J. Meas. Sci. Instrum., 2024, 15(3): 285-291.
DOI: 10.62756/jmsi.1674-8042.2024029

http://jmsi.publish.founderss.cn/
jmsi@nuc.edu.cn

Ultrasensitive stretchable patches for joint motion monitoring

LI Tengteng[1,2], LI Ziwei[1,2], HAO Yafeng[1,2], WU Huijia[1,2], ZHU Pu[1,2], MA Fupeng[1,2], LI Fengchao[1,2], YU Jiangang[1,2], LIU Meihong[1,2*], LEI Cheng[1,2*], LIANG Ting[1,2]

1. School of Semiconductor and Physics, North University of China, Taiyuan 030051, China;
2. State Key Laboratory of Dynamic Measurement Technology, North University of China, Taiyuan 030051, China

***Corresponding author:** LIU Meihong (liumeihong@nuc.edu.cn); LEI Cheng (leicheng@nuc.edu.cn)

Received: August 4, 2024 **Revised:** September 5, 2024 **Accepted:** September 9, 2024

Abstract: Wearable devices have great application potential in the next generation of smart portable electronics, especially in the fields of medical monitoring, soft robotics, artificial intelligence, and human-machine interfaces. Piezoelectric flexible strain sensors are key components of wearable devices. However, existing piezoelectric flexible strain sensors have certain limitations in weak signal monitoring due to their large modulus and low sensitivity. To solve this problem, the concept of Kirigami (paper-cutting) was introduced in this study to design the sensor structure. By comparing the Kirigami structures of different basic structures, the serpentine structure was determined as the basic configuration of the sensor. The serpentine structure not only provides excellent tensile properties, but also significantly improves the sensitivity of the sensor, which performs well in monitoring weak signals. On this basis, the adhesion properties of the flexible sensor were analyzed and tested, and the optimal ratio of the substrate was selected for preparation. In addition, a low-cost and rapid prototyping process for stretchable patches was established in this study. Using this technology, we prepared the sensor device and tested its performance. Finally, we successfully developed a flexible sensor with a sensitivity of 0.128 mV/με and verified its feasibility for wrist joint motion monitoring applications. This result opens up new avenues for the recovery care of tenosynovitis patients after surgery.

Key words: flexible sensor; piezoelectric film; motion monitoring; wearable sensor

0 Introduction

Nowadays, millions of people around the world suffer from joint inflammation[1]. Traditional joint treatment methods use heavy and expensive equipment such as limb protractors[2], optical motion capture systems, and X-ray films. Moreover, the assistance of professional medical personnel is required. However, faced with limited medical resources and the patient's own mobility difficulties, it is still a huge challenge to conduct simple and effective evaluation of rehabilitation treatment. In recent years, flexible and stretchable electronics have developed rapidly in the fields of activity monitoring, medical rehabilitation and remote communication, providing humans with a healthy and convenient lifestyle[3-6]. Among them, flexible strain sensors have drawn great attention due to their ability of monitoring human breathing, pulse beats, blood pressure, and physical motion[7], which provide reliable treatment basis and early warning methods for patients with joint damage.

Flexible strain sensors can convert external stimulation into electrical signals, the sensor is mainly divided into resistive strain sensor, capacitive strain sensor, and piezoelectric strain sensor. The resistance of resistive strain sensors will change under the action of force since slippage or breakage between conductive materials will arise. However, for resistive strain sensor, smaller strain change causes slight change in resistance value. Moreover, the temperature compensation is required when ambient temperature changes. The capacitance of capacitive sensor will change under external pressure. It can be easily affected by the external electromagnetic environment. Piezoelectric sensors have been widely used owing to their advantages of wide frequency band, high sensitivity, high signal-to-noise ratio, simple structure, reliable operation and light weight. With the piezoelectric effect, a strain sensor based on a single ZnO nanowire was fabricated. It was reported that the gauge factor is 200 times higher than that of a commercial strain sensor. However, It can only withstand small strains[8]. Although researchers have made a lot of efforts in the development of ceramic-based flexible piezoelectric sensors, their inherent hard and brittle properties make them unsuitable for use in health monitoring. A key challenge in making stretchable piezoelectric devices is the fabrication process. An annealing process is required for most of ceramic-based piezoelectric materials[9]. In addition, directly depositing and patterning brittle solid piezoelectric materials onto flexible substrates is challenging. The more complex the pattern of the device, the higher the production cost. Moreover, the photolithography-based fabrication process for these devices is complex, costly, and time-consuming.

