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Development of a PDMS/ZnO/Gr piezoelectric tactile sensor and its exploration in intelligent applications

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ABSTRACT

This study investigates the preparation, performance, and intelligent applications of a composite piezoelectric sensor based on polydimethylsiloxane (PDMS), zinc oxide (ZnO) nanoparticles, and graphene (Gr). The effects of different component ratios on the piezoelectric and mechanical properties of the material were systematically studied. The results showed that the maximum voltage reached 21.30 V when the PDMS:ZnO:Gr ratio was 5:2:0.3%, demonstrating excellent piezoelectric performance. Additionally, the influence of ZnO and Gr filling on the mechanical properties of the material was assessed, revealing a trade-off between piezoelectricity and flexibility. Scanning electron microscopy was used to characterize the morphology of the composites, providing insights into the dispersion of ZnO and Gr within the PDMS matrix. Furthermore, the developed piezoelectric sensor was explored for its potential in smart applications, including tactile pressure and frequency recognition, as well as tactile recognition based on convolutional neural networks. The sensor was able to detect and differentiate between various materials, demonstrating its feasibility for intelligent interaction and recognition systems. These findings lay a foundation for the development of high-performance, flexible piezoelectric sensors and open up new avenues for the application of piezoelectric materials in the field of intelligence.

1. Introduction

Piezoelectric materials [1] are a unique class of smart materials that exhibit broad application prospects in sensors [2], actuators [3], energy harvesting [4], and smart structures due to their ability to convert mechanical energy into electrical energy and vice versa [5]. This characteristic stems from the non-centrosymmetric crystal structure of piezoelectric materials [6], which causes a relative displacement of the positive and negative charge centers within the material under external mechanical stress, leading to polarization and the formation of a potential difference across the material.

In recent years, with the rapid development of nanotechnology and composite materials science, the preparation processes and properties of piezoelectric materials have been significantly enhanced [7]. Particularly, the emergence of flexible piezoelectric materials has enabled their application in wearable devices, biomedicine, human-computer interaction, and other fields [8]. Flexible piezoelectric materials not only retain the high sensitivity and fast response characteristics of traditional piezoelectric materials but also possess good flexibility and processability, making them better suited to complex application environments [9]. Among the various flexible piezoelectric materials, composites based on polydimethylsiloxane (PDMS), zinc oxide (ZnO) nanoparticles, and graphene (Gr) have attracted considerable attention due to their unique properties and preparation processes. PDMS [10] is a common flexible polymer material that also exhibits excellent biocompatibility and chemical stability; ZnO nanoparticles, a typical piezoelectric material [11], possess a high piezoelectric coefficient and superior electrical properties; and Gr, a novel two-dimensional material [12] that offers extremely high electrical conductivity and mechanical strength, effectively enhances the electrical signal transmission efficiency and mechanical properties of the composite.

Therefore, this study aims to explore the preparation process, performance testing, and potential intelligent applications of a composite piezoelectric material based on PDMS, ZnO nanoparticles, and Gr. By systematically investigating the effects of different component ratios on the piezoelectric and mechanical properties of the material, this study aims to provide theoretical and technical support for the development of high-performance, flexible piezoelectric sensors. Furthermore, based on the developed piezoelectric sensors, this study will explore their smart potential in tactile pressure and frequency recognition, as well as tactile recognition based on convolutional neural networks (CNNs) [13,14], thereby opening new avenues for the application of piezoelectric materials in the field of intelligence.

