REVIEW ARTICLE



Fibre-based stretchable electrodes for flexible metamaterial electronics: A review

Yunzhao Bai^{1,2}, Chao Hou^{1,2}, Wenna Cheng^{1,2}, Zijian Xu^{1,2}, Kan Li⁽¹⁾, and YongAn Huang^{1,2}

¹State Key Laboratory of Intelligent Manufacturing Equipment and Technology, Huazhong University of Science and TechnologyWuhan, China.

²Flexible Electronics Research Center, Huazhong University of Science and Technology, Wuhan, China.

Corresponding authors: Kan Li, YongAn Huang; Email: kanli@hust.edu.cn, yahuang@hust.edu.cn

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Abstract

Flexible electronics researchers have been conducting studies to explore the response of flexible stretchable electrodes to strain. The regulation of strain response in current flexible stretchable electrodes relies primarily on altering the material system, interfacial adhesion, or electrode structure. However, modifying the material system or interfacial adhesion can negatively disrupt the stretchable electrode preparation process, making commercialization a significant challenge. Additionally, the material system may be inadequate in extreme environments such as high temperatures. Hence a systematic structural design approach is crucial for effective response modulation of stretchable electrodes. One potential solution is the design of fibre structures from the micro to macro scale. This article focuses on discussing how the response of stretchable electrodes can be modulated by fibres in different states. The discussion includes fibres on elastic films, fibres directly constituting fibrous membranes at the microscopic level, and fibres constituting metamaterials at the fine level. The modulation can be achieved by altering the orientation of the fibres, the geometrical structure of the fibres themselves, and the geometrical structure formed between the fibres. Additionally, the article analyses the current situation of stretchable electrodes in extreme environments such as high temperatures. It also reviews the development of ceramic fibre membranes that can be stretched in high-temperature environments. The authors further discuss how the stretchability of ceramic fibre membranes can be improved through the structuring of ceramic fibre membranes with metamaterials. Ultimately, the goal is to realize stretchable electrodes that can be used in extreme environments such as high temperatures.

Introduction

Flexible electronics has experienced explosive growth in recent years due to multidisciplinary crossover. It has shown great promise in various fields such as epidermal electronics (Kim et al., 2011; Wang et al., 2020; Bai et al., 2023), robotic e-skin (Chortos et al., 2016), intelligent skin of aircraft (Xiong et al., 2021), micro-aircraft (Kim et al., 2021), flexible displays (Yokota et al., 2016), and biomedical devices (Noh et al., 2018; Zhao et al., 2020). However, unlike traditional silicon-based electronics, flexible electronics face challenges such as a complex material system, ever-changing processing technology, high cost, poor stability, and limited market demand (Zhou et al., 2023). Scientists from different disciplines are proposing mature, efficient, and inexpensive solutions to advance the commercialization of flexible electronics. To broaden the application track and apply to

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a wider range of scenarios and more extreme use environments, flexible electronics need to develop new technological routes and efficient design methods suitable for commercialization.

Stretchable electrodes have given a significant boost to the capabilities of flexible electronics. A crucial aspect of such electrodes is their response regulation that determines their stretchability and performance as sensors or actuators. The primary objective of regulating the response of stretchable electrodes is to adjust their sensitivity level when subjected to strain. This can be achieved by making them sensitive to strain and controlling the degree and range of sensitivity, or by making them insensitive to strain and controlling the range of insensitivity. The selection of a stretchable electrode's response should be based on the intended function of the electrode and the required stretchable electrode during its use as a flexible wire, electromechanical sensing, or actuating unit.

