Flexible strain sensors fabricated using aligned carbon nanofiber membranes with cross-stacked structure for extensive applications

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ABSTRACT

Carbon nanofibers (CNFs) with excellent conductivity and stability have become a promising material to design the strain sensing network. To date, however, the effect of the stacked structure of CNF membrane on the sensing performance has rarely been studied. In this work, we reported a high-performance sensor based on the cross-stacked aligned CNF membrane. The effects of cross-stacked structures on the sensing characteristics were systematically investigated. The flexible strain sensor could capture low detection limit (<0.1%) with a gauge factor (GF) of 4.24 and wide strain range up to 130%. The uniform GF value reached 2050 when the strain was in the range of 100–130%. In addition, the high linearity under 40% strain (>0.998), excellent durability and quick response time (<200 ms) were demonstrated. The excellent comprehensive performances were simultaneously obtained. The sensor could be used in extensive applications, such as monitoring body movements and distinguishing the track of writing.

KEYWORDS

Electrospinning; carbon nanofiber membrane; cross-stacked structure; strain sensor; extensive applications

1. Introduction

Strain sensors could be widely used in many fields, such as transportation, aerospace, medical healthy and intelligent manufacturing [1,2]. Though some sensors with high stability and durability were obtained by using semiconductor materials and metal foil, they exhibited low strain range (<5%) and poor sensitivity (~2) [3,4]. Additionally, these materials with high stress and hardness could not meet the requirements of flexible
sensing fields. Therefore, flexible strain sensors fabricated using conductive nanomaterials and other carbon materials have gained widespread attraction [5–7]. At present, carbon nanomaterials, metal nanoparticles and nanowires were used to design the resistance-type sensing networks with the simple structure and processing method. For example, a pressure sensor composed of three-dimensional polyolefin elastomer nanofibers and silver nanowires (Ag NWs) showed an ultrahigh sensitivity [8]. Although the sensors fabricated using metal nanomaterials exhibited high sensitivity, they had certain limitations in high price and poor chemical stability. In contrast, carbon nanomaterials (carbon nanofibers (CNFs), graphene, carbon nanotubes (CNTs), etc.) had more advantages [9]. Among them, CNFs with excellent conductivity and electrochemical stability could be simply prepared by electrospinning and then carbonization [10]. Compared with other carbon nanomaterials, CNFs were easier to disperse uniformly in the polymer matrix, and assembled to form CNF membrane [11] and yarn [12]. Therefore, CNFs were promising to fabricate flexible strain sensors with lower cost compared with CNTs and graphene.

CNFs could be directly and uniformly mixed with polymers to design strain sensors [13,14]. For example, the conductive sponge prepared using polycrylonitrile (PAN) nanofibers, polyimide and CNFs could be compressed to 80% due to the porous structure [15]. Unfortunately, the sensitivity was very low, and decreased with increasing compressive strain. To optimize the conductive network, Wu et al. [16] prepared the porous materials by curing polydimethylsiloxane (PDMS) on the surface of CNF-coated sugar particles and washing using deionized water. The wide strain range, high linearity and durability were obtained. However, the sensitivity was poor (<6.5). The above results indicated the strain sensors based on uniform CNF composites still faced a challenge to obtain simultaneously high sensitivity and strain range. Some researchers have also designed strain sensors by fabricating carbon nanofiber yarns [17,18] and fabric [19]. They found these sensors had excellent sensitivity and stability and could effectively detect small deformation (0.1%) because of the brittleness of CNFs. However, the strain ranges were poor (<30%). It is necessary to further improve the assembly structure of CNFs for wide deformation.

