

Flexible Porous CNT/PDMS-PVDF Sensor with Hybrid Triboelectric-Piezoelectric Sensing

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Abstract. This study presents the development of a flexible porous CNT/PDMS-PVDF hybrid sensor (PCPS) that integrates triboelectric and piezoelectric sensing mechanisms for enhanced performance. The sensor is fabricated using a lightweight and porous CNT/PDMS composite film, combined with a PVDF layer, to achieve high sensitivity and flexibility. The porous structure is created by incorporating NaCl particles during the PDMS curing process, which evaporate to form irregular pores, increasing the contact area and improving the sensor's mechanical and electrical properties. The hybrid sensor leverages both triboelectric and piezoelectric effects, resulting in a significant increase in output voltage compared to traditional PDMS-based sensors. Experimental results demonstrate that the porous PVDF/CNT/PDMS hybrid film exhibits a 280% increase in voltage output under applied pressure, highlighting its superior performance. The sensor's ability to convert mechanical energy into electrical signals without an external power supply makes it a promising candidate for applications in wearable electronics, tactile sensing, and energy harvesting. This work provides a novel approach to designing high-performance flexible sensors with hybrid sensing capabilities.

Keywords: Flexible Sensor; Hybrid sensor; Porous CNT/PDMS; Piezoelectric Effect; Triboelectric Effect.

1. Introduction

Flexible sensors have garnered significant attention due to their potential in wearable electronics, soft robotics, and human-machine interfaces [1]. Among these, piezoelectric and triboelectric sensors are particularly promising, as they convert mechanical energy into electrical signals without external power sources, making them ideal for lightweight, flexible, and energy-efficient applications [2], [3]. Polydimethylsiloxane (PDMS) is widely used in such sensors due to its flexibility, biocompatibility, and ease of processing. However, PDMS alone often exhibits limited triboelectric performance, restricting its effectiveness in practical applications [4], [5].

Despite progress in flexible sensor development, challenges remain. Conventional piezoelectric and triboelectric sensors often suffer from structural complexity [6], low signal output, limited sensitivity, and inadequate mechanical properties, hindering their real-world deployment [7]. For instance, PDMS-based sensors, while flexible, typically exhibit poor conductivity and weak piezoelectric response, limiting their performance [8]. To address these limitations, researchers have integrated carbon nanotubes (CNTs) into PDMS composites to enhance conductivity. However, these CNT/PDMS composites often lack sufficient porosity, which is crucial for maximizing the contact area needed to enhance triboelectric effects [9], [10].

Combining triboelectric and piezoelectric mechanisms into hybrid devices has emerged as a promising approach. This hybrid configuration enables simultaneous surface electrification and internal polarization, synergistically enhancing sensor performance [6], [11]. Nevertheless,

achieving flexible hybrid sensors that balance structural integrity, mechanical robustness, and high electrical performance under dynamic conditions remains challenging [12]. Single-mechanism sensors often suffer from limited sensitivity and adaptability, while hybrid systems must address complexities such as material compatibility, fabrication processes, and performance optimization under varying mechanical stresses [12], [13].

Recent advancements in hybrid nanogenerators have demonstrated the benefits of combining multiple energy harvesting mechanisms. For example, Wang et al. developed a flexible triboelectric-piezoelectric hybrid nanogenerator using P(VDF-TrFE) nanofibers and PDMS/MWCNT, achieving high output power [11]. Similarly, Fan et al. created an ultrathin, rollable paper-based TENG for acoustic energy harvesting, showcasing its potential for self-powered sound recording and wearable applications [14].

Building on these advancements, this study proposes a novel flexible porous CNT/PDMS-PVDF hybrid sensor (PCPS) that integrates triboelectric and piezoelectric mechanisms. The porous structure, created by incorporating NaCl particles during PDMS curing, increases the contact area, enhancing sensitivity. The evaporation of NaCl forms irregular pores, improving mechanical and electrical properties. Additionally, the PVDF layer amplifies electrical signal generation during mechanical deformation, addressing the limitations of single-mechanism sensors.

