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Surface Strain Redistribution on Structured Microfibers to Enhance Sensitivity of Fiber-shaped Stretchable Strain Sensors

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Abstract

Fiber-shaped stretchable strain sensors with small testing areas could be directly weaved into textiles. This paves the way for the design of integrated wearable devices capable of obtaining real-time mechanical feedback for various applications. However, for a simple fiber which undergoes uniform strain distribution during deformation, it is still a big challenge to obtain high sensitivity. Herein, we report a new strategy, surface strain redistribution, to significantly enhance the sensitivity of fiber-shaped stretchable strain sensors. A new method of transient thermal curing was used to achieve the large-scale fabrication of modified elastic microfibers with intrinsic microbeads. The proposed strategy is independent of the active materials utilized and could be universally applied for various active materials. Our strategy would shift the vision of the sensitivity enhancement method from the active materials design to the mechanical design of the elastic substrate, and the proposed strategy could also be applied to non-fiber-shaped stretchable strain sensors.

Stretchable strain sensors, as one of the critical components in wearable electronics, are developed to obtain the real-time mechanical feedback for applications in healthcare, rehabilitation, soft exoskeleton suits and so forth.^[1-6] Commonly, the reported stretchable strain sensors are based on flat films,^[7-38] while fiber-shaped stretchable strain sensors with small testing areas could be directly weaved into textiles becoming more accessible for wearable electronics in terms of wearability and integrability.^[39, 40] Currently, several kinds of fiber-shaped stretchable strain sensors were developed based on the conductive materials, such as poly(3,4-ethylenedioxythiophene), silver nanowires, carbon nanotubes, ionic conductive ink, and liquid metal.^[41-45] It is noted that these materials are distributed along the smooth fibers with identical surface structures and had homogenous strain distribution during the deformation (**Figure 1a**). Under this homogenous strain distribution, the gauge factor reflecting the sensitivity of stretchable strain sensors is quite limited (Table S1).

According to the structural mechanical analysis, when a certain work, W , is applied to a system, the energy is absorbed/stored as the strain in the materials. The work can be written as $W = \int E \varepsilon^2 dV$, where E is the modulus, ε is the strain and V is the volume of the materials. For the same work applied, the more significant change in the active materials is equivalent to the higher sensitivity of this sensor. For a smooth fiber, the energy efficiency is quite low, because V keeps constant for each differential length of this fiber and the strain is dispersed uniformly in the fiber. In contrary, if the strain could be accumulated at certain positions, it would induce more significant change in the active materials, i.e., the higher sensitivity. Therefore, how to redistribute the surface strain of the fiber to achieve the strain accumulation is the critical issue. According to the energy conservation principle, although the average strain is constant for a given volume under the same applied work, the local strain distribution still can be enlarged by relatively decreasing the local volume. Therefore, for the resistive stretchable strain sensors, the sensitivity could be significantly improved if the stretchable conductive materials are deposited in the region(s) where the local strain is magnified. By

employing finite element modeling (FEM), we find that the strain is indeed redistributed by proper design. For example, the maximum achievable local strain almost doubled by introducing an uneven distribution of microstructures along the fiber (Figure 1a, b). In addition, if high-modulus materials are employed in the low-strain (large volume) regions, the maximum strain can be further enhanced (Figure S1).

Following this way of reasoning, we suggest a new strategy, surface strain redistribution, to significantly enhance the sensitivity of fiber-shaped stretchable strain sensors. In order to demonstrate this strategy, we firstly propose a new method of transient thermal curing to achieve large-scale fabrication of PDMS microfibers with uniform predefined diameters. Compared to the well-established technologies of the flat film fabrication, it is still a challenge to introduce micro structures to one single fiber. Here we utilize the principle of Plateau-Rayleigh instability to modify microfibers with uniformly distributed microbeads. These microbeads can tune the strain distribution along the fiber by rearranging the strain from the beads to the areas in-between the beads – a strategy that effectively results in strain concentration. The local strain is magnified by this process resulting in longer micro-cracks in stretchable gold films/CNT films deposited on the surface of the fiber, and the sensitivity is significantly improved compared to fibers without microbeads. It is demonstrated that the beads-on-fiber stretchable strain sensor can be employed to reliably monitor sports activity. In addition, it is found that the sensitivity enhancement methods for flat-film stretchable strain sensors are currently limited to the active materials design (Table S2). We envision that the proposed strategy of strain redistribution can be applied to non-fiber-shaped stretchable strain sensors and draws attention to the elastic substrate design.