To obtain simultaneously high sensitivity and wide strain range, many studies have directed toward the CNF membranes. The study found the ultra-thin monolayer CNF membrane could monitor the subtle pressure strain, but the strain range was low [20]. The thick CNF membrane exhibited large strain range, but the sensitivity was not satisfactory under the low strain [21]. The random arrangement of CNFs made the whole conductive network isotropic. When the stress was applied to the sensor, it was difficult to distinguish the direction of stress according to the change in resistance. Therefore, the aligned and regular CNFs were helpful for the conductive network to detect multiaxial strains. However, the dramatical difference of sensing performances was only found between the stretching direction perpendicular to the aligned direction of CNFs and the stretching direction parallel to the aligned direction in our previous work [11]. Lee et al. [22] demonstrated the forced angle direction could be identified when the two aligned CNF membranes were vertically stacked. In addition, the sensitivity could reach to 157 at a strain of 30%. However, the ability to monitor the low strain (<0.1%) was not demonstrated, and the linear relationship between strain and relative resistance change was not perfect. Sengupta et al. [23] found the strain sensor fabricated using monolayer bundled CNFs showed a high linear response. However, the strain range was only up to 50%. These
results also indicated the thickness of CNF membrane and the stacked structure of aligned CNF membrane had the great influence on sensing performance. The comprehensive performances were still not simultaneously achieved, including wide strain range, high sensitivity and linear response. Therefore, it is meaningful to systematically analyze and discuss the effect of the stacked structures on the sensing performance.

Herein, we reported the strain sensor with wide monitoring range and highly sensitivity fabricated using the cross-stacked aligned CNF membranes for intelligent wearable applications. CNF membranes were fabricated by simply electrospinning and then carbonization, and sealed using TPU film with a sandwich structure. The main contribution of this work was that the effects of cross-stacked structures of aligned CNF membranes on strain sensing performance were discussed by varying the thicknesses of monolayer CNF membrane, cross angles and the number of stacked layers. The comprehensive performances were analyzed. The strain sensors could be stretched up to 130% with high sensitivity. The subtle deformation (0.1%) could be also detected. In addition, high sensitivity and linear relationship were obtained. The good dynamic stability and durability were demonstrated under both small and large strains. Finally, we discussed the potential applications of the strain sensor in human body motion and physiological detection.

2. Materials and methods

2.1 Materials

PAN (Mw = 150,000 g mol⁻¹) was supplied from J&K Chemical Ltd. N’N-Dimethylformamide (DMF, purity >99.5%) was obtained by Sinopharm Group Chemical Reagent Co., Ltd. TPU (80 A) was obtained from Bayer Company. Graphene (thickness ~0.8 nm, the diameter of the nanosheet 0.5 ~ 5 μm) was purchased from Nanjing Xianfeng Nanomaterials Technology Co., Ltd. Copper wire (diameter 0.1 mm) and zinc film (thickness 10 μm) was obtained from Haoxuan Metal Materials Co., Ltd. (Qinghe County, China).

2.2 Fabrications of CNF membrane

Graphene was first dispersed in DMF, and then sonicated for 3.5 h. Subsequently, the PAN powder was fully dissolved in the graphene suspension. Finally, the PAN/graphene mixed solution was further sonicated for 3.5 h before spinning. The mass fractions of graphene and PAN in the composite solution were 0.1 wt.% and 10 wt.%, respectively. A small amount of graphene was used to improve the conductivity of CNFs.

The aligned PAN/graphene composite nanofibers were prepared using the electrospinning method reported in our previous work [11]. During electrospinning, zinc foil was used to wrap the rotating drum to collect the nanofibers. The diameter of the drum was 76 mm, and the rotational speed was 1000 r min⁻¹. The solution flow rate, applied voltage, and the distance from the metal needle to the drum were 1 ml h⁻¹, 18 kV, and 30 cm, respectively. The cross-stacked structures of the composite membranes were adjusted by changing the orientation of the zinc films. The electrospinning time was controlled to obtain PAN/graphene composite nanofiber membranes with different thicknesses.
The PAN/graphene composite nanofiber membrane with zinc foil were stabilized in air at 270°C for 1.5 h with a heating rate of 2°C min⁻¹. The subsequent carbonization process was carried out at 1100°C for 3 h in a pure argon atmosphere with a heating rate of 5°C min⁻¹. The zinc film was evaporated at 1100°C to obtain the intact CNF membrane.

