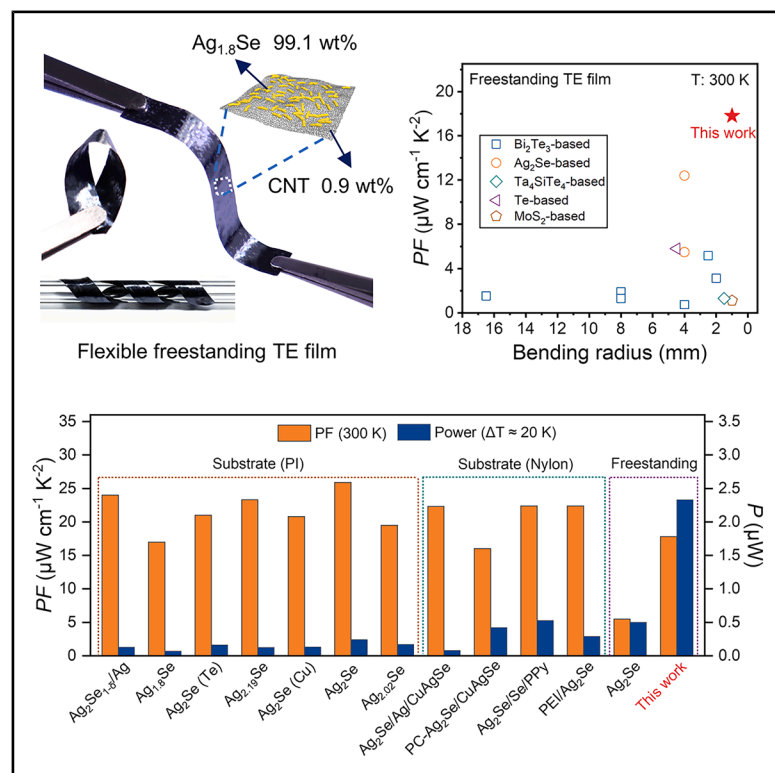


# Flexible freestanding $\text{Ag}_2\text{Se}$ thermoelectric films with high performance for sap flow monitoring

## Graphical abstract



## Authors

Gang Li (李钢), Xiang-Rui Guo (郭祥瑞), Tao Wang (汪涛), Jing Wang (王晶), Yu Wang (王瑜), Guangming Chen (陈光明), Yan-Qing Lu (陆延青)

## Correspondence

yuwang87@nju.edu.cn (Y.W.), chengm@szu.edu.cn (G.C.), yqlu@nju.edu.cn (Y.-Q.L.)

## In brief

Ultralow-content CNT (0.9 wt %) enables freestanding flexibility in  $\text{Ag}_{1.8}\text{Se}$  while enhancing thermoelectric performance via energy filtering. The  $\text{Ag}_{1.8}\text{Se}/\text{CNT}$  films exhibit a high room-temperature power factor and small bending radius. The  $\sim 10\text{-}\mu\text{m}$ -thick freestanding films enable flexible devices with substantial power output. This strategy provides a sustainable route to high-performance inorganic thermoelectrics.

## Highlights

- Freestanding  $\text{Ag}_{1.8}\text{Se}/\text{CNT}$  films endure 20,000 bending cycles at a 2-mm radius
- A power factor of  $20.9 \mu\text{W cm}^{-1} \text{K}^{-2}$  at 380 K is achieved with ultralow CNT content
- A flexible TE generator delivers  $4.6 \mu\text{W cm}^{-1} \text{K}^{-2}$  normalized power density
- A highly sensitive, self-powered TE sensor enables reliable plant sap flow monitoring

Article

# Flexible freestanding Ag<sub>2</sub>Se thermoelectric films with high performance for sap flow monitoring

Gang Li (李钢),<sup>1</sup> Xiang-Rui Guo (郭祥瑞),<sup>1</sup> Tao Wang (汪涛),<sup>1</sup> Jing Wang (王晶),<sup>1</sup> Yu Wang (王瑜),<sup>1,3,\*</sup> Guangming Chen (陈光明),<sup>2,\*</sup> and Yan-Qing Lu (陆延青)<sup>1,\*</sup>

<sup>1</sup>National Laboratory of Solid State Microstructures, Key Laboratory of Intelligent Optical Sensing and Manipulation, College of Engineering and Applied Sciences, and Collaborative Innovation Center of Advanced Microstructures, Nanjing University, Nanjing 210023, China

<sup>2</sup>College of Materials Science and Engineering, Shenzhen University, Shenzhen 518055, China

<sup>3</sup>Lead contact

\*Correspondence: [yuwang87@nju.edu.cn](mailto:yuwang87@nju.edu.cn) (Y.W.), [chengm@szu.edu.cn](mailto:chengm@szu.edu.cn) (G.C.), [yqlu@nju.edu.cn](mailto:yqlu@nju.edu.cn) (Y.-Q.L.)

<https://doi.org/10.1016/j.joule.2026.102487>

**CONTEXT & SCALE** Thermoelectric materials that can directly convert small amounts of heat into electricity are attracting growing interest for powering the Internet of Things and wearable electronics. The development of flexible thermoelectrics is particularly appealing because of their ability to conform to diverse heat sources, enable stable energy harvesting, and integrate seamlessly with soft electronic systems. However, most flexible thermoelectric materials today struggle to reconcile mechanical flexibility with high thermoelectric performance and substantial power output. In particular, flexible inorganic thermoelectric composites face a long-standing trade-off: high filler content enhances flexibility but sacrifices performance, whereas low filler content preserves performance at the expense of flexibility. Overcoming this inherent contradiction remains a central bottleneck in the field.

In this study, we overcome this limitation by engineering the microstructure of a new type of freestanding composite film composed of Ag<sub>1.8</sub>Se with a minimal amount of CNTs. Although the CNT content is very low, it forms an interconnected network that provides mechanical support and enhances electrical transport. This design enables the film to combine strong flexibility with high room-temperature thermoelectric performance. Surprisingly, the material retains nearly all of its performance after 20,000 bending cycles at a small radius of 2 mm, demonstrating its suitability for long-term use in wearable or deformable systems. When assembled into simple devices, these films can generate meaningful power from modest temperature differences and can be integrated onto biodegradable supports. This unlocks opportunities for sustainable and bio-interfaced electronics. We further show that the devices can attach directly to plant surfaces, enabling both high-sensitivity sap flow monitoring and self-sufficient energy supply. This design principle can be applied to other thermoelectric materials, offering a scalable and sustainable route toward high-performance flexible thermoelectrics.

## SUMMARY

Flexible inorganic thermoelectrics are promising for the Internet of Things and wearable electronics. While solution processing offers a facile route to such materials, simultaneously achieving mechanical flexibility and substantial power output remains challenging. Here, we report a microstructure-engineering strategy to fabricate freestanding Ag<sub>1.8</sub>Se/carbon nanotube (CNT) composite films with ultralow CNT content (0.9 wt %). An interwoven CNT network establishes dense, conductive interfaces with Ag<sub>1.8</sub>Se nanowires, enabling efficient carrier transport and exceptional flexibility. The ~10- $\mu$ m films achieve a power factor of 20.9  $\mu$ W cm<sup>-1</sup> K<sup>-2</sup> at 380 K and retain >95% of their performance after 20,000 bending cycles at a 2-mm radius. An assembled flexible device delivers an output power of 15.4  $\mu$ W and a normalized power density of 4.63  $\mu$ W cm<sup>-1</sup> K<sup>-2</sup> under a 50 K temperature gradient. Integrated with biodegradable supports, the devices demonstrate recyclability and enable sap flow monitoring. This facile and sustainable approach is generalizable to other systems such as Bi<sub>2</sub>Te<sub>3</sub> and Cu<sub>2</sub>Se.