As discussed previously, the sensitivity of the fiber-shaped stretchable strain sensor can be enhanced as long as a methodology to modify the fibers with an uneven distribution of microstructures can be devised. It is well known that when oil flows along one fiber it will automatically split into droplets because of Plateau-Rayleigh instability (**Figure 2a** and

Supplemental Video 1). Inspired by this natural phenomenon, we produced PDMS beads on the PDMS fibers with the desired uneven distribution of microstructures that could take advantage of the strain redistribution effect as previously discussed (Figure 2a). As the very first step, we need to develop a feasible method for large-scale fabrication of PDMS microfibers. We propose a new method, transient thermal curing, to achieve the efficient fabrication of these micro fibers (See Supporting Notes for details). The basic principle is that after proper control of the viscosity of PDMS precursor, the microfibers can be fabricated by applying traction and then immersed into hot oil bath immediately to be fully cured (Figure S2, 3 and Supplemental Video 2). The micro beads modification also follows this process (Figure 2b).

We obtained PDMS fibers with good uniformity along the length direction and with predefined diameters by adjusting the viscosity of PDMS and the traction time. The dynamic viscosity of PDMS was tuned by pre-curing process (Figure S4a) and it influenced the fiber diameter (Figure 2c). In order to achieve uniform fibers, for a given viscosity, the period over which the traction strain was applied need to be strictly controlled. Considering the traction process as a viscous catenary flow under gravity, the fiber morphology could be simulated (See simulation details in Supplemental Notes). The diameter of the fiber remained uniformly distributed during the first 3 seconds (Figure S4b-d). Then they gradually turned thicker in the middle and thinner near the traction point (Figure S4c,d, S5 and S6a). The reason behind is that the viscous PDMS precursor actually is not fully cured during the traction and still can flow from the ending to the middle under the gravity. The lower the viscosity is, the better the mobility is. Therefore, all fibers were immersed into the hot silicon oil bath in the first few seconds of the traction in order to obtain the fibers with relatively uniform diameter. The fibers were cut at different position along the length direction and the diameter was measured to ensure reproducible uniformity (Figure S6b). Moreover, the diameter of the fiber, R , also can be controlled by the amount of added PDMS, its viscosity and the traction length. A

minimum diameter of about 10 μm with 4 cm in length was obtained by rapidly increasing the traction range. The length-to-diameter ratio achieved was around 4000 (Figure S7). The maximum fiber diameter was around 1 mm with 8 cm in length through decreasing the traction range. Importantly, by repeating this fabrication process, meters of the fiber can be fabricated in a short time (Figure S7).

After the fiber fabrication process, we tried to modify beads on the fiber to enhance the sensitivity as discussed above. Beads-on-fiber phenomena are commonly found for the nano-size fibers during the electro-spinning process,^[46-48] but only a few reports have been published for micron-sized fibers.^[49-51] Basically, beads automatically form when the viscous liquid flows along a vertically aligned fiber. The formation mechanism has been studied in detail,^[52-55] and the competition and balance between the surface tension and gravity, also called Plateau-Rayleigh instability, could explain this behavior (Figure 2b, Figure S8 and Supplemental Video 1 and 3). Here, the dimensionless Bond number:

$$G = \frac{\rho g R^3}{8\gamma h_0} \quad (1)$$

is used to predict the occurrence of bead formation, where ρ is the density of the liquid, g is the gravitational acceleration constant, R is the diameter of the fiber, γ is the surface tension and h_0 is the undisturbed film thickness of the PDMS precursor.^[53] According to this principle, the fiber diameter significantly affects the attachment and distribution of the beads. Normally, 0.7 is regarded as a critical value for bead formation, i.e., when G is larger than 0.7, the viscous liquid is in steady flow and beads cannot form, but for smaller G values beads do form. In order to tune the diameter of the bead, D , as discussed above, the viscosity, traction time and distance must be controlled. By applying the same traction time and distance, we investigated the effect of the viscosity on the diameter (Figure 2c). According to theory the beads diameter can be predicted as follows:^[53]

