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论文题目：A Low-Cost Graphite Interlayer Enables a Parallel
 $\text{Cu}_2\text{Se}/\text{Ag}_2\text{Se}$ Sandwich Architecture with Enhanced
Thermoelectric Current Output

A Low-Cost Graphite Interlayer Enables a Parallel Cu₂Se/Ag₂Se Sandwich Architecture with Enhanced Thermoelectric Current Output

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Abstract

Thermoelectric generators convert low-grade heat directly into electricity, yet conventional series (π -type) modules are heavy, rigid, and spatial inefficient. This study proposes a parallel sandwich architecture in which a p-type Cu_2Se leg and an n-type Ag_2Se leg share a low-cost graphite interlayer made from a 2B pencil-core coating. This interlayer decouples the conductive path from the generating layers. At a temperature difference of 70 K, a 360 cm^2 flexible film delivered a stable output of 30 mA and a peak near 350 mA, approximately six times the device-level current of an in-house π -type reference under the same ΔT . The output increased with the number of pencil-core layers, and microscopy revealed ordered graphite lamellae that are consistent with a low-resistance bridge for the simultaneous collection of electrons and holes. This study frame ambipolar carrier collection as a hypothesis that requires direct transport confirmation and report current output rather than a ZT or efficiency claim. The novel architecture offers a lightweight, inexpensive route toward wearable thermoelectric harvesting.

Keywords

wearable thermoelectrics, parallel architecture, graphite interlayer, Cu_2Se , Ag_2Se , ambipolar carrier collection

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1. Introduction

In recent years, growing demand for renewable energy sources and increasing concerns about greenhouse gas emissions have driven efforts to reduce dependence on fossil fuels. Generating 30–31 GT of CO₂, the vast pollution engendered by combusting gasoline, coal, and natural gas is putting the environment at risk.^{1, 2} Ways to generate renewable energy were at need.

Hopefully, thermoelectric materials generate electricity directly from a temperature gradient through the Seebeck effect, offering a compact path to recover low-grade heat.^{3–6} Thermoelectric materials contain two distinct types: n-type and p-type, categorized by the different electric carriers. In n-type thermoelectric semiconductors, including Ag₂Se, electron (e^-) travel from hotter end to cooler end; in p-type thermoelectric semiconductors, such as Cu₂Se, holes (h^+) travel from hotter end to cooler end. The performance of thermoelectric materials can be evaluated by ZT values. $ZT = \sigma S^2 T / \kappa$, where S is Seebeck coefficient, σ is electrical conductivity, T is temperature, and κ is thermal conductivity. A higher ZT value indicates better thermoelectric performance.⁶

Though TEG had already been assigned as one of the most promising power candidates for wearable devices,^{3, 7, 8} low efficiency and structural heaviness impedes TEG's competitiveness. With a direct conversion path that seems to circumvent energy loss, current commercial TEGs only achieve conversion efficiencies of approximately 5–7% under a temperature difference of 200–250 K,^{9, 10} and the theoretical ceiling for the best systems is below 33%.⁹ The gap to fossil-fuel systems (about 30% overall) remains large,¹¹ so thermoelectric power is still not competitive for most uses. This study therefore provides an improvement to TEG design that could enhance its competitiveness.

Conventional generators connect alternating p-type and n-type legs in a π -type series layout.³ The metal interconnects between legs occupy space and add mass without contributing to power generation, a “parasitic” loss that is especially severe in compact modules. This problem is particularly prominent in compact module designs, occupying substantial space within the device while increasing its mass. Moreover, this architectural burden compounds the intrinsic trade-off between electrical conductivity and Seebeck coefficient,¹² which limits the thermoelectric figure of merit ZT.

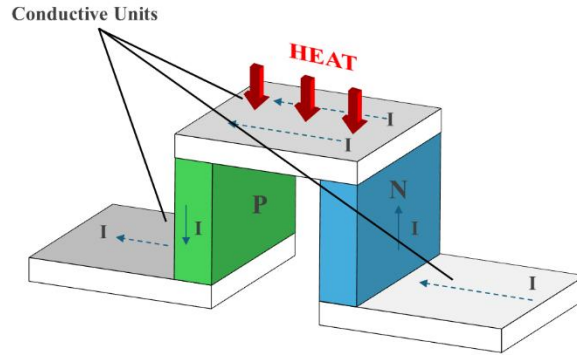


Figure 1. The metal conductive units in conventional thermoelectric generators evoke considerable issues, including spatial occupation and a coupled transmission path.

Low efficiency of TEGs is commonly attributed to the difficulty of simultaneously optimizing the parameters affecting ZT , heat losses through radiation and convection, and the limited thermoelectric performance of existing materials.^{9, 12-14} Yet the device architecture itself is often treated as fixed, while most effort targets new materials. This study reasoned that an alternative connection scheme could raise output without requiring a new high- ZT material.

