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Highly sensitive and wearable capacitive pressure sensors based on PVDF/$BaTiO_3$ composite fibers on PDMS microcylindrical structures

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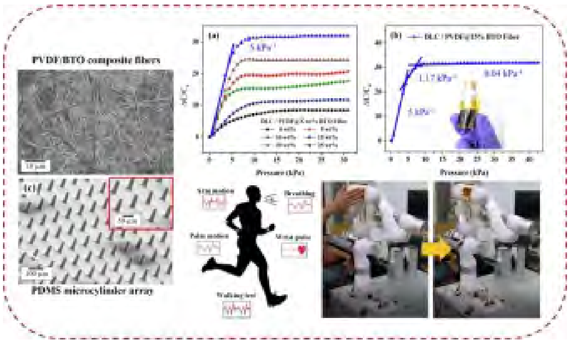
Abstract

This study developed a novel high-sensitivity flexible capacitive pressure sensor by combining $BaTiO_3$-doped polyvinylidene fluoride electrospinning fibers and polydimethylsiloxane microcylindrical structures as the dielectric layer. The flexible electrode formed by the graphene/PI film was assembled into a flexible capacitive pressure sensor, and formed a sandwich-like structure. The proposed sensor could obtain more capacitance variations and improve its sensitivity through composite deformation of electrospinning fiber and microcylindrical structure under pressure. The developed flexible capacitive pressure sensor has a high sensitivity of 5 kPa^{-1}, fast response and release time of 25 and 50 ms, ultralow detection limit of 0.11 Pa, and more than 10000- and 5000-times compressions/bending cycling test without any signal attenuation for the high durability and high reliability. The results of this study proved that the sensors have excellent performance, and can be applied on wearable devices for human pulse monitoring and acoustic detection.

Graphical abstract



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Introduction

With the development of information technology, the requirements for sensor performance have been increasing. In recent years, flexible wearable sensors have been widely applied to the fields of electronic skin, human-machine interface, soft robotics, healthcare monitoring, and biomedical diagnosis [1], [2], [3], [4], [5], [6], [7], [8], [9], [10]. Many studies have developed various flexible pressure sensors, including piezoresistive [11], [12], [13], [14], piezoelectric [15], [16], [17], capacitive [18], [19], [20], [21], [22], and triboelectric [23], [24], [25], based on different physical conversion mechanisms. Among them, capacitive pressure sensors, commonly consisting of top/bottom electrodes and dielectric layers, have become one of the most attractive flexible pressure sensors due to the features of simple structure, low power consumption, high reliability and durability, negligible temperature drift, and fast response.

However, the low sensitivity and narrow detection range still limit the overall performance and further applications of flexible capacitive pressure sensors. Therefore, to improve sensor performance, the common method is to fabricate three-dimensional structures on electrodes or dielectric layers, in order to improve compressibility. To improve the performance of capacitive pressure sensors, several single-sided or double-sided identical microstructures have been developed, such as microbumps [26], [27], micropyramids [28], [29], microcylinders [30], microwaves [31], porous structures [32], and biomimetic structures [33], [34]. These proposed microstructures have been proven to be effective in improving the sensitivity of capacitive pressure sensors. Nevertheless, the operating pressure range mentioned above in studies using single microstructures is often limited to below 2 kPa because these structures collapse rapidly even at low pressures. The results make this sensor unsuitable for most applications, such as joint movements in high-pressure situations and electronic skins of robotic arms. Some studies used composite structures to improve the sensitivity and operating pressure range of capacitive pressure sensors [35], [36], [37]. For example, Yang et al. [38] prepared microholes in the micropyramid structure, and greatly improved the sensitivity of the sensor and the rigidity of the pyramid structure. Ko et al. [39] integrated a carbonized cotton fabric electrode with a dielectric layer with an inclined hole structure on the capacitive pressure sensor. Compared with the vertical structure, the inclined hole structure has higher flexibility and a wider detection range of 1 MPa. Wu et al. [40] prepared poly-acrylamide (PAM)/*N*-isopropyl di-acrylamide (BIS)/graphene oxide (GO) as a nanocomposite hydrogel dielectric layer. It has ultra-high deformability and could greatly improve the sensitivity of the capacitive pressure sensor.