In recent decades, there has been a significant progress in developing stretchable electrodes that can be regulated in response, and this is achieved through electrode design. This includes 'resistancelocked' lead electrodes (Cho et al., 2021), high-sensitivity strain-sensing electrodes (Kang et al., 2014; Araromi et al., 2020), and large-range strain-sensitive electrodes (Ma et al., 2019). The electrical response of stretchable electrodes depends on how well the electroactive material is dispersed during the stretching process (Jiang et al., 2018). The mainstream methods to regulate the dispersion rate of an electroactive material mainly involve changing the material system, interfacial adhesion, or the mechanical structure. However, it's difficult to regulate the response of stretchable electrodes consistently through material system tuning and interfacial adhesion control, which hinders the commercialization of flexible electronics. Furthermore, in extreme environments such as high temperatures, it's challenging to stabilize conventional flexible electronic materials for a long period, and the methods of changing the material system and treating the material interface will become limited. Changing the mechanical structure to modulate the response of stretchable electrodes overcomes these drawbacks and can effectively adjust the electrical response of stretchable electrodes from the perspective of structural engineering. Fibre is an ideal structure for regulating the electrical properties of stretchable electrodes. It's an approximately 'one-dimensional' simple structure with a large 'aspect ratio' that can withstand strain by bending without structural destruction. Additionally, it has a wealth of three-dimensional geometries that can be processed. In this way, it can effectively modulate the electrical response of stretchable electrodes. To better understand this rapidly growing field of response modulation of flexible stretchable electrodes through fibre structure engineering, recent results and modulation mechanisms are summarized. Then, future application scenarios are explored from this perspective.

Stretchable electrodes based on fibre structure

The method of engineering fibre structures enables easy attainment of different electrode responses by designing geometrical parameters of the structure. The continuously changing geometrical parameters facilitate systematic and continuous modulation of the electrode response. The forms of fibres can be classified into the following categories: (a) fibres distributed on elastic films, (b) fibres directly comprising the fibrous membrane, (c) mechanical metamaterial fibre networks, and (d) others.

Fibres distributed on elastic films

Polyimide (PI) fibre devices integrated on 3D elastic films by Rogers's group have successfully achieved continuous conductance under large stretching conditions as shown in Figure 1a (Xu et al., 2014). Furthermore, Rogers's group has investigated the design of fractal fibre structures (stretchability >30% strain) of fibres on elastic films and their conformal attachment to the surface of human skin for precise measurement of electrophysiological signals (Figure 1b) (Fan et al., 2014). To overcome the behaviour of metal films that fail by uncontrolled cracking under small strains, Wang's group, inspired by cochlear stereocilia bundles, assembled nanowires in layers at the interface of an elastic film to retard penetration cracking and to significantly improve the stretchability of metal film-based



Figure 1. Fibres distributed on elastic films. (a) Images of a representative 3D multifunctional integumentary membrane (3D-MIM) integrated on a Langendorff-perfused rabbit heart. The white arrows highlight various functional elements in this system. The electronics can cover both anterior and posterior surfaces of the heart (inset). Scale bars, 6 mm. Magnified views of the functional elements in conformal contact with the epicardium. The images are recorded from the back side of the devices. Scale bars, 500 μ m (Xu et al., 2014). (b) Six different patterns of metal wires fully bonded to elastomer substrates demonstrating the application of deterministic fractal designs as general layouts for stretchable electronics. These patterns include line, loop, and branch-like geometries. Image of metal wires with Peano layouts, with an overall geometry that spells out the characters in 'ILLINOIS', mounted on skin. Here each letter consists of a series of first- and second-order Peano curves. Optical and scanning electron microscopy images of third-order Peano-based wires on skin and a skin-replica (colorized metal wires) (Fan et al., 2014). (c) SEM images of cracking induced by micro-voids (MVs) (yellow arrow) and nano-voids (NVs) (red arrow) of Pt films (scale bar, 2 μ m). C_{MV} (yellow arrow) and C_{NV} (red arrow) represent cracks initiated from MVs and NVs. Key features of nanowire assembly

sensors (stretchability 130% strain, gauge factor 107.45, minimum detection 0.005% strain) as shown in Figure 1c (Miao et al., 2019). Meanwhile, Cheng's group has reported a method to continuously modulate electrode response (gauge factors from 10 to 1360 in the typical strain range of 9–79%) by 'growing' gold nanowires on an elastic film as shown in Figure 1d (Gong et al., 2019). Huang's group has reported a method to achieve serpentine meanders on elastic PDMS films by near-field electrostatic spinning and the processing of self-similar serpentine meanders on thin films by prestretching with stretchability >300% (Figure 1e) (Huang et al., 2017). These fibre structures can transform deformations on a thin film substrate into angular shifts or bending of the fibres, thus avoiding large deformations and failures due to axial strain in the fibres.