$$H_{\max} = 1.2860c^{2/3} - 1.413 - 0.2904c^{-1/3} + O(c^{-2/3}) \quad (2)$$

$$G = 0.5960 + 0.33c^{-2/3} + 0.19c^{-1} + O(c^{-4/3}) \quad (3)$$

where H is the protrusion thickness of the PDMS film (Figure S9), H_{max} is the maximum value of H and $H_{max} = \frac{(D-R)}{2}$; c is the migration speed of the bead. Thus, for a given Bond number, we can predict the bead diameter and the obtained values were in agreement with those found experimentally (Figure 2d).

One typical sample of the fiber with beads is shown in **Figure 3a**. Various active layers can be deposited on this platform including stretchable carbon nanotubes film and stretchable gold films (Figure 3a). The stretchability of this fiber-shaped stretchable strain sensor can reach ~ 125 % (Figure 3b). Besides, it possesses good linear response with the applied strain. This sensor still remains stable after more than 5000 strain cycles at 30% strain (Figure 3c). The initial resistance of the fiber is monitored (Figure S10a). It would increase a little bit in the stabilized cycles and keep constant after that. The cyclic performance under 50%, 100% and 120% is also checked for this sensor showing the good stability for different strain range (Figure S10b-d). We also employed FEM to investigate the strain redistribution effect induced by the beads (Figure 3d). It is evident that the beads tune the strain distribution and induce the strain concentration, which will further determine the crack distribution of the deposited gold film. This stretchable gold film can still be conductive under the large mechanical deformation. The underlying mechanism relies on numerous randomly-distributed initial nano-cracks that develops into micro cracks to release the applied tensile strain.^[56, 57] Therefore, these micro-cracks can be taken as the tracking signal for the surface strain distribution. We observed that cracks indeed became longer and wider at the strain concentration areas compared to those found in areas exposed to a smaller strain (Figure 3d). The statistical analysis also confirmed that cracks became longer than those observed in fiber without beads (Figure 3e). It is also worth mentioning that the resistance change was determined by the longest cracks that were more abundant of the fibers with beads. As a result,

the fiber with beads possessed larger resistance change, i.e., higher gauge factor, than that without beads (Figure S11 and Figure 3f). Other stretchable conductive materials can also be applied for this sensitivity-enhanced platform to fabricate fiber-shaped sensors. For example, we also employed carbon nanotubes as the conductive layer, and the sensitivity enhancement was observed and the gauge factor increased to a value around 100 (Figure S12). And the performance comparison of our sensors with others recently reported is shown in Table S1. Besides, we also investigate the effect of beads spacing on the sensitivity. The sensitivity is significantly enhanced with the decrease of the beads spacing (Figure S13). The reason is that with the decrease of the spacing, the number of the bead on the fiber actually increases and the strain redistribution effected will be enhanced. Moreover, since the strain can be tuned by using our platform, any active materials can be utilized to enhance the sensitivity. Thus, it opens up a new perspective to tune the sensitivity by employing a hybrid approach where the supporting elastic substrate is different from the active material itself.