2.3 Preparation of strain sensor

A similar process to previous work [11] was used to fabricate the flexible strain sensor. Copper wires were bonded with conductive silver paint as electrodes at both ends of the rectangular CNF membrane with a width of 5 mm. The CNF membrane and electrodes were then co-encapsulated in 2.5 g TPU/DMF solution (15 wt.%). By cutting, an H-shaped sensing device was obtained. The final width of the sensor was 10 mm, and the effective sensing distance was 30 mm.

2.4 Experimental design

Three important factors about the cross-stacked structures of CNF membranes, including the thicknesses of monolayer CNF membrane, cross angles and the number of stacked layers, were investigated to evaluate the sensing performance. The experimental matrix is shown in Table 1. In total, nine CNF membrane samples were prepared. Samples 1 to 5 were fabricated using two layers of CNF membranes with a crossing angle of 90° to discuss the effect of thickness of monolayer CNF membrane prepared by controlling the electrospun times on the sensing performance. Samples 5 to 7 were used to analyze the effect of the number of layers of CNF membranes on the sensing performance, and the total electrospin time was constant for 60 min. Samples 2, 8 and 9 were prepared using two layers of CNF membranes to analyze the effect of the crossing angles on the sensing performance, and the electrospin time of monolayer CNF membrane was 15 min. Sample 9 was stretched with two directions to analyze the anisotropy of the sensing performance.

2.5 Characterization and electrical signal measurement

The morphology of CNF membranes was analyzed using a scanning electron microscope (S-4800, Hitachi, Japan). The diameters of nanofibers were measured based on the SEM images using an analysis software (Image-Pro Plus 5.0). The orientation degree of CNF

| Sample number | The number of layers of CNF membrane | The crossing angle between adjacent layers | Electrospin time of monolayer CNF membrane (min) | The total electrospin time (min) |
|---------------|-------------------------------------|------------------------------------------|-----------------------------------------------|-------------------------------|
| 1             | 1                                   | 10                                       | 20                                            |
| 2             |                                     | 15                                       | 30                                            |
| 3             | 2                                   | 20                                       | 40                                            |
| 4             |                                     | 90°                                      | 25                                            | 50                            |
| 5             |                                     | 30                                       | 60                                            |                               |
| 6             | 4                                   | 15                                       | 60                                            |
| 7             | 6                                   | 10                                       | 60                                            |
| 8             | 2                                   | 0°                                       | 15                                            | 30                            |
| 9             | 2                                   | 45°                                      | 15                                            | 30                            |
membrane was measured based on the SEM images using a nonwoven fabric orientation analysis system (BD-11, Donghua University). The structural changes of CNF membranes in the process of stretching were observed using an optical biological microscope with an industrial lens (CX33, Olympus Corporation, Japan). The mechanical properties of strain sensors were controlled using a universal testing machine (Instron 3365, Instron Corporation). The strain sensors were mounted onto the testing machine, and the distance between two clamps was 30 mm. The electrical signals were recorded using an electrochemical workstation (CHI 760E, CH Instruments, Inc. USA). Amperometric i-t curve was selected. A constant voltage of 0.1 V was used to generate an electrical current signal.

The relative resistance change (ΔR/R₀) and gauge factor (GF) were used to evaluate the sensitivity of strain sensors. GF was calculated according to Equation (1) [24]:

\[ \text{Gauge factor} = \left( \frac{(R - R_0)}{R_0} \right) / \left( \frac{(L - L_0)}{L_0} \right) \]  

where ΔR is the resistance change between final resistance R and initial resistance R₀, L and L₀ were the final length and initial length of strain sensor, respectively.

