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A Stretchable Self-Healing Temperature Sensor Based on Pu-Ta@Mxene Fiber for Body Surface Temperature Monitoring

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Abstract: Sensitivity, repeatability, and mechanical properties have been the focus of research on flexible temperature sensors. However, existing temperature sensors suffer from low sensitivity and untimely array sensing. In this paper, a core-sheath structured fiber based on modified tannic acid (meta), carbon nanotubes (CNT), Mene, and polyurethane (PU) is investigated, which is made of a sheath consisting of a layer of PU wrapped around a core consisting of TA, CNT, and Mene, and exhibits excellent mechanical properties and self-healing ability. Through experimental testing, the tensile strength of the fiber reaches 6.35 MPa and the elongation at break is 66.43%, showing good elasticity and flexibility. Utilizing the characteristics of the core-sheath structure, the fibers were able to achieve self-healing at 80°C when cut by the knife. The sensitivity of the fibers to temperature changes was different in different temperature zones. In the low temperature range (-5°C to 20°C), the TCR was high at 5.58 %°C-1, while in the higher temperature range (20°C to 60°C), the TCR was 2.08 %°C-1. The TCR of the fibers was also higher in the low temperature range (-5°C to 20°C). In addition, human breath cycle tests were conducted and showed excellent reliability. In addition, an array sensor circuit was developed and designed to accurately measure the spatial temperature distribution, thereby increasing the sensor's usefulness in a variety of applications.

Keywords: Wearable sensors, Mene-based sensors, Modified nanomaterials, Self-healing

1. Introduction

As technology continues to advance, flexible temperature sensors are increasingly in demand in a variety of fields. Among them, multifunctional sensors with self-healing properties, high sensitivity and good mechanical properties are especially in demand. These flexible temperature sensors have a wide range of applications in fields such as smart textiles [1,2], wearable electronics [3], human-computer interaction [4,5] and health care-monitoring [6].

In these applications, the mechanical properties, self-healing properties and sensitivity of flexible temperature sensors are key factors. Mechanical properties determine the durability and reliability of flexible temperature sensors in service; self-healing properties can extend the service life of flexible temperature sensors and reduce maintenance costs; and sensitivity determines the responsiveness of flexible temperature sensors to environmental changes. Therefore, the development of flexible

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temperature sensors with high sensitivity, self-healing properties and high mechanical properties is a hot spot in current research.

MXene is a two-dimensional material obtained by chemical stripping of transition metal carbides, nitrides, or carbon-nitrides with good electrical conductivity and mechanical properties, and has attracted wide attention because of its unique structure and excellent performance. MXene has good electrical conductivity and mechanical properties, and can be chemically compounded with other materials to prepare multifunctional materials with excellent performance [7]. For example, compounding MXene with PU can improve the tensile strength and elongation at break of the material; compounding MXene with CNT can further improve the conductivity and sensitivity of the material.

mTA is a natural polyphenol compound with good antioxidant properties and biocompatibility. By compounding tannic acid (TA) with MXene, the mechanical and self-healing properties of the material can be further improved. In addition, mTA can interact with PU and CNT through hydrogen bonding to form a stable composite structure. In recent years, researchers have developed materials with self-healing properties and high sensitivity through a variety of methods. By combining MXene with other materials, multifunctional materials with excellent properties can be prepared. Liu et al. proposed an interface engineering strategy to encapsulate MXene into a stable TA@MXene nanosequence by introducing a high-density hydroxyl TA, while increasing the hydrogen bond interaction between TA@MXene and the polymer network. The obtained organic hydrogels have comprehensive properties such as high tensile property, low hysteresis property, excellent fatigue resistance and good adhesion [8]. Yuan et al. fabricated a serpentine stretchable conductor (SSC) with high conductivity, stretchability, and ultra-stability by spontaneously wrapping elastic waterborne polyurethane (WPU) sheath on the surface of the multi-walled carbon nanotube (MWCNT)/ WPU nanocomposite conductive yarn, which was prepared by a simple wet-spinning method [9].