Compared to solid piezoelectric materials, piezoelectric polymers such as polyvinylidene fluoride (PVDF), have mechanical strength, and biocompatibility. In addition, they have lower manufacturing and engineering requirements than solid piezoelectric materials. Despite their low piezoelectric coefficients, they can be used in human medical applications that require high sensitivity and low power. A key challenge in manufacturing stretchable piezoelectric devices, such as strain sensors, pressure sensors, and energy harvesters[10,11], is the manufacturing process. Since piezoelectric polymers can be solution processed, various manufacturing techniques such as spin coating[12], electrospinning[13], and inkjet printing can be applied, which makes devices based on piezoelectric polymers with high mechanical strength possible. For example, researchers invented a skin-conformal piezoelectric generator made of PVDF copolymer. Polyvinylidene fluoride trifluoroethylene (PVDF-TrFE) was cast on a composite of carbon nanotubes (CNTs), which was then covered with graphene nanosheets. The device has a stretchability of up to 30%, which is comparable to that of human skin[13]. However, the modulus of straight PVDF is relatively large (~3.6 GPa), and its application in human health monitoring may result in poor wearing comfort or even monitoring failure. To solving this problem, we proposed the cut-and-paste technique. Combined with the concept of Kirigami technology, a stretchable strain sensor with ultrasensitive and ultra-low modulus was developed. In addition, good monitoring results were obtained by attaching the sensor to wrist joint, indicating that through novel structural features, it is possible for PVDF to achieve truly bio-integrated, self-powered devices for bio-integrated applications[15-19]. This will pave the way for providing personalized telemedicine rehabilitation services.

1 Principle and design

1.1 Piezoelectric principle

Piezoelectric strain sensors have fast response speed, low hysteresis, and excellent stability[20-23]. The response characteristics of piezoelectric strain sensors are mainly derived from the piezoelectric effect. When a piezoelectric material is subjected to a certain pressure, polarization occurs inside it, resulting in opposite charges on its two opposite surfaces. The piezoelectric constitutive equation is usually used to describe the coupling relationship between the forces applied to the sensor and electrical signals[24].

$$\boldsymbol{D}=\boldsymbol{\varepsilon}^{\mathrm{T}}\times\boldsymbol{E}+\boldsymbol{d}\cdot\boldsymbol{T},\quad \boldsymbol{S}=\boldsymbol{d}^{\mathrm{T}}\times\boldsymbol{E}+\boldsymbol{s}^{\mathrm{E}}\cdot\boldsymbol{T},\quad (1)$$

where the variables in the formula above are all in tensor form, $\boldsymbol{D}$ is the electric displacement vector, $\boldsymbol{E}$ is the electric field tensor, $\boldsymbol{T}$ is the stress tensor, $\boldsymbol{S}$ is the strain tensor, $\boldsymbol{d}$ is the piezoelectric strain tensor, $\boldsymbol{d}^{\mathrm{T}}$ is its transposed matrix, $\boldsymbol{\varepsilon}^{\mathrm{T}}$ is the dielectric constant tensor under constant stress, and $\boldsymbol{s}^{\mathrm{E}}$ is the compliance tensor under constant electric field.