Finally, we demonstrate that the fiber-shape stretchable strain sensor can be used to monitor strain induces during sports activity. A practical issue in wearable stretchable strain sensing relates to proper strain transfer from the object (muscle/joint) to the sensor. The default hypothesis assumes that the object properly interfaces with stretchable strain sensor, and that the only issue worthwhile considering is the sensitivity of the sensor. However, the adaptability of the wearable sensor to the object upon deformation must be considered to ensure that the strain is accurately transferred to the sensor to avoid the signal distortion. Herein, we report that the interlocking effect between PDMS and textile (e.g., kinesiology tape) provides a reliable adhesion mechanism that guarantees proper strain transfer (**Figure 4a**). The PDMS precursor was half-cured and then poured onto the porous fibrous textile. Through the mechanical test, the strength of the junction area of PDMS and textile is larger than that of PDMS itself (Figure 4a and Supplemental Video 4). Moreover, the kinesiology tape is a good candidate to support the stretchable strain sensor. It adheres tightly and

conformably to skin and is insensitive to the sweat. Integrated with the customized chip, the data was recorded wirelessly (Figure 4b,c). The gesture of squatting and leg lifting was successfully monitored (Figure 4d,e and Supplemental Video 5,6). We can also correspond the strain change to the leg bending angle (Figure S14). Also, the advantage of fiber-shaped stretchable strain sensors is that they can be more conveniently weaved into the clothes, compared to the flat-film one. Here, we add one more demonstration to show this advantage. One fiber-shaped stretchable strain sensor was weaved into one piece of elastic clothes by hand (Figure S15). The bending action can be monitored by wearing this clothes (see Supporting Video 7). For wearable electronics, actually, fiber-shaped sensors could be more important because they can be weaved into clothes directly based on the well-developed textile machinery, which will benefit the low-cost and large-scale process. Besides, they can definitely cooperate with other sensors, like acceleration sensors and temperature sensors, to obtain the full monitoring of the sports activity. We envision that our fiber-shaped sensors can be further integrated with other fiber-shaped batteries/super-capacitors to achieve the smart textile in the near future for potential healthcare, rehabilitation and sports monitoring and so forth.

In summary, the novel strategy of surface strain redistribution through delicate structure designs to enhance the sensitivity of the fiber-shaped stretchable strain sensor is proposed and the new beads-on-fiber stretchable strain sensor is reported based on the principle of Plateau-Rayleigh instability. We also report a new method, thermal transient curing, to achieve large-scale PDMS fiber fabrication. The fiber diameter and the diameter uniformity along the length direction can be well controlled. Our fiber-shaped stretchable strain sensors can be reliably used to monitor the sports activity. Following our strategy, the sensitivity of the fiber-shaped stretchable strain sensor could be further enhanced and other thermosetting polymer based fiber sensors can be fabricated. Moreover, our proposed strategy, surface strain redistribution, is independent of the active materials utilized and opens up a new

perspective of the sensitivity enhancement. It also could be applied to non-fiber-shaped stretchable strain sensors.

Experimental Section

Fabrication of PDMS fibers: PDMS 184 (Sigma Aldrich) with 1:10 weight ratio of the prepolymer and the cross linker was prepared following the normal procedure, and then pre-cured in 60 °C oven for around 25 minutes, lastly, kept in room temperature for different time to obtain the PDMS with different viscosity. Meanwhile, the clean silicon oil was heated up to 170 to 180 degree centigrade. During this process, it should be emphasized that the more uniform the temperature field of the silicon oil is, the better the quality of the fiber is, because the micro fibers were cured within 3 seconds in the hot oil and the temperature vibration would induce different thermal expansion in the same fiber inducing the abnormal deformation. When the oil bath was ready, certain amount of PDMS precursor was stretched in a short time and then put into the bath (see details in Supporting Notes). After few seconds, the fiber was ready and taken out followed by the washing process by using daily-used cleaning fluid and deionized (DI) water. Finally, fibers were immersed in the DI water overnight and then dried in 60 °C oven.

Fabrication of beads onto the fibers: We used the prepared PDMS fibers to further do the modification with PDMS beads. The viscosity tuning process of the PDMS precursor was the same with that described in the fabrication of the fiber. Then fibers were immersed into the precursor for few seconds and then suspended vertically along the length direction. After tens of seconds, beads will appear and then the fiber with beads was put into the hot oil following the same process described before. The beads diameter could be further regulated by using the PDMS precursor with different viscosity.