In the future, designing fibre structures on top of thin films will be an important research direction. Huang's group has also reported the Buckling-driven self-assembly of self-similar micro/nanofibres for ultra-stretchable nanofibres by controlling the fibre's aspect ratio (Figure 1f) (Bian et al., 2017). Rogers's group has reported the modulation of local deformation of elastomers by different thicknesses (Figure 1g) (Nan et al., 2017). Meanwhile, Zhang's group has reported the arrangement of low Young's modulus elastomers underneath rigid islands on elastic thin films to isolate the strain transfer (Figure 1h) (Li et al., 2022).

The aforementioned work shows that the elastic fibres distributed on thin films are not only subjected to radial strain but also affected by the elastic substrate in the vertical direction of the fibres. Therefore, in addition to studying the angle of the fibres, the strain transfer in the vertical direction of the fibre substrate will be the focus of future research. Besides the near-field electrostatic spinning process mentioned in the text, photolithographic processes (Singh et al., 2012), reactive ion etching (Matsui et al., 2014), lasers (Lei et al., 2020), or inverted films (Long and Wang, 2021) can also be used to achieve the preparation of fibres on thin films. These well-established methods provide strong support for the processing of fibre structures on thin films and offer a predictable future for commercialization.

Fibres directly comprising the fibrous membrane.

Fibre mats that consist of micro/nanofibres can be obtained with ease through electrostatic spinning. Shen's group collected aligned wavy stretchable fibre mats by a high-speed rotating drum, realized the strain-sensing function by decorating them with carbon nanotubes, and compared the mechanical and electrical properties of the sensors along the fibre direction and along the perpendicular direction of the fibres (gauge factor is about 5.25–20 when the strain is 0–900%) as shown in Figure 2a (Ren et al., 2019). Zhao's group reports a fibre mesh processed by near-field electrostatic spinning to achieve anisotropic modulation of the electrical properties of stretchable electrodes with gauge factor 338.47 under 200% strain (Figure 2b) (Huang et al., 2020). Huang's group has employed a mechanical strategy to modify the orientation of fibres in the surface layer of disordered fibre membranes (Figure 2c)

Figure 1. (Continued)

in FE modelling. Evolution of hierarchical assembly of 4-µm-long nanowires as a function of strain (Miao et al., 2019). (d) Fabrication of V-AuNWs embedded in PDMS substrate and cross-sectional SEM of V-AuNWs/PDMS composite (Gong et al., 2019). (e) Fabrication of self-similar nano/microfibres (Huang et al., 2017). (f) Simulation results of in-plane buckling mode and out-of-plane buckling mode of the fibre on the elastic film after releasing the pre-stretched fibre are compared with the real picture. The semi-elliptical cross-section's critical strain for both in-plane and out-of-plane buckling (Bian et al., 2017). (g) The strain field distribution of the elastic film is not uniform due to its uneven thickness, and the finite element analysis of 3D structure assembly is realized by this method (Nan et al., 2017). (h) Schematic diagram when the rigid island is fixed to the pre-stretched elastic substrate. The finite element analysis of the strain field distribution in the X direction before and after the release of the pre-stretched buffer layer (Li et al., 2022).



Figure 2. Fibres directly comprising the fibrous membrane. (a) Schematic of the preparation equipment of the pure aligned TPU fibrous mat. Illustration of the pure aligned TPU fibrous network and CNTs/aligned TPU fibrous network. The mechanical and electrical responses of the sensor were compared along the fibre arrangement direction and vertically along the fibre arrangement direction (Ren et al., 2019). (b) Schematic diagram of the conductive PU/AgNW network prepared by near-field spinning method, physical diagram of strain sensor, display of different tensile states, and the simulated cloud image under 100% strain state (Huang et al., 2020). (c) Comparison between the mechanical strategy to change the orientation of surface fibres and the traditional process flow. The statistical methods (Bai et al., 2023). (d) Graphene-based composite fibre with SEM diagram of 'compression spring' architecture and its theoretical modelling as strain sensor (Cheng et al., 2015). (e) Optical photographs, electrical response curves, strain cloud images, and SEM images of different positions of a beaded fibre strain sensor (Liu et al., 2018).