This study replace the series connection with a parallel sandwich framework: a p-type Cu_2Se leg and an n-type Ag_2Se leg sit side by side and share a mutual interlayer (Figure 2a,b). This removes the interconnect material in series design and separates the conductive layer from the generating layer, therefore raising current without sacrificing Seebeck response accordingly. This paper therefore do not claim a ZT improvement; the advance the study report is architectural.

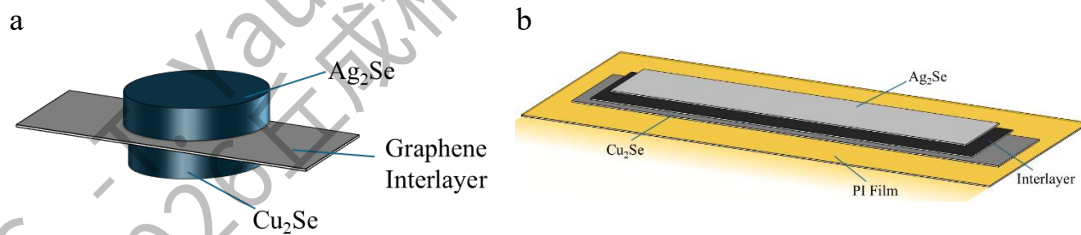


Figure 2. The novel sandwich thermoelectric generator in (a) bulk structure, (b) membranous structure.

Electron-hole recombination normally limits bipolar transport.¹⁵⁻¹⁷ It can be suppressed by physical and chemical processes, as seen in silicon (Si), perovskites (CaTiO_3), gallium nitride (GaN), etc.¹⁸⁻²⁰ Simultaneous e^- and h^+ transport is usually achieved through ambipolar field-effect transistors that need an external gate voltage,^{21, 22} or through ambipolar diffusion driven by mutual electrostatic attraction without a gate voltage.^{23, 24} The proposed device has no gate control, so the interlayer must

support ambipolar carrier collection on its own via ambipolar diffusion.

A flat, two-dimensional structure in which carbon atoms are arranged in a honeycomb-like lattice is called graphene, which possesses ambipolar field effect.²⁵⁻²⁷ However, the paucity in the study of ambipolar diffusion of this material remains to be studied. This study hypothesized the existence of ambipolar diffusion in graphene, thereby applying several graphene-related interlayer materials: graphite paper, carbon nanotube (CNT), carbon cloth, and graphite coatings derived from a 2B pencil core. 2B pencil paste on paper was found to possess ambipolar field effect.²⁸ Via scanning electron microscopy (SEM), high-resolution transmission electron microscope (HRTEM), and selected area electron diffraction (SAED) characterization, this paper provides potential substantiations that ambipolar diffusion is present in the material as well.

In the parallel architecture, the carbon interlayer acts as a common pathway. Under a temperature gradient, h^+ from Cu_2Se leg and e^- from Ag_2Se leg converge at central layer, forming a p-n junction bridged by the interlayer. Yet due to high longitudinal resistance and lateral conductivity of the interlayer, the interface forms a heterojunction similar to a Schottky barrier. This correction follows the verified carrier polarity (Cu_2Se is p-type, Ag_2Se is n-type) and suppresses interfacial recombination while preserving high lateral conductivity. Previous studies had discovered using ambipolar transport to transmit carriers enhances performance of thermoelectrics.^{9, 10, 29, 30} The approach of this study is therefore viable in a way.

After confirming feasibility in a bulk sandwich (Figure 2a), the study transferred the design to a 20 μm polyimide-supported film (Figure 2b), far thinner than the 1–2 cm π -type device of Zhang et al.³ The film bends readily (Figure 3) and is suited to wearable use.

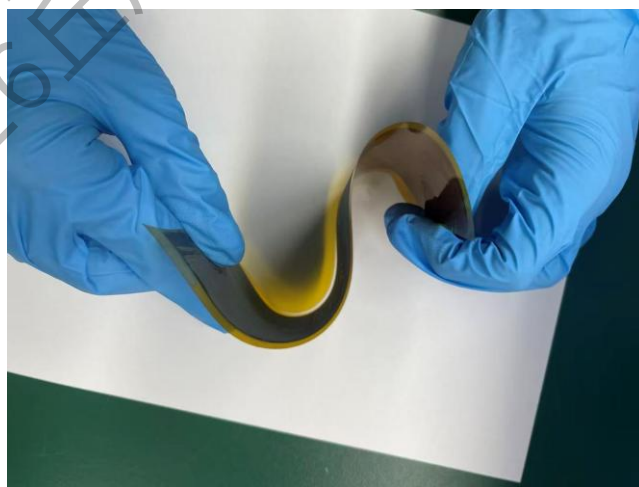


Figure 3. The membranous device possesses considerable bending capacity.