2. Results and discussion

Figure 1a illustrates the detailed preparation process of the piezoelectric composites. Initially, a PDMS precursor was mixed with a curing agent at a ratio of 10:1. Subsequently, an appropriate amount of ZnO nanoparticles with an average diameter of 30 nm and Gr were added to the mixture. After thorough stirring and vacuum defoaming to remove bubbles, the prepared mixture was poured into a cylindrical concave mold made of PTFE with a depth of 0.1 mm. The sample was then placed in an oven and cured at 120 °C for 12 minutes, ultimately yielding a piezoelectric film. It is noteworthy that in the PDMS/ZnO/Gr composite, ZnO serves as the piezoelectric filler, generating a piezoelectric potential under dynamic pressure stimulation, while Gr acts as the conductive filler, potentially enhancing the overall piezoelectric response by improving the efficiency of electrical signal transmission. Figure 1b presents all the prepared samples. Firstly, blends

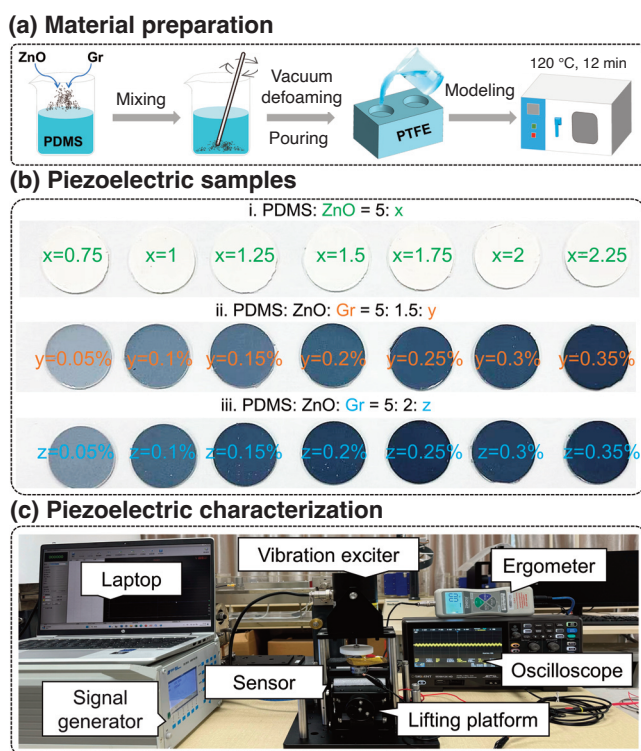


Fig. 1. Principle of the work: (a) material preparation, (b) piezoelectric samples, and (c) piezoelectric characterization.

of PDMS and ZnO were considered, where the composition ratio of PDMS was fixed at 5, and the composition ratio of ZnO ranged from 0.75 to 2.25. Subsequently, two Gr-filled systems with different ZnO ratios were studied, namely PDMS:ZnO at 5:1.5 and 5:2, with Gr composition ratios ranging from 0.05% to 0.35%. It can be clearly observed that the overall material appears white after doping PDMS with ZnO; however, as Gr is incorporated, the color of the material gradually deepens to dark gray. Figure 1c showcases the home-made highly integrated piezoelectric sensor testing platform. This platform applies a vibration signal to the exciter via a signal generator, which then exerts periodic pressure on the piezoelectric sensor. Precise control of the pressure magnitude is achieved by adjusting the lifting platform, while an external force gauge monitors the exact pressure value (in this study, the pressure is constant at ~ 0.5 N). A digital oscilloscope is used to detect the piezoelectric signal (i.e., open-circuit voltage), and the dual signals of pressure and voltage are aggregated and analyzed in real time by a host computer, enabling the monitoring and evaluation of the sensor's piezoelectric performance.

For this work, we have selected the maximum voltage and average voltage as indicators to evaluate the piezoelectric performance of the aforementioned samples. Figure 2a presents the influence of ZnO content in PDMS on the overall piezoelectric performance. As illustrated, the maximum voltage reaches 18.68 V when the PDMS:ZnO ratio is 5:1.5. However, excessive filling of ZnO leads to a decline in piezoelectric performance, with the voltage dropping to 5.84 V at a PDMS:ZnO ratio of 5:2.25. Figure 2b analyzes the effect of Gr filling on the piezoelectric performance of the optimal PDMS:ZnO = 5:1.5 system. It can be observed that the overall piezoelectric performance fluctuates as the Gr content increases. Specifically, the maximum voltage reaches 18.46 V when the Gr filling is 0.15%. Concerning Fig. 2c, it examines the impact of Gr filling on the piezoelectric performance of the PDMS:ZnO = 5:2 system. The overall piezoelectric per-