(Bai et al., 2023). This strategy enables precise regulation of performance as strain sensors. The underlying mechanism is that fibres that are aligned with the tensile direction experience greater tensile strain and are prone to damage on the electrodes during processing. Conversely, fibres that are oriented perpendicular to the tensile direction undergo less tensile strain and may even face compressive strain that does not result in electrode damage. By controlling the direction and magnitude of the applied strain, the article achieves systematic modulation of the properties of stretchable electrodes. The gauge factor decreased from 18.45 to 3.33 when modulating the strain sensor measurement range from 0.25 to 0.9. However, this approach is only effective for elastic materials. For plastic or brittle materials, stretching leads to permanent changes or destruction of fibre morphology and structure, making it impossible to rebound for fibre orientation modulation.

In addition to mechanical strategies, methods such as substrate movement speed collection (Persano et al., 2013; Ren et al., 2019), far-field electrostatic spinning method alteration (e.g., pneumatic electrostatic spinning) (Varesano et al., 2007), electric field control (Robinson et al., 2021), or self-assembly (Shi et al., 2010) provide a basis for regulating fibre orientation on fibrous membranes. Nonetheless, all these methods change the fibre orientation of the fibrous membrane as a whole, making it challenging to achieve local regulation of surface fibre orientation. Furthermore, the macroscopic mechanical properties of highly uniformly aligned fibre alignment and reduces it in the direction that is perpendicular to the fibre alignment. This is not favourable for the application of fibrous membranes and the preparation of complex conductive sensing networks. Therefore, developing methods that can directly perform local fibre orientation modulation is essential for the future to achieve the modulation of local electrical properties of stretchable electrodes.

In exception of the fibre orientation, the structure of the fibre itself can also influence the response of the electrode. Gao's group proposed a fibre that achieves large stretching capacity and high sensitivity using a secondary helical structure on the outer surface of the fibre (gauge factor decreases from 10 to 3.7 when strain is increased from 1% to 50%) as shown in Figure 2d (Cheng et al., 2015). Chen's group realized local strain regulation by adding bead structures to the fibres to achieve the purpose of improving the sensitivity of strain sensing, and the strain sensors so prepared have a gauge factor of up to 100 (Figure 2e) (Liu et al., 2018). It appears that the response of the electrode can also be effectively regulated by changing the structure of the fibre itself is quite complicated and it is difficult to achieve accurate and continuous regulation. In the future, researchers are expected to develop a simple and effective process to achieve continuous and accurate regulation of the fibre structure in order to further precisely regulate the response of the stretchable electrode.

Mechanical metamaterial fibre networks

A mechanical metamaterial fibre network is a periodic network structure consisting of finely sized fibre segments. In contrast to fibres on an elastic film, the mechanical metamaterial fibre network is mainly free-standing. Modulating the stretchability and Poisson's ratio of metamaterials has long been a focus of scientific research.

Zhang's group investigated the nonlinear stress-strain curves and elastic stretchability of a class of fractal horseshoe microstructures and improved the elastic stretchability of traditional horseshoe designs by more than four times (up to 1400% stretchability) through fractal structure design. Different orders of fractal structures correspond to different tensile capacities, and the team finally demonstrated a mechanistic metamaterial fibre network using fractal horseshoe structures, pointing out advantages such as its high elastic stretchability as an electrophysiological electrode (Figure 3a) (Ma and Zhang, 2016). Therefore, the introduction of fractal structure design into the structural design of mechanistic metamaterial fibre networks can effectively regulate their tensile capabilities. Wang's group created lattice metamaterials with tensile capabilities of more than 50% and negative Poisson's ratios by using sinusoidally shaped beams instead of regular straight beams. In addition, four types of sinusoidally



Figure 3. Mechanical metamaterial fibre networks. (a) Geometric construction of the fractal horseshoe microstructures for fractal order from 1 to 3. Theoretical predictions and optical images of deformed configurations for two second-order fractal horseshoe microstructures, with arc angles $\theta = 240^{\circ}$, under different levels of applied strain. The normalized width and unit cell number are fixed as m = 5. The scale is 5 mm. Normalized stress–strain curves (with the stress in logarithmic scale) for fractal order from 1 to 3. Schematic illustration of a representative EP electrode constructed with the generalized second-order fractal horseshoe microstructures (Ma and Zhang, 2016). (b) Schematics and deformation behaviour of the sinusoidally architected lattice material. Effect of the topology on the stress–strain curves and Poisson's ratio (Chen et al., 2017). (c) Schematic of typical re-entrant negative Poisson's ratio structures: mono chirality and constrained rotational symmetry, non-chirality and constrained rotational symmetry, non-chirality and constrained rotational symmetry, and non-chirality block and relaxed rotational symmetry (Hou et al., 2015). (d) Geometrical illustration of different zero Poisson's ratio structures: zero Poisson's ratio in one direction and two directions (Naghavi Zadeh et al., 2020).