Fabrication of the stretchable gold film and carbon nanotube film onto the fiber: In order to demonstrate that our beads-on-fiber PDMS can act as the platform to enhance the sensitivity

of the strain sensor, we employed two kinds of stretchable conductive materials as the active layer. One was the stretchable gold film reported by our previous study;^[56] this film was with initial nano cracks without strain applied and these nano cracks will grow into micro cracks releasing the strain in the first strain cycle. Then subsequently, micro cracks opened and closed during the next strain cycles inducing the resistance change and achieving the stretchability of the gold film. The other one was the stretchable carbon nanotube film reported by our previous study.^[10] This film followed the same principle to achieve the stretchability. MTS model 42 was employed to apply the tensile strain. And the liquid metal (Gallium-indium eutectic, Aldrich) acted as the wire bonding adhesive which could be adaptive to the tensile strain and keep the stable wire bonding under deformation. Keithley 4200-SCS was used to do the measurement of the resistance using a sampling rate of 125 samples per second.

Demonstration of the fiber sensor in textile: The textile was purchased in the sports shop and was initially used for the joint protection (Pro-Tech tape). Half-cured PDMS was used as the connection glue and then fully cured at two ends of the fiber-shaped stretchable strain sensors. The strong connection was proven by the tensile testing (Figure 4a). The integrated customized chip was used to connect with the sensor to wirelessly collect the data.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Figure 1. Proposed strategy of surface strain redistribution to enhance the sensitivity of fiber-shaped stretchable strain sensors. a) FEM simulation to investigate the strain regulation effect of the micro-structured fiber as compared to the flat fiber. b) Strain distribution along the fiber surface for different structures in a).

Figure 2. Fabrication of PDMS fibers with beads. a) Inspired from the phenomenon of oil droplet splitting, the structured fiber could be fabricated accordingly to regulate the strain distribution. b) After fibers are cleaned and dried, beads are formed according to the principle of Plateau-Rayleigh instability, by controlling the Bond number. c) By keeping other fabrication parameters constant, the fiber diameter can be tuned by adjusting the dynamic viscosity. The red line shows the fitting curve. The related formula is in Supporting Note and Adj. R-Square is 0.99. d) Influence of the dynamic viscosity of the PDMS precursor on the bead diameter. The fiber diameter used is around 250 μm . The inset red curve shows the predicted value by the theory (Equation 1 and 2).

Figure 3. Performance and related principles of beads-on-fiber stretchable strain sensors. a) The stretchable gold film was deposited onto the structured fiber. b) Resistance changes as a function of applied tensile strain. c) Long-term stability of stretchable strain sensors upon exposure to 5000 tensile strain cycles at an applied strain of 30%. d) SEM images of the stretchable gold film at different locations along the fiber. e) Statistic analysis of the crack length in d). Dots beside the box present the crack length analyzed. f) Quantitative analysis of the effect of the bead diameter on the strain sensitivity. The red line shows the fitting curve. The related formula is in Supporting Note and Adj. R-Square is 0.96.

Figure 4. Demonstration of the beads-on-fiber stretchable strain sensor for sports monitoring. a) Adhesion test of the PDMS film and the kinesiology tape. Reliable attachment was achieved by interlocking effect of the PDMS and the tape. b) The sensor with the tape was adhered onto the joint of lower limb to monitor the squatting. c) Detailed description of the customized wireless resistance monitoring chip. Also, the sensor can be used to monitor the leg lifting. d,e) The typical raw data obtained by the fiber-shaped sensor when detecting squatting and leg lifting. The noise was induced by the limb natural vibration.

The table of contents entry

Surface strain redistribution in substrate is proposed to significantly enhance sensitivity of fiber-shaped stretchable strain sensors fabricated by thermal-transient-curing and Plateau-Rayleigh instability. PDMS microfibers with beads are produced in large scale. Beads regulate strain distribution. Novel sensors are well adhered to textiles monitoring sports process. Our study opens up a new perspective of fiber-shaped sensors and method to enhance sensitivity.

Keywords: stretchable strain sensors, fiber shape, beads, strain redistribution, enhanced sensitivity

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Surface Strain Redistribution on Structured Microfibers to Enhance Sensitivity of Fiber-shaped Stretchable Strain Sensors

ToC figure