Crack engineering boosts the performance of flexible sensors

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Funding information
National Key R&D Program of China, Grant/Award Number: 2021YFB3200700; National Natural Science Foundation of China, Grant/Award Numbers: 51925503, 52188102; Xplorer Prize, Grant/Award Number: 2020-1036

Abstract
With exceptional performance, flexible sensors have found broad applications, including human health monitoring, motion detection, human–machine interaction, smart wearable technology, and robot control. Crack-sensitive structures based on animal bionics have also caught increasing attention because of their extraordinary sensitivity. Crack-based flexible sensors, which combine the flexibility of the flexible sensors and the high sensitivity of the crack sensing structures, have seen rapid development in recent years. In this review, we summarize the sensing mechanisms of the flexible sensors based on the crack disconnection–reconnection process. The effects of crack type, depth, and density on sensor performance are explored in detail. We also discuss the performance characteristics and applications of the crack-based flexible sensors with various materials, design structures, and crack generation procedures. Finally, the main challenges of the crack-based flexible sensors are also reviewed, and several research directions are proposed.

KEYWORDS
crack engineering, flexible sensors, flexible pressure sensors, strain sensors

1 | INTRODUCTION

Consumer electronics have shown tremendous potential for applications in wearable electronics,1 electronic skin,2 fitness and health monitoring,3 and flexible display4 in the foreseeable future. Flexible sensors with excellent performance can achieve high-perception analysis of external signals. Compared with conventional rigid sensors, they have shown the advantages of low modulus, small bending stiffness, and elastic response to strain deformation.5
Therefore, flexible sensors have tremendous potential in sensing external environmental parameters on non-planar surfaces and in deformable configurations.6

Numerous efforts have been paid to the development of skin-attachable, conforming, and miniaturized health-monitoring sensors.16 However, the development of flexible sensors still faces considerable challenges. In comparison with matured rigid sensing devices, flexible sensors with revolutionary techniques and materials often sacrifice their electrical performance while satisfying mechanical requirements such as bending and folding.17 In recent years, the dual enhancement of mechanical and electrical characteristics of flexible sensors is a hot research topic.18 In 2014, Choi and coworkers19 first introduced nano-cracks into flexible sensors and inspired by the spider sensory system accomplished tremendous improvement in the electrical performance of flexible sensors. Generally, cracks are recognized as defects or failures of a material, while the cracks in flexible sensors play a positive role in the performance of a crack-based flexible sensor. Systematically studies have been conducted on the applications of nano-cracks in flexible sensors and have achieved fruitful results.20 Zhu and coworkers20b studied the generation mechanisms of cracks and first proposed the regulation methods of channel cracks and isolated microcracks, demonstrating the engineering of the connection channels between metal film cracks as an effective way to enhance sensitivity. Wu et al.21 introduced channel cracks into pressure sensors through the pre-compression of foam piezoresistive unit, achieving ultralow minimum pressure detection limit. Moreover, some recent studies have extended the application of highly sensitive cracks in detecting chemicals,20c humidity,20d magnetic field,11 and temperature.20e In summary, the introduction of cracks into the sensor field promoted sensor performance and opened a new avenue for applications in motion detection, pulse monitoring, human–machine interface, medical rehabilitation, robot tactile perception, and environmental perception, as shown in Figure 1.

In this manuscript, we reviewed the applications of crack engineering in flexible strain sensors, flexible pressure sensors, and other functional flexible sensors. The crack formation mechanism and sensing mechanisms of these sensors are discussed, and the influence of fabrication methods and material on crack construction are examined in detail. Table 1 summarizes the crack-based flexible sensors of key materials, main mechanism, sensitivity, sensing range, and cyclic stability. We conclude this review by analyzing the current challenges and future developments of cracks in flexible sensing applications.

2 | CRACK-BASED FLEXIBLE STRAIN SENSORS

Directly spurring, transforming or depositing a conductive layer onto a flexible substrate are the most common methods for strain sensor preparation. Since Choi and coworkers19 first introduced nano-cracks to strain sensors in 2014, crack-based strain sensors show great development in a decade. In this section, we will take a comprehensive discussion on the mechanism of crack-based strain sensors, and the influence of crack depth and density on sensor performance.

2.1 | The mechanism of crack-based strain sensors

2.1.1 | Generation mechanisms of crack patterns and their influence on sensor performances

Cracks on strain sensors with single-layer conductive materials have two major categories: channel crack (Figure 2A) and isolated microcrack (Figure 2D). Channel crack strain sensors often have mismatched modulus between the conductive layer and the substrate. Under large strain, cracks and delamination both form in the sensor. After the pre-strain is released, the partly delaminated crack edges are reconnected and overlapped. The crack structures, which are initialized under large pre-strain, can be divided into three stages under repetitive strain loading as shown in Figure 2B.22 In the first stage, most of the edges are overlapped, and the cross-sectional areas of overlapped parts are much smaller than the interlayer ones of the conductive layer, bringing on much larger resistance. Therefore, only the resistances of overlapped parts should be considered for calculating the whole resistance. Following Ohm’s law, the resistance can be calculated by

\[ R = R_0 = \frac{d \rho_0}{lw} \]  

where \( \rho_0 \) is the resistivity of overlapped part, \( d \) is the distance between overlapped layers, \( l \) is the length of the overlapped section, and \( w \) is the width. According to Equation (1) and the inverse proportion between strain and \( w \), \( R \) should be approximately proportional to strain. In the second stage, the overlapped areas begin to decrease, and some of the initially overlapped edges separate while the distance of the crack edges formed is in the range of a certain tunneling cut-off distance. The resistance is now
FIGURE 1 Schematic illustration of crack-based flexible sensors in development today across a broad range of applications. Reprinted with permission.7 Copyright 2017, American Chemical Society. Reprinted with permission.8 Copyright 2020, Springer Nature. Reprinted with permission.9 Copyright 2022, American Chemical Society. Reprinted with permission.10 Copyright 2019, American Chemical Society. Reprinted with permission.11 Copyright 2021, WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. Reprinted with permission.12 Copyright 2020, PNAS. Reprinted with permission.13 Copyright 2020, WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. Reprinted with permission.14 Copyright 2022, American Chemical Society. Reprinted with permission.15 Copyright 2021, Royal Society of Chemistry.

-dependent on both the overlapped model and the tunneling effect. The third stage refers to fully drawn-apart crack edges, and the resistance is completely determined by the tunneling effect. The resistance rises speedily with the increased strain, and can be expressed as

\[ R = \frac{h^2d}{Ae^2 \sqrt{2m\lambda}} \exp \left( \frac{4\pi d}{h} \sqrt{2m\lambda} \right) \]  

where \( h \) is the Planck constant, \( e \) is the electron charge, \( m \) is a single electron’s mass, \( \lambda \) is the height of the energy barrier, and \( d \) is the tunneling distance. With the increase of strain, the distance between two crack edges raises, and thus the \( R \) increases with \( d \), according to Equation (2).

Moreover, the crack edges present a sawtooth shape, owing to the asperity of fracture surface on the metal layer. As the applied strain increases, the crack structure suffers from a disconnection–reconnection–disconnection process (Figure 2C). While initially applying a strain, the cracks are broken up. But with strain increasing, the substrate will perform a small necking because of the Poisson effect, leading to the swelling and approach of adjacent crack edges and producing several connected points on the conductive layer. When the tensile strain finally increases larger than the sum of adjacent swelled peaks, the cracks are totally taken apart.19 The resistance can be estimated as

\[ R = 2 \left( 1 - \text{erf} \left( \frac{\ln(\varepsilon/\varepsilon_0)}{\mu} \right) \right)^{-1} \]  

where “erf” refers to the error function, \( \varepsilon \) refers to the strain, and \( \mu \) refers to the Poisson ratio.

The isolated microcrack strain sensors, with pre-tensile loading, will generate network cracks on the composite of conductive layer and substrate. The network crack
### TABLE 1  Summary of the crack-based flexible sensors of key materials, mechanism, sensitivity, sensing range, and cyclic stability