architected lattice metamaterials with hexagonal, Kagome, square, and triangular topology were investigated for their stress and Poisson's ratio variations with strain. The results show that the tensile ability and Poisson's ratio can be effectively regulated by adjusting the structure of lattice metamaterials as shown in Figure 3b (Chen et al., 2017). Xiaonan Hou et al. summarized different negative Poisson's ratio metamaterials, including typical re-entrant negative Poisson's ratio structures: an arrowhead, star shape and missing rib structure and typical chiral Negative Poisson's Ratio structures: mono chirality and constrained rotational symmetry, non-chirality Negative Poisson's Ratio structures: mono chirality and constrained rotational symmetry, non-chirality and constrained rotational symmetry and non-chirality block and relaxed rotational symmetry (Figure 3c) (Hou et al., 2015). Iman Dayyani et al. summarized different zero Poisson's ratio metamaterials containing zero Poisson's ratio in one direction. hybrid honeycomb, semi re-entrant design and Silicomb, and zero Poisson's ratio in two direction: a star shape design and reverse semi re-entrant design (Figure 3d) (Naghavi Zadeh et al., 2020). Although a large number of mechanistic metamaterial fibre network structures have been developed, there is a need for a more systematic and continuous design approach to quantify the tensile capacity and Poisson's ratio properties of customized mechanistic metamaterials.

Recent reports have shown a high demand for mechanical metamaterial fibre networks for flexible devices. There has been a significant amount of research work on negative Poisson's ratio metamaterial fibre networks for applications such as artificial stents (>85% tensile capacity) and cardiac electrophysiology patches (>12.6% tensile capacity) as shown in Figure 4a (Jiang et al., 2022). These structures can effectively enhance the tensile strength of the material and have a smaller equivalent Young's modulus. This is achieved by translating in-plane tensile or compressive deformations into rotations of the in-plane fibre structure and local out-of-plane bending. As a result, mechanical metamaterial fibre networks can achieve large tensile and conformal attachment capabilities. These networks have been widely used in fields such as implant electronics and wearable electronics. Park et al. (2016) designed an electrically conductive mechanistic metamaterial fibre network around the heart, capable of efficiently delivering electrical impulse signals to the heart for the treatment of cardiac arrhythmias (Figure 4b). Li's group designed a dual-phase metamaterial (chiral-horseshoes), using which wearable strain sensors (gauge factor 2, tensile capacity 35%) and wearable stretchable displays were prepared (Figure 4c) (Deng et al., 2023). The use of mechanistic metamaterial fibre networks as stretchable electrodes has become a trend in the development of stretchable electronics. There are a large number of research works to achieve the application of stretchable electrodes (which are strain insensitive) by using the strong stretching ability of their metamaterial fibre network, and a small number of research works to achieve the measurement of large strains by using the axial strain on the fibre segments of the metamaterial fibre network during stretching. However, achieving higher sensitivity and more accurate measurements requires the consideration of strain field distribution on the mechanical metamaterial fibre network. Lu's group systematically investigated the effect of the geometry of serpentine-shaped ribbons on the strain field distribution and gave design guidelines for the tensile and flexible properties of serpentine-shaped ribbons (Figure 4d) (Widlund et al., 2014). Su's group explored the non-uniform distribution of stresses in the tensile process of the bent structure and realized the measurement of large strains (0–50% strain) as shown in Figure 4e (Li et al., 2022). Some researchers have examined the distribution of the local strain field in metamaterials, but only with the aim of designing the tensile capacity of metamaterial fibre networks. However, there is still much work to be done in the future to incorporate the strain field distribution of mechanistic metamaterials in the modulation of electromechanical responses.

Others

Chen's group increased the sensitivity of a conventional sensor by a factor of 24 (gauge factor ≈ 835) by incorporating a negative Poisson's ratio auxetic metamaterial into a stretchable strain sensor (Figure 5a) (Jiang et al., 2018). In the future, the use of a network of mechanical metamaterial fibres acting as an auxiliary support medium to modulate the response of the electrode is a very promising development.