## INTRODUCTION

In recent years, the rapid advancement of self-powered Internet of Things (IoT) and wearable electronics has driven growing interest in sustainable energy-harvesting technologies.<sup>1,2</sup> Flexible inorganic thermoelectric materials, which can directly convert low-grade body heat into usable electricity, represent a promising power solution for such applications because of their capacity for lightweight, sustainable power generation; close adherence to arbitrary heat sources; and silent operation.<sup>3–5</sup> Ideally, these materials should combine high thermoelectric performance, substantial power output, robust mechanical flexibility, low processing temperatures, low toxicity, and long-term stability.<sup>1</sup> Nevertheless, simultaneously fulfilling these criteria remains a significant challenge with current material systems. To improve output performance, it is crucial to optimize the figure of merit ( $zT = S^2\sigma T/\kappa$ ) and the power factor ( $PF = S^2\sigma$ ), where  $S$ ,  $\sigma$ ,  $T$ , and  $\kappa$  represent the Seebeck coefficient, electrical conductivity, absolute temperature, and thermal conductivity, respectively.<sup>6,7</sup> Beyond intrinsic thermoelectric properties, the effective power generation of thermoelectric devices is strongly determined by the material's thickness,<sup>8,9</sup> which governs internal resistance across the device. This interdependence underscores that the design of flexible thermoelectric materials requires optimizing their thermoelectric properties and thickness in a coordinated manner while ensuring sufficient mechanical flexibility to maximize power output.

The general method for creating flexible inorganic thermoelectric materials involves depositing inorganic layers (typically less than 1  $\mu\text{m}$  thick) onto flexible polymer substrates through techniques such as magnetron sputtering, pulsed laser deposition, and screen printing.<sup>5,8,10</sup> However, these methods have inherent drawbacks; the submicron-thick films show high internal resistance and low power output (Figure 1A, left; Note S1), while the polymer substrates contribute to heat loss, ultimately reducing power density.<sup>7,11,12</sup> To address these issues, freestanding inorganic thermoelectric films have gained considerable interest as an alternative platform.<sup>13</sup> Recently, silver chalcogenides Ag<sub>2</sub>X (X = S, Se, Te) have been demonstrated to be processable into ductile, freestanding thermoelectric films.<sup>4,14,15</sup> Still, their fabrication often depends on energy-intensive procedures. Despite progress in reducing metalworking temperatures, these methods are still limited by the essential step of high-temperature melting (Figure 1A, right).<sup>16–18</sup> An effective approach to bypass this problem is to create inorganic-carbon composite thermoelectric materials via solution-processing techniques such as vacuum filtration, drop-casting, and layer-by-layer assembly.<sup>19–21</sup> While adding conductive carbon frameworks can improve both mechanical flexibility and thermoelectric performance to some degree, current strategies still struggle to optimize both simultaneously.<sup>22,23</sup> Generally, a higher carbon filler content helps build a strong mechanical network but significantly hampers thermoelectric performance because of increased interfacial scattering and reduced carrier mobility.<sup>24</sup> On the other hand, low filler amounts can enhance the  $S$  and  $PF$  through interfacial energy filtering, but insufficient filler connectivity results in weak mechanical support and brittleness. Therefore, achieving a balance of excellent flexibility and high

thermoelectric performance at ultralow carbon loadings remains a key challenge.

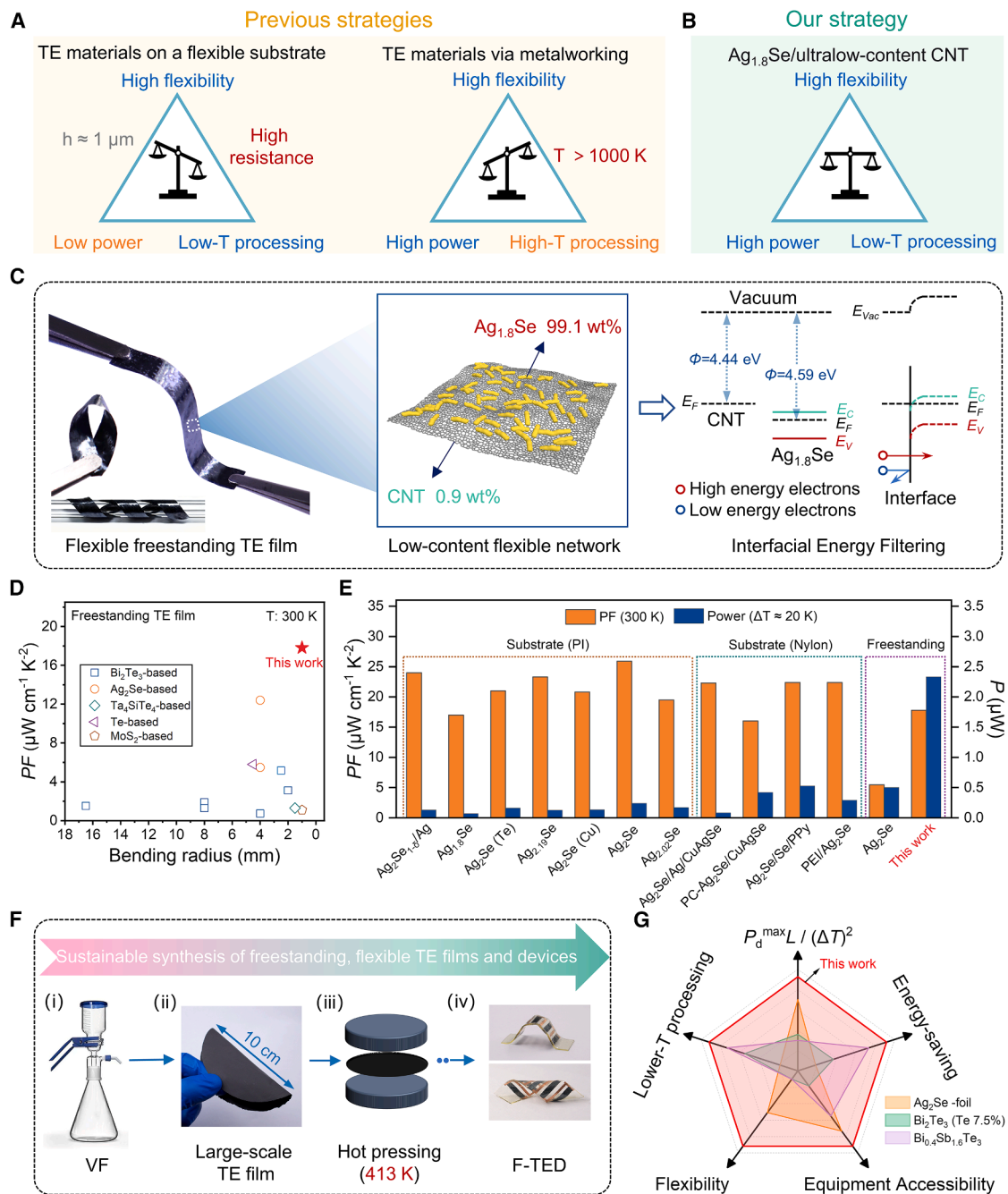
In this work, we report the facile fabrication of freestanding Ag<sub>1.8</sub>Se/carbon nanotube (CNT) composite films, through vacuum filtration followed by hot pressing, which exhibit excellent flexibility, high  $PF$ , enhanced power output, and reduced processing temperatures (Figure 1B). We propose that synergistic enhancement of flexibility and thermoelectric performance at an ultralow CNT content (0.9 wt %) is achieved by precisely controlling the Ag/Se molar ratio and facilitating the stable, uniform dispersion of Ag<sub>1.8</sub>Se with CNT, using *N,N*-dimethylformamide (DMF) as a solvent. We show that the resulting film with a thickness of  $\sim 10$   $\mu\text{m}$  achieves a high  $PF$  of 20.9  $\mu\text{W cm}^{-1} \text{K}^{-2}$  at 380 K, while it exhibits remarkable durability—maintaining over 95% of its performance after 20,000 cycles at a 2-mm bending radius. We fabricated an in-plane flexible thermoelectric generator (F-TEG) comprising five Ag<sub>1.8</sub>Se/CNT legs connected in series, which delivers an outstanding output power ( $P$ ) of 15.4  $\mu\text{W}$  under a temperature difference ( $\Delta T$ ) of 50 K. With biodegradable poly( $\alpha$ -lipoic acid) (PLA) films as encapsulation layers, the developed flexible device showcases excellent recyclability, while the recovered materials retain outstanding performance stability. Finally, we successfully demonstrated that this device functions as a dual-purpose unit for both sensing and power generation in sap flow monitoring, establishing a versatile, efficient, and cost-effective solution for bio-interfaced applications.

