective working strain levels. Each mechanism corresponds with a different sensing parameter. The proportion of tunneling resistance in total resistance determines the sensitivity, while the proportion of interface contact resistance governs the linearity. Thus, the sensing performances of piezoresistive sensors are highly related to the morphology of the conductive network.

Selections of conductive materials and substrate materials are crucial to electrical conductivity, sensing properties and stretchable range in flexible piezoresistive sensors. Hybrid conductive materials usually perform better than single-type materials because the conductive networks and the mechanical properties are improved by the complementation of different materials.

The sensing performances of flexible sensors can be further enhanced through strategies such as the proper combination of different types of conductive fillers, strong bonding and cooperative deformation between the fillers and the substrates, and assistances of microcracks or microstructure. Nevertheless, the incompatibility of high sensitivity, high stretchability, satisfactory linearity and durability is still a great challenge in developing high-performance strain sensors.

Applications of piezoresistive strain sensors in civil SHM mainly focused on tracking concrete cracks and detecting damage in FRP structures. Feasible strategies for selecting suitable piezoresistive strain sensors for civil SHM are provided. Although carbon-based flexible piezoresistive sensors have been well-developed in recent years, challenges such as complex fabrication, performance degradation during long-term use and overall integrated systems still exist for further field application. More attempts are required to develop advanced piezoresistive sensors targeted for civil SHM.

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