3. Results and discussions

3.1 Structure of CNF membrane and characterization

Figure 1(a) illustrates the procedure of the CNF membrane, including electrospinning, stabilization and carbonization. The CNF membrane stacked with a crossing angle of 90° in the diagram was shown as an example. The raw PAN/graphene composite nanofiber membrane is shown in Figure 1(b). The monolayer CNF membrane was prepared with an electrospun time of 30 min. The diameter for nanofibers was 244 ± 54 nm. The orientation

![Figure 1](image_url)

**Figure 1.** (a) The preparation process of the carbonized composite nanofiber membrane; (b) SEM image of the raw PAN/graphene composite nanofiber membrane; (c) real photo of nanofiber membrane after carbonization; (d) SEM image of the composite nanofiber membrane after carbonization.
of composite nanofibers in the monolayer membrane reached to 61.7%. The perfect CNF membrane with excellent bendability and flexibility was prepared under the action of two carbon plates (Figure 1(c)). Its thickness was less than 10 µm. The diameter of nanofibers shrunk to 111 ± 25 nm after carbonization. However, the microstructure of CNF membrane has hardly changed as shown in Figure 1(d). More SEM images for different cross-stacked CNF membranes are shown in Fig. S1 (ESI).

The carbonized CNF membranes also obtained excellent electrical conductivity. The resistance for the stacked structures of CNF membranes is shown in Table S1. We found the resistance decreased from 7.98 to 1.36 kΩ/cm² with increasing the thickness of CNF membrane. The number of layers of CNF membranes had no great influence on the resistance. Notably, the resistance changed due to the change of crossing angles, which was ascribed to the anisotropy of conductivity for aligned CNF membrane [25].

3.2 Preparation and characteristic of strain sensors

Figure 2(a) shows the preparation process of the strain sensor. Firstly, TPU solution was uniformly spread out on a glass substrate to obtain the first layer of film. Subsequently, an oblong CNF membrane with a crossing structure of 60 mm × 5 mm was fixed. A small amount of TPU solution (5 wt.%) was used by a needle to infiltrate the CNF membrane from an electrode to the other one. The method could effectively solve the damage of CNF membrane due to the interfacial tension of TPU solution in the packaging process. Finally, the device was sealed using TPU solution to form a sandwich structure. The width of strain sensor was 10 mm, and the effective sensing distance was 30 mm. The thickness of the sensor was 239 ± 16 µm. Figure 2(b) shows a photo of the strain sensor fabricated using sample 2. The semitransparent feature was exhibited. The fonts under the strain sensor could be observed. The thin device can be easily bent and twisted as shown in Figure 2(c,d). The flexible electronic device can be attached on the surface of skin and integrated with textiles.

![Figure 2. (a) The schematic diagram for preparation process of strain sensor; (b) The real image for strain sensor; (c) Bending and (d) twisting of strain sensor.](image)
The previous studies indicated the research on the effects of stacked structures of CNF membranes on the sensing performance was significant [22]. Firstly, we studied the effects of thickness of CNF membrane on the sensing performance. The structural diagrams are shown in Fig. S2 (ESI). Five samples were prepared by controlling the electrospun times. Two layers of CNF membranes with a crossing angle of 90° were used to form the crossing stacked composite membranes. The results indicated the maximum strains were 99.5%, 95.1%, 93.3%, 90.9% and 138.4%, when the electrospun time for each layer was 10, 15, 20, 25 and 30 min (samples 1 to 5), respectively (Figure 3(a)). The strain range was slightly decreased when the electrospun time for each layer increased from 10 min to 25 min, and then sharply increased to 138% when the electrospun time was 30 min. The CNF membrane parallel to the stretching direction prevent them from deforming, and the strain range of cross-stacked CNF membrane decreased. However, when the thickness of the aligned CNF membrane perpendicular to the stretching direction increased, more CNFs were overlapping each other, leading to the greater deformability of CNF membrane. This was because there were always additional fibers bridging to maintain the conductive network during fiber stripping. The sensitivity for five sensors was small under the low deformation, and dramatically increased under the large strain. The result was attributed to that the structure of CNFs perpendicular to the stretching direction was only deformed, and the CNFs parallel to the stretching direction were broken in the initial phase of stretching. The most conductive paths were maintained. The large cracks