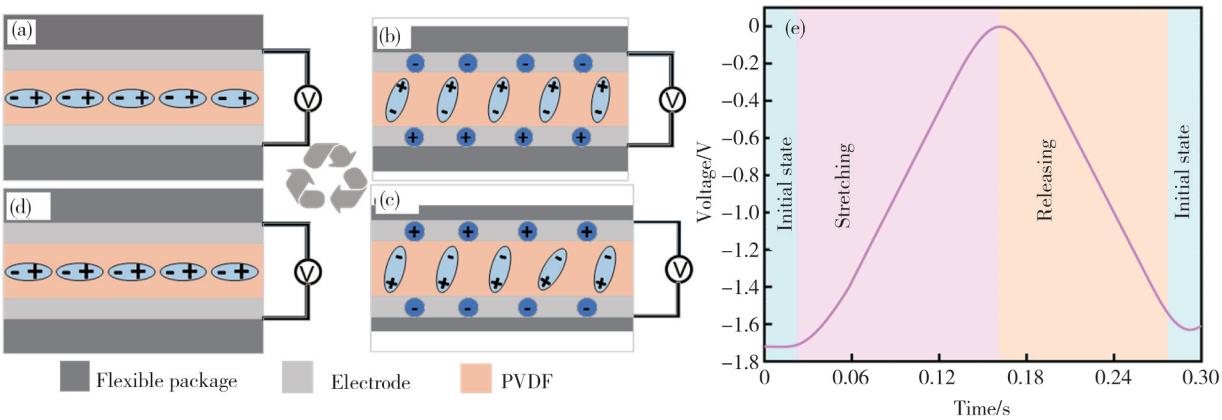

Fig. 1 Schematic diagram of charge generated by stretchable piezoelectric sensor during the cyclic stretching. (a–b) The strain sensor is in the stretching stage; (c) The sensor is stretched to the maximum strain; (d) The sensor is released; (e) The output voltage signal during the stretching process

When strain is transferred from the wrist joint to the strain sensor, the strain-induced charge signal can be obtained from the flexible piezoelectric strain sensor. Fig.1 shows the change in surface charge generated by the sensor under cyclic stretching. When the flexible piezoelectric strain sensor is stretched, the induced charge on the top electrode decreases, and the current flows from the top electrode to the bottom electrode, and the voltage begins to increase (Fig. 1(a) & Fig. 1(b)). When the sensor is stretched to the maximum strain, the current in this circuit is zero and the voltage is maximum (Fig.1(c)). On the contrary, when the sensor is released, the voltage decreases (Fig.1(d)).

1.2 Design of stretchable sensor

Since the skin is flexible and easily deformable, stretchable piezoelectric strain sensors should be flexible enough to facilitate the transfer of micro-strain of the skin to the sensor. However, the PVDF film has a large modulus and cannot meet the requirements of wearable devices for comfort and softness. As a traditional paper-cutting art, Kirigami has developed into a manufacturing framework that can give hard and difficult-to-stretch functional materials higher stretchability and bendability. The design concept of the Kirigami provides a platform for heterogeneous integration of electronic devices with different functions. This solution has paved the way for the construction of the next generation of flexible metamaterials, physiological signal monitoring sensors and intelligent soft robots. Therefore, in order to construct sensors with less restrictions on biological tissues, we proposed to use the Kirigami concept to design the stretchable sensor. Rectangular, circular, and serpentine structures were used as the basic units of Kirigami structures. To avoid the sensor's performance degradation due to excessive local stress during the stretching, the local strain of devices with different basic structures was compared under the same overall strain.

Fig.2 shows finite element analysis (FEA) results of local strain for three different structures (rectangular, circular, and serpentine).

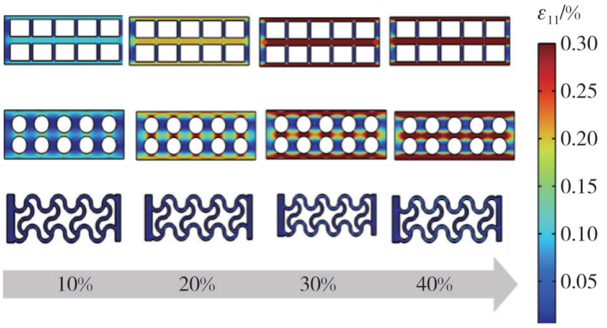

Fig. 2 FEA results of local strain with three different sensors (rectangular, circular, and serpentine)

In order to ensure the reliability of the results, the overall size and duty cycle of the three different structures are the same. It can be concluded that under the same overall strain, the local strain of sensor with serpentine structure is the smallest. Therefore, the sensor with a serpentine structure is chosen to ensure that the device will not be damaged due to stress concentration during the monitoring process.