| Type of sensors                  | Key materials          | Mechanism                        | Sensitivity (max) | Sensing range (max) | Cyclic stability | Ref. |
|---------------------------------|------------------------|---------------------------------|-------------------|---------------------|-----------------|------|
| Single-layer strain sensor      | Au/PDMS                | Channel crack sensing            | 5000 GF           | 1%                  | 1000            | 20b  |
|                                 | Pt/PU                  | Isolated microcrack sensing      | 30 GF             | 150%                | 500             | 7    |
|                                 | Pt/PUA                 | Crack depth control              | 16,000 GF         | 2%                  | No exact data   |      |
|                                 | CNTs/PDMS              | Crack density control            | 87 GF             | 100%                | 1500            |      |
|                                 | Pt/PUA                 | Programmable crack patterning    | 2 × 10^6 GF       | 10%                 | 5000            |      |
| Hierarchical strain sensor      | PEDOT:PSS-CNF/PDMS     | Multi-units mixture in CL\(^\text{0}\) | 278.1 GF         | 201%                | 2000            | 50   |
|                                 | Au/SWCNTs/PDMS         | Multi-layer sensing              | 3.4 × 10^6 GF     | 100%                | 1000            | 45   |
|                                 | Pt/phage/PDMS          | Interfacial adhesion intensification | 845.6 GF        | 24%                 | 1000            | 46   |
| Single-fiber strain sensor      | g-MWCNTs/WPU           | Annular crack sensing            | 4600 GF           | 10%                 | 2000            | 64   |
| Textile-based strain sensor     | CNTs/PU                | Parallel effect in cracked CN\(^\text{0}\) | 1344.1 GF       | 200%                | 10,000          | 65   |
| Encapsulated strain sensor      | SHP/Pt/PUA             | Protection of encapsulation      | 2102 GF           | 2%                  | 10^6           | 78   |
| Planar pressure sensor          | Pt/PUA                 | Pressure-induced strain effect    | 3.1 × 10^6 MPa\(^{-1}\) | 10 MPa              | 10,000          | 15   |
|                                 | Cu/PDMS                | Metal wire network sensing       | 76.1 kPa\(^{-1}\) | 10.4 Pa             | 1000            | 91   |
| Sandwich-structured pressure sensor | PANIH/rGO@PU          | Crack effect/CN\(^\text{0}\) contact effect | 0.0109 kPa\(^{-1}\) | 25 kPa              | 10,000          | 96   |
|                                 | Carbon foam            | Tunneling effect in discontinuous CN\(^\text{0}\) | 100.29 kPa\(^{-1}\) | 10 kPa              | 11,000          | 98   |
| Hydrogen sensor                 | PdNPs/PU               | Swell effect of CL\(^\text{0}\) | -27.3%            | 10% H\(_2\)         | -              | 12   |
| Alcohol sensor                  | PEDOT:PSS/PDMS         | Swell effect of CL\(^\text{0}\)/condensation effect of vapor | 10^6       | 300 ppm             | -              | 108  |
| Humidity sensor                 | Ni@PU                  | Swell effect of polymer          | 14.96 Ω/1% RH    | 95% RH              | -              | 20d  |
| Temperature sensor              | PEDOT:PSS/PDMS         | Swell effect of polymer          | 0.042°C\(^{-1}\) | 55°C                | -              | 109  |
| Magnetic field sensor           | Graphene/magnetic substrate | Deformation effect of magnetic substrate | RRC of 4.0 × 10^10 | 43 mT             | 10,000          | 11   |

Abbreviations: CL\(^\text{0}\), conductive layer; CN\(^\text{0}\), conductive network; CNTs, carbon nanotubes; GF, gauge factor; MWCNTs, multi-walled carbon nanotubes; PANIH, polyaniline nanohair; PDMS, polydimethylsiloxane; PdNPs, Pd nanoparticles; PEDOT:PSS, poly(3,4-ethylenedioxythiophene) polystyrene sulfonate; rGO, reduced graphene oxide; PU, polyurethane; PUA, polyurethane acrylate; RRC, relative resistance change; SHP, self-healing polymer; SWCNTs, single-walled carbon nanotubes; WPU, water-based polyurethane.

Structure can be simplified to a resistance model with several gaps, isolated islands, and connections, as shown in Figure (2D,E). Different from channel cracks, network cracks only emerge with initially applied large strain, and will not expand or newly appear during the following strain-release process. Their resistance model can also be divided into two stages (Figure 2F). The first stage corresponds to mostly bridged crack edges. The gaps between cracks increase with the increased strain, resulting in a decrease of the conductive path and a correspondingly rise
of sensor resistance. The resistance in this stage can be written as:

$$ R = \frac{R_1 R_2 + 2R_1 R_c + R_2 R_c}{R_1 + 2R_2 + R_c} $$ \hspace{1cm} (4)$$

where $R_1$, $R_2$, and $R_c$ refer to the resistances of islands, connected bridges, and gaps, respectively. In the second stage, the crack edges are majorly disconnected from each other, in which the gap resistance $R_c$ is much larger than the island resistance $R_1$ and the bridge resistance $R_2$. And also, the $R_2$ increases in linearity with strain increasing. Then, Equation (4) can be simplified to

$$ R = 2R_1 + R_2 $$ \hspace{1cm} (5)$$

Generally speaking, channel crack sensors always possess excellent sensitivity, while isolated microcrack sensors exhibit a wide strain range. Choi and coworkers produced channel crack by depositing 20-nm-thick Pt layer onto polyurethane acrylate (PUA) substrate and bending, and thus achieved an outstanding sensitivity up to 2000 gauge factor (GF) within 0%-2% strain range. GF represents the value of the sensitivity, which is calculated as:

$$ GF = \frac{\Delta R}{R_0 \varepsilon} $$ \hspace{1cm} (6)$$

where $\Delta R$ is the change in resistance, $R_0$ is the initial resistance, and $\varepsilon$ is the strain on the sensor. Zhu and coworkers deposited Au thin film on polydimethylsiloxane (PDMS), which exhibited weak interfacial bonding. Channel crack was generated by pre-loading tensile strain, and the developed sensor attained a high GF of over 5000 in the 0%-1% strain range. Furthermore, with the direct laser writing carbonization technique, the polyimide (PI) precursor film could be pyrolyzed/carbonized as a dense,
uniform, and ultrathin carbon film without crack. The as-prepared film was transferred to a flexible polymer film and stretched to induce the formation of straight-line and parallel channel cracks, which are vertical to the stretching direction. The high uniformity of conductive layer led to stress concentration on cracks, further improving the sensitivity to an incredible 450,000 GF in the 1.8% strain range.24 Basically, strain sensors based on channel crack perform superior sensitivity with a relatively small strain range mostly not greater than 5%, while the isolated microcrack sensors attain a wider strain range. By controlling the thickness of Pt deposited onto polyurethane (PU) substrate to 10 nm, a network crack sensor composed with isolated microcrack was obtained, reaching a strain range as wide as 150%.7 To summarize, channel crack sensors can achieve an ultrahigh sensitivity within a small strain range, thus are sensitive enough for capturing various subtle signals; while isolated microcrack sensors can reach a wide sensing range with relatively low sensitivity, which are suitable for whole-body motion monitoring.

The crack type has a significant influence on the performance of strain sensors, and considerable researches have been conducted on the mechanism of crack formation and the methodology of their controllable preparation. Zhu and coworkers20b studied the influence of strain localization on the type of crack in detail. When the conductive film and the substrate show weak interfacial bonding, the strain localization caused by local geometry defects or nonuniform interlayer binding will lead to between-layer traction and induce the delamination of conductive layer. This results in a larger strain concentration on the neck structure generated by delamination (Figure 3A,B), and the initial microcracks are finally expanded to create the channel crack architecture.25 On the contrary, strong interlayer bonding will delocalize the strain and diminish the delamination, and help to form network cracks composed of isolated microcracks. Two types of crack sensors were prepared through adjusting the interfacial bonding strength between Au conductive film and PDMS elastomer. With weak bonding force, the microcracks were extended to channel cracks after tensile loading, and the GF reached as high as 5000 within 1% strain. By adding a 10-nm-thick titanium adhesion layer, the bonding strength was greatly improved and network cracks formed by isolated microcracks existed. Although the GF decreases to 10, the sensing strain range was significantly broadened to 0%–15%.20b

Moreover, dual optimization of sensitivity and strain range for the crack sensors can also be realized by the controlling of crack geometry. MXene film could be combined with thermoplastic polyurethane (TPU) by drop-casting, and their strong combination as well as the uniformity of MXene would bring about strain delocalization and the generation of isolated microcracks on the conductive MXene layer (Figure 3C). The as-prepared network crack sensors could be applied to finger joints and monitor large-strain human body motions such as finger bending. Further oxidation of MXene would induce the growth of TiO2 nanoparticles at the edge of MXene flakes and build isolation among the homogeneous MXenes, constructing a heterogeneous and partially oxidized MXene film. The strain localization was initialized at the incompact TiO2 and brought channel crack structure (Figure 3D), resulting in a superior sensitivity as high as 76,000 GF in 0%–3.6% strain range. The optimized sensors, when pasted to the throat, could detect the vibration of vocal cords and distinguish every vowel in a complex word. As utilized on the wrist, the sensors were apposite to pulse measurement and could supply information for cardiovascular diseases and clinical diagnosis.26 Oxygen plasma was also an effective solution for regulating the geometry of cracks by changing the hydrophilicity of substrates like PDMS. According to the capillary theory for heterogeneous nucleation, different hydrophilicity of substrate surfaces would lead to differences in the way metal growth. As shown in Figure (3E), while a 50-nm-thick Au film was directly grown on a substrate by thermal evaporation, a denser and more uniform Au film was exhibited on hydrophilic fresh PDMS (F-PDMS), which benefited for the formation of neck structures and expanded channel cracks after tensile loading. Conversely, Au deposited on hydrophobic fresh PDMS (F-PDMS) was grown with randomly distributed and intrinsic cracks, and isolated microcracks were constructed due to tensile concentration on these initial cracks. With the adjustment of hydrophilicity, channel crack sensors with a GF greater than 10,000 and network crack sensors with more than 100% strain range were both obtained (Figure 3F). In detail, channel crack sensors were available for the measurement of weak pulse signals and for the perception of sound vibration to recognize notes in a melody. And taking advantage of five wide-range network crack sensors attached to finger joints could help to track and distinguish gestures.27