## RESULTS

### Ag<sub>1.8</sub>Se/ultralow-CNT-content composites

To achieve synergistic enhancement of thermoelectric performance and mechanical flexibility at low CNT filler contents, it is essential to precisely control the Ag<sub>x</sub>Se-to-CNT ratio while ensuring uniform distribution of both components within the composite system. To accomplish these goals, we propose a strategy that combines optimization of the Ag/Se molar ratio with selection of an appropriate solvent to promote homogeneous dispersion of the nanocomponents. Following this strategy, we synthesized Ag<sub>1.8</sub>Se nanowires and prepared a composite solution in which Ag<sub>1.8</sub>Se and CNTs were uniformly dispersed using DMF as the solvent. We then successfully fabricated freestanding Ag<sub>1.8</sub>Se/CNT composite films via a facile vacuum filtration approach followed by mild-temperature hot pressing (Figure S1).

By incorporating an ultralow-content CNT network (0.9 wt %) as a flexible conductive scaffold within an Ag<sub>1.8</sub>Se (99.1 wt %) matrix, the resulting composites exhibit high thermoelectric performance and excellent flexibility simultaneously (Figures 1C and S2). The uniform distribution of Ag<sub>1.8</sub>Se and CNT facilitates the formation of abundant, well-established Ag<sub>1.8</sub>Se-CNT interfaces, leading to a pronounced interfacial energy-filtering effect that effectively decouples  $\sigma$  and  $S$  (Figures 1C, S3, and S4; Note S2),<sup>22</sup> yielding a remarkable  $PF$  at room temperature. Meanwhile, the percolating CNT network enhances charge transport and mechanical compliance, enabling the film to maintain stable thermoelectric performance even after repeated bending to a radius as small as 1 mm.



**Figure 1.  $\text{Ag}_{1.8}\text{Se}/\text{ultralow-CNT}$ -content composite films and devices**

(A and B) Schematic illustration comparing limited-power or high-temperature (high-T) processing via conventional approaches (A) with high-power and low-T processing via our proposed strategy (B) for preparing flexible inorganic thermoelectric (TE) films.

(C) Schematic illustration showing the structure and electron transport mechanism of  $\text{Ag}_{1.8}\text{Se}/\text{CNT}$  films. The inset shows a bent  $\text{Ag}_{1.8}\text{Se}/\text{CNT}$  film.

(D) Comparison of the  $PF$  and bending radius of freestanding TE films via solution processing, as reported in previous studies and this work.

(E) Comparison of  $PF$  of  $\text{Ag}_2\text{Se}$ -based TE materials and power of five devices (uniform length and width for each leg), as reported in previous studies and this work.

(F) The fabrication steps of the freestanding  $\text{Ag}_{1.8}\text{Se}/\text{CNT}$  film and the flexible TE device (F-TED): (i) vacuum filtration of the mixture of  $\text{Ag}_{1.8}\text{Se}$  and CNT, (ii) the formation of a freestanding and flexible  $\text{Ag}_{1.8}\text{Se}/\text{CNT}$  film, (iii) hot-pressing processing at 413 K for 30 min to improve its density, and (iv) the assembling of the F-TED on a flexible and recyclable PLA film.

(G) Comparison of the multidimensional performance of this work with representative TE materials and devices. The plot's source data are provided in [Table S4](#).

Owing to this compelling combination of high  $PF$  and mechanical flexibility, the Ag<sub>1.8</sub>Se/CNT freestanding film surpasses all flexible freestanding thermoelectric materials via solution processing (Figures 1D and S5; Tables S1 and S2).<sup>19,25–33</sup> A F-TEG constructed with five series-connected Ag<sub>1.8</sub>Se/CNT legs delivers a  $P$  substantially exceeding that of its counterparts fabricated from thermoelectric materials deposited on flexible substrates. Despite the excellent  $PF$  of the materials used in these devices, their submicron-thickness structural design—necessary to maintain flexibility—limits the conversion of high material  $PF$  into effective device output, resulting in extremely low power levels (Figure 1E; Table S3).<sup>30,34–44</sup> Moreover, the fabrication of freestanding Ag<sub>1.8</sub>Se/CNT composite films employs a mild hot-pressing annealing temperature (413 K) and requires neither complex nor costly equipment, thereby reducing energy consumption and enabling the fabrication of large-area films (Figures 1F and S6). The assembled devices can be readily transferred onto diverse substrates, providing enhanced design flexibility and facilitating integration with additional functionalities. A comparative analysis of state-of-the-art flexible thermoelectric materials, assessed based on power density, energy savings, equipment accessibility, flexibility, and processing temperature, underscores the advantages of our Ag<sub>1.8</sub>Se/CNT composite platform (Figure 1G; Table S4).<sup>5,16,45</sup> This approach effectively balances flexibility, power generation, and mild-temperature processability (Figure 1B), providing a strong foundation for future use in self-powered wearable sensors and IoT devices.

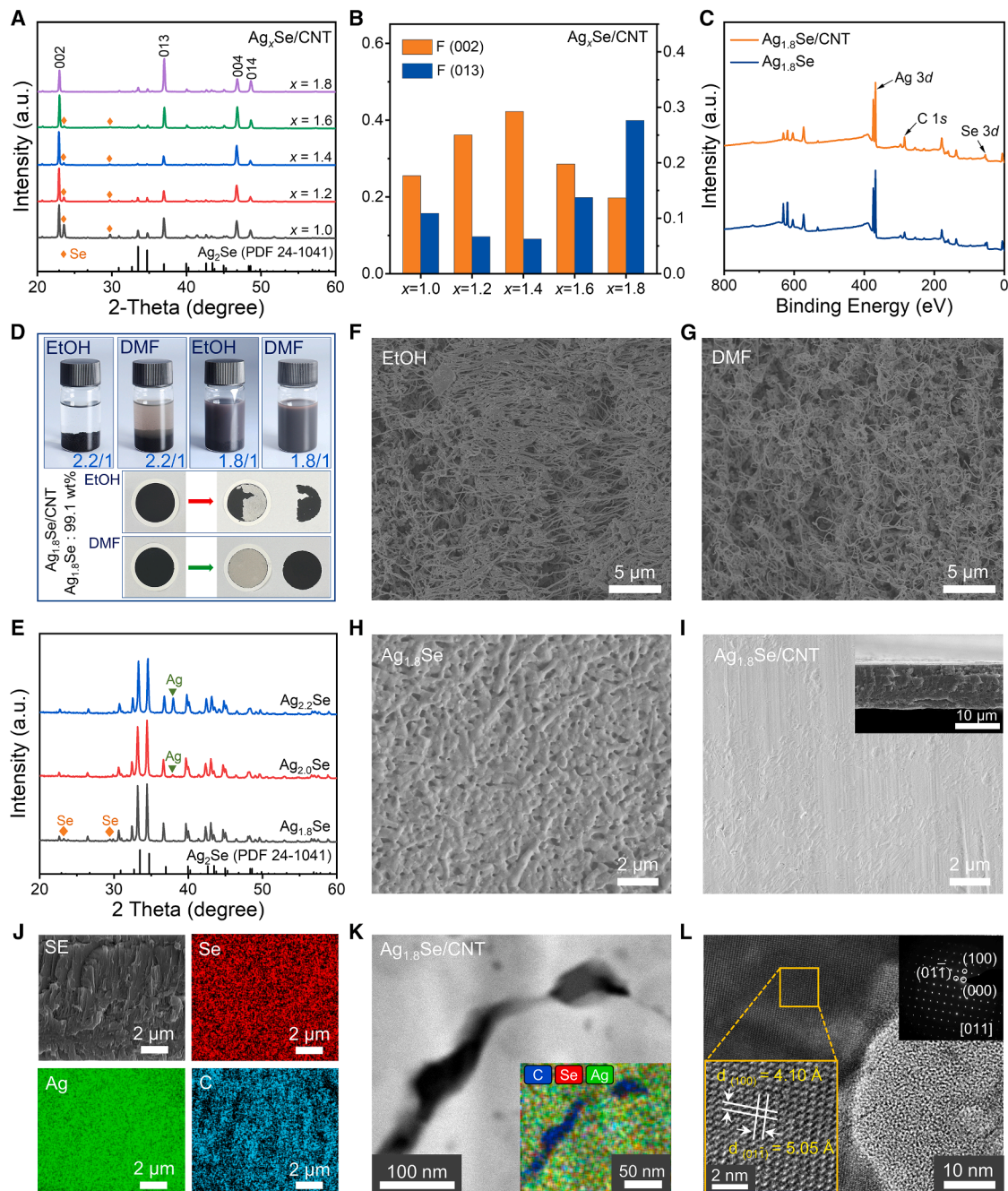
### Characterization of the composite films