2. Materials and Methods

2.1 Experimental materials and instruments

A. R. standard sodium sulfide nonahydrate, copper selenide powder, N,N dimethylformamide (DMF), and CNT were all purchased from Aladdin. Selenium powder, nano-silver powder, and Ag₂Se solid were obtained from MACKLIN. The nano-copper paste and nano-silver paste were purchased from Shenzhen Saiya Electronic Paste Co., Ltd. Experimental reagents and materials were directly used without further purification. Additionally, the sandwich-type parallel thermoelectric film samples were supported by 20 μm thick polyimide (PI) films, and 2B pencil, graphene paper (Louis HCP010N hydrophilic type, 1 mm thickness), and carbon cloth (240 g/m², 3K twill) were used in the experiment. Carbon cloth is made of polyacrylonitrile (PAN)-based carbon fiber.

Table 1 summarizes the apparatuses used in reading outputs of sandwich TEGs.

Apparatus	Version
Electrochemical Workstation	CHI760E
Semiconductor TEC Temperature Control System	TLTP-TEC2415D-WB80-80-Z
Seebeck Coefficient Measurement System	LSR-3, Linseis
SEM	ZEISS Sigma 300
HRTEM	JEOL JEM-F200
SAED	JEOL JEM-F200

Table 1. Apparatuses applied in the experiment and their corresponding versions.

2.2 Construction of sandwich structure thermoelectrics

A relatively large block-shaped sandwich-type parallel thermoelectric structure (bulk structure) was first prepared to verify its feasibility. Then, this structure was simplified into a layered sandwich-type parallel thermoelectric film to enhance the wearable performance.

Cu₂Se (4.00 g, 0.019 mol) and β -phase Ag₂Se (2.00 g, 0.014 mol) were ground into powder in a mortar, respectively. Then, 1.50 g of each was taken and pressed in a 13.0 mm diameter circular mold under an 8.00 T pressure for 30.0 seconds. Two samples of copper selenide disks and Ag₂Se disks with a thickness of approximately 1.50 mm were obtained, respectively. Cu₂Se samples underwent α -to- β phase transformation using a Seebeck coefficient measuring instrument at high temperatures for a comparative experiment.

The nano-membrane structure was synthesized by the *in-situ* metal selenization

method (Figure 4).³ 1.20 g of $\text{Na}_2\text{S}\cdot 9\text{H}_2\text{O}$ and 0.850 g of Se were added to 90.0 mL of deionized water, and the solution was stirred at 500 r/min for 15 minutes. The solution quickly turned reddish-brown. Nano-silver paste and nano-copper paste were printed onto the PI membrane using a screen-printing method, and then placed in the selenization solution. After selenization, the samples were rinsed and dried to obtain nano-membrane-like samples. After coating the intermediate layer, a second layer of nano-selenized membrane was prepared similarly by the *in-situ* metal selenization strategy. CNT coating was obtained from a carbon nanotube solution using DMF as the dispersant and was dried after coating.

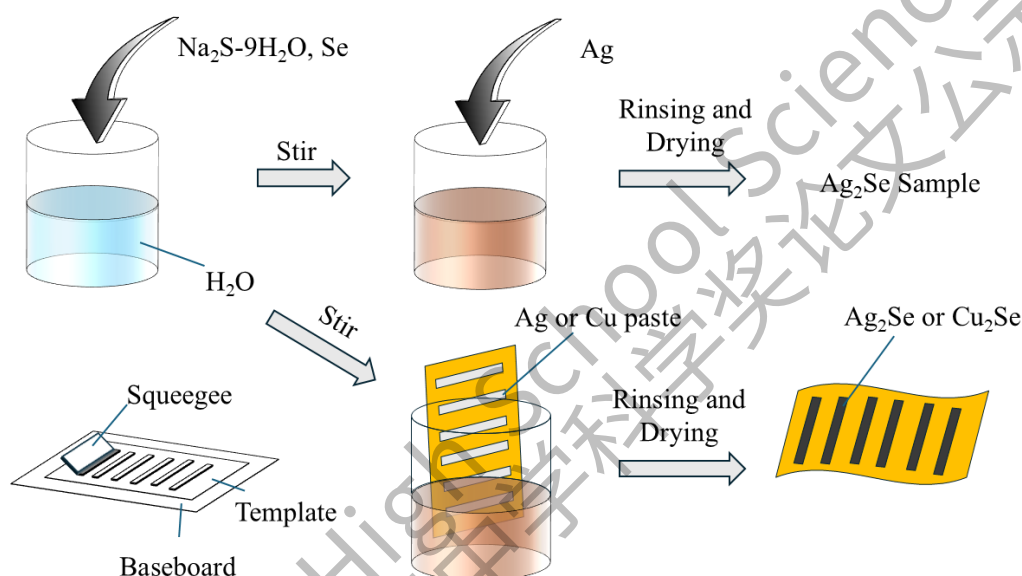


Figure 4. *In-situ* synthesis for metal selenides.