### 2.1.2 Influence of crack depth on strain sensor performance

In addition to the geometry distribution type of cracks, the specific morphology of a single crack also has a crucial influence on the characteristics of strain sensors. As for crack depth, a deeper crack is always accompanied by larger resistance variation and higher sensitivity of the sensor. Park et al.28 prepared a 20-nm-thick Pt deposition on PUA substrate, and introduced pre-cracks with a certain density by a controlled bending. Subsequently, the
FIGURE 3 Formation mechanism of different types of cracks. (A) Scanning electron microscopy (SEM) images of dislocation slip of 500-nm-thick Cu films deposited on polyimide (PI) under 70% strain. Reprinted with permission. Copyright 2010, Elsevier. (B) SEM images of cross-section of 500-nm-thick Cu deposited on PI with debonding and neck structure under 70% strain. Reprinted with permission. Copyright 2010, Elsevier. (C) Schematic diagram of structure, sensing mechanism, and characteristics of MXene/thermoplastic polyurethane (TPU) sensor. Reprinted with permission. Copyright 2020, Elsevier. (D) Schematic diagram of structure, sensing mechanism, and characteristics of MXene/TPU sensor. Reprinted with permission. Copyright 2020, Elsevier. (E) Manufacturing process diagram of Au/fresh polydimethylsiloxane (F-PDMS) and Au/plasma-treated polydimethylsiloxane (P-PDMS) strain sensors. Reprinted with permission. Copyright 2021, American Chemical Society. (F) Resistance–strain response of the Au/F-PDMS and Au/P-PDMS sensors. Reprinted with permission. Copyright 2021, American Chemical Society.

The depth of cracks could be regulated by adjusting the applied pre-stretching force. With the establishment of an analytical model about crack geometry (Figure 4A), the increase of crack depth was found to provide a positive impact on sensitivity, and a superior GF as high as 16,000 in 2% strain range could be achieved through crack depth changing.

Besides the degree of pre-strain, the type of substrate materials shows a close relationship with crack depth as well. When indium tin oxide (ITO) was, respectively, deposited on polyethylene terephthalate (PET) and PDMS, the cracks of ITO were not extended to the substrate on PET film, while the cracks were penetrated and further propagated to substrate on PDMS film, which improved the crack depth. Thus, pre-constructing an easy-to-crack layer between the conductive material and the substrate is a considerable method for the sensitivity enhancement of sensors. For instance, when depositing Pt nanoparticles (PtNPs) onto alumina/Kapton composite film with natural cracks, the crack depth could be regulated by changing the thickness of the alumina layer. As mentioned above, the cracks would penetrate the alumina layer, and thus deeper cracks, for thicker alumina, could induce larger crack gaps while strain applied, indicating more obviously conductive path variation for the attached PtNPs and higher sensitivity for the whole sensor (Figure 4B).

Apparently, the crack depth is also dependent on the thickness of the conductive layer such as Au/Cr. Controlling the thickness of Au as a constant of 20 nm, while the thickness of Cr was increasing below 60 nm, a sensitivity enhancement was shown due to the crack depth increase. However, when the thickness of Cr was larger than 60 nm, the decrease of crack density with the increase of Cr thickness would induce a crack dense effect to oppositely diminish the sensitivity. Therefore, the influence of crack density on the performance of crack sensors is also crucial enough for discussion.

2.1.3 Influence of crack density on strain sensor performance

Plenty of researches focus on crack density modulation for improving sensing range or balancing the contradiction between strain range and sensitivity. For nanocrystal
materials like Pt, the cracks were extended along the grain boundaries with low atomic density and weak atomic bonding, so decreasing the thickness of Pt film would lead to the decrease of crystal dimension and thus attain higher crack density. As shown in Figure (4C), sensors with high crack density performed large space capacity and thus the measuring range was remarkably enhanced to 150%. As for low-dimensional materials such as carbon nanotubes (CNTs), the crack density could be controlled through their layer number. Briefly, lower crack density as well as an increasing crack length was shown on the composite film with more CNT layers, resulting in the partial sacrifice of sensing range but an improvement of sensitivity. Therefore, by tuning the layers of CNT, it was possible to realize relatively high sensitivity in the required strain range. Immersing the prepared multilayer CNT film into a PDMS-based solution was a common method for strain sensor fabrication, and thus CNTs were embedded in PDMS substrate to provide a firm bonding. In this situation, no new cracks (NCs) would be generated even under large strain, accompanying with sensitivity diminishing. On the contrary, directly spray coating CNT on TPU substrate benefited for crack elongation and generation. The ratio of crack area continuously augmented with the increase of tensile strain, resulting in a still distinct resistance variation even when applying large strain. The as-prepared sensor reached both a large sensing range to 300% and an incredible high GF to 83,982.8, supplying a solution to the low sensitivity defect of high crack density sensors. Additionally, a predesigned geometric structure directly accomplishes to controllable crack production. For instance, the designed microgrooves on an electrical sensitive layer would cause stress concentration and crack expansion. Consequently, the highly manageable dense crack network was attained, which benefited to the effective regulation of sensor stretchability. After
sputtering Au/Cr on the substrate with designed V-type grooves, a stress localization would exist at the grooves when tensile loading. Through programming the quantity and density of the V-type groove array, straight-line cracks with corresponding numbers, accurate intervals, and length were available within a reasonable strain range (Figure 4D). A three-dimensional (3D) printing technique was introduced to print substrate with aligned groove patterns. After Pt sputtering and tensile loading, the bottom of grooves performed as stress concentrated zones, and thus self-aligned crack arrays with high density were generated (Figure 4E), acquiring a high GF of 184.4 

Other than V-type grooves, any structures that can induce regular or cyclic stress concentration are suitable for preparing microcracks with specific density. For example, introducing an equidistant array of holes with a distance of 20 μm on the surface of PUA would prompt the origination of straight cracks between holes, owing to stress concentration. The morphology of specific induced straight cracks, compared with naturally prepared ones, diminished the crack reconnection due to the Poisson effect, and induced a distinctive resistance rising. The strain sensor maintained an up to $2 \times 10^6$ GF within 10% strain range, which provided enough resolution for accurate pulse monitoring. Similarly, a patterned PUA substrate with a nanowire array and nanoscale gaps could be prepared by molding on a specific Si template. Cu was deposited on PUA nanowires to establish a Cu nanowire array, and various conductive materials (Pt, ITO, and Cu) were vertically deposited onto it. The stress localized structure with highly parallel Cu nanowires served as an effective solution to overcome the influence of interfacial bonding strength and generate highly self-aligned and dense channel crack array regardless of the materials. An enhancement of durability to 20,000 cycles as well as a high GF of 670 in 0.3% strain range could be observed on the as-prepared sensor, and subtle variation of neck pulse could be detected. Kim et al. further exploited linearly polarized light to prepare a liquid crystal polymer network (LCN) with aligned monomers. As shown in Figure (4F), the deposited Cr layer on LCN enabled controllable cracks parallel to the nematic director of liquid crystals, which attributed to the interaction between the inner tensile stress of Cr and the oriented structure of LCN. Higher sputtering power results in faster deposition and higher tensile stress in Cr, leading to an increased crack density of later LCN (Figure 4F). Therefore, changing the sputtering power enables the control of crack density effectively. More interestingly, the cracks could even be programmed to express patterns like circles and letters on LCN substrate, providing a potential solution for the programmable design and characteristic improvement of single-layer crack sensors. Differently, low-dimensional materials like CNT performed mostly independently on substrate morphology, thus directly laser cutting single-walled carbon nanotube (SWCNT) membrane to produce microgrooves supplied as an alternative. After that, the SWCNT film with PDMS encapsulation produced channel cracks with manageable density during a repeatedly roll-to-roll process, and the different crack density referred to a variety of strains ranging from 60% to 150%. Likewise, laser-induced strip patterned graphene also attained microcracks with programmable density. The structures of cracks, such as their morphology and dimension, were regulated with the parameters of the laser, and finally a strain range of 50% was obtained accompanied by a maximum GF of 191.55.

### 2.2 Crack-based strain sensors with hierarchical structure

The crack strain sensors with different conductive materials possess different advantages and defects. Brittle metal membranes are ultra-sensitive to strain, but their conductive paths are easily broken by cracks, always exhibiting a small sensing range. The composite conductive films based on low-dimensional materials possess superior conductivity under large strain, benefiting from huge specific surface area and bridged structure of conductive components. Thus, they express opposite characteristics with a large sensing range and relatively low sensitivity. To pursue a large enough sensitivity and strain range, some of recent researches mixed several sensitive materials or produced hierarchical sensitive structures to acquire crack strain sensors with excellent electrical performances.

Composite conductive materials with multiunits can achieve capacity improvement on strain sensors by enhancing the mechanical and electrical performance of conductive layers. Nanowire-based materials can twine and bridge together to form the conductive network, which is commonly too robust and stable to reach high sensitivity except for purposely breaking the intrinsic structure by adding other materials. The composite conductive layer with Ag nanowires and graphene oxide formed a nonuniform interface because of weak interfacial strength with the TPU substrate. With tensile strain, stress concentration occurred accompanying with swift crack generation
and expansion. The interfacial uniformity would decrease with a rising ratio of graphene oxide in the mixture conductive material, thus inducing more cracks. The sensor achieved a GF as high as 4000 in 1% strain, which showed an outstanding sensitivity improvement. By doping brittle 3-aminopropyltriethoxysilane (KH550) polymer into stable CNT, fragile “sensitive points” were introduced into the conductive network and induced stress localization for crack production and expansion.45 Similarly, the content of KH550 in the mixture could influence the mechanical performance of the conductive network, further regulating the length, density, and other morphology of cracks and balancing the sensitivity (5–1000 GF) with the strain range (2%–250%).