To determine the optimal Ag/Se ratio for maximizing thermoelectric performance, we prepared a series of films with nominal compositions Ag<sub>*x*</sub>Se/CNT ( $x = 1.0, 1.2, 1.4, 1.6, 1.8$ ). We focused on  $x < 2$ , as freestanding films are difficult to fabricate at  $x \geq 2$  (see below for details and Figure S7). The CNT content in the Ag<sub>*x*</sub>Se/CNT films was fixed at 0.9 wt % (if not otherwise specified), which was identified to be the minimum loading required to produce flexible freestanding films (Figure S8).

The phase structure of the films was characterized using X-ray diffraction (XRD, Figure 2A). For all samples, the dominant diffraction peaks can be indexed to the  $\beta$ -Ag<sub>2</sub>Se phase (PDF #24-1041). Notably, the phase composition is highly sensitive to the Ag/Se feeding ratio. For samples with lower Ag content ( $x = 1.0, 1.2, 1.4$ , and  $1.8$ ), additional diffraction peaks corresponding to elemental Se are observed alongside the Ag<sub>2</sub>Se phase. The intensity of these Se peaks gradually decreases with increasing  $x$  and nearly disappears at  $x = 1.8$  (Table S5). This trend indicates that when the Ag feeding ratio is below the stoichiometric requirement for Ag<sub>2</sub>Se formation ( $x < 1.8$ ), the reaction remains incomplete due to kinetic limitations associated with Ag diffusion into the Se nanowires, resulting in a two-phase composite of Ag<sub>2</sub>Se and residual Se. Accordingly, the notation “Ag<sub>*x*</sub>Se” used in this work refers to the nominal feeding ratio; the resulting products consist of Ag<sub>2</sub>Se/Se mixtures for  $x < 1.8$  and predominantly single-phase Ag<sub>2</sub>Se at  $x = 1.8$ . As the Ag/Se molar ratio increases, the intensity of the (002) diffraction peak in the Ag<sub>*x*</sub>Se/CNT composite films initially rises and subsequently declines, whereas the (013) diffraction peak exhibits the

opposite trend, decreases at first and then increases. In symmetric  $\theta$ - $2\theta$  XRD measurements, only lattice planes parallel to the film surface are detected. Therefore, enhanced intensity of the (002) or (013) reflection indicates a preferred orientation with these planes aligned parallel to the film plane. As thermoelectric properties are measured along the in-plane direction, charge transport occurs predominantly within these crystallographic planes. The orientation factor analysis reveals that at  $x = 1.8$ , F (013) increases to 0.27, surpassing F (002) as the dominant orientation, which favors enhanced carrier mobility and electrical conductivity (Figure 2B; Note S3). Elemental compositions and oxidation states of Ag<sub>1.8</sub>Se/CNT and Ag<sub>1.8</sub>Se were further analyzed (Figures 2C and S9). The enhanced C 1s intensity observed in Ag<sub>1.8</sub>Se/CNT relative to Ag<sub>1.8</sub>Se supports the successful integration of Ag<sub>1.8</sub>Se and CNT. The binding energy positions of Se 3d<sub>5/2</sub> and Se 3d<sub>3/2</sub> confirm the  $-2$ -oxidation state of Se, characteristic of metal-selenide bonding. The binding energy of Ag 3d in Ag<sub>1.8</sub>Se/CNT composites matches that of pure Ag<sub>1.8</sub>Se, indicating that the incorporation of CNT does not alter the chemical environment of the matrix (Figures S9C and S10). The high-resolution C 1s spectrum (Figure S9D) indicates that carbon is predominantly present as C–C bonds, with a minor contribution from C–O–C species, suggesting slight surface oxidation of the CNTs.

For flexible, freestanding thermoelectric films, achieving a uniform force-bearing morphology is critical, particularly at low CNT loading. This requires well-dispersed components, controlled by both the Ag/Se ratio and the choice of solvent. As illustrated in Figures 2D and S7, when  $x \geq 2$  (such as 2.2), Ag<sub>2.2</sub>Se exhibits poor dispersibility in both EtOH and DMF, resulting in strong adhesion of the film to the substrate and ineffective co-dispersion with CNTs. As evidenced by the contact-angle and X-ray photoelectron spectroscopy (XPS) analyses (Figure S11), excess Ag increases the surface energy and alters the surface chemical states, thereby weakening solvent stabilization and promoting agglomeration (Figures 2E and S12). Reducing  $x$  to 1.8 markedly improves the dispersion of Ag<sub>1.8</sub>Se in DMF, although partial agglomeration still occurs in EtOH. In EtOH, adding CNTs further disrupts dispersion, causing film fracturing during peeling from the nylon substrate. This behavior is attributed to the limited dispersion capability of EtOH for both Ag<sub>1.8</sub>Se and CNT, where van der Waals interactions among CNT promote mutual aggregation, thereby intensifying the agglomeration phenomenon. In contrast, the higher polarity and strong coordination of DMF enable uniform dispersion of Ag<sub>1.8</sub>Se and CNTs, producing flexible composite films that can be easily peeled from the substrate. Scanning electron microscopy (SEM) of the as-prepared vacuum-filtrated Ag<sub>1.8</sub>Se/CNT films further illustrates this effect. EtOH-dispersed films display loose, non-uniform porous structures, arising from inadequate dispersion that promotes particle agglomeration. The formation of large clusters impedes close packing during vacuum filtration, leading to structural heterogeneity (Figures 2F, S13, and S14). DMF-dispersed films exhibit a more uniform morphology than those obtained with EtOH (Figures 2G, S13, and S14), owing to the superior dispersing ability of DMF, which ensures homogeneous distribution of CNT and Ag<sub>1.8</sub>Se and promotes the formation of a densely interconnected, continuous network after vacuum filtration. We further



