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Flexible piezoelectric energy harvester based on graphene macro-film electrode enabled by exploiting auxetic mechanical property

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ABSTRACT

The flexible piezoelectric energy harvester (f-PEH) has attracted wide attention due to its potential use for autonomous wearable electronics. However, the amount of power from the existing f-PEH is insufficient for practical applications. Hence, this paper aims at developing a novel electrode material, i.e. graphene macro-films (GAMF), to improve the electrical output performance of the f-PEH. Theoretical results indicate that the f-PEH of GAMF with absolute value of greater negative Poisson's ratio (NPR) and a larger Young's modulus shows better electrical output performance. Demonstrative experiment shows that the voltage output performance of f-PEH can be significantly increased by improving its mechanical properties. The load characteristic test shows the maximum output power of the optimized sample is 296.86 nW. The feasibility of the sample in practical application is also verified by the stability test. In addition, the energy harvesting circuit is designed to demonstrate high efficiency of this novel f-PEH, which was able to lighten LEDs after the motivation of finger bending to charge the capacitor. This paper indicates that the f-PEH based on the NPR GAMF has great application potential in the wearable field.

1. Introduction

The depletion of fossil energy globally requires the development of renewable energy technology. Many researchers have recently focused on flexible piezoelectric energy harvesters. However, the relatively small amount of energy harvested from existing f-PEHs prevents the practical application of the equipment. To address these issues, previous studies mainly focused on piezoelectric material improvement [1,2], circuit optimization [3] and structural design [4], etc. However, stability issues remain the key points about the viability of f-PEHs for practical applications.

In contrast, our previous work has developed a graphene macro-film (GAMF) with good conductivity and negative Poisson's ratio (NPR), which is attributed to its unique micro-structure referred to paper [5]. This new material with NPR effect has been applied to design f-PEH, providing a new way to improve the electrical output performance of the device [6]. However, a few issues remain with this approach. Firstly, little information on the influence of mechanical properties of GAMF on the electrical output performance of devices has been presented. Secondly, the electromechanical behaviors of f-PEH based on GAMF with NPR lack systematic exploration.

In this study, we conducted a detailed analysis of derived analytical expression of the relationship between the open circuit voltage and the electrode mechanical properties. According to analysis results, f-PEHs with GAMF of different mechanical properties were prepared as electrodes, the experimental electrical output is found to fit the theoretical results. In addition, the novel f-PEHs combined with storage device could light LED bulb, showing great potential in the wearable field.

2. Materials and methods

The GAMFs were prepared from graphene oxide (GO, purchased from Wuxi Chengyi Education Technology Co., Ltd.). For higher film formation efficiency, we disperse graphene oxide in deionized water, and rapidly stirred at 400 rpm for 4 h to form graphene oxide suspension with the solid content of 2%-4%. The GO film was prepared by introducing the graphene oxide suspension into a glass mold and dry naturally at room temperature. The obtained graphene oxide film is then annealed at 1300 °C for 2 h and at 2850 °C for 1 h under the protection of argon to graphitize graphene oxide. After cold pressing treatment, the highly conductive GAMF was obtained. During the preparation of the graphene oxide film, adjusting the cold pressing pressure can change the

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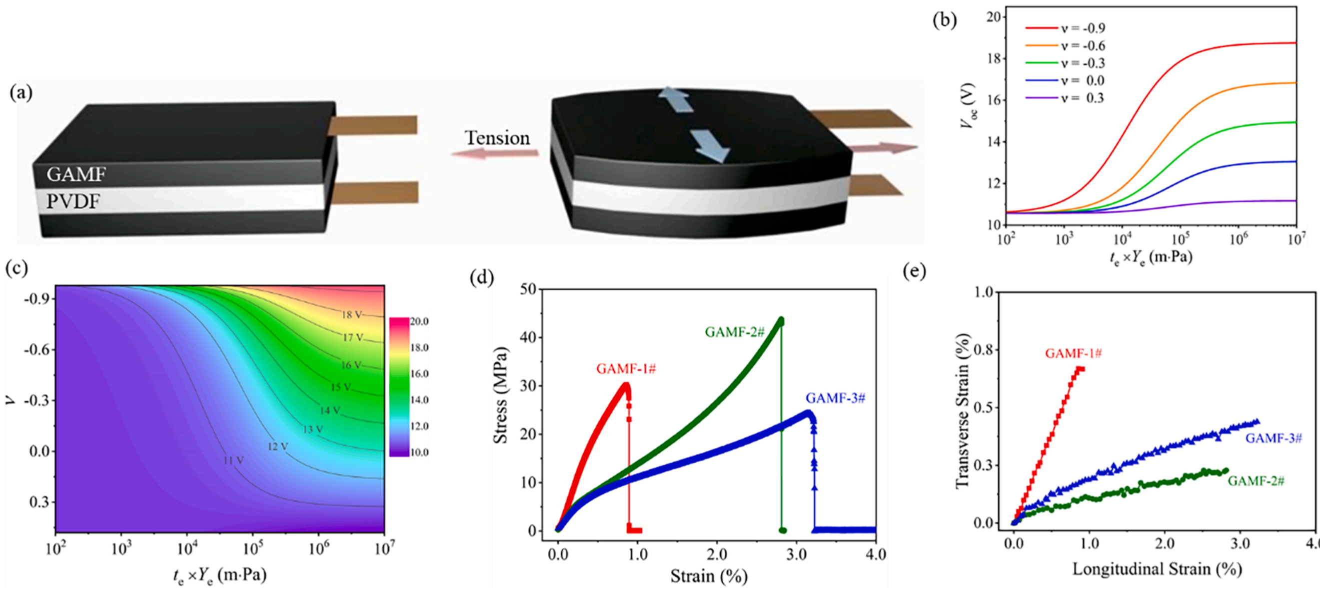