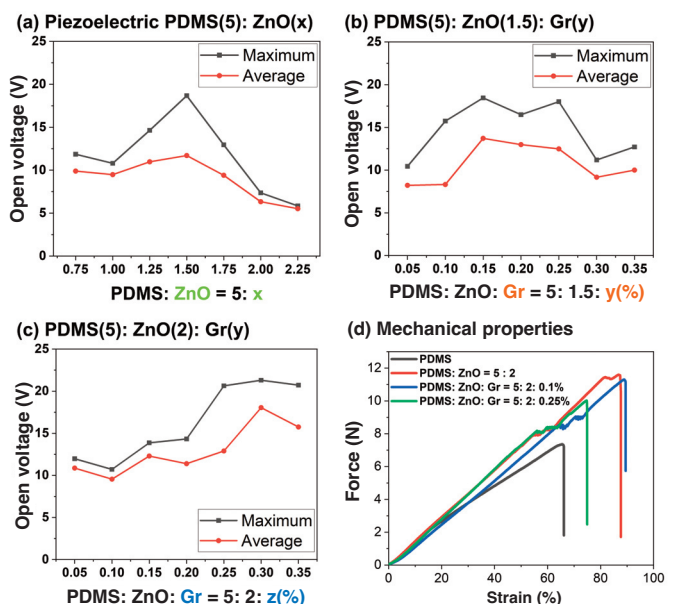


Fig. 2. Piezoelectric and mechanical properties of the prepared sensors.

formance exhibits fluctuations with increasing Gr content, reaching a maximum voltage of 21.30 V at a Gr filling of 0.30%. Subsequently, we assessed the influence of ZnO and Gr filling on the overall mechanical properties. Figure 2d demonstrates that the initial elastic moduli of the different filling systems are similar. The pure PDMS system exhibits a fracture strain of 65.43%. Upon filling with 40% ZnO (i.e., PDMS:ZnO = 5:2), the overall fracture strain and maximum fracture stress significantly increase to 86.71% and 11.6 N, respectively. One can notice that when Gr filling is 0.1%, the overall maximum fracture strain slightly increases to 88.75%, but the maximum fracture stress decreases to 11.3 N. However, excessive Gr filling (0.25%) results in a decrease in the overall fracture strain to 74.61%. These findings indicate that, although Gr filling can enhance the piezoelectric performance of certain composite sensor systems, it may lead to a decline in overall mechanical properties.

Further works regarding the influence of Gr on the morphology of PDMS/ZnO composites have been done. We focused on examining two doping systems, namely PDMS:ZnO:Gr = 5:2:0.1% and PDMS:ZnO:Gr = 5:2:0.25%, following the piezoelectric tests. It can be observed from the scanning electron microscopy (SEM) images of the cross-sectional morphology in Fig. 3 that when the Gr filling is 0.1%, particle sedimentation due to gravity leads to phase separation. Specifically, a uniform and smooth PDMS protective layer forms on the upper part of the blend, while a uniform dispersion of the ZnO particles is evident in the lower layer. The bottom surface morphology reveals that, although some ZnO particles exhibit aggregation, the overall dispersion is relatively uniform. On the other hand, when the Gr filling is increased to 0.25%, the cross-sectional morphology shows less distinct layering between the upper and lower layers. This is attributed to the high filling of Gr, which

hinders the sedimentation of piezoelectric particles such as ZnO, thereby reducing the morphological differences at the two-phase interface (this explains why the piezoelectric performance of the 0.25% Gr-filled system is optimal). However, the bottom surface morphology indicates that excessive Gr filling can impair the overall elasticity of the sensor, with noticeable microcrack structures observable under high-resolution imaging. This further illustrates a partial trade-off between the piezoelectricity and flexibility of the piezoelectric sensor, which necessitates further optimization in future work.