2.3 Microscopic examinations of 2B pencil core coating

The microscopic examinations of 2B pencil core coating—including SEM, HRTEM, and SAED—was mandated to be conducted by Scientific Compass under Hangzhou Yanqu Information Technology Co., Ltd.

3. Results and Discussion

3.1 Bulk Feasibility Check

To validate the concept before film fabrication, this study built block sandwiches (Table 2): 1-1 (β -Cu₂Se/ β -Ag₂Se with graphene paper), 1-2 (α -Cu₂Se/ β -Ag₂Se with graphene paper), and 1-3 (β -Cu₂Se / β -Ag₂Se with carbon cloth).

Experiment Number	Crystal Form	Interlayer
1-1	β -Cu ₂ Se β -Ag ₂ Se	Graphene Paper
1-2	α -Cu ₂ Se β -Ag ₂ Se	Graphene Paper
1-3	β -Cu ₂ Se β -Ag ₂ Se	Carbon Cloth

Table 2. Experiment 1 explored the effect of utilizing different crystalline forms of Cu₂Se and various interlayers in a bulk sandwich TEG.

Figure 5a shows that α -Cu₂Se gave higher output than β -Cu₂Se, consistent with the high-temperature α phase having better thermoelectric properties,³¹ and that graphene paper outperformed carbon cloth, whose mesh voids raise lateral resistance. This bulk test supported the subsequent film simplification.

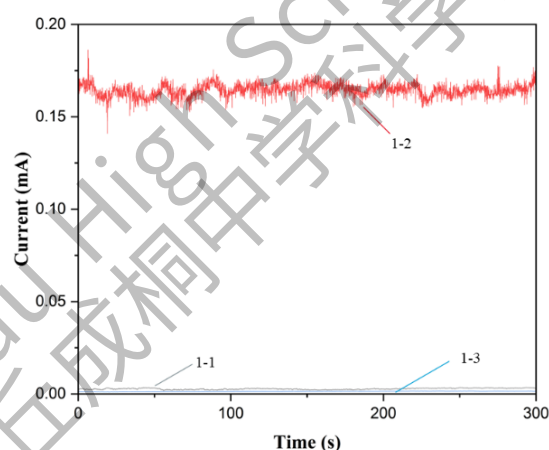


Figure 5. Current output of experiment set 1.

3.2 Interlayer Screening on Flexible Films

This study simplified the bulk sandwich to a nanoscale film (Figure 2b) and compared carbon nanotube (CNT) and 2B pencil-core coatings as interlayers (Table 3). Intermediate layer was replaced by a CNT coating (Figure 6a) or 2B pencil core coating (Figure 6b), and comparative experiments were conducted.

The interlayer number provided accounts only an approximate number of the actual coating layers. It was hard to control the actual number of interlayers printed, but difference among experiments 2-2 to 2-5 was guaranteed.

Experiment Number	Interlayer	Interlayer Number
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2-1	CNT	1
2-2	2B Pencil Coat	1
2-3	2B Pencil Coat	3
2-4	2B Pencil Coat	6
2-5	2B Pencil Coat	9

Table 3. Experiment 2 applied different materials as the interlayer with various numbers of interlayers.

Figure 6c shows the outputs of structures 2-1 to 2-5. The 2B pencil coating at six layers outperformed the CNT coating, and the output rose with the number of pencil-core layers (2-2: 1 layer; 2-3: 3 layers; 2-4: 6 layers; 2-5: 9 layers). The 2B advantage can be attributed to its clay-graphite mixture³², which may form additional sp^2 carbon networks or defect states that help separate electrons and holes. CNT coatings suffered from poor dispersion and contact, giving higher contact resistance. Output fluctuations in samples 2-3 and 2-5 tracked instability of the temperature-control device. Sample 2-3 even showed current reversal during long tests; the cause is not yet understood and may relate to carrier drift induced by temperature fluctuation.