2 Experiment

2.1 Adhesion performance test of substrate

It is important to select a suitable elastomer substrate for flexible sensors. One reason is that substrate can isolate the interference of pollutants in the environment and increase the durability of the sensor. Furthermore, flexible substrate can prevent the functional layer from direct contact with the skin, avoiding leakage current. Polydimethylsiloxane (PDMS) has the characteristics of good mechanical flexibility, stretchability and biocompatibility. Here, it was selected as the substrate of the stretchable piezoelectric strain sensor.

As a wearable sensor, the device should maintain good adhesion to the skin. In order to explore the effect of different mixing ratios (A:B=5:1, 10:1, and 15:1, where A is poly (dimethyl-methylvinylsiloxane) and B is poly (dimethyl-methylhydrogenosiloxane)) on the adhesion performance of the device and test the peeling force of the device, we adopted the standard 90-degree peeling test method to measure the peeling force of the prepared elastomer sample (14 mm×10 mm×0.1 mm) attached to the human forearm. The top surface of the elastomer is attached to a thin polyimide (100 μm thick) to prevent the elastomer from stretching along the peeling direction, as shown in Fig.3(a).

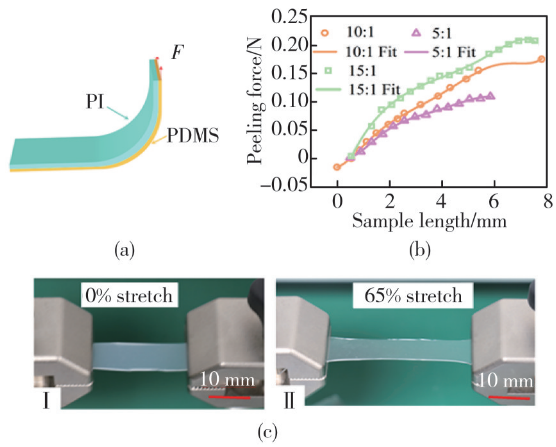

Fig. 3 Device adhesion test. (a) Schematic diagram of peeling force test; (b) PDMS materials with mixing ratios of 5:1, 10:1, and 15:1; (c) Optical images of PDMS encapsulation material with a mixing ratio of 15:1 at 0% (I) and 65% (II) strain

The sample to be tested was laminated to the forearm, and a tensile tester (ESM303, Mark-10) was used to clamp one end of the sample, pull and peel, and record the peeling force. The peeling force curve with different peeling displacements can be obtained. It is worth mentioning that the peeling energy can be determined by integrating the peeling force over the sample length.

The PDMS with mixing ratios of 5 :1, 10 :1, and 15 :1 was selected for peeling experiments, and the corresponding moduli of PDMS films are listed in Table 1.

Table 1 Moduli of PDMS films of various proportions

Materials	Modulus/MPa
PDMS 5:1	1.64
PDMS 10:1	0.97
PDMS 15:1	0.31

Fig. 3 (b) shows the relationship between the peeling force and displacement of PDMS with different mixing ratios (5: 1, 10: 1, and 15: 1). Under the same sample length, the peeling force required for PDMS with a mixing ratio of 15 :1 is the largest, and the peeling force required for PDMS with a mixing ratio of 10 : 1 is the second largest. The integral of the peeling force curve on the horizontal axis in Fig. 3 (b) represents the peeling energy of the test sample. It can be seen that the peeling energy of the PDMS encapsulation material with a mixing ratio of 15 :1 is the largest. PDMS with a mixing ratio of 15 : 1 was selected as the packaging material of the sensor. Fig. 3 (c) shows the optical images of the PDMS film with a mixing ratio of 15 : 1 under 0% and 65% stretching states. It can be seen that PDMS does not break under 65% stretching state and can be used as a packaging material for wearable sensors.