**Figure 2. Phase, structure, and composition analysis of  $\text{Ag}_x\text{Se}/\text{CNT}$  films**

- (A) XRD patterns of  $\text{Ag}_x\text{Se}/\text{CNT}$  films prepared by hot pressing. Elemental Se is indicated by rhombus symbols.  
 (B) Orientation factors  $F(002)$  and  $F(013)$  of  $\text{Ag}_x\text{Se}/\text{CNT}$  films.  
 (C) X-ray photoelectron spectroscopy (XPS) full spectra for  $\text{Ag}_{1.8}\text{Se}/\text{CNT}$  and  $\text{Ag}_{1.8}\text{Se}$  films.  
 (D) Images of  $\text{Ag}_x\text{Se}$  and CNT dispersions with different Ag/Se molar ratios in EtOH and DMF after ultrasonic treatment for 1 h (top) and corresponding  $\text{Ag}_{1.8}\text{Se}/\text{CNT}$  films peeled off nylon substrates (bottom).  
 (E) XRD patterns of  $\text{Ag}_{2.2}\text{Se}$ ,  $\text{Ag}_{2.0}\text{Se}$ , and  $\text{Ag}_{1.8}\text{Se}$  nanowires.  
 (F and G) Cross-sectional SEM images of  $\text{Ag}_{1.8}\text{Se}/\text{CNT}$  films prepared using EtOH (F) and DMF (G) dispersions.  
 (H and I) Surface SEM images of  $\text{Ag}_{1.8}\text{Se}$  (H) and  $\text{Ag}_{1.8}\text{Se}/\text{CNT}$  (I) films after hot pressing. Inset: SEM cross-section morphology of  $\text{Ag}_{1.8}\text{Se}/\text{CNT}$  film.  
 (J) Secondary electron (SE) cross-sectional SEM image of  $\text{Ag}_{1.8}\text{Se}/\text{CNT}$  film and the corresponding EDS maps of Se, Ag, and C.  
 (K) HAADF-STEM image of the cross-section of  $\text{Ag}_{1.8}\text{Se}/\text{CNT}$  film and corresponding EDS elemental mapping.  
 (L) HRTEM image of the  $\text{Ag}_{1.8}\text{Se}/\text{CNT}$  film, with the insets showing the corresponding enlarged image and the SAED pattern along the [011] direction.

investigated the morphologies of Ag<sub>1.8</sub>Se/CNT films after hot pressing. Compared with the hot-pressed Ag<sub>1.8</sub>Se films (Figures 2H and S15), the hot-pressed Ag<sub>1.8</sub>Se/CNT films exhibit reduced porosity and enhanced density (Figures 2I and S16). This improvement is attributed to the CNT network, which enhances interparticle contact and facilitates effective filling between Ag<sub>1.8</sub>Se nanowires.

To assess the distribution of CNTs within the hot-pressed film, energy-dispersive X-ray spectroscopy (EDS) mapping was investigated (Figure 2J). Ag, Se, and C mapping confirm uniform dispersion of CNTs with Ag<sub>1.8</sub>Se (Figure S17), which is further supported by the SEM image of the fractured sample obtained at room temperature (Figure S18). Additionally, the cross-sectional and surface EDS spectra reveal Ag/Se atomic ratios of 2.0183:1 and 1.9948:1, respectively (Figures S17 and S19). This deviation from the initial feed ratio (1.8:1) is attributed to the volatilization of Se during the hot-pressing process. To further elucidate the microstructure of the Ag<sub>1.8</sub>Se/CNT composite, the samples were systematically characterized by transmission electron microscopy (TEM). High-angle annular dark-field scanning TEM (HAADF-STEM) images show close contact between amorphous and crystalline regions, with CNTs occupying the amorphous phase and Ag<sub>1.8</sub>Se the crystalline phase (Figures 2K and S20), which is further confirmed by EDS elemental mapping analysis. Additionally, high-resolution TEM (HRTEM) images of Ag<sub>1.8</sub>Se near CNTs reveal well-defined lattice fringes with spacings of 4.10 and 5.05 Å, corresponding to the (100) and (011) planes, respectively (Figure 2L). Selected area electron diffraction (SAED) patterns further confirm alignment along the [011] zone axis, thereby verifying the crystal structure of the region.

### Thermoelectric properties and flexibility

To investigate the thermoelectric performance of Ag<sub>x</sub>Se/CNT films, measurements of  $\sigma$ ,  $S$ , and  $PF$  were conducted at 298 K along the in-plane direction. The dependence of thermoelectric properties on the Ag/Se molar ratio is presented in Figures 3A and 3B. With an increasing Ag/Se ratio,  $\sigma$  rises markedly, whereas  $S$  initially shows a slight decrease followed by an increase, resulting in an overall improvement in  $PF$ . Notably, Ag<sub>1.8</sub>Se/CNT films achieve a maximum  $\sigma$  of  $836.7 \pm 47.0$  S cm<sup>-1</sup>, corresponding to a maximum  $PF$  of  $16.8 \pm 1.4$   $\mu$ W cm<sup>-1</sup> K<sup>-2</sup>. The role of CNT content in determining the thermoelectrical performance was further examined. At loadings above 0.9 wt %, the  $PF$  of Ag<sub>x</sub>Se/CNT films declines sharply, demonstrating that excess CNT incorporation degrades thermoelectric performance (Figure S21).

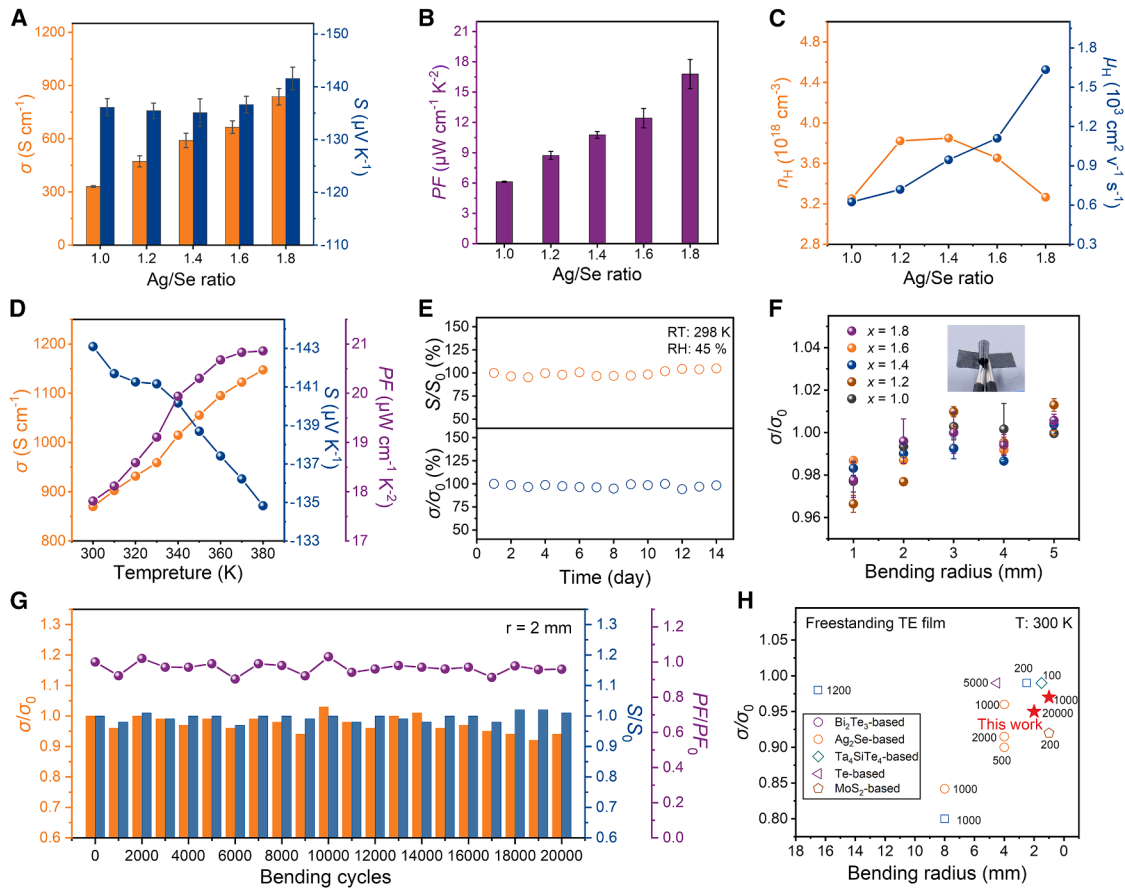
To elucidate the origin of the variations of  $\sigma$ ,  $S$ , and  $PF$  with Ag/Se ratio, Hall effect measurements were performed to determine the carrier concentration ( $n_H$ ) and carrier mobility ( $\mu_H$ ). As shown in Figure 3C,  $n_H$  initially increases and then decreases with increasing Ag/Se ratio, confirming the variation in  $S$ . This behavior is attributed to the phase transition from a mixed Ag<sub>2</sub>Se and Se system to a single-phase Ag<sub>2</sub>Se, where the Se diffraction peak gradually disappears and eventually vanishes as the Ag/Se ratio increases, which is further corroborated by EDS analysis (Table S5). The EDS results show that the actual Ag/Se atomic ratio increases progressively with the nominal composition,