a



b



c

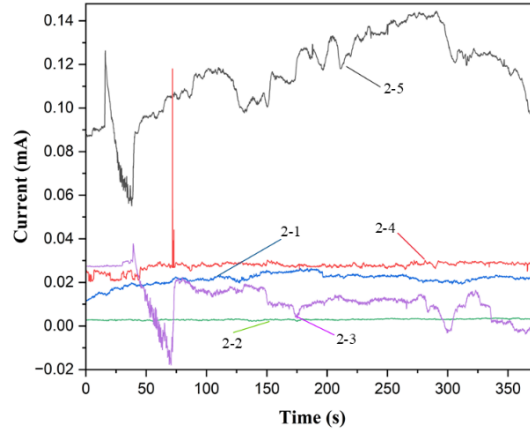


Figure 6. (a) Structure 2-1 applying CNT as the interlayer. (b) Structure 2-2 with 2B pencil core coating as the interlayer. (c) Current output of experiment set 2.

3.3 Multi-film output and benchmark

This study then fabricated three larger parallel films (Figure 7a) and compared four connection schemes (Table 4; Figure 7b–d): parallel (3-1), double-layer parallel (3-2), single-layer parallel (3-3), and a conventional π -type series. All membranous sandwich architecture applied 2B pencil paste coatings with approximately 9 layers.

Experiment Number	Connection Method
3-1	Parallel-connected Method
3-2	Double-layer Parallel-connected Method
3-3	Single-layer Parallel-connected Method
Π -type Structure	Conventional Series Connection

Table 4. Experiment 3 used three different connection methods between three film sandwich TEGs.

The single-layer parallel scheme (3-3) reached a peak near 350 mA with a stable average of about 30 mA, fluctuating between 15 and 45 mA (Figure 7e). This stable 3-3 output greatly exceeded the π -type reference (slightly above 5 mA) and the 3-1 (about 0.21 mA) and 3-2 (about 2.05 mA) schemes (Figure 7f). The single-layer parallel strategy performed best, probably because more transmission paths reduced the temperature drop between legs; the thin films are very sensitive to temperature, so this effect is pronounced.

Normalized to a 360 cm^2 device area, 3-3 film delivered about $0.083 \text{ mA} \cdot \text{cm}^{-2}$. The roughly sixfold increase is a relative device-level current under the same ΔT in comparative test, not a normalized figure of merit. A fair performance comparison requires matched area, mass, and measured power density, which the study now states as a limitation. Output voltage or load resistance weren't measured, so power density remains to be determined.