Although the above-mentioned studies have improved the sensitivity and effective pressure range of the capacitive pressure sensor, they still use more complex fabrication techniques or highly polluting etching processes. These shortcomings further result in higher costs and environmental hazards for the fabricated sensor, which is not conducive to the popularization of flexible wearable devices. Therefore, this study proposes a novel fabricating method of a dielectric layer for the capacitive pressure sensor that can improve the sensor sensitivity and working pressure range and can be produced in large quantities. By adding $BaTiO_3$ (BTO) to polyvinylidene fluoride (PVDF) for electrospinning, the BTO doped PVDF fibers were integrated on casted polydimethylsiloxane (PDMS) with double-sided microcylindrical structures to form a double-sided dielectric layer. The fiber layer with a fluffy structure formed by the air gap becomes a new type of double-sided dual-structure dielectric layer through the support of a microcylinder. Compared to a single-structure dielectric layer, the two heterostructures cause the synergistic effect, which is complementary to each other's shortcomings. When the sensor is pressed, the fiber layer can expel air from the pores, and even slight pressure can cause changes in the electrode spacing. As the pressure is increased, the cylindrical structure sinks into the fiber layer and undergoes a large pressure to produce bending deformation. After the pressure is released, the fiber layer thinned by the pressure is quickly supported. Hence, the sensors can have a larger distance variation and faster rebound time, increase the sensitivity of the sensor, and reduce the response time. The process is simple, fast, large-area, and cost-effective, achieved by electrospinning and casting methods. Furthermore, a high-performance flexible capacitive pressure sensor can be completed without complex and expensive equipment. The sensor developed in this study has a high sensitivity of 5.0 kPa^{-1}, a wide detection range of 0–5 kPa, a fast response time of 25–50 ms, an ultra-low detection limit of 0.11 Pa, a load/unload of more than 10,000 times, and bent/unbent cycle life of more than 5,000 times without any signal degradation during the test. In addition, practical tests were conducted on the sensors, including pulse monitoring, acoustic vibration detection, respiration monitoring, and real-time testing of body movement monitoring. The sensor is also successfully integrated into the robotic arm as electronic skin, and the robotic arm can perform the function of safety protection through self-shutdown by touch sensing. This study contributes to the applications of different fields, including wearable devices, biosensors, human-machine collaboration, etc., thereby improving the microstructure dielectric layer and enhancing the sensor performance.

Section snippets

Fabrication of flexible polyimide (PI)/graphene electrodes

The PI film has the advantages of high insulation and high flexibility; therefore, it is suitable as a substrate for flexible sensors. In this study, graphene was coated on top to form a conductive film, and was used as the electrode substrate of the sensor. The process is detailed as follows. First, 2 wt% polydimethyldiallylammonium chloride solution was used as a graphene dispersant, and then 2 wt% multilayer reduced oxide graphene was added to form a graphene slurry, which was stirred at

Sensing mechanism of the capacitive pressure sensors

Fig. S2 is the schematic diagram of the sensing mechanism of the capacitive pressure sensors with (a) pure fiber dielectric layer, (b) pure microcylindrical dielectric layer, and (c) single-sided and (d) double-sided dielectric layer with fiber and microcylinder when applying and releasing pressure. During the applying pressure, the fiber structure has a large number of holes, as shown in Fig. S2(a). Even under small pressure, the air can be squeezed out so that the capacitance value of sensors

Conclusion

In this study, the electrospun fibers doped with BTO and PVDF were developed and integrated with PDMS double-sided microcylinders as a double-layered dielectric layer. The flexible electrode formed by the graphene/PI film was assembled into a flexible capacitive pressure sensor by a sandwich-like structure. Using the higher dielectric constant of BTO and PVDF formed the fiber structure, a special fluffy layer produced an air gap and was supported by the PDMS microcylinder array structure. These

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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