![Graph](image-url)

**Figure 3.** The relative resistance change–strain curves of strain sensors based on different CNF membranes: (a) Samples 1 to 5; (b) Samples 5 to 7; (c) Samples 2, 8 and 9; (d) Sample 9 stretched at different stretching directions.
occurred when the deformation of the CNF membrane reached its limit, and the resistance sharply increased. In addition, the high linear relationship between the relative change in resistance and strain (0 ~ 40%) was obtained, and the linearity reached 0.994 for the sensor based on sample 4 (ESI, Fig. S3). It was higher than that obtained by Sengupta [23].

We also studied the effect of the overlapping times on the sensing performance. The structural diagrams of three samples are shown in Fig. S4 (ESI). The electrospun layers were 2, 4 and 6, respectively. The total electrospun time for each sample was 60 min. The result indicated the strain sensing range decreased for the sensor based on sample 6 as shown in Figure 3(b). This may be due to the fact that the multilayer structure limited the strain capacity of CNF membrane layer, whose alignment was perpendicular to the stretching direction. The interaction force between adjacent membranes was strong, which led to the higher sensitivity under a strain of 60% (ESI, Fig. S5). However, the strain range for the sensor based on sample 7 slightly increased to 118%, and the sensitivity was small under the low strain, attributing to that the interactions between adjacent ultrathin monolayer CNF membranes were homogeneous, and more tiny cracks were formed resulting in increased deformability. As shown in Fig. S6 (ESI), the breaking points of the conductive network were dispersed in multiple positions of the CNF membrane during the stretching process. Besides, the linearity of sample 7 reached 0.996 (ESI, Fig. S5). The results showed that increasing the number of stacked layers in the CNF membrane was not conducive to improving the strain performance of the sensor.

The effect of the stacked crossing angles between two layers of CNF membranes on the sensing performance was examined. The structural diagrams of three samples are shown in Fig. S7 (ESI). Two layers of CNF membranes were stacked, and the crossing angles were 0° (sample 8), 45° (sample 9) and 90° (sample 2), respectively. The electrospun time for each layer was 15 min. Specially, the orientation of a layer in the stacked CNF membrane was abidingly perpendicular to the stretching direction. The results found that the strain range gradually decreased with the increase in the crossing angle (Figure 3(c)). This was attributed to that the crossing layers for 45° and 90° inhibited the structural change of the CNF layer perpendicular to the stretching direction. Meanwhile, the crossing layer for 0° could improve the strain capacity of the CNF layer perpendicular to the stretching direction. However, the sensitivity was very small for stacked CNF membrane with a crossing angle of 0° (ESI, Fig. S8). The sensitivity increased under 60% strain with the increase in the crossing angle. The main reason was that the structural deformation without fracture damage was not conducive to the change in resistance of conductive network. The linearity for sensor based on sample 9 (0.998) was larger than that of other two samples. High linear relationship was beneficial to the precise monitoring of strain.

The effect of the stretching direction on the sensing performance was also studied using sample 9. The structural diagram is shown in Fig. S9 (ESI). Two layers of CNF membranes with a crossing angle of 45° were prepared, and the electrospun time for each layer was 15 min. Two stretching directions, named as X axis and Y axis, were described. The results found that the strain range was markedly large when the stretching direction was along the Y axis (Figure 3(d)). However, the sensitivity was low compared with that obtained along the X axis under 60% strain as shown in Fig. S10 (ESI). The significant anisotropy was obtained at the different stretching directions. This was attributed to that the strain sensing performance of aligned CNF membrane was anisotropic.
Previous research has reported the strong mechanical and sensing anisotropy of the highly aligned fiber membrane [26]. The aligned membrane has a higher sensitivity parallel to the aligned direction and a wider strain range perpendicular to the aligned direction. The above results demonstrated that the stacked structure of CNFs had great influence on the performance of sensors. The crossing structure was beneficial to obtain wide strain range and improve the sensitivity of the sensor.