2.2 Processing of sensor

We cut the PVDF film with 70 nm thick Cu electrodes on both sides into 4 cm×3 cm films for later use (Fig.4 (a)). Since the piezoelectric properties of the PVDF film are easily affected by temperature, thermal processing (such as infrared laser processing) is avoided and machining is used to process the PVDF film. During the machining process, PVDF film needs to be adhered to a temporary base. Ultraviolet (UV) film can be degummed well after 2 min of UV lamp irradiation, Therefore, it was selected as the temporary substrate during PVDF film processing. Then, the PVDF film was patterned using a engraving machine (Yitu pattern engraving machine), and the entire cutting process could be completed within 5 min (Fig.4(b)). After the functional layer was cut, functional material together with the UV substrate was irradiated with a UV lamp for 2 min (Fig.4 (c)).

To ensure that the electrical signal generated by PVDF can be stably transmitted to the data acquisition card and signal processing circuit during the monitoring process, 10 mm×4 mm Cu strips were used as the top and bottom electrode leads of the sensor (Fig.4 (d, e)). Firstly, we mixed the precursors A and B of PDMS at a mass ratio of 15 :1, and used a magnetic stirrer to stir the mixture for 5 min to ensure that the two precursors is mixed evenly. Then, we put the fully stirred mixed solution into a vacuum pump and evacuate it for 5 min. The vacuum pump pressure was maintained at 10^{-5} Pa and the valve of the vacuum pump was slowly opened to fully remove the bubbles in the PDMS solution. Afterwards, we poured the PDMS mixture into the Petri dish, and then let the Petri dish stand in a vacuum environment for 15 min to ensure that the PDMS solution was spread flat on the functional layer. Finally, the colloidal PDMS was baked in a vacuum oven at a temperature of 50 ℃ for 24 h to completely solidify the PDMS and the sensor was processed (Fig.4 (f)).

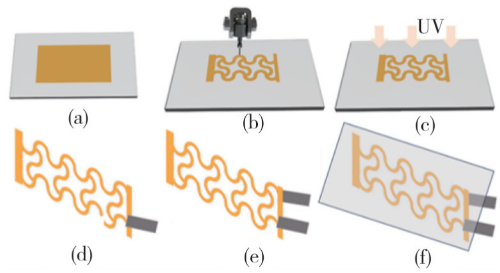

Fig. 4 Processing of flexible piezoelectric strain sensor. (a) PVDF film; (b) Patterned film; (c) Irradiated film; (d) Cu strip as top electrode lead; (e) Cu strips as top and bottom electrode leads; (f) Solidified PDMS.

Fig.5 shows the images of the device at 0% and 25% stretching states after processing. It can be seen that the device remains intact at 25% stretching state.