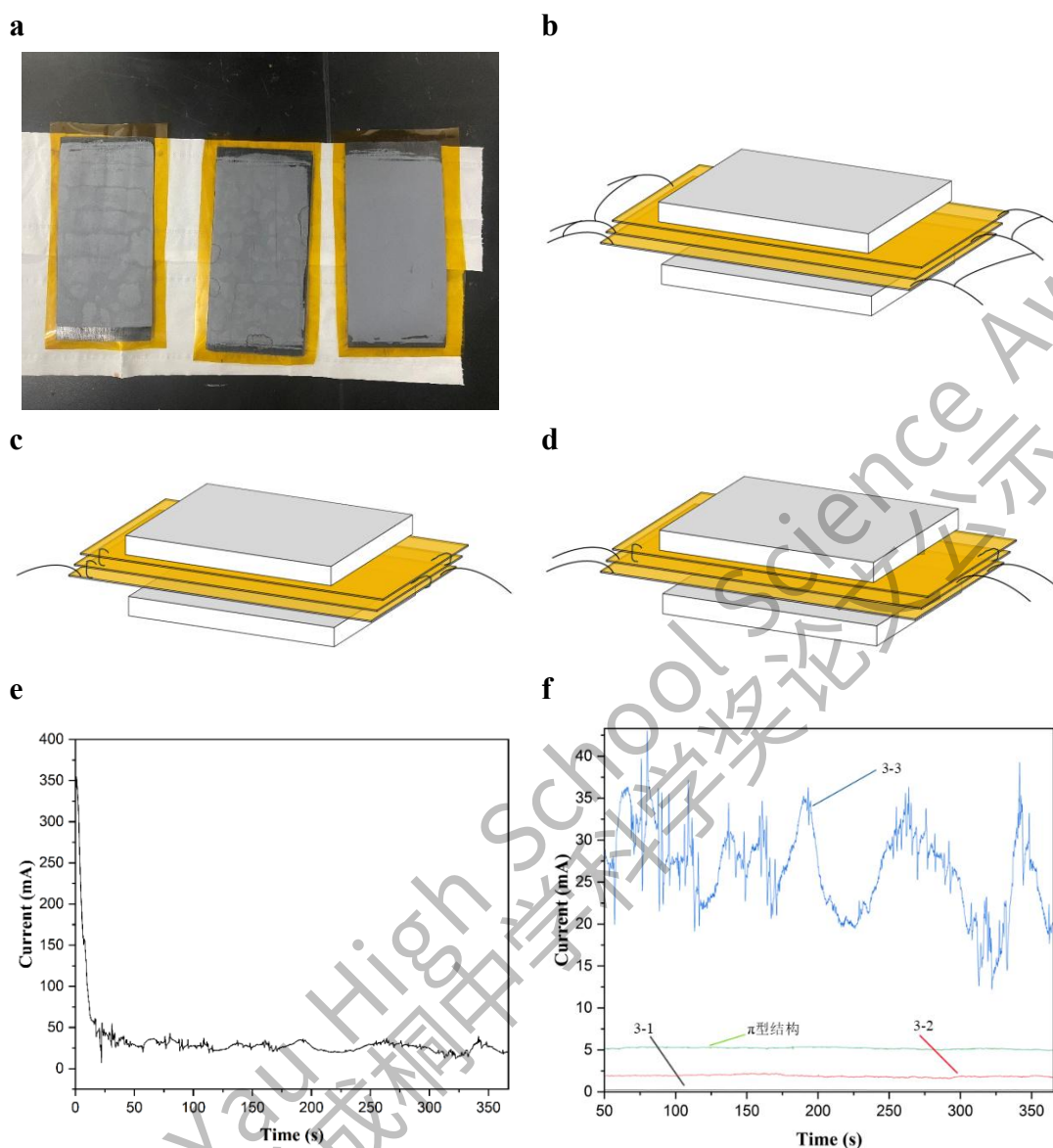


Figure 7. (a) Larger sandwich films developed through *in-situ* synthesis. (b) The thermoelectrics were connected in parallel with each other. (c) Carbon conductive layers of the upper and middle films with those of the middle and lower films were connected. (d) Carbon conductive layers of the upper and middle films were connected. (e) Current output of structure 3-3. (f) Current output of experiment set 3.

3.4 Microstructure of the 2B pencil coating

SEM images (Figure 8a–c) show the large lamellar structure of the 2B pencil coating. Continuous layered frameworks may provide a built-in path for electron and hole transport, and larger grains can reduce grain-boundary barriers that promote recombination.^{33, 34} HRTEM images (Figure 8d,e) reveal clear lattice fringes, and the SAED pattern (Figure 8f) shows an ordered, monocrystalline framework. These structural features are consistent with a low-resistance interlayer that could facilitate

simultaneous electron and hole collection³⁵⁻³⁷, but they do not by themselves prove ambipolar diffusion. Though ambipolar field effect exists in 2B pencil paste²⁸, no study has yet confirmed a synchronize occurrence of ambipolar field effect and ambipolar diffusion. Direct evidence, such as Seebeck polarity, Hall effect, or temperature-dependent transport, is still required and is planned.