A conductive composite could be improved by introducing an addition of carbon nanofibers with excellent conductivity into poly(3,4-ethylenedioxythiophene) polystyrene sulfonate.50 The crack sensors were prepared by coating the composite onto PDMS and stretching. While tensile loading, the carbon nanofibers could not only bridge two adjacent microcracks to maintain a conductive path, but also prevent microcracks to expand to channel cracks with their tenacity, highly improving the stretchability of the crack sensor. The as-prepared strain sensors were available for various applications which required high sensitivity and large stretchability. For instance, they could be attached to different parts of body and detect body motion and physiological signals such as wrist pulse, breathing rate, facial expression, finger bending, etc., expressing huge potential in health monitoring and human–machine interaction. By blending 1D CNTs with 2D graphene nanoplatelets (GNP), a GNP/CNT hybrid film, which possessed the characteristics of low-dimensional structures and initiated cracks among GNPs during stretching, was prepared.46 Differently, the CNT dispersion showed relative sliding and functioned as the bridges among cracks, which could reconnect the broken GNPs under large strain and maintain a conductive path (Figure 5A), overcoming the defect in highly sensitive graphene strain sensor that the conductive network could be irreversibly damaged under small strain (7%). As a result, the optimized sensor realized a GF as high as 197 in a strain range of 10%, and possessed an excellent stretchability of over 50%. The weak interaction between graphene flakes, because of the short and disordered cracks with pre-tensile, limited the sensitivity improvement of graphene-based strain sensor. The conductive composite film, composed of reduced graphene oxide (rGO) and polydopamine (PDA)/Ni, presented the synthetic effects of the chemical binding and physical blocking of PDA, as well as the ionic and coordination bonding of Ni2+, enhancing the interactive force between rGO flakes.47 As a consequence, the cracks only occurred at the weak positions of the rGO layer, diminishing the formation of squamous cracks and their disordered expansion. The cracks were majorly for eliminating internal stress, and thus were long and vertical to tensile direction (Figure 5B), which benefited for swift changing of conductive channels under tensile loading and highly improved the sensitivity.

In addition to taking advantage of conductive composites, directly preparing hierarchical structures with different functional material layers is another crucial approach for sensor performance regulation. According to the buckling theory of rigid film on a flexible substrate, while tensile loading, cracks along stretching direction initially occur to release strain energy. The wavelength of cracks increases with film thickness, but the releasable strain energy decrease with a worsened stretchability and a rising brittleness. Based on the above theory, Liu et al.52 fabricated a SWCNT conductive film with gradient thickness on a PDMS substrate. The thicker part, which possessed higher brittleness, initialized cracks with small applied strain and induced a significant resistance variation, while the thinner part, with a smaller wavelength, could withstand the higher strain and meanwhile maintain conductive access. Therefore, combining the brittleness of thick parts and the stretchability of thin parts, the sensor could simultaneously reach a high sensitivity (up to 161 GF) and a wide strain range (0%–60%). By subsequently depositing Au film with high sensitivity and graphene film with a wide sensing range on the elastomer, the sensors with parallel hierarchical structures were prepared.20b Due to the crack effect, Au film could access an ultrahigh sensitivity in a small strain range of 0%–2%. On the other hand, graphene film, although possessed relatively low GF, could withstand up to a 10% strain range. Taking advantage of both the characteristics, the Au layer performed dominantly and possessed high sensitivity at small strain, while, with large strain, the penetrating cracks of Au film led to an infinite resistance, but the parallel graphene layer was still conductive, broadening the sensing range of the whole sensor. The resulted sensors possessed both superior sensitivity at small strain and large sensing range (Figure 5C). SWCNT performed similarly with graphene and could constitute multilayer crack sensors in cooperation with Au film.48 The superior conductivity as well as the high Young’s modulus of Au incited crack production and distinctive resistance variation under subtle strain, while the outstanding flexibility of SWCNT film provides extra conductive access by bridging Au cracks under the large strain (Figure 5D), thus realizing both high sensitivity and wide sensing range. By reducing the Au/SWCNT line width, the staggered crack network originated on Au film would expand to the whole conductive composite, highly enhancing the sensitivity although with the sacrifice of stretchability.
Moreover, the compatibility and the adhesion between the substrate and sensitive unit are crucial for sensor performance. With weak interfacial adhesion and poor compatibility, it is harder to produce expected cracks. As a solution, the interlayer, which acts as a medium, can be introduced to form a multilayer structure with conductive materials, and thus generates expected crack patterns for satisfying the requirement. Cho et al. demonstrated the induction of interlayer on cracks. They introduced atomically thin 2D interlayers, such as graphene, molybdenum disulfide, and hexagon boron nitride, between Au film and PDMS. The 2D interlayer modified the interfacial adhesion and increased the film/substrate modulus ratio, inducing transformation of the crack pattern on Au film from channel crack to serpentine network microcrack. Introducing a bacteriophage (phage) film between Pt film and PDMS had a similar function. While stretching, the rippled surface structure of phage film as well as its good adhesion with Pt film assisted the stress redistribution and transformation from PDMS to Pt film. The redistribution effect of stress constrained the formation of channel cracks on Pt film, and the randomly distributed network microcracks were generated instead (Figure 5E). The sensor performed high stretchability to 24% strain and an excellent sensitivity up to 845.6 GF, which was available for monitoring both large human motion like finger and knee joint movement and weak physiological signals like breathing and pulsing.

Ineffective adhesion also occurred between Au and PET films, and an addition of Cr/MoO$_3$ metal film between the two layers was an effective solution to improve sensor performance. MoO$_3$, because of its strong adhesion and good compatibility with PET, is majorly performed as the attached layer to ensure the stability and the durability of sensors. The Cr film, which benefited from high brittleness, could form highly sensitive channel cracks under stretching. Owing to the strong adhesion between Cr and Au, the crack patterns on Cr film could be directly mapped to Au conductive layer and thus the stable and expected crack structures were acquired. These multilayered sensors achieved a sensitivity as high as 1600 GF and possessed relatively high durability for 5000 stretching cycles, showing excellent reversibility. Besides, interlayer could also be obtained with the treatment of substrate materials. Amjadi et al. treated the surface of Ecoflex with oxygen plasma, resulting in the generation of a hard, oxidized polymer layer on the elastomer. With tensile loading, the oxidized layer cracked and produced parallel aligned microgroove patterns on Ecoflex. Due to the excellent adhesion between the graphite conductive layer and brittle oxidized polymer, as well as the localized deformation and the stress...
concentration, the cracks were generated along the edges of microgrooves on the conductive layer, which were similar to the patterns of microgrooves. The length and density of microgrooves on the oxidized layer were controllable by regulating the processing time of oxygen plasma, and thus the related crack pattern could be obtained on graphite. The outstanding sensors with expected crack patterns reached a 50% strain range with up to 100 GF. The sensors could be attached to the actuator of the pneumatic soft robot to monitor the touching condition, contact force and bending position of grasp fingers, which could be applied for the feedback control of the soft robot and artificial skin, realizing the intelligent perception between robot and environment or between robot and the interactive target.

2.3 Crack-based fabric strain sensors

Fiber-shaped electronics have become a hotspot in the field of intelligent wearable devices due to their superior flexibility, weavability, and comfort. Thanks to the intrinsically low elastic modulus and the intertwined net-like structure, the fabrics usually possess excellent stretchability and the fabric-based sensors always express large sensing ranges. Therefore, the introduction of strain-sensitive crack structures on the fabrics is conducive to the production of simultaneously highly sensitive and wide-range strain sensors. Fabric-based sensors are usually prepared by employing conductive nano materials, such as CNTs, graphite nanoplatelets, Pd nanoparticles (PdNPs), silver nanowires (AgNWs), etc., on them to form a fabric conductive network by spray coating, drop coating, or spin coating and introducing crack structures with pretensile loading. In this section, single-fiber crack strain sensors and textile-based crack strain sensors are discussed sequentially.

2.3.1 Singer-fiber strain sensors

Single-fiber strain sensors are in small size and flexible enough for strain measurement in microscale environments as well as possessing the capacity to be further knitted to large-scale textile sensors. Under large strain, conductive fiber sensors without cracks exhibit low sensitivity due to the gradually increased distance between the flakes of 2D conductive materials and the slowly rising resistance. With the introduction of cracks, the expansion of microcracks during stretching induces the crack effect, leading to the significant resistance changes and the highly improved sensitivity. However, the microcracks randomly generated on single-fiber conductive sensors still result in a relatively small change of conductive path and relatively low sensitivity under large strain. To improve the sensitivity, some structure designs that could induce stress concentration and related crack generation were introduced into the fiber sensors. For example, the fibers could be dip-coated into conductive solution with PDMS microbeads. With stretching, the stress concentrated on the microbeads of the conductive layer and the microcracks generated nearby (Figure 6A). Therefore, the crack density could be regulated to improve sensitivity by adjusting the content of PDMS microbeads in the solution. Simultaneously, the fiber sensors could still maintain conductivity under relatively large strain because no cracks occurred at the places with no microbeads. The resulting sensors could work under 400% strain with the highest sensitivity of 863 GF.