The performance of the sensor prepared using sample 9 was systemically studied. The stretching direction was perpendicular to the orientation of CNFs (Y axis, ESI, Fig. S9). Figure 4(a) shows ∆R/R₀ of the sensor during single-cycle stretching of 60% at a stretching rate of 40 mm min⁻¹. Due to the plastic deformation of TPU polymer during the cyclic loading-unloading process, the sensor was bent when the clamping retracted back to the original length (ESI, Fig. S11). When the clamping moved to 9% during the loading process, the bending device was in a straight state. This was presumably attributed to the irreversible deformation of TPU molecular segments. When the device was gradually stretched to 60% strain, the ∆R/R₀ increased. However, the resistance did not return to the original value when the clamping retracted back to the original length. The resistance kept going down until the sensor was stretched up to 9% again. The result demonstrated the larger response hysteresis for this sensor. In addition, the resistance change during stretching was obviously discrepant to that during retracting, indicating a large hysteresis at the retracting stage.

The sensitivity and stability of sensor device were important factors in evaluating strain-sensing performance. Figure 4(b) shows the ∆R/R₀ responses in the subtle strain range of 0.1–2% at the stretching rate of 1 mm min⁻¹, and Figure 4(c) shows the ∆R/R₀ responses in the strain range of 10–100% at the stretching rate of 40 mm min⁻¹. The response increased with the increase in strain levels. The response showed high

![Figure 4. The performance of the sensor prepared using sample 9. (a) Resistance changes during single-cycle stretching (stretching rate 40 mm min⁻¹); strain response in the strain range of (b) 0.1–2% (stretching rate 1 mm min⁻¹) and (c) 10–100% (stretching rate 40 mm min⁻¹); (d) relative resistance change in different strain ranges. (e) Gauge factors in different strain ranges; (f) the effect of stretching rate on sensitivity (strain 60%).]
repeatability at different strain ranges. However, the resistance change during stretching was unsymmetric to that during retracting, indicating a conspicuous hysteresis. Gratifyingly, the wide strain monitoring range was obtained from 0.1% to 100%. Notably, the $\Delta R/R_0$ of the sensor under the low strains showed two peaks (Figure 4(b)). The appearance of double peaks was mainly caused by the competition between the destruction and reconstruction of the conductive network during stretching process [27,28]. On the one hand, the competition between disruption and reorganization of the nanofiber conductive network was intense under low strains. When large strain was applied, the crack effect became the dominant mechanism for the conductive network. On the other hand, attributing to the Poisson effect, the sensor was subjected to contract perpendicular to the stretching direction. The resistance had a decreasing trend under compressive deformation. In addition, the hysteresis of the TPU matrix and the weak interfacial bonding force between the CNF membranes and TPU were also the reasons for the formation of the double peaks. Similarly, the asymmetry of $\Delta R/R_0$ between the loading and unloading process in Figure 4(c) could also be attributed to a weakened bimodal effect. During unloading, the hysteresis of the elastic matrix was still severe, but the faster strain rate promoted the rapid recovery of the conductive network. Therefore, the resistance change during releasing was asymmetric with that during stretching.