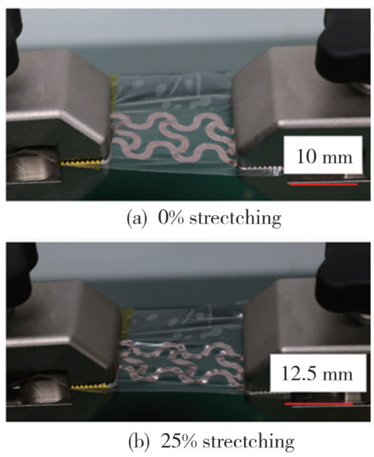

Fig. 5 Images of the sensor under 0% and 25% stretching

2.3 Sensor's performance test

In practical applications, flexible sensors are required to have good electrical output characteristics, so that they have good response and sensitivity during the sensing process and ensure reliable signal capture. In order to study the electrical output characteristics of the serpentine sensor, when the sensor was uniaxially stretched, the top electrode and the bottom electrode were connected to a data acquisition device (DAQ, NI—6225, National Instruments) to measure the output voltage in the thickness direction of the PVDF film. Thus, the output voltage of the device under different displacements can be obtained. The relationship between the displacement and stretching force of the sensor is shown in Fig.6 (a). Fig.6 (b) shows the output voltage of the serpentine - shaped structure under 3 mm tensile displacement. Fig. 6 (c) shows the output voltage of the sensor after 760 uniaxial stretch-release cycles when the maximum stretching displacement of the device is 7.5 mm, indicating that the output voltage of the patch remains stable during the stretching cycle, which confirms the robustness of the stretchable patch. The output voltage - strain curves under different strains are plotted as shown in Fig.6 (d). From the slope of the curve in Fig.6(d), it can be seen that the strain sensitivity of the sensor is 0.128 mV/με.

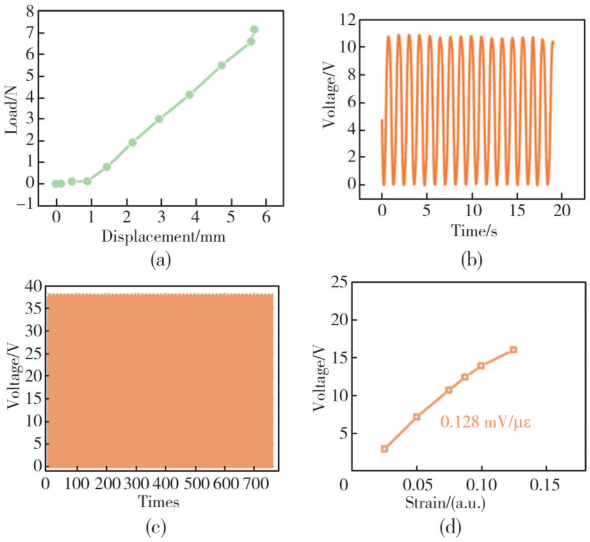

Fig. 6 Test results of sensor's performance. (a) Relationship between force and displacement; (b) Output voltage at 3 mm displacement; (c) Output voltage after 760 cycles of stretching at 7. 5 mm displacement; (d) Voltage-strain curve.

2.4 Wrist motion test

In order to measure the wrist joint movement, the sensor was attached to the outside of the wrist, as shown in Fig.7 (a) and (b). When the wrist moves in one direction, the sensor is in a stretch-relax cycle. When the wrist moves in two directions, the sensor is in a stretch-compression cycle. The corresponding voltage output is shown in Fig. 7 (c) and (d). It can be seen from that the output voltage remains stable during multiple measurements, proving that the developed device has the ability to measure wrist joint movement.

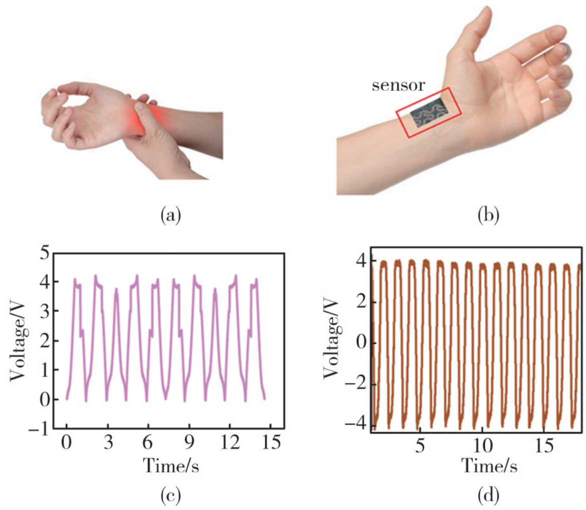

Fig. 7 Wrist motion test. (a–b) Schematic diagram of the sensor attached to the wrist joint; (c) The output voltage of the device when the wrist joint moves in one direction; (d) The output voltage of the sensor when the wrist joint moves in two directions.