The PCPS converts mechanical energy into electrical signals without external power, making it suitable for wearable electronics, tactile sensing, and energy harvesting. Its robust, flexible structure, enabled by precise material ratios and fabrication processes, supports adaptability in dynamic environments. This study advances flexible sensor technology, emphasizing the importance of structural engineering and hybrid mechanisms for next-generation sensor development [15], [16], [17], [18].

2. Design and Analysis

2.1 Porous CNT/PDMS Hybrid Film Preparation

Lightweight and flexible CNT/PDMS hybrid films were synthesized by precisely controlling material ratios and optimizing the fabrication protocol. Initially, a PDMS stock solution (Dow Corning 184 Silicone Elastomer, 5 g) and curing agent (0.5 g) were mixed in a 10:1 mass ratio, combined with multilayer carbon nanotube (CNT) powder (0.05 g). To achieve uniform CNT dispersion within the PDMS matrix, ultrasonication was performed for 30 minutes. Subsequently, NaCl solution (0.2 g) was added, and the mixture was magnetically stirred for 20 minutes before introducing the curing agent. Stirring continued for an additional 30 minutes to ensure homogeneity.

The well-mixed solution was transferred to a 3D-printed rectangular mold (30 mm × 15 mm × 3 mm) and thermally cured in an oven at 65 °C for 5 hours. The resulting CNT/PDMS film weighed approximately 0.92 g, exhibiting a porous microstructure attributed to concentration gradients caused by NaCl solution evaporation during curing. This process created irregular pores and surface depressions, as illustrated in Fig. 1(a). Comparative analysis indicated that a 0.25 g NaCl particle content provided optimal pore volume, balanced morphology, and enhanced mechanical properties, avoiding issues such as solid inclusions observed at higher concentrations, shown in Fig. 1(b).

2.2 Tactile Sensor Fabrication

The fabrication of the tactile sensor involved the use of copper foil as the electropositive triboelectric material for electrode creation. A 50 μm thick copper foil was immersed in deionized water and subjected to a hydrothermal oxidation reaction at 80 °C for 2 hours. This process facilitated the formation of irregular nanostructures on the copper surface, resulting from the interaction between the heated water and the oxide layer present on the copper foil. These nanostructures enhance the surface roughness, which is critical for improving the sensor's triboelectric performance.

The porous CNT/PDMS composite film was then integrated with the treated copper foil, with the pore-exposed side facing the copper surface. External copper wires were connected to the copper foil to serve as electrical leads. On the opposite side of the composite film, a polyvinylidene fluoride (PVDF) layer was placed, and the conductive, non-woven fabric was employed as the corresponding electrode. This configuration ensures efficient electrical contact and signal transduction. Finally, the entire sensor assembly was encapsulated using a commercially available Kapton film with a thickness of 80 μm , providing mechanical stability and environmental protection. Fig. 1(c) illustrates the detailed fabrication process of the hybrid sensor.

2.3 PCPS Working Principle

The hybrid sensor operates based on the dual-layer principle, combining triboelectricity and piezoelectricity to achieve high voltage output and rapid response [19]. In the piezoelectric mechanism, mechanical stress applied in the direction opposite to the material's polarization induces the separation of positive and negative charge centers within the piezoelectric PVDF layer, generating an electric dipole moment governed by the constitutive piezoelectric equation,

$$D = d \cdot T + \varepsilon \cdot E \quad (1)$$

Where D is the electric displacement, d is the piezoelectric strain coefficient, T is the applied mechanical stress, ε is the permittivity, and E is the resulting electric field [15].

This mechanism enables the PVDF layer to convert mechanical deformation into electrical signals efficiently [11]. Simultaneously, the triboelectric mechanism relies on contact-separation actions between the electropositive copper foil and the electronegative CNT/PDMS composite film. Upon contact, electrons transfer from the copper foil to the CNT/PDMS surface due to their difference in electron affinity, and upon separation, an electric potential is generated, driving a current through the external circuit. The amount of charge transferred is given by

$$Q = \sigma \cdot A \quad (2)$$

Q is the transferred charge, σ is the surface charge density, and A is the contact area—the effective contact area [20]. The porous microstructure of the CNT/PDMS film significantly increases the contact area, enhancing triboelectric charge generation, while the irregular nanostructures on the copper foil further improve charge transfer efficiency [15]. The synergistic combination of these mechanisms not only amplifies the overall voltage output but also ensures rapid response times, making the sensor highly suitable for dynamic environments such as wearable electronics and soft robotics [16]. Moreover, the self-powered operation of the sensor eliminates the need for external power sources, enhancing its practicality for real-world applications [17].