Figure 4(d) shows the dependence of $\Delta R/R_0$ on strain. The results indicated that the linear relationship between $\Delta R/R_0$ and strain was observed when the strain was in the range of 0.1–2% (stretching rate 1 mm min$^{-1}$) and 10–100% (stretching rate 40 mm min$^{-1}$). The linear response suggested that the electric resistance increased in equal proportion to strain. The $\Delta R/R_0$ value sharply increased when the strain was more than 100% (Figure 3(d)), indicating the structure of conductive network was rapidly damaged at the large strain. Although the strain range will be slightly reduced compared with that the orientation of CNF membrane was completely perpendicular to the stretching direction (Figure 3(c)), the low strain (0.1%) can be monitored.

Gauge factor (GF) was calculated based on relative change in resistance at a specific strain to quantify sensitivity of sensor device. In the strain range of 0.1–2% (stretching rate of 1 mm min$^{-1}$), the GF was stabilized at 5.5 with a variation within ±1.3 (Figure 4(e)). The GF firstly decreased and then increased when the strain changed from 10% to 100%, and the uniform GF value was only 2.48. In addition, the uniform GF value reached 2050 when the strain was in the range of 100–130%, attributing to the dramatical crack variation [21].

Figure 4(f) shows the effect of the stretching rates on the sensing performance. At a strain of 60%, the $\Delta R/R_0$ had no apparent change when the stretching rate varied in the range of 20–80 mm min$^{-1}$. Meanwhile, the high stability was obtained at the different stretching rates (ESI, Fig. S12).

The durability of strain sensor was also studied using a self-made reciprocating motion device. The durability at the subtle strain of 0.1% with a stretching rate of 1 mm min$^{-1}$ over 300 loading-unloading cycles and at the large strain of 100% with 450 mm min$^{-1}$ over 2000 loading-unloading cycles were outstanding (ESI, Fig. S13). It was observed that the structure of the nanofiber membrane did not change significantly after multiple cycles of stretching (ESI, Fig. S14). The result indicated that this sensor device could be used to detect the wide strain range with high durability. However, the static stability was perfect as shown in Fig. S15(a) (ESI). The main reason was the creep behavior of TPU matrix.
The response time and recovery time were also important for real-time monitoring. The result found the response time was less than 200 ms (ESI, Fig. S15(b)). Nevertheless, the recovery time was long, and more than 10 s. The performance may be improved using the other elastic matrix. In summary, the sensing performances (including strain range and gauge factor) of our sensor were compared with other CNF-based strain sensor in the literature (ESI, Fig. S16).

In order to understand the structural change of the stacked conductive network, the optical images of CNF membrane fabricated using two layers with a crossing angle of 45° are analyzed at different strain levels as shown in Figure 5(a). The electrospun time for each layer was 15 min. The stretching direction was perpendicular to the orientation of CNFs in the one layer (under layer). The angle between the orientation of the other layer and the stretching direction was 45° (top layer). The stacked structure was very conspicuous. It could be observed that the structure of the CNF membrane changed significantly during the stretching-recovery process. The CNFs at the top layer firstly broken, and the CNFs at the under layer underwent the bending deformation. The conductive paths were still connected because of the interaction of the two layers of CNF membranes. The resistance change was not prominent at this strain stage. With the increase in strain, the cracks of CNF membrane aggrandized, and the number of cracks also increased. At 100% strain, many fibers were broken, and the blank area in the membrane increased. A few obvious band cracks could be found. However, upon careful observation, it could be found that many nanofibers parallel to the drafting direction or forming a small crossing angle with the drafting direction were broken, while the nanofibers perpendicular to the drafting direction were significantly deformed. Finally, CNFs at the under layer broken at the large strain range. The conductive network lost its function when these cracks joined together. These cracks were not easy to be found when the external stress was removed.