3 Conclusions

In summary, we proposed a concept for preparing flexible piezoelectric strain sensors. The bending strain sensor adopts the Kirigami structure and the preparation process adopts a simple cut-and-paste technique. Through a simple and low-cost manufacturing method, the stretchable strain sensor achieves high sensitivity. When the sensor is attached to the wrist, it can well distinguish between unidirectional and bidirectional motions, showing its great application potential in intelligent wearable devices for human-computer interaction. In future research, the pressure of the sensor on the skin should be studied to achieve the purpose of non-sensing application. In addition, a fully flexible signal conditioning circuit should be used to enhance the wearing comfort of the patient.

Acknowledgement

This work was supported by National Natural Science Foundation of China (Nos. 62301509, 62304209), Key Research and Development Program of Shanxi Province

(No. 202302030201001), and Fundamental Research Program of Shanxi Province (Nos. 202203021222079, 0210302123203, 202103021223185).

Declaration of conflicting interests

The authors have no conflict of interests related to this publication.

References

[1] CHOY E H, PANAYI G S. Cytokine pathways and joint inflammation in rheumatoid arthritis. The New England Journal of Medicine, 2001, 344(12): 907-916.

[2] LIEBERTHAL J, SAMBAMURTHY N, SCANZELLO C R. Inflammation in joint injury and post-traumatic osteoarthritis. Osteoarthritis and Cartilage, 2015, 23(11): 1825-1834.

[3] LUO Y F, ABIDIAN M R, AHN J H, et al. Technology roadmap for flexible sensors. ACS Nano, 2023, 17(6): 5211-5295.

[4] SASSANELLI C, ARRIGA T, ZANIN S, et al. Industry 4.0 driven result-oriented PSS: an assessment in the energy management. International Journal of Energy Economics and Policy, 2022, 12(4): 186-203.

[5] MISHRA T, WANG M, METWALLY A A, et al. Pre-symptomatic detection of COVID-19 from smartwatch data. Nature Biomedical Engineering, 2020, 4(12): 1208-1220.

[6] LUKAS H, XU C H, YU Y, et al. Emerging telemedicine tools for remote COVID-19 diagnosis, monitoring, and management. ACS Nano, 2020, 14(12): 16180-16193.

[7] MA Z, CHEN P, CHENG W, et al. Highly sensitive, printable nanostructured conductive polymer wireless sensor for food spoilage detection. Nano Letters, 2018, 18(7): 4570-4575.

[8] ZHAO H, LIU X J, CHEN L Z, et al. Fabrication of ultra-high-sensitivity flexible strain sensor based on single ZnO nanowire. Microsystem Technologies, 2017, 23(6): 1703-1707.

[9] KIM B H, LI K, KIM J T, et al. Three-dimensional electronic microfliers inspired by wind-dispersed seeds. Nature, 2021, 597(7877): 503-510.

[10] KIM D H, XIAO J L, SONG J Z, et al. ChemInform abstract: stretchable, curvilinear electronics based on inorganic materials. ChemInform, 2010, 41(30): 1-17.

[11] ROGERS J A. Semiconductor devices inspired by and integrated with biology//2012 IEEE 25th International Conference on Micro Electro Mechanical Systems, January 29-February 2, 2012, Paris, France. Piscataway, N. J.: IEEE, 2012: 51-55.

[12] TYONA M D. A theoritical study on spin coating technique. Advances in Materials Research, 2013, 2(4): 195-208.

[13] TEO W E, RAMAKRISHNA S. A review on electrospinning design and nanofibre assemblies. Nanotechnology, 2006, 17(14): R89-R106.

[14] CHO K Y, PARK H, KIM H J, et al. Highly enhanced electromechanical properties of PVDF-TrFE/SWCNT nanocomposites using an efficient polymer compatibilizer. Composites Science and Technology, 2018, 157: 21-29.

[15] WISE K D, ANDERSON D J, HETKE J F, et al. Wireless implantable microsystems: high-density electronic interfaces to the nervous system. Proceedings of the IEEE, 2004, 92(1): 76-97.