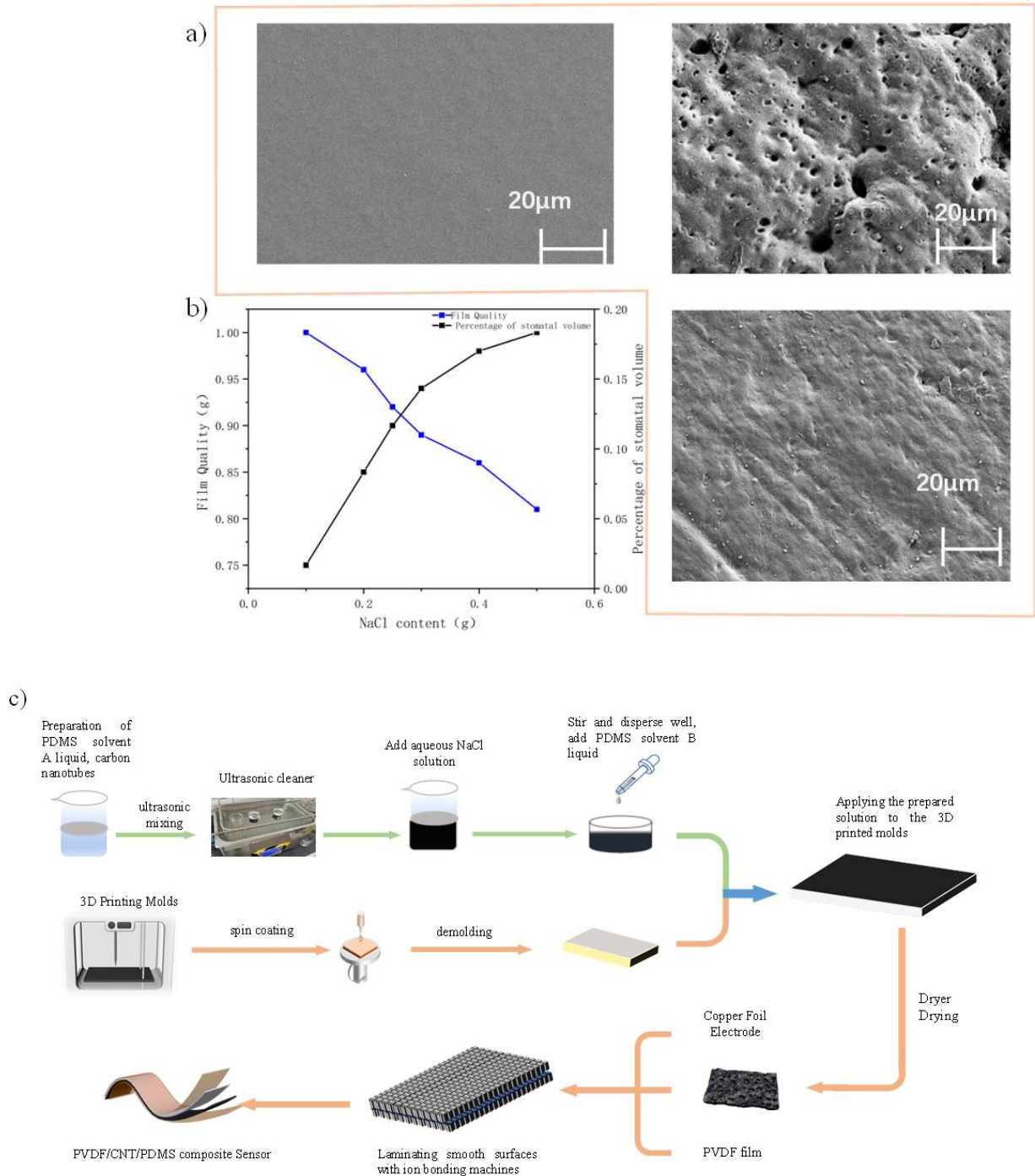


Figure 1 a) PDMS, CNT/PDMS, CNT/PDMS porous hybrid film optical microscope magnification, b) physical plots of sensitive film mass and stomatal volume occupancy at different mass ratios of NaCl particles to PDMS corresponding to different NaCl masses, c) PCPS fabrication process