Fig. 1. Theoretical analysis ad characterization of the GAMF. (a) Schematic diagram of deformation. (b, c) Visualization of Eq. (1) showing the effect of mechanical properties of GAMF on the open-circuit voltage. (d) The stress–strain and (e) transverse strain and longitudinal strain diagram of the GAMF under uniaxial stretching.

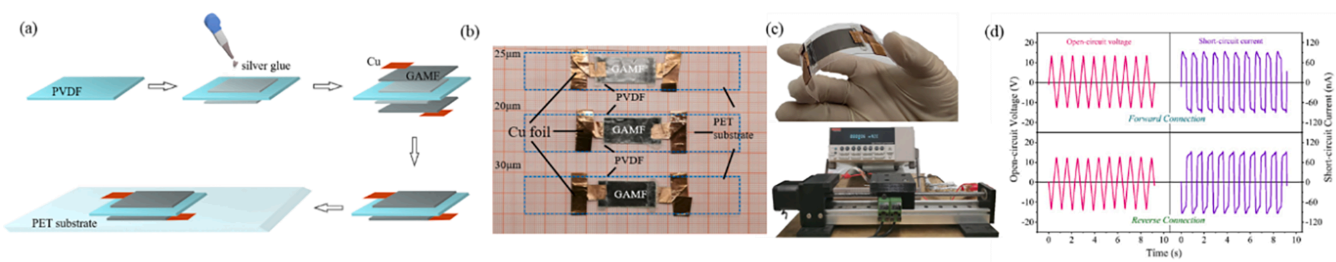

Fig. 2. Fabrication and test of the f-PEH. (a) Schematic process diagram of the fabricating processes. (b) Electronic images the samples. (c) Flexibility display and bend test chart. (d) Open-circuit voltage and short-circuit current waveforms of the f-PEH in forward and reverse connections.