One of the potential smart applications is illustrated by Fig. 4a, which represents the scenario of piezoelectric signals in tactile pressure and frequency recognition. For this case, four sensors were affixed to a copper sheet and mounted on the thumb, index finger, middle finger, and ring finger of a glove, respectively. The piezoelectric signals generated by pressing the mouse with the fingers were first converted from analog to digital signals. The digital signals were then sent to a microcontroller development board for processing. The processed signals were transmitted to a computer and displayed as tactile pressure and frequency through custom-developed software. The test results demonstrated that when the mouse was clicked, the bar graph on the software successfully showed a significant signal peak corresponding to the index finger position. This verifies the feasibility of the piezoelectric sensor in the field of tactile pressure and frequency recognition and highlights its application prospects in smart interaction. Figure 4b presents a smart use of tactile recognition based on CNN. The sensor was worn on the hand, and different materials (glass, copper, paper, and plastic) were tapped with similar force, generating distinct piezoelectric potentials that were captured by an oscilloscope. Tapping on glass produced the highest positive voltage of 6.60 V and a negative voltage of -2.67 V. Tapping on plastic yielded the lowest positive voltage of 1.44 V; however, it is noteworthy that its negative voltage reached -2.88 V, the highest among the four materials. This indicates that the output voltages of different materials not only vary in magnitude but also in the ratio of positive to negative voltages. The output voltage range for copper was -2.81 to 5.08 V, while for paper it was -1.75 to 3.96 V. Based on the above findings, we employed a CNN-based framework for analyzing pulsed AC voltage signals in piezoelectric materials. The dataset comprised 40 000 samples categorized into four distinct classes (10 000 samples per class), which were split into training (80%) and testing sets (20%) to ensure balanced representation of class-specific features. To mitigate signal distortion caused by environmental noise and piezoelectric instability, we first applied noise reduction techniques, followed by data augmentation strategies. StandardScaler was used for normalization (mean = 0, variance = 1) to stabilize the input distribution, after which raw signals were converted into 5-length sequential segments for temporal feature extraction. The CNN architecture consisted of an input layer, two convolutional layers with MaxPool1d downsampling, and two fully connected layers (Fig. 1). Specifically, the first Conv1d layer processed 4-channel inputs through 16 kernels

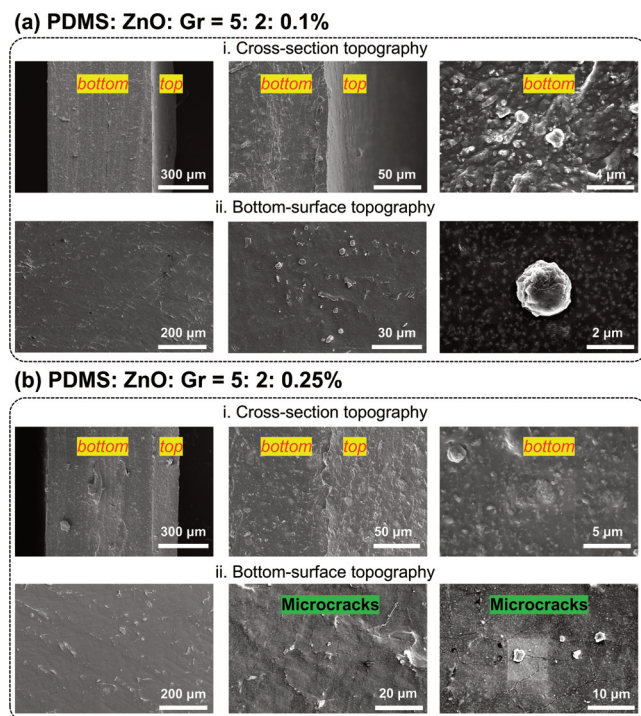


Fig. 3. SEM characterization of piezoelectric sensor of (a) PDMS:ZnO:Gr = 5:2:0.1% and (b) PDMS:ZnO:Gr = 5:2:0.25%.