reaching  $\sim 1.995$  for the Ag<sub>1.8</sub>Se/CNT film, corresponding to nearly stoichiometric Ag<sub>2</sub>Se. In contrast,  $\mu_H$  increases steadily as the Ag/Se ratio rises (reduction in high-resistance phase Se), reaching an exceptional value of  $1,634$  cm<sup>2</sup> v<sup>-1</sup> s<sup>-1</sup> at room temperature for the Ag<sub>1.8</sub>Se/CNT film. This remarkable  $\mu_H$  arises from a single Ag<sub>2</sub>Se phase combined with the incorporation of CNTs, which exhibit high intrinsic mobility and form a continuous conductive network with Ag<sub>1.8</sub>Se nanowires, thereby strengthening inter-nanowire connectivity and enabling efficient carrier transport. The synergy between high  $\mu_H$  and moderate  $n_H$  ensures enhanced  $\sigma$  while maintaining a sufficiently high  $S$ , thereby contributing to the excellent  $PF$  of the composite film. It is worth noting that the monotonic increase in  $\mu_H$  and  $PF$  with Ag/Se ratio, in contrast to the non-monotonic evolution of the (002) or (013) orientation, suggests that the enhanced  $\mu_H$  and  $PF$  are predominantly governed by the optimization of the Ag<sub>2</sub>Se stoichiometry, rather than by the orientation change.

The temperature-dependent performance of the Ag<sub>1.8</sub>Se/CNT film was analyzed over the range of 300–380 K. As shown in Figure 3D,  $\sigma$  increases while absolute  $S$  decreases with temperature, yielding a maximum  $PF$  of  $20.9$   $\mu$ W cm<sup>-1</sup> K<sup>-2</sup> at 380 K. To assess the practical applicability of the Ag<sub>1.8</sub>Se/CNT composite films, we examined their long-term stability and mechanical flexibility. Stability tests over 15 days show negligible changes in  $\sigma$  and  $S$ , confirming excellent performance stability (Figures 3E and S22). Mechanical flexibility was analyzed by measuring the normalized resistance at different bending radii. As demonstrated in Figures 3F and S23, the Ag<sub>x</sub>Se/CNT films display superior mechanical flexibility. In particular, the  $\sigma$  of the Ag<sub>1.8</sub>Se/CNT film decreased by less than 3% relative to its initial value after 1,000 bending cycles at a bending radius of 1 mm, demonstrating excellent durability for flexible thermoelectric applications. Moreover, after 20,000 cyclic bending tests at a radius of 2 mm, the thermoelectric performance remains stable, with  $\sigma$ ,  $S$ , and  $PF$  retaining over 94%, 101%, and 95% of their initial values, respectively (Figures 3G and S24). Compared with previously reported flexible freestanding materials, the Ag<sub>1.8</sub>Se/CNT films exhibit highly competitive flexibility (Figure 3H; Table S2).<sup>19,25–33</sup> Additionally, to evaluate practical durability under real-world conditions, we further investigated the material's performance under multi-axis bending, twisting, and coupled thermo-mechanical loading. The results demonstrate excellent stability of the material properties under these conditions (Figure S25).

Furthermore, the influence of film thickness on thermoelectric performance and flexibility was characterized (Figures S26 and S27). Decreasing the film thickness enhances  $\sigma$ ,  $S$ , and  $PF$ , owing to the larger grain size and denser structure that make the hot-pressed films more similar to the properties of two-dimensional materials. The variation in flexibility stems from the cubic relationship between bending stiffness and thickness, which inherently makes thinner films more flexible. When reduced to below 10  $\mu$ m, the films become difficult to peel from the nylon substrate owing to insufficient mechanical strength. Thus, a thickness of  $\sim 10$   $\mu$ m represents the optimal compromise between thermoelectric performance and flexibility.

Overall, these observations underscore three key requirements for achieving an optimal balance between high thermoelectric efficiency and mechanical flexibility: (1) achieving a



**Figure 3. TE performance and flexibility of Ag<sub>x</sub>Se/CNT films**

(A and B) Room-temperature  $\sigma$ , and  $S$  (A) of Ag<sub>1.8</sub>Se/CNT films with different Ag/Se molar ratios and their corresponding  $PF$  (B). The error bars represent three independent measurements.

(C) Room-temperature  $n_H$  and  $\mu_H$  of Ag<sub>1.8</sub>Se/CNT film with varying Ag/Se molar ratios.

(D) TE properties of Ag<sub>1.8</sub>Se/CNT film between 300 and 380 K.

(E) Variations of Seebeck coefficient ( $S/S_0$ ) and electrical conductivity ( $\sigma/\sigma_0$ ) of Ag<sub>1.8</sub>Se/CNT film at 298 K and 45% RH over 15 days.

(F) Conductivity changes ( $\sigma/\sigma_0$ ) of Ag<sub>x</sub>Se/CNT films under different bending radii over 1,000 cycles. The error bars represent three independent measurements.

(G) TE property variations of Ag<sub>1.8</sub>Se/CNT film at a 2-mm bending radius after 20,000 bending cycles.

(H) Relative conductivity ( $\sigma/\sigma_0$ ) of freestanding TE films as a function of bending radius after different numbers of bending cycles (with data values indicated near the corresponding points).

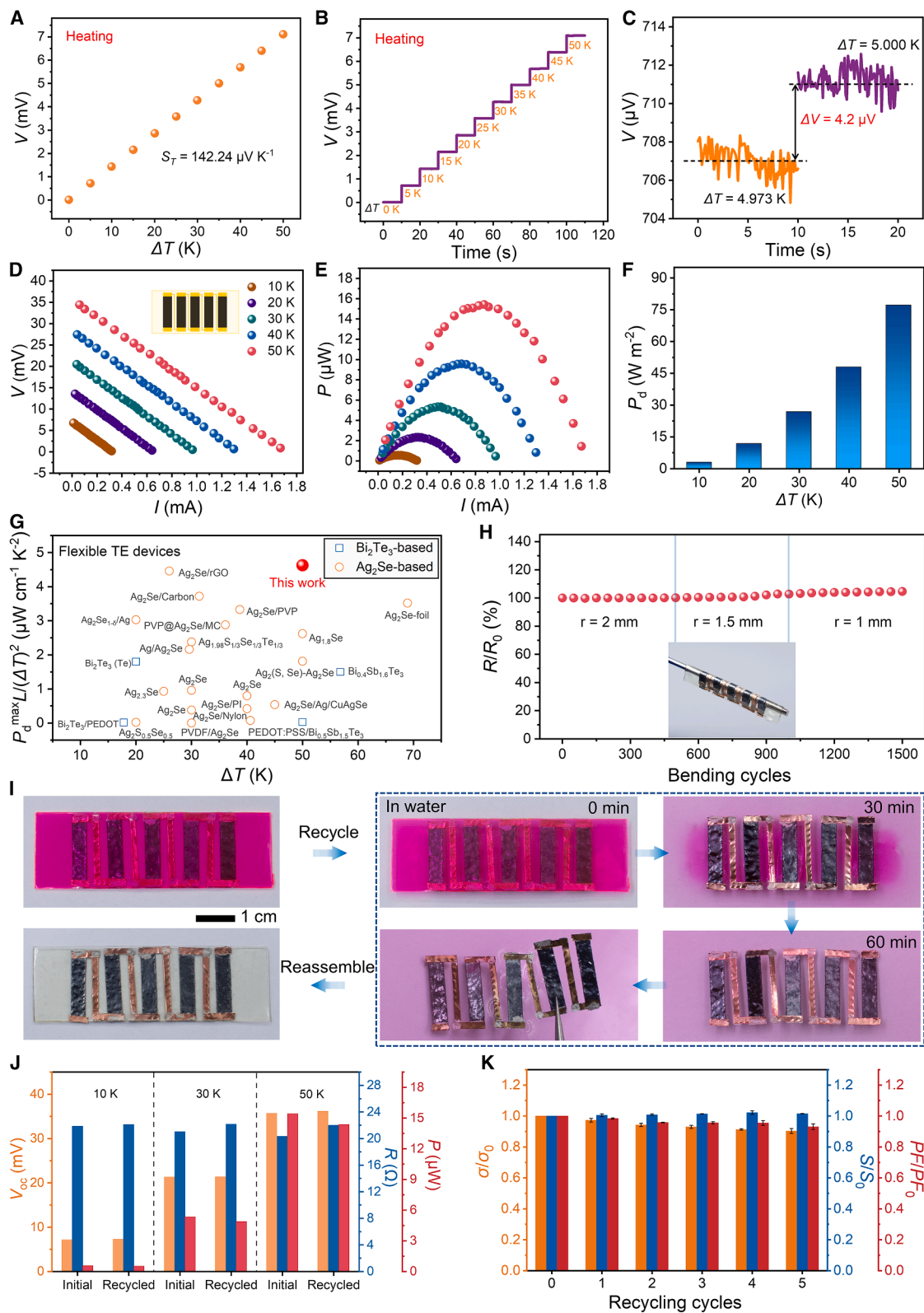
high Ag/Se ratio while avoiding nanowires agglomeration, (2) ensuring uniform distribution of all components to facilitate the formation of abundant and stable interfacial contacts between them, and (3) minimizing the CNT filler concentration and film thickness without compromising the integrity of the freestanding structure.