3. Results and Discussion

3.1 PCPS Experimental Platform

The experimental setup for evaluating the PCPS (Porous CNT/PDMS-PVDF Hybrid Sensor) involved several key components and processes to ensure precise fabrication and accurate performance testing. Initially, the carbon nanotubes (CNTs) were uniformly dispersed within the

PDMS matrix using an ultrasonic cleaner (SCIENTZ, SB800 DTD). The CNT/PDMS film solution was then dried and air-cooled in an ultra-clean bench to prevent contamination and ensure consistent film quality. Molds for the CNT/PDMS films were fabricated using a 3D printer (FEM A81, IG Maker), and the resulting porous films were examined using a scanning optical microscope (ICX41, Sunnyoptical) to analyze their microstructure and pore distribution.

A comprehensive pressure testing platform was established to evaluate the performance of the hybrid sensor. The system included a pressure transducer (SBT641, Simbatouch) to measure the applied pressure, a vibration exciter (Modal25, MB Dynamics) to generate controlled mechanical stimuli, and a signal generator (HDG2102B, Hantek) to provide input signals for the exciter, enabling precise control over the magnitude and frequency of the applied pressure. The output signals from the PCPS were captured using a dynamic signal tester (DH8303, DongHua), which operated with an internal resistance of 60 M Ω and a sampling frequency of 50 Hz. This setup allowed for the accurate detection and analysis of the sensor's voltage output under varying mechanical loads.

3.2 Comparative Analysis of Sensor Performance

The performance of the hybrid PCPS (Porous CNT/PDMS-PVDF Hybrid Sensor), which integrates both triboelectric and piezoelectric mechanisms, was evaluated using a constructed pressure testing system, as illustrated in Fig. 2(a). The experiment compared the output voltage signals of four PDMS-based films—pure PDMS, PVDF/pure PDMS, non-porous PVDF/CNT/PDMS hybrid film, and porous PVDF/CNT/PDMS hybrid film under a consistent applied pressure of 1 N. The results, shown in Fig. 2(b), demonstrate that the porous PVDF/CNT/PDMS hybrid film exhibits a significant increase in output voltage, with voltage peaks 280%, 60%, and 39% higher than those of pure PDMS, PVDF/pure PDMS, and non-porous PVDF/CNT/PDMS hybrid film, respectively. This remarkable enhancement in performance can be attributed to several key factors. First, the synergistic effects of hybrid mechanisms combine triboelectric surface electrification and piezoelectric internal polarization, significantly amplifying the sensor's output compared to single-mechanism sensors [11], [14]. Second, the porous structure of the CNT/PDMS film increases the effective contact area between the PDMS and the electrode, enhancing charge transfer during triboelectric interactions [16]. Third, the addition of carbon nanotubes (CNTs) to the PDMS matrix improves its conductivity, further boosting the sensor's electrical performance [17].

Additionally, the porous structure allows for greater mechanical flexibility, enabling the sensor to respond more effectively to dynamic mechanical stimuli. Fig. 2(c) shows the voltage output signal measured during the pressing and releasing of the PCPS, confirming its ability to capture dynamic mechanical stimuli accurately. The sensor's rapid response and high sensitivity highlight its potential for applications in real-time tactile sensing and energy harvesting, where reliable performance under varying mechanical loads is critical [18], [21]. Furthermore, the self-powered operation of the PCPS eliminates the need for external power sources, making it highly suitable for integration into wearable electronics, soft robotics, and other energy-efficient systems [21].