![Figure 5](image-url)

**Figure 5.** (a) The optical images of CNF membrane fabricated using two layers with a crossing angle of 45° at different strain levels (electrospun time 15 min for each layer); (b) the diagram for the structural change of conductive network in the loading-unloading process.
The sensing mechanism is described in more details as shown in Figure 5(b). The resistance perpendicular to the orientation of the nanofibers is relatively high because there are only scattered contact points between the nanofibers. With the increasing cross angle of CNF membranes, the number of the conductive path increases. Besides, the electrical conductivity of the CNF membranes varies with the stacking density of nanofiber membranes. CNFs parallel to the stretching direction and formed a small angle along stretching direction firstly fracture at a low strain due to the brittleness of CNFs. Subsequently, the sloping CNFs fracture with increasing the strain level. The resistance gradually increases. Meanwhile, CNFs perpendicular to the stretching direction were bent, and the distance between the adjacent CNFs increased. However, the conductive paths are connected due to the effect of broken CNFs as bridge. The points of function are maintained in the stretching process. Therefore, the resistance change is not remarkable. The curved CNFs are also broken with the increase in the strain level. In addition, the original cracks gradually increased, and the more breaking points are formed. Finally, a complete crack is formed in the weak position, leading to a remarkable change in resistance. These cracks decreased under the action of elastic TPU matrix when the loading force is removed. However, the cracks cannot lose, leading to the noteworthy resistance change between the first stretching cycle and the subsequent cycles. The number of cracks and their structure are not remarkable change in the further loading-unloading cycles. The outstanding stability and durability are obtained.

We further showed that the strain sensors can be used to detect different deformations such as bending and stretching as shown in Figure 6(a,b). The characteristic peaks were obtained. When the device was attached to a human wrist, it could detect the bend freely up and down (around 90° in the photo, Figure 6(c)). The obvious peaks were obtained. However, the peaks were low when the wrist swung from side to side (see in the photo). The significant difference was attributed to the anisotropy of sensing performance. The sensor could be used to distinguish the moving direction of human joint and detect the bending of joint by attaching it to the index finger. The resistance

![Figure 6. The practical applications of strain sensor. (a) Bending; (b) twisting; (c) bending and swing of wrist; (d) hold the objects with different diameters (the sensor was fixed on the index finger); (e) deglutition; (f) writing three letters on the surface of strain sensor.](image)
responses are different to grab the pen and pen holder as shown in Figure 6(d). The device could also detect deglutition caused by muscle movement (Figure 6(e)) and sense the change of facial expressions (Fig. S17). In addition, the sensor could also monitor the dynamic sliding pressure of single point. The relative resistance changes are recorded when three letters (a, b, c) are written on the surface of the sensor as shown in Figure 6(f). The curves of resistance changes were different for these three letters. There were two characteristic peaks for letter ‘a’ and letter ‘b,’ and one peak for letter ‘c.’ The curves of resistance changes had no significant difference for the same letter in the same position. The result indicated that the handwriting had a certain repeatability, which could provide a new idea for the development of handwriting recognition system with higher sensitivity.

4. Conclusion

In this work, the effects of cross-stacked structures on the sensing performances were investigated, including thickness of CNF membrane, the cross angles between two layers of membrane and the number of layers of CNF membranes. The strain range was firstly decreased and then increased for two layers of CNF membranes with a crossing angle of 90° when the electrospun time for each layer increased, and the sensitivity was small under the low deformation, and dramatically increased under the large strain. The sensing strain range had a slight decrease with increasing the overlapping times of CNF membranes, and the sensitivity was higher for two layers of CNF membranes under the low strain. The strain range decreased with the increase in the crossing angle, and the sensitivity gradually increased under 60% strain. The significant anisotropy was obtained at the different stretching directions for two layers of CNF membranes with a crossing angle of 45°. The flexible strain sensor can capture low detection limit (<0.1%) with a gauge factor of 4.24. The uniform GF value reached 2050 when the strain was in the range of 100–130%. In addition, the high linearity under 40% strain (>0.998), excellent stability, durability (100% strain for 2000 cycles) and quick response time (<200 ms) were obtained. The sensor showed the excellent comprehensive performances. The sensor could be used to monitor human body movements and distinguish the track of writing.

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Disclosure statement

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