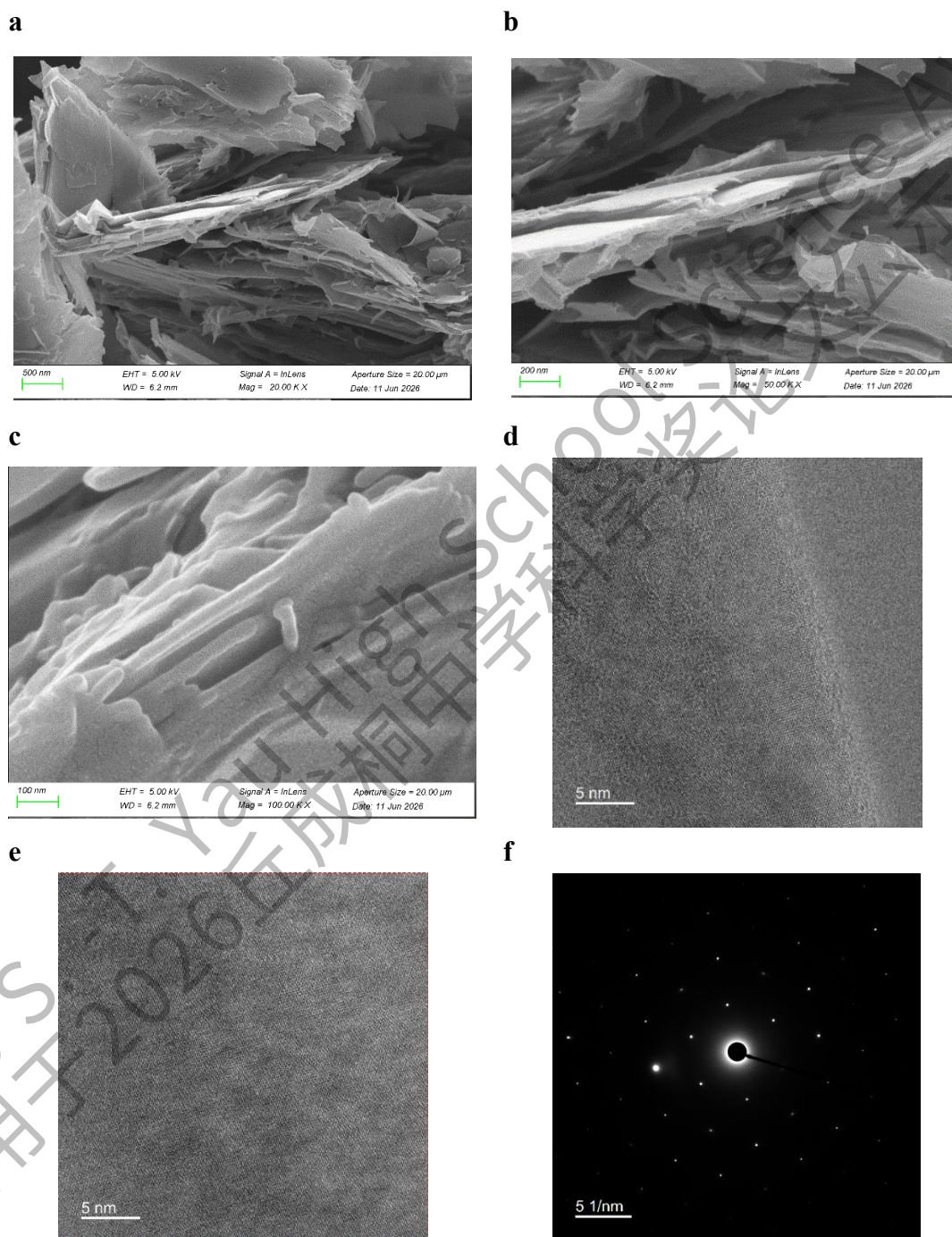


Figure 8. Characterization of 2B pencil paste. (a,b,c) SEM images. (d,e) HRTEM images. (f) SAED image.

4. Conclusion

This study constructed a flexible parallel sandwich thermoelectric generator that uses an everyday 2B pencil-core graphite interlayer between p-type Cu_2Se and n-type Ag_2Se legs. At a 70 K temperature difference over 360 cm^2 , the device delivered about 30 mA stable and 350 mA peak, roughly six times the current of an in-house π -type reference. Output increased with the number of interlayer coatings, and structural microscopy is consistent with a low-resistance bridge for electron-hole collection. The work shows that device architecture, not only new materials, can raise thermoelectric current output at low cost. This study do not claim a ZT or efficiency improvement. Confirming ambipolar carrier collection and reporting Seebeck coefficient, electrical and thermal conductivity, and power density are the necessary next steps. To determine whether increasing 2B pencil paste layers, sandwich membranes, and device surface area enhance the performance to an extent are also planned.

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6. Acknowledgements

I decided to research in the topic of next-generation TEGs because of my experience once during hiking. In the mountains of Yunan, I was poured wet by a heavy rain. I was even scared when finding my mobile phone of out power. Therefore, I sought for an approach to provide a light and portable energy supply (that's easy to carry with even in an eleven-hour-trek).

Professor Zeng introduced me to thermoelectric generators when I pursued a non-paying internship at Institute of Chemical Engineering, Guangdong Academy of Sciences. This idea inspired me.

At first Prof. Zeng suggested me with a topic of building TEGs in fabric structure comparable to cloths. Yet finding this approach to be studied previously, I soon aspired to try another approach. With my background research I found the drawbacks of orthodox TEGs, and as I was having lunch one day, I thought of this idea I subsequently examined: a sandwich structure. Prof. Zeng supported me to examine this crazy idea of combining two thermoelectric legs together. With the help of his fellow colleagues and his students in the group, I was able to utilize multiple apparatuses to substantiate the viability of my device.

The major obstacle in my experiment is the design of an ambipolar-transportable layer. As discussed in the paper, electrons and holes don't typically transport simultaneously on the same mediate. Yet in my design such interlayer is required. To overcome this barrier, I consulted previous literatures, and finally reached Novoselov et al.'s 2004 essay on the topic of finding graphene. In their study the ambipolar field effect—a way in which electrons and holes can transport selectively (but not simultaneously) on a specific material by gate voltage—of few-layer graphene (FLG) was found. With further research I discovered that ambipolar diffusion in a material can achieve simultaneous ambipolar carrier transportation. Yet too often such materials are hard to fabricate.