### High-performance recyclable thermoelectric device

Leveraging the properties-optimized Ag<sub>1.8</sub>Se/CNT films, we constructed flexible thermoelectric devices to assess their potential utilities for sensing and power generation. We first fabricated a thermoelectric sensor comprising a single thermoelectric leg. The thermoelectric sensor exhibits a strong linear relationship between the output voltage ( $V$ ) and the  $\Delta T$ , with a slope of 142.24  $\mu V/K$  corresponding to the  $S$  of the film (Figure 4A). At a fixed  $\Delta T$ , the device provides a stable  $V$  output,

confirming reliable temperature sensing (Figure 4B). Additionally, when  $\Delta T$  is introduced through cooling, the device maintains similar performance, demonstrating good usage stability (Figure S28). Notably, under proportion integration differentiation (PID)-controlled temperature regulation, the device delivered a  $V$  of 4.2  $\mu V$  at an ultra-small  $\Delta T$  of 0.027 K (Figure 4C), highlighting its remarkable sensitivity and suitability for high-precision temperature sensing under minimal thermal gradients.

To further evaluate the potential for power generation, we fabricated an F-TEG comprising five thermoelectric legs connected in series (inset, Figure 4D). For this study, the F-TEG was integrated with a soft, comfortable, and biodegradable PLA film to demonstrate its recyclability and favorable utility for bio-interfaces. With this device design, we systematically studied its output performance.



(legend on next page)

The open-circuit voltage ( $V_{oc}$ ) of the generator increases linearly with rising  $\Delta T$ , reaching 35.65 mV at a  $\Delta T$  of 50 K, in close agreement with the theoretical prediction (Figure S29A). The actual output performance of a thermoelectric generator under a fixed  $\Delta T$  can be evaluated by measuring the  $V$ , the loading current ( $I$ ), and the resulting  $P$  ( $P = VI$ ) when connected to different external load resistors in series. As shown in Figures 4D and 4E, the  $V$  decreases proportionally with the  $I$ , while the  $P$  demonstrates a parabolic dependence on  $I$ . Moreover, both  $V$  and  $I$  increased with rising  $\Delta T$ . At a  $\Delta T$  of 50 K, the generator can deliver a maximum  $P$  of 15.4  $\mu\text{W}$  at an external load resistance of 20  $\Omega$  (Figures S29B and S29C). For comparative evaluation of device performance, the power density ( $P_d$ ) and the normalized  $P_d$  were calculated. As shown in Figure 4F, the device achieves a  $P_d$  of 77.14  $\text{W m}^{-2}$  at a  $\Delta T$  of 50 K. Moreover, the normalized  $P_d$  exhibits a slight increase with rising  $\Delta T$  (Figure S30, which can be attributed to a gradual reduction in device resistance at higher  $\Delta T$ ). The maximum normalized  $P_d$  reaches 4.63  $\mu\text{W cm}^{-1} \text{K}^{-2}$ , offering a marked advantage over previously reported flexible thermoelectric devices (Figure 4G; Table S6).<sup>5,16,17,23,26,29–33,39,44–55</sup> This superior performance is primarily attributed to the high  $PF$  of the Ag<sub>1.8</sub>Se/CNT composite and the low interface resistance of the device.

We then characterized the flexibility and recyclability of the device. The device was subjected to repeated mechanical bending with different radii. As illustrated in Figure 4H, the device exhibits a resistance increase of less than 7% after 500 bending cycles, confirming its excellent mechanical flexibility and structural stability. In addition, the recyclability of the device was evaluated through water-based testing. After 60 min of immersion, the PLA film is fully degraded, thereby releasing the F-TEG device from the matrix (Figures 4I and S31). After retrieval from the solution, the device remains structurally intact and is suitable for reuse. The PLA solution can be recovered and reprocessed into films, exhibiting negligible changes in both the structures and mechanical properties before and after recycling (Figures S32 and S33). Using these recycled components, F-TEG devices were remanufactured, and their output performance as well as thermoelectrical properties were subsequently evaluated (Figures 4J and 4K). To provide a more quantitative understanding of the recycling process, we further investigated the evolution of material properties. As shown in Figure S34, the morphology and thickness of the films remain virtually unchanged after multiple recycling cycles. However, while  $\mu_H$  is largely maintained, the  $n_H$  gradually decreases with cycling,

which accounts for the observed slight reduction in  $\sigma$  and the corresponding increase in  $S$ . Notably, the  $PF$  degradation rate remains below 7% even after 5 cycles. This also explains why recycled devices exhibit an increased  $V_{oc}$  and internal resistance, along with a reduced  $P$  (<7%), compared with initial devices. Furthermore, the entire recycling loop—for both the device and the PLA substrate—employs pure water as the sole solvent (Figure S32), relying exclusively on dissolution and evaporation without any toxic organic additives. This protocol effectively avoids secondary chemical pollution, ensuring high sustainability and minimal environmental impact. These results highlight the substantial potential of our Ag<sub>1.8</sub>Se/CNT-based flexible devices for sustainable power generation applications.