Figure 4. Application of mechanical metamaterial fibre networks. (a) Schematic of several mechanical metamaterial structures, stretchable metamaterial devices, and stretchability display (Jiang et al., 2022). (b) Elasticity of the epicardial mesh and photograph of the epicardial mesh implanted in a control heart (Park et al., 2016). (c) The designed dual-phase metamaterial. Flexible display attached to the arms. Optical image showing the laughing faces on display before stretching the structures and after stretching the structures (Deng et al., 2023). (d) Schematics of a serpentine unit cell. Schematics of systematically varied serpentine shapes. Contour plots of strain fields obtained from CB theory and FEM (Widlund et al., 2014). (e) Schematic illustrations and images of a contact resistance-free stretchable strain sensor. Schematic illustration of the sensing mechanism (Li et al., 2022).

Ma's group proposed the concept of using a honeycomb basis as the support medium and a serpentine structure as the device (Figure 5b) (Chen et al., 2018). This suggests that composite mechanical metamaterial structure networks also have some potential for development in the field of performance modulation of stretchable electrodes. Three-dimensional fibre structures can provide superior tensile capabilities such as mechanically guided self-assembled three-dimensional structures (Figure 5c and d) (Xu et al., 2015; Jang et al., 2017). However, such three-dimensional structures require more three-dimensional space for placement and are quite difficult to encapsulate. There is an urgent need to compress the spatial height of 3D fibre structures and to seek optimized packaging solutions. To make flexible stretchable electronics more comfortable to wear, Xu's group has designed the fibre membrane with a metamaterial structure (Figure 5e) (Liu et al., 2022). Fibrous membranes have a higher tensile



Figure 5. (a) Stretchable strain sensors based on auxetic mechanical metamaterials (Jiang et al., 2018). (b) Three representative shapes of the serpentine interconnects, with different lengths of the straight part, bonded to the cellular substrate along (top) X and (bottom) Y directions, respectively. The elastic stretchability of the serpentine interconnect (Chen et al., 2018). (c) Average curvature components and mode ratio of a 3D mesostructure (3D wavy ribbon) that involves only bending, as a function of prestrain in the stretched assembly platform. Similar results for a 3D mesostructure (3D single-helical coil) that involves both bending and twisting (Xu et al., 2015). (d) Optical image of the system at a bi-axially stretched state of 50%, showing ~250 3D helices, ~500 bonding sites, ~50 component chips, and elastomers for full encapsulation. Inset: optical image of the device under a complex state of deformation. Scale bar, 5 mm (Jang et al., 2017). (e) Schematic and physical drawings of Kirigami metamaterial structure based on fibre membrane with strong tensile capability. Multifunctional electronic devices, including LED, bioelectrodes, and temperature sensors, are integrated on the metamaterial fibre membrane (Liu et al., 2022).

capacity than the intrinsic material film properties. On top of that, the design of the metamaterial structure further enhances its tensile ability and also reduces the equivalent Young's modulus and provides a larger hollowing area, which demonstrates a strong advantage in wearable adaptability and comfort. Fibrous membranes are micro/nanoscale fibrous interwoven structures at the microscopic

level, and the deformation of the microstructure during stretching leads to the enhancement of the macroscopic tensile ability. Mechanical metamaterials structuring is the design of stretchable structures at the fine level, and the design of such cross-scale fibrous structures from the microscopic to the fine level greatly enhances manoeuvrability in the regulation of tensile ability. In future research, the idea of cross-scale fibre structure design provides endless imaginative space for the modulation of stretchable capacity and electrode response of materials.

Stretchable electrodes for high-temperature environments

Fibre structure engineering to modulate electrode response has been used in a large number of different application scenarios, but its development is still on the rise. On the one hand, there is still a lack of detailed, systematic, and theoretical research to form a perfect fibre structure engineering electronics system. On the other hand, for high temperatures and other extreme working conditions, the traditional flexible electronic materials are difficult to apply, and so there is need for high-temperature-resistant materials (including insulating substrate materials and conductive materials) through the structural design to regulate stretchabilities. A typical application scenario in such extreme working conditions is a hypersonic vehicle surface, which is used to detect the aerodynamic parameters (including strain, pressure, temperature, heat flow, friction, pulsation, separation point, and transition point) of the vehicle smart skin is usually realized on PI substrate, which possesses the strongest temperature-resistant performance among organic materials, with the highest temperature-resistant temperature not exceeding 400°C. However, hypersonic vehicle surfaces typically reach temperatures of 500°C or more, and current flexible material systems can no longer be applied.