Knowing that graphene has certain ambipolar characteristics, I hypothesized that it also possesses inherent ambipolar diffusion effect. With this premise, I constructed the sandwich TEG. I first tried with a bulk structure (the p-n legs were in massive bulks) for a simple substantiation of my design. When it worked, I tuned to making a lightweighted, membranous architecture. In their 2025 work, Prof. Zeng and his team published a method to synthesize metal selenides under room temperature—known as *in-situ* synthesization. I adopted their method in my study, and I give thanks to them sincerely.

I applied various graphene-related materials as the interlayer for a comparison test. This strategy can help decide which interlayer has the best performance. Apart from graphene paper and CNT, it came so natural to me trying the coating of a 2B pencil core

as it was posited right next to me on the desk.

Finding out the performance of 2B pencil paste to be outstanding among all, I applied it on a new set of TEGs that I later used for multi-layer examinations. This set of experiments was used for determining the how connection strategy between TEG structures impacts the output. The greatest among all peaked at around 350 mA and steadied at around 30 mA. This output was approximately a sixfold increase compared to conventional TEG architectures.

With the help of Prof. Zeng and his team, I used electrochemical workstation to examine the electric current output of my proposed TEG and analyzed the data with Origin.

I wrote the first draft of the paper myself, and in each revision I add in the advice from Prof. Zeng and my school tutor, Ms. Chen. They helped construct my paper with increased academic rigor as well as accuracy. It was with their support I am able to present this work now, and I cordially thank them for their efforts.

致谢

我之所以决定研究新一代热电发电机（Next-generation TEGs）这一课题，是因为有一次徒步旅行的经历。在云南的高山中，我在被一场暴雨淋得浑身湿透后发现手机没电而感到十分害怕。因此，我开始寻找一种轻便、可携带的能源供应方案（一种哪怕是在长达十一个小时的徒步中也能使用的器件）。

在广东省科学院化学工程研究所进行非营利性实习期间，曾教授向我介绍了热电发电机。这个想法深深打动了我。

起初，曾教授建议我以类似布料的织物结构来构建热电发电机。但考虑到该方法此前已有研究，我很快萌生了尝试其他方案的想法。通过前期的研究，我发现传统热电发电机存在诸多不足。一天中午吃午饭时，我突然想到一个构想，并进一步深入探讨：采用类似“三明治”的夹层结构。在后续的研究中，曾教授支持我尝试将两个热电腿结合在一起的这个“疯狂”设想。在他与同事和团队研究所的帮助下，我利用多种实验设备验证了该装置的可行性。

我实验中的主要障碍在于设计一种双极性可迁移层。如论文中所述，电子和空穴通常不会在同一介质上同时传输。然而，在我的设计中，这种双极性传输是必需的。为克服这一难题，我查阅了相关文献，最终找到了诺沃谢洛夫等人 2004 年关于寻找石墨烯的研究文章。在他们的研究中，发现了一种双极场效应——即通过栅极电压使电子和空穴在特定材料上选择性地（但非同时）进行传输的现象——该效应在多层石墨烯（FLG）中得到了验证。进一步研究后，我发现材料中的双极性扩散可以实现双极性载流子的同步传输。然而，这类材料往往难以制备。鉴于石墨烯具有一定的双极性特性，我推测它也具备固有的双极性扩散效应。基于这一假设，我构建了三明治 TEG 结构。首先，我尝试使用块体结构（p-n 结构的两部分为大块材料）来验证设计方案的可行性。当该结构成功后，我进一步优化设计，转向轻质化、膜状结构的构建。在 2025 年的研究中，曾教授及其团队发表了一种室温下合成金属硒化物的方法，即原位合成法。我在自己的研究中采用了他们的方法，并向他们致以诚挚的感谢。

我将多种与石墨烯相关的材料作为夹层用于对比测试，这一策略有助于确定哪种夹层性能最佳。除了石墨烯纸和碳纳米管外，尝试用 2B 铅笔芯作为涂层也显得非常自然，因为它就摆放在我的桌面上。

经过测试发现，2B 铅笔芯涂层在所有材料中表现尤为出色，于是我将其应用于一组新的 TEG（热电发电机）器件上，这些器件随后被用于多层结构的实验。该系列实验旨在研究 TEG 结构之间的连接方式对输出性能的影响。结果显示，最大输出电流约为 350 mA，稳定输出约为 30 mA，相比传统 TEG 结构，输出功率提高了约六倍。

在曾教授及其团队的帮助下，我利用电化学工作站测试了 TEG 的电流输出，并使用 Origin 软件对数据进行了分析。

我亲自撰写了论文的初稿，而在后续的每次修改过程中，我都加入了曾教授和我的学校导师陈老师的意见。他们帮助我提升了论文的学术严谨性和准确性。

正是在他们的支持下，我才能将这项成果呈现出来。衷心感谢他们的付出。

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