In addition, introducing annular cracks could also improve the sensitivity. But similar with channel crack structures in single-layered crack strain sensors, 3D annular crack structures had great negative impact on strain range, although leading to an obvious resistance variation under stretching. The contradiction between sensing range and sensitivity could be effectively solved by designing the crack morphology of multilayered conductive materials. By employing multi-walled carbon nanotubes (MWCNTs)/water-based polyurethane (WPU) composite as the conductive material, a multilayered structure with two different conductive layers dip-coated onto elastic fibers was prepared. The bottom one was a conductive layer without cracks (10 wt% g-MWCNTs/WPU) and the top one was a sensing layer with annular cracks (40 wt% g-MWCNTs/WPU). With tensile loading, the annular cracks on the sensing layer opened and the resistance rose rapidly, while the non-crack layer remained conductive. As shown in Figure (6B), the two layers formed an island–bridge structure which broadened the sensing range and diminished the negative influence of annular cracks on strain range, improving the measurement range by over five times (10%) and maintaining sensitivity up to 4600 GF. The sensors could be attached to the radial artery of wrist for accurate measurement of pulsing, and had a promising application in clinical field because of its simple fabrication process, portability, and excellent performance. A single cracked conductive fiber could be prepared with AgNWs and PU, and a fiber sensor array could be fabricated with several cracked fibers arranged orthogonally and encapsulated in PDMS elastomer. The assembled sensor array could measure multidimensional force and strain, which worked steadily for 5000 cycles within 30% strain (with a GF of 3.2). Benefiting from the relatively large sensing range as well as the multidirectional strain sensing ability endowed by the orthogonal sensing array, only a single sensor attached to the back of hand could monitor and approximately
distinct different hand movements by detecting the resistance variation caused by skin strain on different joints, illustrating its potential for wearable artificial skin.

2.3.2 Textile-based strain sensors

Textile-based strain sensors and the intertwined fabrics network composed of fascicular or reticular fibers can maintain a stable conductive path under larger tensile strain and achieve a wider sensing range, which are accessible for full-range detection and sensing of human motions and physiological signals.\(^{71}\) For example, the bundled fiber (yarns)-based crack sensors with PdNP conductive network generated cracks in high density through repeated pre-stretching, achieving a measurement strain of 70% with an excellent sensitivity of 2040 GF, which was significantly broader than that of the sensors formed by single fiber.\(^{12}\) As shown in Figure (6C), the conductive network model of parallel cracks on yarns showed that the cracks on a single fiber increased and expanded when stretching, leading to a rapid variation of conductive path. But the parallel conductive network in yarns could reconnect the thoroughly broken conductive islands and maintain the conductive access with high-density cracks, effectively preventing crack propagation and catastrophic failure and thus retaining a relatively high sensitivity with a wide sensing range.\(^{65}\) To further enhance the sensing range, the wrinkle structure and the crack structure were constructed in the CNTs/PU-based yarn crack sensor.\(^{72}\) Due to the function of the wrinkle structure, the strain range of the sensor reached 200% with a highest sensitivity of 1344.1 GF, achieving a large sensing range and high sensitivity simultaneously. The sensors could be directly sewn to the gloves, wristbands or knees on pants to monitor the related human motions with large strain. The sensors could also accurately detect the pulse, vocalization, facial expressions, breathing, and other weak human
physiological activities when attached to corresponding human body parts, demonstrating the prospect of highly wearable devices for full-range physiological detection.

The mesh-like textile structures have the highest compatibility with conventional textiles and play a significant role in the preparation of wearable sensors. With a low content of conductive material, a discontinuous conductive network was formed on the fibers and the conductive accesses were determined by the interconnection of the conductive fiber network. Thus, a small strain would cause the detachment of cross-linked conductive fibers and lead to the significant variation of resistance and the circuit failure, resulting in a sensor with high sensitivity and relatively small sensing range. With a higher content of conductive material, a uniform and complete conductive network was generated on the fibers and the fiber detachment under small strain would not obviously change the conductive path or cause the circuit failure. While under large strain, the strong adhesion between the fibers and the conductive coatings restrained strain localization and enabled the generation of uniform microcracks (Figure 6D). Moreover, the increased proportion of conductive material reduced the modulus difference between the coatings and the fibers and enhanced their compliance, thus increasing the crack density and expanding the sensing range. However, it should be noted that for strain sensors with high crack density, although possessing a larger sensing range, the slower opening process of cracks caused a smaller resistance variation and a decreased sensitivity. In order to improve sensitivity, some studies directly employed conductive materials as the conductive mesh layers. Li et al. embedded a mesh-like graphene woven fabric (GWF) composed of graphene microribbons (GWRs) into a PDMS elastomer and introduced cracks through stretching to produce a GWF-based crack sensor with a conductive network (Figure 6E). Due to the strain-sensitive property of the fabric network, the cracks on GWF would increase rapidly under stretching and generate a crack structure with high density, resulting in an as high as 1000 GF with a strain range of 2%–6%. While under larger strain, the cracks would expand with a swiftly increased resistance until the GWRs were completely broken, reaching an ultrahigh sensitivity of over 10^6 GF. Since the parallel fibers of GWF were broken separately or alternately, the sensing range could be broadened to 10% by increasing the amount of parallel GWR fibers.

Another distinctive advantage of the mesh-like textile is the air permeability and excellent wearing comfortability. Li et al. dip-coated cellulose nanocrystal (CNC)/MXene hybrid conductive material on the flexible TPU non-woven fabric and developed the crack-based mesh fabric sensors through stretching. The as-prepared sensors exhibited tremendous advantages in long-term collections of human body signals exploiting their excellent flexibility, breathability, and comfortability. Real-time monitoring of human motions and physiological signals was realized with the sensors. For example, the sensors could be attached to the wrist to accurately observe the three typical peaks of the pulse waveform, corresponding to the percussion (P), tidal (T), and diastolic (D) waves (Figure 6F), providing a low-cost, simple, and convenient method for a long period of human signs monitoring and demonstrating the potential as next-generation, comfortable, and wearable flexible sensors. The mesh-like textiles could also improve the durability of sensors. For film-based crack sensors, the differences of mechanical and surface properties between the substrate and the conductive film induced the debonding and delamination and diminished the durability. However, as shown in Figure (6G), the porous structure of mesh-like textiles could form an interlocked strong interpenetrating structure with the conductive network, which tremendously enhanced the durability to up to 10,000 cycles. The content of each materials in the conductive solution could also influence the durability. Carbon-based ink consisting of environment-friendly carbon black, gelatin binder, and elastic PU could penetrate into textiles and develop a uniform coating on the fiber surface through a simple dip-coating technique. After stretching, the contraction of the woven yarns prompted the partial delamination on the surface, thus inducing the formation of macroscopic cracks in micrometer scale and the preparation of triaxial woven yarn sensors. PU played an important role in improving the sensor durability, as PU could improve the adhesion between carbon black and the yarns. On the sensors with an increased PU mixing ratio of 10 wt%, carbon black and PU were densely integrated into the yarns with a strong bonding force. The resulting sensor expressed superior stability and durability for 5000 testing cycles.

2.4 Influence of encapsulation on crack-based strain sensor

The cracks of conventional strain sensors are directly exposed to air and the conductive layer is susceptible to friction, water, chemical substances, and other external factors. For example, a humid environment will easily induce the delamination, corrosion, and oxidation of sensors. In addition, due to the fatigue effect of the polymers, the stress concentration can easily deepen the cracks, leading to a continuous rising of resistance and resulting in the degeneration of the crack sensors. Encapsulation on the sensor can effectively protect the sensor from water, chemicals, and other environmental influences and thus improves its mechanical resistance.
to further enhance its durability. Kim et al.\textsuperscript{76} employed fluorinated ethylene propylene (FEP) to encapsulate Au/Cr/PI sensors through a high-temperature adhesion technique (Figure 7A) and explored the reasons for the durability enhancement of the sensors. The low water permeability and high chemical resistance of FEP resulted in the water and corrosion resistant properties of the encapsulated sensors. Meanwhile, the FEP encapsulation layer possessed a smaller Young’s modulus than that of the PI substrate. The stress on FEP was smaller than that on the PI and thus the FEP layer was tightly attached to the sensitive metal during elastic deformation, which diminished the stress concentration on cracks and enhanced the mechanical resistance. The 50-μm-thick FEP encapsulated sensor could still retain 93% of its initial GF after being immersed in water for 6 days and perform a GF retention of 90% after the 4-day immersion in chromium corrosive agent, with superior durability of 15,000 testing cycles under 0%–12% strain loading. In summary, the FEP encapsulation distinctively improved the durability of sensors, which could withstand human secretions such as perspiration, saliva, and gastric fluid and illustrated a potential in wearable and medical applications. PDMS is also performed as a common encapsulation material for the crack strain sensors.\textsuperscript{77} The hydrophobic property of PDMS enabled the stable utilization of crack strain sensors in various of ionic liquids. Besides, the Cr/SiO\textsubscript{2} chemical bonds between Pt conductive layer and PDMS enhanced their adhesion and further improved the stability of sensors. No change in GF because of fatigue effect was observed with over 5 million heartbeats, achieving long-term and highly sensitive measurement of the cardiomyocytes. Moreover, the PDMS encapsulation of the crack strain sensor provided isolation to prevent the direct contact and the formation of parallel circuit between the metal layer and the conductive solution in the culture medium, eliminating the interference of liquid resistance and contributing to more accurate experimental results. Self-healing polymer (SHP) possessed the characteristic of low-temperature polymerization, which could heal at room temperature and was developed to encapsulate crack strain sensors.\textsuperscript{78} With repeated stretching, the accumulation of stress would cause a continuous expansion and propagation of cracks from the sensitive layer to SHP and a gradually diminished sensitivity. After being heated at 50°C for 10 min, the recovery of SHP chemical bonds would induce the close of SHP/Pt cracks to the initial state (Figure 7B). This self-healing property warranted...
the repairing and maintaining of the crack gaps, which tremendously enhanced the durability of the sensors to over 1 million cycles with an almost constant sensitivity.