[16] WANG C H, LI X S, HU H J, et al. Monitoring of the central blood pressure waveform via a conformal ultrasonic device. Nature Biomedical Engineering, 2018, 2(9): 687-695.

[17] SUN H Y, YE C, ZHAO G, et al. Ultrasensitive micro/nanocrack-based graphene nanowall strain sensors derived from the substrate′s Poisson′s ratio effect. Journal of Materials Chemistry A, 2020, 8(20): 10310-10317.

[18] ZHANG Q X, YU A B, GUO L H, et al. RF MEMS switch integrated on printed circuit board with metallic membrane first sequence and transferring. IEEE Electron Device Letters, 2006, 27(7): 552-554.

[19] WU J F, FEDDER G K, CARLEY L R. A low-noise low-offset capacitive sensing amplifier for a 50 μg/rtHz monolithic CMOS MEMS accelerometer. IEEE Journal of Solid-State Circuits, 2004, 39(5): 722-730.

[20] WANG Z Y, WANG C, LIU L T. Design and analysis of a PZT-based micromachined acoustic sensor with increased sensitivity. IEEE Transactions on Ultrasonics, Ferroelectrics, and Frequency Control, 2005, 52(10): 1840-1850.

[21] POLCAWICH R G, SCANLON M, PULSKAMP J, et al. Design and fabrication of a lead zirconate titanate (PZT) thin film acoustic sensor. Integrated Ferroelectrics, 2003, 54(1): 595-606.

[22] DEFAŸ E, MILLON C, MALHAIRE C, et al. PZT thin films integration for the realisation of a high sensitivity pressure microsensor based on a vibrating membrane. Sensors and Actuators A: Physical, 2002, 99(1/2): 64-67.

[23] SCHEEPER P, GULLOV J O, MUNCH KOFOED L. A piezoelectric triaxial accelerometer. Journal of Micromechanics and Microengineering, 1996, 6(1): 131-133.

[24] STARR M B, WANG X D. Coupling of piezoelectric effect with electrochemical processes. Nano Energy, 2015, 14: 296-311.

用于关节运动监测的超灵敏可伸缩贴片

李腾腾[1,2], 李子唯[1,2], 郝亚峰[1,2], 武慧嘉[1,2], 朱　璞[1,2], 马富鹏[1,2], 李丰超[1,2], 佘建刚[1,2], 刘美宏[1,2*], 雷　程[1,2*], 梁　庭[1,2]

1. 中北大学 半导体与物理学院, 山西 太原 030051;

2. 中北大学 动态测试技术国家重点实验室, 山西 太原 030051

摘　要: 可穿戴设备在新一代智能便携电子领域,特别是在医疗监测、软体机器人、人工智能以及人机交互等领域展现出巨大的应用潜力。压电柔性应变传感器是可穿戴设备的关键部件,但现有压电柔性应变传感器由于模量大、灵敏度低,在微弱信号监测方面具有一定的局限性。为解决这一问题,引入了Kirigami(剪纸)概念来设计传感器结构。通过对不同基本结构进行比较,最终确定传感器结构为蛇形结构。蛇形结构不仅提供了出色的拉伸性能,还显著提升了传感器的灵敏度,在监测微弱信号方面表现出色。在此基础上,进一步分析和测试了柔性传感器的粘附性能,并选择了最优的基底材料比例进行制备。此外,还建立了一种低成本、快速的可拉伸贴片制备工艺,并利用这一技术,制备了传感器设备并对其性能进行测试。最终,成功开发出一种灵敏度为0.128 mV/με的柔性传感器,并验证了其在手腕关节动作监测应用中的可行性。这一成果为腱鞘炎患者手术后的恢复护理开辟了新的道路。

关键词: 柔性传感器; 压电薄膜; 运动监测; 可穿戴传感器

引用格式: LI Tengteng, LI Ziwei, HAO Yafeng, et al. Ultrasensitive stretchable patches for joint motion monitoring. Journal of Measurement Science and Instrumentation, 2024, 15(3): 285-291.