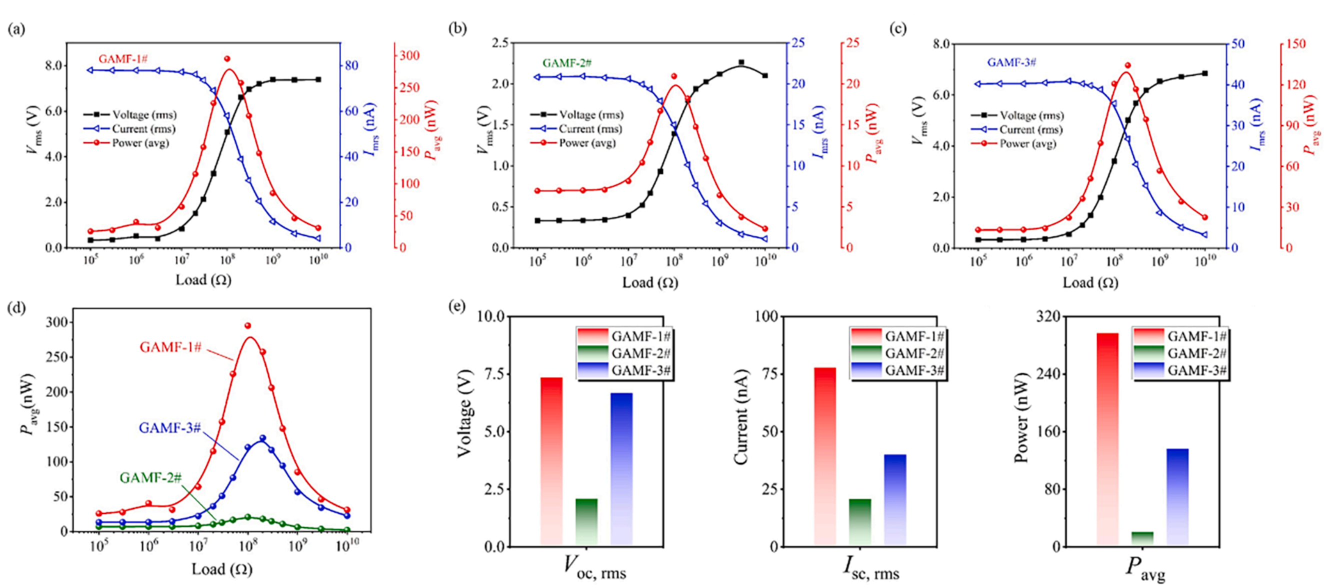

Fig. 3. Load characteristics of the f-PEH. Load characteristics of electricity output for (a) 1#, (b) 2# and (c) 3# f-PEH. (d) Comparison of the load characteristics in terms of output power. (e) Performance comparison histogram.

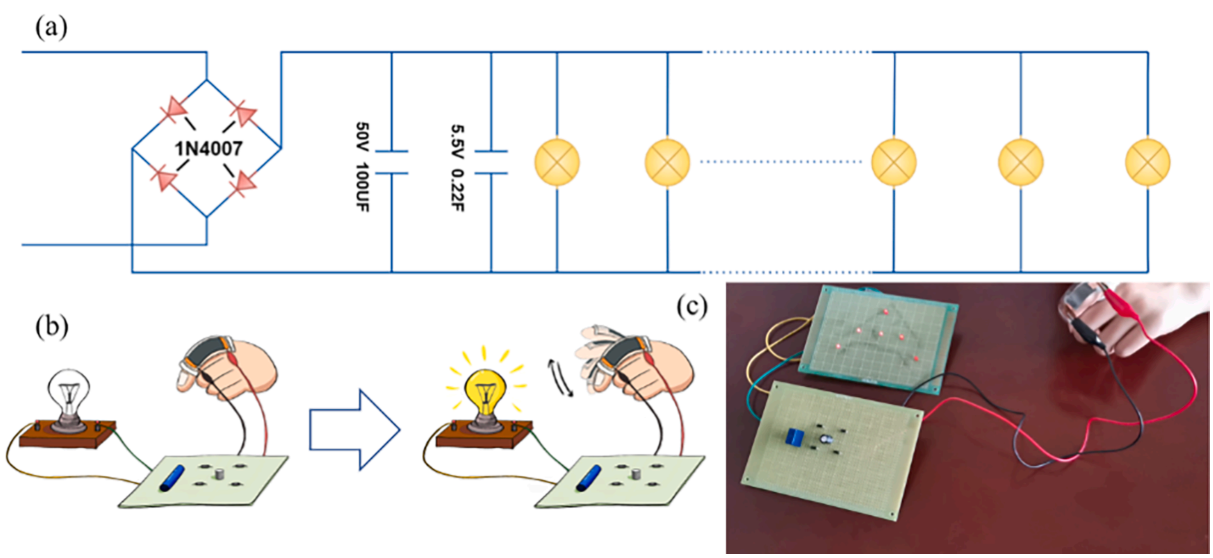

Fig. 4. Wearable application display. (a) Energy harvesting circuits for the application of f-PEH. (b) Schematic diagram of wearable applications. (c) Photograph of lightening LEDs by finger bending.

degree of compaction of the graphene sheet stacking in the graphene film, and then adjust its macro-mechanical properties. In this study, we prepared three kinds of GAMF with different mechanical properties. For convenience, these three films (and corresponding devices) with the cold pressing pressure of 0 MPa, 60 MPa and 10 MPa at room temperature for 10 min are numbered 1#, 2# and 3#, respectively.

3. Results and discussion

Fig. 1(a) illustrates the NPR effect of GAMF on the PVDF film of piezoelectric devices. That is to say, the PVDF film will be deformed with the deformation of GAMF. This principle has been proved in our previous work [6]. According to the structure, the relationship between open-circuit voltage V_{oc} and the mechanical properties of eletrode materials is [6].

$$V_{oc} = \left(\frac{\left(\upsilon_P^D - \upsilon_e\right)\left(\upsilon_P^D g_{31} + g_{32}\right) t_e Y_e}{\left(1 - \left(\upsilon_P^D\right)^2\right) t_e Y_e + \left(1 - \upsilon_e^2\right) t_p Y_p^D} + g_{31} \right) \cdot Y_p^D S_1 t_p,$$