Although encapsulations substantially improve the durability and cyclic stability, the introduction of them will to some extent diminish the sensitivity of the sensors in most cases. As shown in Figure (7C), the 25 or 50-μm-thick FEP encapsulated sensors exhibited lower resistance variations under strain than the ones without encapsulation, and the GF decreased with an increased sensor thickness. The comparison of sensors with and without SHP encapsulation illustrated that the encapsulation effect would cause a 5% resistance decrease on the SHP encapsulated sensors, reducing the sensitivity to some extent. However, rational design of the sensor structure could offset the adverse effect of the encapsulation on the sensitivity, allowing the sensor to exhibit a remarkable GF, such as the PDMS encapsulated Ni@graphene-coated PU sponge (GPUS) strain sensors. Within 20% strain, the graphene sheet provided stable conductive path and the resistance change mainly attributed to the gradual expansion of electrodeposition ICs and NCs generated during PDMS encapsulation, which resulted in a relatively low sensitivity with a GF of 36.03 as shown in Figure (7D). While in 20%-65% strain range, the sliding of graphene sheet produced a microcrack network, which could maintain conductive under large strain. Meanwhile, the microcracks on the graphene layer promoted the generation of derived cracks around the ICs and NCs. This dual-microcrack mechanism boosted the resistance change and tremendously enhanced the sensitivity to as high as 3360.09 GF (Figure 7D).

In some cases, the encapsulations could broaden the measurement range of sensors as well. The sensors realized encapsulation by coating uncured liquid PDMS onto the SWCNT-sensitive layer and introduced the channel crack structure by pre-stretching. Under low strain, the average gap distance of channel crack increased with an elevated resistance, reaching superior sensitivity up to 107 GF. The electrical conduction between cracks was accessible through the intertwined CNT at this stage, but the connection between CNTs was broken under larger strain. Profiting from the partial penetration of PDMS precursor into SWCNT during encapsulation, the embedded CNT could still connect the cracks and retain electrical transportation while the delamination occurred at the interface of PDMS/SWCNT under large strain. This residual conduction mechanism at the delaminated interface distinctively broadened the sensing range to 50%, realizing the stretchable sensor with outstanding sensitivity. Similarly, Xin et al. transferred laser engraved SWCNT onto the PDMS substrate and poured the PDMS precursor on them. The SWCNT layer was completely encapsulated to form a sensor after curing at 70°C, with some PDMS precursor penetrating into SWCNT. Under large strain, the delamination of SWCNT/PDMS left obvious CNT fragments on the interface, and the conductive access through the residues synergized with the laser-regulated high-density cracks further expanded the sensing range to 100%.

3 | CRACK-BASED FLEXIBLE PRESSURE SENSORS

Flexible pressure sensors have recently shown great promise as important components in the fields of touch screen, wearable electronics, human–machine interface, and real-time physiological detection. Particularly in soft tissues such as soft robotics and human skin, flexible pressure sensors cannot only perform as a major method for tactile interaction to perceive the pressures generated through physical contact between or inside the body, but also carry physiological information under various deformations, illustrating tremendous significance in electronic skin, human–machine interaction, and medical monitoring. Nowadays, extensive researches have been conducted on flexible pressure sensors and a variety of transformation mechanisms and structure designs are introduced for their development with conformality on arbitrary surfaces. Pressure sensors with planar cracked-sensitive units possess extremely simple structure and ultrahigh sensitivity under subtle pressure. Similarly, introducing cracks to original sandwich-like capacitance/resistance pressure sensors can tremendously enhance sensor properties. In this section, we give a comprehensive description of recent planar-structured and sandwich-structured flexible crack pressure sensors.

3.1 | Crack-based pressure sensor with planar structure

Planar crack sensors can transform the applied pressure into a resistance signal through the pressure-sensitive cracked structure on the conductive layer. Similar to strain sensors, crack pressure sensors possess the superiorities of simple preparation, excellent sensitivity, and plain structure. For instance, the preparation of a planar sensor could be achieved through repeatedly pencil-drawing on paper with a cantilever structure, which formed a firm adhesion between graphite and the paper. Under compression, microcracks were introduced by the tensile strain resulting from the deflection of the cantilever structure, constituting an electronic whisker (Figure 8A). While pressing, microcracks occurred between the graphite flakes on
the surface of the electronic whisker, which realized highly sensitive pressure sensing. Lee et al. study the mechanism of planar crack pressure sensors in detail and provided the modulation method for sensitivity. As shown in Figure (8B), with an applied positive pressure, an upward deflection of the PDMS membrane induced the tensile strain on the sensing film. The strain-induced crack (IC) opening on the sensitive layer enabled prompt resistance increase on silver nanoparticles film, which was calculated to 0.35 relative variation under 1 kPa, exhibiting high sensitivity. Moreover, the rigidity of films, to some extent namely their thickness, had a significant effect on their deformation ability under pressure and further influenced the sensitivity. The 100-μm-thick PDMS-based sensor possessed a sensitivity 28 times higher than the 500-μm-thick one, demonstrating that a membrane with lower elastic modulus expressed larger strain and enhanced sensitivity under the same pressure.

The sensitivity could also be modulated through the shape of conductive units. While applying a pressure, the generated strain would induce the opening of micro-cracks on the serpentine Au thin film pressure sensor. The cracks were not completely broken to cut off the conductive pathway but remained a sub-nanoscale metal layer at the bottom, the GF simply becomes

$$GF = \frac{\Delta R}{R} = \frac{T_0}{T_c}$$

where $T_0$ and $T_c$ are the thickness of the original metal film and the residual film at the bottom. The GF calculation formula showed that sensitivity was proportional to the thickness ratio of the metal film to the residual film at microcracks, which led to an ultrahigh sensitivity because of the ultrathin residual film. Moreover, the serpentine patterns could elongate the length of the Au thin film resistor with a minimum sensing area, which also had a positive effect to the sensitivity. The as-prepared sensor demonstrated a superior sensitivity of 0.23 kPa–1 within 0–50 mmHg. In addition, the sensor possessed the flat (200 μm thickness) and small-area (3 × 3 mm²) structure as well as a simple fabrication process and good compatibility, which was explored with the opportunities for large-area and high-resolution artificial skin applications. To further elevate the sensitivity of planar crack pressure sensors, photolithography was also introduced for preparing orderly microstructures in conductive films. The angles of triangles were aligned orderly on the film, expressing a relatively low sensitivity only related to the strain effect of conductive films under small pressure. While the applied pressure increased, the stress concentrated at the angles and IC initiation and penetration, which promoted the length and depth increase of cracks. The phenomenon resulted in a steep increase of resistance and assisted the accurate pressure detection, reaching a sensitivity up to 24.2482 kPa–1. The crack pressure sensor accomplished pulse-induced cyclic pressure monitoring around the wrist, and the consequence highly matched.
with the electrocardiogram, demonstrating potentials in wearable devices and medical treatment.

The Poisson effect showed influence on planar strain under pressure, illustrating the Poisson’s ratio of substrates as a factor to tremendously affect the sensitivity. Two-layer structures based on TPU with different elastic moduli and different patterns could be prepared through 3D printing. The difference of mechanical Young’s moduli in lateral and longitudinal directions caused the anisotropic Poisson’s ratio, which was 0.67 in the lateral direction (the direction crack width expanded) and 0.11 in the longitudinal direction. Compared with conventional sensors with isotropic Poisson’s effect, the novel sensor presented a more obvious strain-induced elongation in a specific direction of the elastomer while pressure loading. In detail, the substrate was elongated for 5.9% and 0.98%, respectively, laterally and longitudinally with an 8.9% vertical compression, manifesting a remarkably increased strain in the width expansion direction of cracks and an enhanced sensing ability. The sensor illustrated a superior sensitivity of 3.1 x 10⁶ MPa⁻¹ in 3 MPa, a maximum sensing range of 10 MPa and the durability of 10,000 pressure cycles at 10 MPa. The ultrathin, highly flexible, and extremely stable properties of the sensors enabled their integration and miniaturization for rehabilitation equipment. For head and neck cancers, approximately 70% of the patients suffered from swallowing and chewing impediments after cancer removal and jaw reconstruction. The generated sensing arrays could detect the unbalanced occlusal pressure and perform as jaw rehabilitation devices for people with swallowing difficulty. In addition, a coupling of cracks and wrinkled structures assisted to accomplish a larger sensing range. Zhou et al. developed Au/Pt sensing layer with nano-to-microscale wrinkled structures on silicone elastomer through sputtering. The sensors presented lateral expansion with pressure loading, which caused the occurrence of cracks. Meanwhile, the wrinkled structures provided an extra strain mitigation effect and their spread along stress direction permitted a wider sensing range.