Conventional electrodes for high-temperature environments

Zhang's group used magnetron sputtering and microelectromechanical systems (MEMS) to prepare a flexible temperature sensor on flexible Hastelloy tapes that can operate at 550°C (Figure 6a) (Shao et al., 2021). Guo's group prepared strain sensors by depositing 80 nm Ba(Ti, Nb)O₃ film on ultrathin flexible mica substrates, and the prepared sensors exhibited excellent thermal stability ranging from 20 K to 773.15 K (Figure 6b) (Yang et al., 2019). Ren's group demonstrated the use of conductive pastes and ultrathin flexible yttrium oxide-stabilized zirconia (YSZ) ceramic substrates to prepare hybrid radio-frequency (RF) antenna devices for wireless communications and high-temperature sensing (Figure 6c) (Li et al., 2020). Liu's group realized strain sensors on mica substrates that can withstand up to 700°C (Figure 6d) (Guo et al., 2020). In terms of other applications, high-temperature-resistant flexible electronics are urgently needed in aero-engines, power generators, nuclear reactors, space equipment, and wearable devices for firefighters, among others. In the future, high-temperature-resistant metals, ceramics, mica, and other materials are expected to be used in high-temperature and other extreme operating environments through fibre structure engineering to regulate their tensile ability and the response of the corresponding electrodes, expanding the range of applications to stimulate the development and commercialization of flexible electronics.

Nanostructured ceramic fibre membranes for high-temperature environments

Nanostructured ceramic fibre membranes, as an emerging high-temperature-resistant material capable of withstanding extreme working conditions, are interwoven with nanoscale fibres with high aspect ratios at the microscopic level. The high aspect ratio of the ceramic nanofibres can bend, which leads to the macroscopic nano-ceramic fibre membrane has a certain deformation ability, so the nanostructured ceramic fibre membrane in the field of high-temperature-resistant flexible electronic devices has an inherent advantage. A variety of ceramic materials including silica, alumina, and zirconia have been used to fabricate ceramic fibre membranes, and the preparation processes mainly include far-field electrostatic spinning, solution blow spinning, and freeze-drying. Ceramic fibre membranes have been



Figure 6. Flexible sensor for high temperature resistance. (a) Image of Pt thin-film RTD on Hastelloy tape and the resistance variation of the sensor with ramp rate of 5° C/min (Shao et al., 2021). (b) Optical image of the flexible BNTO/mica heterostructure in our experiment and resistance as a function of temperature (20–350 K) for BNTO films measured at various in situ bending states (Yang et al., 2019). (c) Schematic representation of the development process of a copper ink for printable electronics, temperature dependence of the relative permittivity for printed Cu-YSZ high-temperature electronics, and S11 for the effect of thickness on the resonant frequency for printed Cu-YSZ antenna devices (Li et al., 2020). (d) Schematic diagram of preparation process of La0.7Sr0.3MnO₃/mica heterostructure and resistance changes ($\Delta R/R0$ (%)) upon different bending radius (Guo et al., 2020).

reported to be used as a substrate to achieve high-temperature-resistant temperature sensors (Figure 7a) (Liu et al., 2023), or as a dielectric layer (Figure 7b) (Fu et al., 2020), or as a piezoresistive layer (Figure 7c and d) (Gao et al., 2021; Wang et al., 2023) to achieve a high temperature-resistant pressure sensor, but none of them has good tensile capabilities.

In order to make the high-temperature-resistant fibre membranes more deformable, researchers have been improving the microstructural level of the fibre membranes. Wang's group combined sacrificial templates and chemical vapour deposition (CVD) to prepare high-performance carbon tube aerogels with a highly cross-linked structure capable of withstanding up to 99% of compressive strain and withstanding up to 2500°C of compressive strain in an argon environment as shown in Figure 8a (Zhuang et al., 2023). This material cannot be used directly in atmospheric environments; however, ceramic-based fibre membranes appear to be effective in overcoming these drawbacks. The fluffy ceramic fibre structure achieves up to 40% plastic deformability (Figure 8b) (Peng et al., 2021). Curled ceramic fibre structures achieve up to 20% elastic deformation (Figure 8c) (Su et al., 2021). 3D interwoven curled nanofibre structures have up to 100% tensile deformation (Figure 8d) (Cheng et al., 2022).