With several stable piezoelectric material parameters and assuming $t_p = 20$ μm, $S_1 = 0.05\%$, the influence of electrode material on the output performance of f-PEH can be determined, which is given in Fig. 1(b) and (c). Fig. 1(b) shows the influence of the product of Young's modulus and thickness on the output voltage of the f-PEH in five cases. The contour map of Fig. 1(c) further proves the importance of Young's modulus and thickness for electrode materials to output performance. Fig. 1(d) and Fig. 1(e) show the mechanical properties curves of three GAMFs, which the Young's modulus and Poisson's ratios of 1#, 2#, 3# GAMFs (with the thickness of 30 μm, 25 μm, 20 μm) is calculated to be 4.00 GPa, 2.53 GPa, 2.39 GPa, and −0.76, −0.08, −0.22, respectively. Fig. S9 shows the surface and cross-sectional SEM image of GAMFs, the micro porous contained in the films explain the negative Poisson's ratio behavior of GAMF laterally.

A set of f-PEH were assembled by using the above three GAMFs with different properties. The schematic diagram of the fabricating processe and the sample are shown in Fig. 2(a) and (b). Fig. 2(c) proves that the flexibility of the device and shows a scene diagram of the bend test. In the experiment, an electrometer (Keithley 6514) was used to collect signal generated by the periodic bending of the f-PEH. As shown in Fig. 2 (d), the voltage and current waveforms in forward and reverse connections confirm that the signal is produced by piezoelectric effect.

Fig. 3(a)–(c) respectively show the load characteristics of the voltage, current, and average power of the f-PEHs under the bending frequency of 1.09 Hz. It can be seen that the trend of the curves of voltage, current, and power are similar for three samples. When the load is 100–200 MΩ, the device reaches the impedance matching state, and the power is maximized, as shown in Fig. 3(d). Fig. 3(e) also shows the histogram of open-circuit voltage, short-circuit current and maximum output power of three devices. The results indicate that 1# sample has better electrical output performance than other devices.

In addition, comparing the output performance of different devices, the 1# device has the largest voltage, current and power, which are 7.35 V, 77.82nA and 296.86 nW, respectively. These results also prove that the GAMF with minimum NPR and maximum Young's modulus has the best output performance. In addition, the voltage data obtained by the experiment is consistent with the changing trend of the theoretical data. The mechanical property data obtained by the test is brought into the Eq. (1), the strain is assumed to be 0.05%, the calculated voltages of 1#, 2# and 3# devices are 8.57 V, 6.30 V and 6.48 V, respectively. Meanwhile, we also carried out stability test, it can be found in supplementary information.

This research further combines f-PEHs based on GAMFs with energy storage devices, as shown in Fig. 4(a) and (b). After bending for several times to charge the capacitor, the lighting experiment of small bulb is realized by connecting capacitor with small LED bulb, as shown in Fig. 4 (c). This experimental result shows that the f-PEH has good applicability in the field of wearable electronic devices.

4. Conclusions

In this study, we revealed the relationship between the output performance of f-PEHs with the Young's modulus, Poisson's ratio and thickness of electrode material through mutual verification between experiment and theory. The results indicate that reducing the NPR or enhancing the Young's modulus and thickness of the GAMF can greatly improve the electrical output performance of the f-PEH. Moreover, the load characteristics and stability of the sample were studied, it concluded that the maximum output power of the sample is 296.86 nW, which has excellent stability. We also carried out the experiment of lighting the small bulbs by loading the energy collection circuit, which proved the durability and practicability of the device.

CRediT authorship contribution statement

Haoyu Zheng: Writing – original draft, Formal analysis. **Huazhang Zhang:** Writing – review & editing. **Pin Wen:** Conceptualization, Supervision. **Daping He:** Resources.

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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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.matlet.2022.132165.

References

[1] D.Y. Hyeon, K.-I. Park, Energy Technol. 7 (10) (2019) 1900638.
[2] C. Ryu, S. Hajra, M. Sahu, S.I. Jung, I.R. Jang, H.J. Kim, Mater. Lett. 309 (2022), 131308.
[3] X. Gao, M. Zheng, X. Yan, M. Zhu, Y. Hou, Mater. Today Phys. 19 (2021), 100424.
[4] J.H. Han, K.-I. Park, C.K. Jeong, Sensors (Basel) 19 (6) (2019) 1444.
[5] P. Li, Z. Wang, R. Song, W. Qian, P. Wen, Z. Yang, D. He, Carbon 162 (2020) 545–551.
[6] H. Zhang, P. Wen, P. Li, Z. Wang, S. Wang, X. Zhao, Y. Xiao, J. Shen, D. He, W. Chen, Appl. Phys. Lett. 117 (10) (2020), 103901.