A large-area, uniform crack network could be generated through spraying solution onto the substrate, and composed a well-distributed, highly conductive metal wire network with metal deposition on it. The nanowire conductive network based on crack structure possessed superb conductivity and mechanical flexibility, not to mention the properties of a simple fabrication process and low cost. This preparation method for conductive electrodes could also be expanded to pressure sensors to acquire superior performance. As shown in Figure (8C), Li et al. coated a crack template layer onto ITO glass and developed a homogeneous network crack template after the crack template dried and cracked. The Cu network was generated in the gaps of network cracks through electroplating, and was transferred onto the PDMS substrate to compose a pressure sensor. The vertical and localized pressure excited abundant Cu wires to fracture, leading to a noticeable decrease in the conductivity of the Cu network. Meanwhile, the sensitivity reached as high as 76.1 kPa⁻¹ with a subtle pressure detection limit to 1.1 Pa (Figure 8D), which even surpassed the capability of human skin. A similar pressure sensor was produced by depositing Ti₃C₂–MXenes into crack gaps on the surface of the PDMS/Mo substrate. The crack-induced net-like MXene conductive layer not only endowed the sensor with outstanding sensitivity of 516.74 kPa⁻¹ and a low tactile detection limit of 24.5 kPa, but also provided the conductive material with favorable embedment and fitness to avoid exfoliation under repeated loading-unloading cycles. The sensor demonstrated remarkable stability and reliability to maintain a barely changed electrical response during 1400 testing cycles.

In order to improve the stability of the network conductive sensing layer, specific microstructures could be utilized to realize the directional cracking and to prepare the conductive network with favorable repeatability for pressure sensors. Shi et al. patterned orderly triangular holes on photoresist and then induced stress concentration and crack expansion at the tips of triangles by changing the temperature of photoresist film. Nano-channel crack network composed of regular hexagonal arrays was generated. By subsequently electroplating Ni and electrochemical depositing graphene oxide onto the crack network, a hexagonal arrayed network conductive layer with highly regular and repeatable Ni/multilayer graphene oxide (MLGO)/Ni sandwich structure was produced and integrated with the protective PDMS coating to develop a pressure sensor. The bottom Ni film of the composite conductive network only underwent strain effect and showed slight resistance variation under pressure, while the sliding and misalignment of adjacent MLGO flakes as well as the tunnel effect in the middle layer resulted in significant resistance increasing. In addition, the rough MLGO induced the formation of numerous convex points on the pleated top Ni layer. Cracks occurred around convex points while tensile loading, which conduced to prominent changes of electron transporting accesses. Therefore, the combination of a well-aligned conductive network synergistically accomplished a large sensing range, high sensitivity, and favorable reproducibility of the network structure. The as-prepared sensor retained as high as 4953.15 kPa⁻¹ sensitivity at 52.27 kPa, and could detect laughter to different extents when fixed to the throat, illustrating potentials in e-skin applications.
3.2 Crack-based pressure sensor with sandwich structure

The most common construction of flexible pressure sensors is a sandwich structure consisting of the upper and lower electrode layers and the middle functional layer. Cracks can be introduced to the initial structure to synergistically obtain flexible pressure sensors with superior comprehensive performance regardless of resistance-based or capacitance-based mechanisms. The piezoresistive pressure sensors rely on the compress-induced resistance variation on the middle conductive layer to accomplish pressure sensing. A highly elastic conductive network based on 3D sponge microporous materials endowed pressure sensors with the pressure detection potential under a large sensing range. However, there was no contact between the skeletons of the 3D conductive network under small pressure, which led to a faint change of conductive accesses and a bare response to low-pressure signals (<10 kPa). The introduction of cracks in flexible pressure sensors could effectively enhance their response to tiny variations. Specifically, subtle bending of the conductive skeletons provided more conductive path and promoted the swiftly increasing current, resulting in a sensitivity improvement from 0.096 to 0.122 kPa⁻¹. Benefiting from the assistance of channel cracks, the sponge sensing network realized a dual enhancement of both sensitivity and response range, satisfying the monitoring requirement of whole-range body motion.

According to the above principle, crack-assist resistance pressure sensors based on many materials, such as AgNWs, carbon black, MWCNT, graphene oxide, etc., were all developed. Zhang et al. reported a piezoresistive pressure sensor fabricated by dip coating the as-prepared cellulose nanofibril/AgNWs mixed solution onto PU foam, drying and finally treated with pre-compression. The high elastic modulus of cellulose contributed to the brittleness of the AgNWs conductive layer, which benefited the crack formation of the sensor. The easy-to-make process enabled large-scale production of this superior sensor with low detection limit, high sensitivity, and wide sensing range. Similarly, Tewari et al. dip-coated MWCNT/rGO ink onto PU foam and developed a MWCNT–rGO foam pressure sensor. The introduction of functionalized MWCNT into highly dispersed rGO ink assisted the electron penetration between rGO flakes and tremendously improved the conductivity of the coating, yielding low-cost, easy-to-make sensors with outstanding performance for real-time monitoring of small and large-scale human body motion. Carbon-based materials attached to sponge structures always suffered from fracture and exfoliation during repeatedly pressure loading, which brought on a gradual reduction in the stability and sensitivity of resistance pressure sensors. In comparison, the gelatinous and non-powdered hydrogel showed a stronger adhesion with polymers. For example, the poly(vinyl alcohol)/sulfuric gel electrolyte (PVA/H₂SO₄) gel electrolyte could be applied as conductive material and formed a PVA/H₂SO₄@PU pressure sensor through the dip-coating method. The crack effect under small pressure only occurred on PU skeleton carbonized by sulfuric acid when heated, while the hydrogel work under large strain. The combination of flexible hydrogel and the cracks on carbonized PU skeleton ensured a relatively large sensing range (16.2 kPa) and stability while promoting sensitivity.

Some double-layered conductive structures were also introduced to resistance pressure sensors to elevate the sensing enhancement effect of cracks. A double-layer conductive structure was prepared by separately coating a GO layer and polyaniline nanohair (PANIH) on the sponge. As shown in Figure (9A), under small pressure, the cracks between graphene oxide flakes opened and thus the conductive path was fractured, leading to a swiftly rising resistance. When applied large pressure, the foam skeletons began to contact and generate interlocked polyaniline nanohair arrays, which allowed more speedily increased conductive path and tremendously improved sensitivity. The static trembling at 4–6 Hz was one of the typical early symptoms of Parkinson’s disease, and the as-prepared sensors could detect this characteristic on fingers and predict early Parkinson’s disease. Numerous conditions should be satisfied for the introduction of cracks in microstructures, and directly preparing a noncontinuous conductive sensing layer as cracks was an effective way for performance enhancement. Kim et al. attached a layer of noncontinuous cracked paddy-shaped MoS₂ flakes on 3D graphene porous network to produce enhanced sensitive units. When the applied pressure exceeded a certain value, the crack gaps of adjacent MoS₂ flakes gradually decreased and provided an extra conductive path that was manifested as resistance reducing. The sensor reached the highest sensitivity of 6.06 kPa⁻¹ within 0.6–25.4 kPa, much larger than 1 kPa⁻¹. By carbonizing commercial melamine foams, plenty of unconnected cracked fibers were also produced in the obtained 3D ultrathin carbon fiber network. The shortest distance (d) between the central lines of adjacent crack fibers was a significant factor for the resistance of the carbon fiber network. When
FIGURE 9  Crack-based pressure sensor with sandwich structure. (A) Schematic of the sensing mechanism of polyaniline nanohair (PANIH)/microcracked reduced graphene oxide (rGO)@PU sponge pressure sensor under small and large pressures; on the right is the sensor’s diagnosis of early Parkinson’s disease. Reprinted with permission. 96 Copyright 2018, WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. (B) Tunneling resistance model of two adjacent cracked fibers. Reprinted with permission. 97 Copyright 2014, American Chemical Society. (C) Current–pressure response of ultrafine carbon fiber networks. Reprinted with permission. 98 Copyright 2018, Royal Society of Chemistry. (D) Schematic diagram of crack deformation and fluid infiltration into the crack for crack-enhanced microfluidic pressure sensors, on the right is the capacitance–pressure response of sensors with different liquid mediums. Reprinted with permission. 99 Copyright 2017, American Chemical Society.