Figure 7. Flexible sensor based on high-temperature-resistant fibre membrane. (a) Optical and SEM images of the temperature sensor under different thermal environments (Liu et al., 2023). (b) A photograph and a SEM image of an ultrathin TiO₂ nanofibrous network. Photograph of the sensor burned in the butane flame (temperature $\approx 1300^{\circ}$ C). Capacitance to pressure sensitivities of the sensor tested at 30°C, 370°C, and 30°C after burning in the butane flame (Fu et al., 2020). (c) Resistive pressure sensor based on SC-Ag/SiO₂ fibre membrane and relative resistance change versus pressure (Wang et al., 2023). (d) Flexible display and mechanical characteristic curve of high-temperature-resistant ceramic fibre membrane. Optical photographs and sensor electrical response curves of resistive pressure sensors with certain bending capability (Gao et al., 2021).

Achieving a ceramic fibre membrane is possible by controlling the microscopic fibre structure and tensile ability modulation. However, the production process is complex and performance stability is poor due to various factors. Purely modulating the microscopic fibre structure is challenging to meet the commercialization of high-temperature-resistant flexible electronics. To achieve high-temperature-resistant flexible and stretchable conductive or sensing ability, it is necessary to continue developing the ceramic fibre structure regulation, combined with the fine level of the serpentine metamaterial fibre structure. This combination is expected to achieve stronger stretching capability, resulting in high-temperature-resistant stretchable electrodes. This development is crucial to achieve high-temperature-resistant large strain sensors (>20% strain).



Figure 8. Fibre membrane for high temperature resistance. (a) Stress versus strain curves of a CTA (12.9 mg cm⁻³) compressed to 10%–99% strain. Inset: Enlarging the onset of the compression curves, demonstrating no permanent deformation. Experimental snapshots of a CTA ($7 \times 7 \times 8$ mm) under uniaxial compression to 99% strain. In situ TEM observations of carbon tube networks during uniaxial loading and unloading (Zhuang et al., 2023). (b) SEM image of nanofibrous membrane after heat treatment at 1300°C and longitudinal stress–strain curves (Peng et al., 2021). (c) Microstructure of ceramic fibre film with elastic stretchability up to 20% and reversible stretching test at 1200°C (Su et al., 2021). (d) Illustration of 3D reaction electrospinning method for preparing ceramic nanofibrous aerogels; physical drawings and tensile stress–strain curve of a prepared ceramic nanofibrous aerogel (Cheng et al., 2022).

The process of structuring the mechanics of metamaterials through the use of ceramic fibre membranes presents challenges. The first challenge arises from the fluffy fibre structure of the membrane, which does not guarantee elastic strain capacity. To achieve the desired elastic deformation capacity, cross-linking of the ceramic fibres is necessary to limit sliding between them. The second challenge is encountered during the cutting of the ceramic fibre membrane into metamaterials, where smooth cutting edges are crucial to avoid penetrating fractures while stretching. This is due to the low

local strength at the cut edges of the metamaterial, which can cause fractures. Lastly, the low breaking stress of ceramic fibre membranes can result in poor load-bearing capacity, which can be addressed by improving the material properties or using composite structures. There is an urgent need for high-temperature-resistant stretchable electrodes to enable applications in extreme environments, and as such, new technical routes and comprehensive research are necessary.

Conclusion

Fibre structure engineering is expected to become a more systematic, theoretical, and universal method for regulating the response of stretchable electrodes in the future. This method will not only solve the problem of matching the electrode performance faced by flexible stretchable electronics in conventional or high-temperature environments but also eliminate the difficulty of unifying the preparation process of flexible stretchable electrodes. It will reduce the risk of the material system not being able to adapt to extreme environments, as well as promote commercialization. The applications of customizable flexible stretchable electrodes are vast, including epidermal electronics, intelligent skins for aircraft, wearable devices for firefighters, electronic skins for robots, micro-vehicles, and space equipment. These diverse applications are exceedingly valuable. Therefore, the method of precisely modulating the response of stretchable electrodes through fibre structure engineering is of profound significance.

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