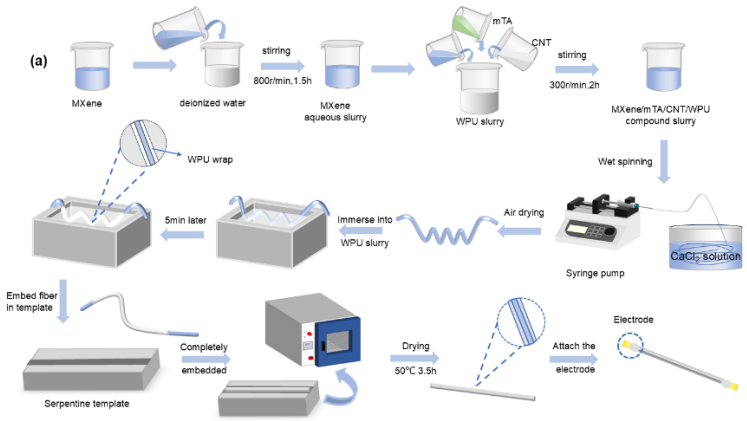

Figure 1: The synthetic strategy of the materials. (a) Preparation process of flexible temperature sensors. (b) Preparation process of mTA. (c) Structural distribution of fibers

In this work, we prepared a novel multifunctional fiber using the core-sheath structure of mTA-CNT-MXene-PU. This fiber not only has good tensile strength (6.35 MPa) and elongation at break (66.43%), but also has excellent elasticity and flexibility, as well as self-healing properties and high sensitivity. By optimizing the formulation and preparation process of the materials, we have successfully prepared fibers with excellent comprehensive performance. This fiber has a broad application prospect in the fields of smart textiles, wearable devices and health monitoring.

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2. Materials and methods

2.1. Preparation of raw materials

The preparation process of mTA is illustrated in Figure. 1b. The mTA was synthesized via electrostatic assembly. Briefly, TA and benzalkonium chloride (BAC) were mixed under alkaline conditions (pH=9) with continuous stirring. The negatively charged TA molecules (due to deprotonation at high pH) and positively charged BAC formed stable nanocomposites through electrostatic interactions, resulting in the final mTA product.

2.2. Preparation of flexible sensors

The preparation process of the flexible temperature sensor is shown in Figure.1a. MXene was added to deionized water and mixed and stirred at 800r/min for 1.5h. Modified tannic acid, CNT, WPU was added and stirred at 300r/min for 2h. Then calcium ions were injected into the coagulation bath through a syringe. The fibers were taken out and dried at room temperature. Then the fibers were impregnated in aqueous polyurethane solution for 5min, after which the polyurethane formed a protective layer on the fiber surface. The fibers were then placed inside the 3D printed template and placed in an oven at 50°C for 3.5h to dry and shape. Finally, Cu electrodes were added to both ends of the fibers.

As shown in Figure. 1c, the core-sheath fiber features a functional core composed of MXene, mTA, and CNT, encapsulated by a PU sheath. Within the core, MXene nanosheets are interconnected via hydrogen bonds formed between their oxygen-containing groups and the phenolic hydroxyl groups of mTA. Simultaneously, mTA bridges MXene and CNT by interacting with defect sites on CNT surfaces, effectively preventing aggregation of both components. The PU sheath provides mechanical robustness and flexibility, while ensuring interfacial adhesion to the core through hydrogen bonding. This hierarchical architecture synergistically enhances the fiber's electrical conductivity, tensile strength, and environmental stability, making it suitable for advanced wearable electronics.