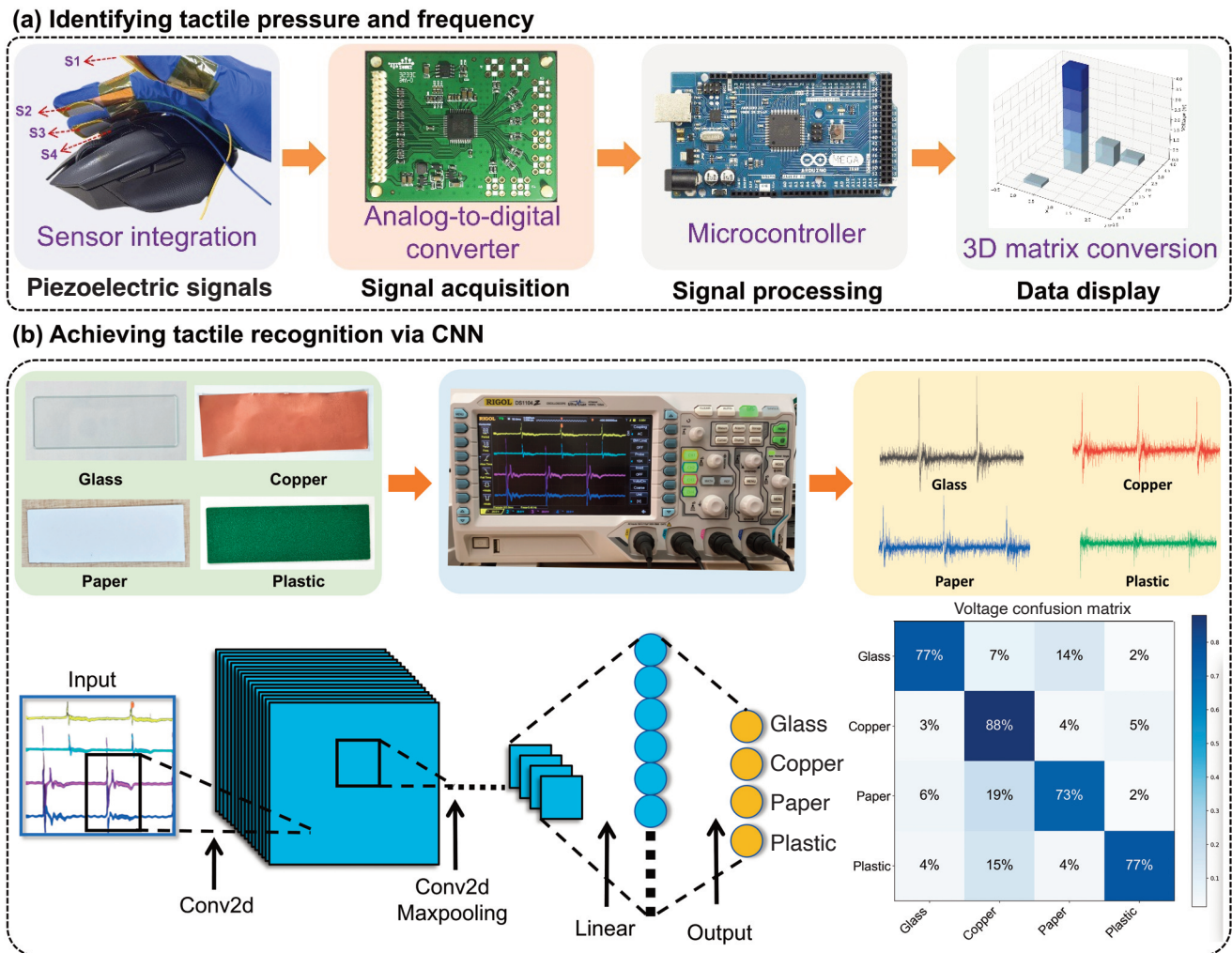


Fig. 4. Exploration of the piezoelectric sensors in intelligent applications.