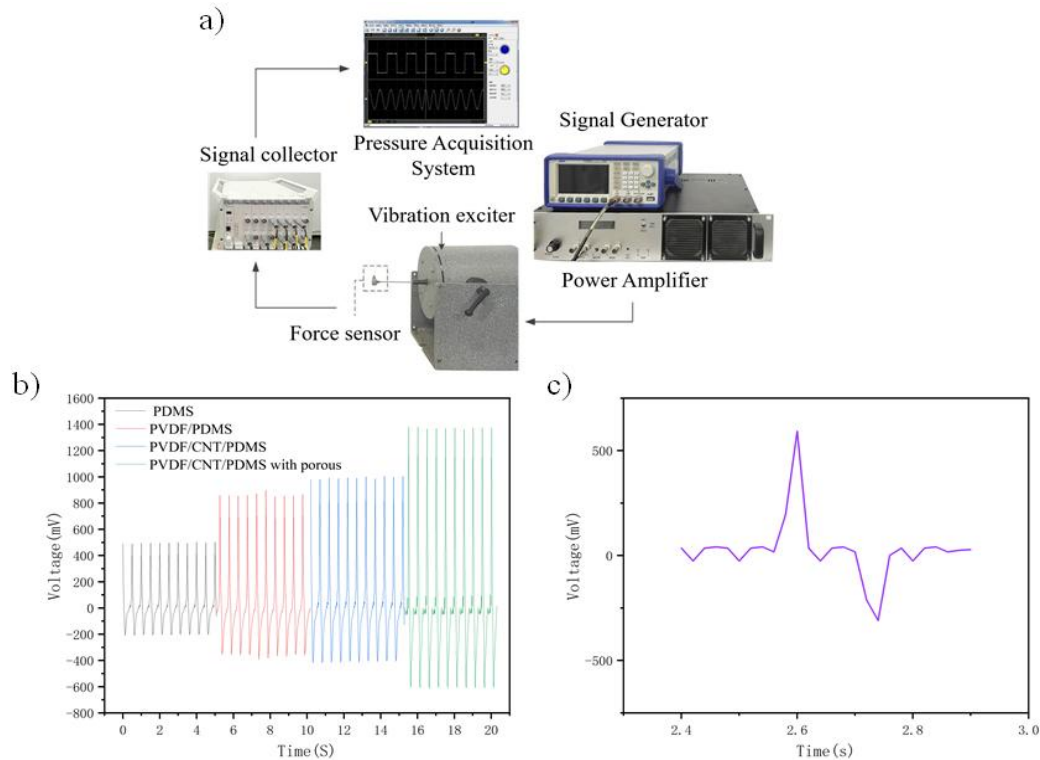


Figure 2 a) Sensor pressure test system, b) comparison of sensor output voltages based on PVDF/PDMS, porous PVDF/CNT/PDMS and non-porous PVDF/CNT/PDMS as sensitive films, c) pressure pattern sensing output voltage detection.

4. Conclusion

In this study, we developed a flexible porous CNT/PDMS-PVDF hybrid sensor (PCPS) that integrates both triboelectric and piezoelectric sensing mechanisms to achieve enhanced performance in terms of sensitivity, flexibility, and energy efficiency. By introducing a porous structure into the CNT/PDMS composite, the sensor's contact area was significantly increased, leading to improved triboelectric charge generation. The incorporation of PVDF as the piezoelectric layer further amplified the electrical signal output during mechanical deformation, addressing the limitations of traditional single-mechanism sensors. Experimental results demonstrated that the porous PVDF/CNT/PDMS hybrid film exhibited a 280% increase in voltage output under applied pressure compared to conventional PDMS-based sensors, highlighting the synergistic benefits of combining triboelectric and piezoelectric effects.

The sensor's robust and flexible structure, facilitated by precise material ratios and optimized fabrication processes, makes it highly suitable for dynamic environments such as wearable electronics, soft robotics, and energy harvesting systems. The self-powered operation of the PCPS eliminates the need for external power sources, enhancing its practicality for real-world applications. Furthermore, the sensor's ability to accurately capture dynamic mechanical stimuli underscores its potential for real-time tactile sensing and energy harvesting, where rapid response and high sensitivity are critical.

This study not only advances the field of flexible sensor technology but also provides a novel material design strategy that emphasizes the importance of structural engineering and hybrid mechanisms. Future research will focus on material optimization, miniaturization, and wireless integration to further enhance the sensor's performance and broaden its applications in next-generation wearable and implantable devices. By addressing the challenges of structural complexity,

low sensitivity, and inadequate mechanical properties, this work paves the way for the development of high-performance, self-powered sensors capable of operating efficiently in diverse environments.

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