3. Results and discussions

3.1. Strain sensing behaviors

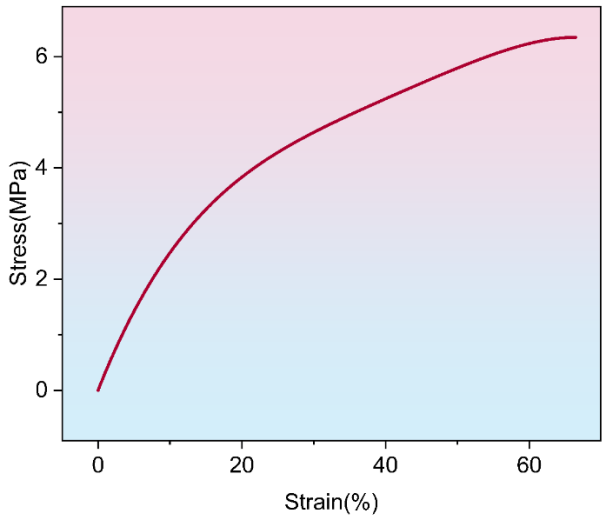

Figure 2: Stress-strain curve of fiber

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The stress-strain curve of the fibers is shown in Figure.2, which shows that the tensile strength of the fibers is 6.35 MPa and the elongation at break is 66.43%. This indicates that the fibers have impressive strength. PU, as the matrix material, imparts good elasticity and flexibility to the fibers. CNT has excellent mechanical properties, which effectively disperse the stress and improve the strength of the composites. MXene, due to its lamellar structure and high specific surface area, also plays a reinforcing role in the composites. mTA further strengthens the intermolecular interactions by forming a large number of hydrogen bonds, which makes CNT and MXene more flexible, which improves the overall mechanical properties of the material. At the initial stage of the stress-strain curve, the material exhibits high stiffness due to the synergistic effect of TA, CNT and MXene. As the strain increases, the reinforcing phase begins to slip or break, resulting in a decrease in overall stiffness. This increase in mechanical properties gives the fiber potential for a wide range of applications where high strength and flexibility are required, such as wearable devices and flexible electronics.

As shown in Figure.3, the self-healing property of the fiber is shown schematically, and the core-sheath structure was cut by the knife to achieve self-healing at 80℃. The yellow part of the figure shows the cut part. From the figure, it can be seen that the fibers were cut at 6 s. After 15 s of healing process, the fibers regained their electrical signals, in which many hydrogen bonding groups in the fibers were connected with each other to realize the connection of the conductive path. This indicates a good self-healing property. This excellent self-healing performance can cope with all kinds of scratches and damage, thus avoiding device failure caused by external mechanical stress.

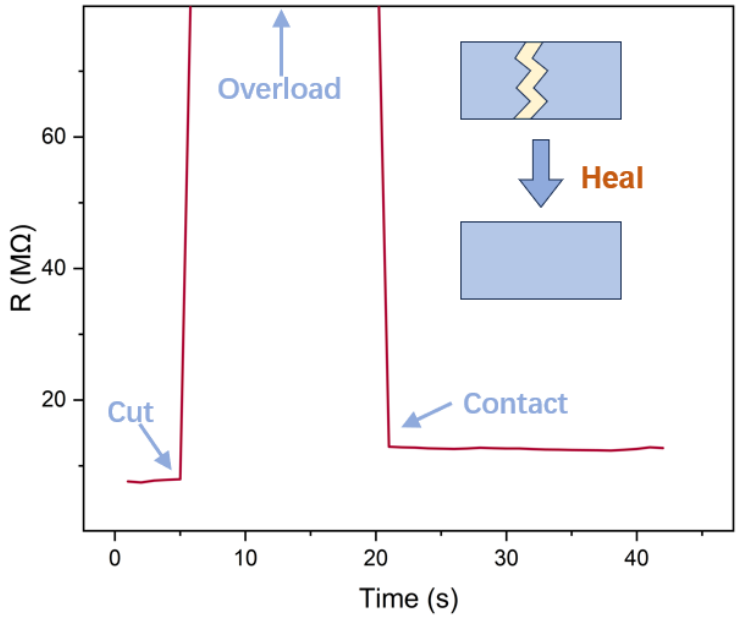

Figure 3: Plot of resistance versus time during sensor healing process

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3.2. Applications in temperature monitoring

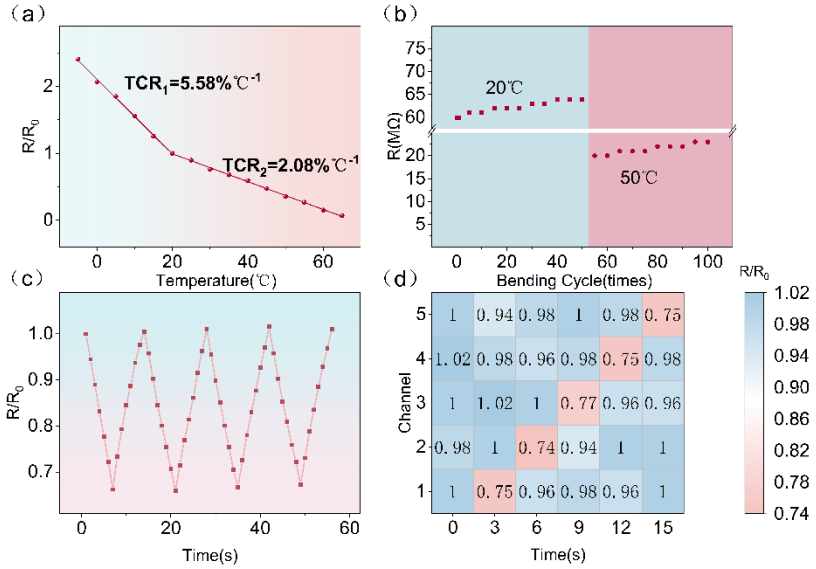

Figure 4: Temperature sensing properties of fiber. (a) The relative resistance curve of the fiber varies with the temperature. (b) The resistance changes corresponding to sensor bending test at 20°C and 50°C. (c) The relative resistance changes corresponding to sensor breathing cycle test. (d) Relative resistance of arrayed flexible temperature fibers as a function of timeigure 3: Plot of resistance versus time during sensor healing process