$d$ was larger than the average diameter of cracked fibers but smaller than a certain tunneling cut-off distance, a tunneling junction was created between two adjacent but unconnected cracked fibers (Figure 9B). 97 Due to the tunnel effect, a slight change in $d$ induced by subtle pressure would cause largely varied tunneling current and resistance in the conductive network, resulting in an ultralow detection limit of 3 Pa. Based on the above mechanism, the pressure sensors based on cracked carbon fiber network accomplished the wide-range sensing of 3 Pa–10 kPa and the highest sensitivity of 100.29 kPa$^{-1}$ (Figure 9C).

The crack structure could also combine with other structures in the pressure sensors to synergistically enhance the comprehensive performance. For example, the incorporation of microcrack-induced high sensitivity and wrinkle-induced wide sensing range accomplished a dual elevation of sensor performance. By pre-loading compression strain on the foam and dip-coated it with conductive solution, microcracks and wrinkles were introduced on the foam skeleton after strain release. 104 According to tunneling theory and contact mechanics, the sensing model formula of this sensor could be further established. It was shown that the sensitivity was in negative correlation with the initial contact area between PU foam and the electrodes, and was in positive correlation with the initial resistance of PU foam. Under pre-strain, the wrinkles and cracks on the conductive layer caused the rising of initial PU resistance and the decrease of initial contact area, resulting in a significantly improved sensitivity of 158.1 kPa$^{-1}$ as well as a wide sensing range of 11 kPa. Also, the crack structure could be introduced into microchannel-based capacitance pressure sensors. 99 The applied pressure induced the increasing of interfacial contact area between liquid and nanowire electrodes, which caused the capacitance change and realized the sensing process. The opening microcracks on the surface of electrodes further increased the contact area and enhanced the sensitivity (Figure 9D). The integration of microcracks and pores enabled the preparation of pressure sensors based on a crack-across-pore composite structure. 105 When applying a small pressure (0–33 kPa), a concentrated stress distribution occurred around the pores and incited a swift variation of the contact area between cracks near the pores, achieving a sensitivity up to 19.77 kPa$^{-1}$. The cracks far from pores kept away from the stress concentration area and continued the changing of contact area and conductive path with continuously increased pressure, tremendously widening the sensing range. Zhu et al. 106 coupled the crack structure with dome arrays to
elevate the sensitivity and sensing range of sensors. Under small pressure (0–40 Pa), microcracks were generated on the top areas covered with dense AgNWs conductive network, inciting a rapidly increased resistance (6.258 kPa⁻¹). Under larger pressure (40 Pa–10.3 kPa), the sparser AgNWs network on the lateral and bottom areas maintained a gradually reduced rising rate of the new conductive path, providing relatively high sensitivity with a wide sensing range.

Microcone-based elastomer structure could increase the sensitivity, but showed large deformation under pressure and caused severe hysteresis. Yao et al. joined the annular crack structure on microcones to provide a solution for low hysteresis and high sensitivity performance. When micro-pyramids were pressed into a soft substrate, the stress initially concentrated on the tips transferred and formed a annularly stress distribution, producing a regular annular crack structure. With external pressures, the annular cracks were closed and the contact area between the electrode and pyramids increased. Their synergistic effect to the conductive path incited a considerable resistance decrease, illustrating an ultrahigh sensitivity over 10⁷ kPa⁻¹ in 0–20 kPa. And the introduction of crack patterns effectively diminished the viscoelastic effect and tremendously reduced the hysteresis (<3%). The novel nanovesicle conductive material with the coupling structure of hollowspheres and crack junctions was reported. With applied pressure, the contact area of pressed microspheres was increased. Under larger pressure, the stress transferred to the interior of microspheres and the hollow structure suffered from entire deformation, compacting the crack junctions. The sensors exhibited a promoted multiscale response ability, a detection limit of 5.5 Pa and a sensitivity of 11.3 kPa⁻¹.

4.2 | Alcohol sensors

The conductive polymer films showed a swelling effect when absorbing alcohol vapor, which could be employed to prepare alcohol concentration crack sensors. Cracks were created on poly(3,4-ethylene dioxythiophene) polystyrene sulfonate (PEDOT:PSS)/PDMS flexible film through external tensile loading, and completely penetrated the film through removing the residual PEDOT:PSS on crack areas via oxygen plasma etching. A condense of vapors was shown at crack areas when the obtained sensor film was exposed to alcohol vapor, which realized the ion-transport conduction and ultra-sensitive detection of alcohol concentration.

4.3 | Humidity sensors

The difference of water absorption extent between polymers and metal conductive materials promoted the crack variation, which could be employed in humidity sensors. By depositing Ni on PU foam, the channel cracks were shown through pre-compression. Under certain humidity, the PU swelled when absorbing water molecules and thus the induced strain on Ni film caused crack expansion. According to the tunneling effect, the resistance exhibited an exponential increase with the width of cracks, acquiring high-level humidity detection. Crack-based flexible humidity sensors also demonstrated the breath distinguishing ability by measuring the humidity variation of breathed gas, which was applicable for studying respiratory rhythm obstruction during sleeping periods.
4.4 | Temperature sensors

Temperature change would also induce the interface variation between polymer and conductive materials, resulting in a changed crack morphology and resistance value. The longer width and higher density of cracks promoted larger temperature sensitivity. By optimizing the crack morphology, the sensor finally performed a sensitivity of 0.042°C⁻¹ and illustrated a calculated skin-attached temperature (28.4°C) extremely close to the temperature measured by an infrared camera (28.5°C), manifesting the potentials for skin temperature measurement in exercising monitoring and medical care.109 Luo et al. thoroughly researched the influence of PDMS substrate swelling/contraction to resistance under different temperatures. Further regulated experiments were designed to eliminate the influence of temperature on conductive materials, achieving an actual sensitivity of 1.44%/°C⁻¹ approaching the measured sensitivity of 1.2%/°C⁻¹. The sensor could monitor body temperature fluctuation and diagnose abnormal temperature during fever with its reliable temperature measurement ability, illustrating a promise in healthcare applications.20c

4.5 | Magnetic field sensors

A type of magnetic-sensitive crack sensor was developed by the deposition of graphene nanoflakes onto a flexible magnetic substrate.31 The magnetic-sensitive substrate bend in the magnetic field, inciting the formation and expansion of cracks in the conductive layer. Plenty of electron transport accesses were destroyed, manifested as a significantly increased resistance and an ultrahigh sensitivity (with $4 \times 10^{10}$ change of relative resistance) in the medium magnetic field (0–43 mT).

5 | CONCLUSION AND OUTLOOK

The integration of the advances from various fields to the crack design has made significant progress in the development of flexible sensors. In this review, the applications of crack engineering in strain sensors, pressure sensors, temperature sensors, magnetic field sensors, hydrogen sensors, and other sensors are summarized. We summarize the sensing mechanisms of various types of cracks in detail: (1) the effects of crack depth and density on sensor performance, (2) the effects of cracks on hierarchical structure sensors and fiber-structured sensors, and (3) the effects of encapsulation layers on sensor performance. In general, engineered cracks can effectively improve the performance of a cracked-based sensor. For example, channel cracks can enhance the sensitivity of a cracked-based sensor, and network cracks can extend the sensing range.

Although numerous progress has been made in the application of crack engineering in flexible sensors, the following challenges still need to be addressed. Firstly, the sensor’s crack pattern needs to be precisely controlled. Channel cracks and network cracks, two main types of crack morphologies, have different effects on sensor performance. In sensing applications, the intermediate state between the two crack morphologies may be more important. The sensitivity and sensing range of a sensor can be precisely controlled by adjusting the intermediate pattern consisting of the two kinds of cracks. Furthermore, by adding conductive materials, the conductive paths can be precisely controlled, and the electrical performance of the sensor can be effectively managed. Secondly, achieving the practical application of flexible sensors requires a highly reliable crack disconnection-reconnection process. Generally, the cracks are recognized as flaws or damage and are avoided in practical application. However, in cracked-based sensor applications, the sensor sensitivity could be tuned through the disconnection and reconnection of cracks. Of cause, the disconnection and reconnection of cracks are not fully reversible, which should be considered when designing a sensor. Therefore, through interface engineering, achieving directed adhesion of fractures to boost sensor stability is a research direction. Finally, the crack generation mechanism of metals such as Pt and Au is relatively well studied, while the crack generation mechanism of low-dimensional materials such as CNT and graphene remains unknown. There is no unified crack generation theory of sensing materials, which are composites and have complex mechanical and electrical properties. Traditional low-dimensional materials are widely used for the fabrication of flexible sensors. Understanding the crack generation theory of classic low-dimensional materials could be a valuable guide to the development of high-sensitivity sensors.

The studies mentioned above all contribute to the development of flexible sensors. We believe that the crack-based flexible sensor will eventually find solutions to overcome these obstacles and outperform conventional rigid sensors in terms of sensitivity, flexibility, and durability, leading to broader applicability. Flexible crack-based sensors will greatly enhance robotic technology and play a vital role in medical health and sports training through personal sensing and real-time monitoring, promoting the quality of life for humans.

ACKNOWLEDGMENTS

This work was supported by the National Key R&D Program of China (2021YFB3200700), the National Natural
The authors declare no conflict of interest.
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How to cite this article: Y. Zhou, H. Lian, Z. Li, L. Yin, Q. Ji, K. Li, F. Qi, Y. Huang. VIEW. 2022, 3, 20220025. https://doi.org/10.1002/VIW.20220025