(kernel size = 3, padding = 1), followed by MaxPool1d (kernel size = 2) to reduce spatial dimensions. This was repeated with 16 input channels feeding 32 output channels. Feature maps were flattened into 64 dimensions through the first dense layer before final classification into four categories. Training employed CrossEntropyLoss with Adam optimizer (learning rate = 0.0001) over 400 epochs on Apple Silicon (M3 Pro) hardware. To ensure experimental reproducibility, we standardized data preprocessing, fixed random seeds (42) for NumPy, PyTorch, and CUDA operations, and maintained consistent software versions (NumPy==1.26.4, torch==2.5.0, and the CUDA pre-compiled package by PyTorch includes MPS support by default). Despite challenges in capturing transient pulsed signals, our multi-run deep learning framework achieved robust classification performance (73–88% accuracy), as validated by the confusion matrix, demonstrating the model’s capacity to generalize across heterogeneous piezoelectric responses.

3. Conclusion

In this work, we systematically investigated the preparation, performance testing, and potential applications of composite piezoelectric materials based on PDMS, ZnO nanoparticles, and Gr in intelligent systems. A series of PDMS composite

piezoelectric films containing varying ratios of ZnO and Gr were successfully fabricated. Detailed experiments were conducted to verify the significant roles of ZnO as a piezoelectric filler and Gr as a conductive filler in enhancing the piezoelectric properties of the material. The experimental results demonstrated that when the ratio of PDMS to ZnO was 5:2, and the Gr content was 0.3%, the material exhibited a maximum voltage of 21.3 V, indicating excellent piezoelectric performance. Furthermore, we thoroughly examined the influence of different filler systems on the mechanical properties of the material. It was found that an appropriate amount of Gr filler could enhance the fracture strain of the material to some extent, whereas excessive filling led to a decrease in overall mechanical performance. Additionally, SEM observations revealed the impact of Gr content on the microstructure of the material, providing a theoretical basis for further optimizing its performance. Finally, based on the developed piezoelectric sensor, we successfully demonstrated its intelligent applications in tactile pressure and frequency recognition, as well as in CNN-based tactile recognition. The experimental results showed that the sensor could accurately identify objects made of different materials and generate corresponding piezoelectric signals. Through processing and analysis by a CNN model, intelligent recognition of tactile signals was achieved. This not only verified the feasibility of

piezoelectric sensors in intelligent applications but also laid a solid foundation for their widespread use in the future.

Data availability statement

All research data are contained within the article and can be shared upon request from the authors.

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PDMS/ZnO/Gr piesoelektrilise taktilise sensori arendamine intelligentsete rakenduste jaoks

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Uuring käsitleb polüdimetüüsiloksaanil (PDMS), tsinkoksiidi (ZnO) nanoosakestel ja grafeenil (Gr) põhineva komposiit-piesoelektrilise sensori valmistamist, jõudlust ja kasutamist intelligentsetes rakendustes. Uuriti erinevate koostisosade suhete mõju materjali piesoelektrilistele ja mehaanilistele omadustele. Tulemused näitasid, et maksimaalne pinge ulatus 21,30 V-ni, kui PDMS:ZnO:Gr suhe oli 5:2:0,3%, mis tõestas suurepäraselt piesoelektrilist jõudlust. Samuti hinnati ZnO ja Gr täiteainete mõju materjali mehaanilistele omadustele, tuues välja kompromissi piesoelektriliste omaduste ja painduvuse vahel. Skaneeriva elektronmikroskoopia abil analüüsiti komposiitmaterjalide morfoloogiat, andes ülevaate ZnO ja Gr jaotumisest PDMS-maatriksis.

Uuriti väljatöötatud piesoelektrilise sensori kasutusvõimalusi nutikates rakendustes, sealhulgas taktilise surve ja sageduse tuvastamisel konvolutsiooniliste närvivõrkude mudelite abil. Sensor suutis tuvastada ja eristada erinevaid materjale, demonstreerides selle sobivust intelligentsete interaktsiooni- ja tuvastussüsteemide jaoks. Saadud tulemused loovad aluse suure jõudlusega ja paindlike piesoelektriliste sensorite arendamiseks ning avavad uusi võimalusi piesoelektriliste materjalide rakendamiseks tehisintellekti valdkonnas.