To investigate the temperature sensing properties of the fibers, the temperature sensing properties of the prepared fibers were measured. The relative resistance curve of the fibers versus ambient temperature is shown in Figure.4a. In the low temperature interval (-5°C to 20°C), the TCR is high (5.58 %°C-1), while in the higher temperature interval (20°C to 60°C), the TCR is 2.08 %°C-1. At low temperatures, the conductive networks of CNT and MXene are more stable, with many and stable conductive pathways through the mTAs to each other, resulting in a higher TCR.As the temperature increases, the contact between CNT and MXene changes due to thermal expansion, and there are fewer overlaps with each other, resulting in a change in the conductive pathways and a thinner conductive network, which leads to a slower decrease in resistance, thus showing a lower TCR at higher temperatures. higher temperatures, thus showing a lower TCR.

To further evaluate the recovery performance, a series of bending tests were performed. As shown in Figure 4b, the resistance of the fibers is plotted against the number of bending cycles at 20°C and 50°C. The fibers show an increase in resistance with the number of bending cycles at both 20°C and 50°C, but the initial resistance is lower at higher temperatures. This is because bending affects the conductive paths within the fiber. As the number of bending cycles increases, the contact point between the CNT and MXene breaks, resulting in an increase in resistance. Temperature has a significant effect on the electrical conductivity of the fiber.

To further evaluate the sensitivity and repeatability of the fibers to temperature, human respiration monitoring was performed as shown in Fig. 4c, where the relative resistance of the fibers is plotted against human sleep time. The fibers were placed on the anterior side of the human nasal cavity, and the curves showed obvious periodic fluctuations during sleep, and the resistance of the fibers varied periodically with time, indicating that the prepared flexible temperature sensor is capable of detecting human respiration conditions in real time. CNT and MXene are known for their excellent electrical conductivity and mechanical properties, and TA can enhance these properties by forming hydrogen

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bonds. Therefore, when the temperature of the fiber changes, which leads to a change in resistance. During inhalation, the temperature decreases and the resistance increases, while during exhalation, the temperature increases and the resistance decreases. The curves show steady periodic changes with no significant drift or noise, indicating good stability and sensitivity of the fibers. R/R0 fluctuates between 0.65 and 1, which is significant for breath detection. The curves show multiple cycles with each cycle having a similar shape, indicating that the fiber has good repeatability and reliability over multiple breathing cycles.

In order to explore the spatial temperature distribution, the array flexible temperature sensor was also prepared in this study, as shown in Figure.4d, which is an array of five fibers, with the change of time, the finger touches different fibers, from the test results, it can be seen that the fibers are able to sense the temperature change caused by the finger in real time. Since the temperature of the finger is higher than the room temperature, it causes the temperature of the fibers to increase, the conductivity to increase and the resistance to decrease. This change in resistance can be used to detect the movement path of the finger. The change in resistance is very noticeable and proves that it has good sensitivity.

4. Conclusion

In summary, we have developed a core-sheath structured fiber composed of mTA, CNT, MXene, and PU. The mTA reduces aggregation of MXene and CNT by forming extensive hydrogen bonds with both components, while the PU sheath provides mechanical robustness and flexibility, ensuring interfacial adhesion to the core through hydrogen bonding. This hierarchical architecture endows the fiber with exceptional mechanical strength (6.35 MPa tensile strength and 66.43% elongation at break), rapid self-healing capability (achieved through dynamic hydrogen bond reconfiguration at 80°C within 15 seconds), and high temperature sensitivity (TCR of 5.58%°C^{-1} in the range of -5–20°C and 2.08%°C^{-1} at 20–60°C). The excellent sensitivity and repeatability of the fiber were validated through bending tests, respiratory cycle monitoring, and sensor array applications. We believe that the material's microscale/nanoscale structural design and high performance make it promising for flexible sensor applications.

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