### Applications for sap flow monitoring

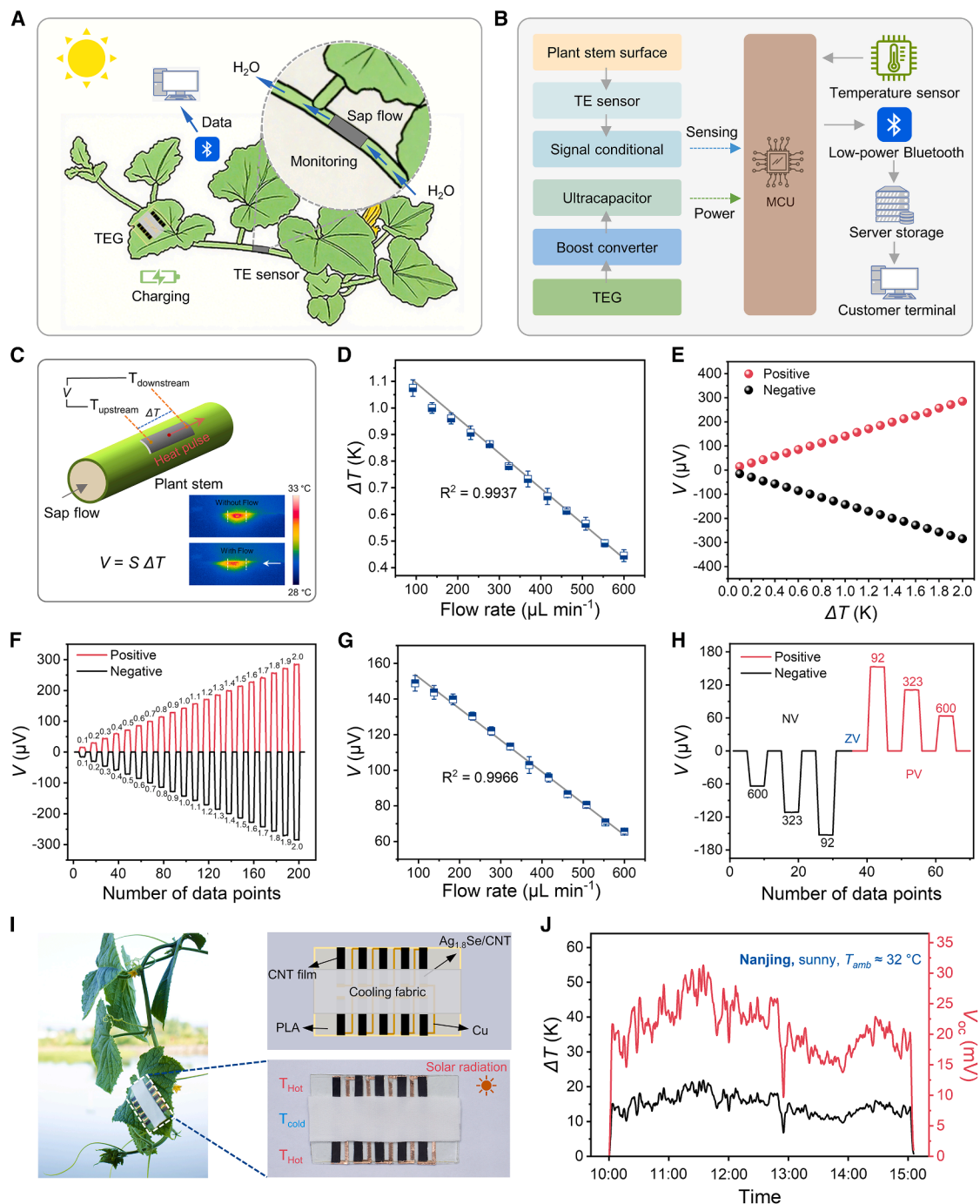
Sap flow functions as the “pulse of life” for plants. Precise and continuous monitoring of sap movement is crucial for understanding plant water status, diagnosing health issues, and enabling precision irrigation in smart farming. Furthermore, high-quality sap flow data provide essential input for ecological hydrology, watershed modeling, and climate-change-related carbon-water coupling studies. However, commercial sap flow monitoring methods, such as thermal diffusion probes and thermal balance techniques, often rely on rigid sensor components that can cause plant tissue damage during installation and often fail to maintain stable performance as plants grow or undergo environmental fluctuations.

In contrast, the flexible Ag<sub>1.8</sub>Se/CNT-based thermoelectric devices developed here offer a mechanically compliant and tissue-friendly alternative. Of particular significance, their highly sensitive sensing capabilities, robust power output, and excellent flexibility enable reliable operation under the subtle thermal gradients associated with plant transpiration. In addition, the Ag<sub>1.8</sub>Se/CNT films display favorable biocompatibility (Figure S35), and encapsulation with biodegradable PLA layers<sup>56</sup> further ensures environmental safety and stable integration with biological tissues. Leveraging these advantages, we validate the feasibility of using these devices as multifunctional bio-interfaces capable of simultaneously sensing sap flow and harvesting energy through a proof-of-concept demonstration (Figure 5A).

Figure 5B describes the working mechanism of the self-powered wearable plant device within the monitoring system. During heater operation (active excitation source), sap flow generates a  $\Delta T$  across the sensor ends, producing a  $V$  that is conditioned and transmitted to the microcontroller unit (MCU) for processing to

### Figure 4. Performance, stability, and recyclability of Ag<sub>1.8</sub>Se/CNT devices

- (A) Dependence of output voltage ( $V$ ) of a single Ag<sub>1.8</sub>Se/CNT device on temperature differences ( $\Delta T$ ).  
 (B) Stable and continuous  $V$  output of a single Ag<sub>1.8</sub>Se/CNT device as a function of time.  
 (C) High-sensitive response of a single Ag<sub>1.8</sub>Se/CNT device to a small temperature change ( $\Delta T = 0.027$  K).  
 (D and E) Output voltages (D) and powers ( $P$ ) (E) at different  $\Delta T$  for the five series-connected devices as a function of output current ( $I$ ).  
 (F)  $P_d$  ( $P_d = P/NWH$ , where  $N$  denotes the number of TE legs,  $W$  is the leg width, and  $H$  is the leg thickness) of the device at different  $\Delta T$ .  
 (G) Comparison of the normalized power density ( $P_d L/(\Delta T)^2$ , where  $L$  represents the effective length of the TE leg excluding the electrode) at maximum  $P$  versus  $\Delta T$  of flexible TE devices, as reported in previous studies and this work.  
 (H) Variation in device resistance after continuous bending under different bending radii.  
 (I) The water-based recycling and reassembling of the TE devices encapsulated with PLA films. For visualization of the recycling process, the PLA film was stained with Rhodamine B.  
 (J) Comparison of initial and recycled  $V$ ,  $R$ , and  $P$  values of the device.  
 (K) Comparison of TE properties of the initial and recycled Ag<sub>1.8</sub>Se/CNT films. The error bars represent three independent measurements.



**Figure 5. Utilities of Ag<sub>1.8</sub>Se/CNT devices for plant sap flow monitoring**

(A) Schematic diagram of a TE sensing device for plant sap flow monitoring and a generator (TEG) for power supply.

(B) Operating principles of the plant sap flow monitoring system.

(C) Schematic of the TE sensor for plant sap flow monitoring, with the inset showing upstream and downstream spatial temperature distributions under fixed heating, both with and without sap flow.

(D) Upstream and downstream  $\Delta T$  as a function of sap flow rate (heating temperature: 34°C). The error bars represent three independent measurements.

(E) Dependence of  $V$  of the TE sensor on  $\Delta T$ .

(F) Continuous  $V$  output under different  $\Delta T$ .

(legend continued on next page)

quantify the sap flow rate. The acquired data are then transmitted to a remote server via a Bluetooth module, enabling real-time monitoring through terminal devices. Meanwhile, the F-TEG harvests electrical energy by exploiting the  $\Delta T$  caused by solar radiation. The harvested energy is regulated by a boost circuit and stored in a supercapacitor, providing a stable, continuous power supply to the MCU and the connected signal conditioning circuit.

Figure 5C illustrates the operating mechanism of the sensor, where activating the heating plate causes a rapid increase in local temperature (Figure S36). When simulating sap flow occurs, convective heat transfer along the flow direction induces anisotropic heat propagation within the plant stem, thereby generating a  $\Delta T$  across the sensor ends (Note S4). Infrared imaging (inset, Figure 5C) shows the anisotropic temperature distribution under conditions with and without sap flow, serving as an indicator of flow direction and rate. Measurements under varying simulated flow rates demonstrate that the  $\Delta T$  across the sensor exhibits a clear linear relationship with flow rate (Figures 5D and S37). Meanwhile, because of the strong linear relationship between  $V$  and  $\Delta T$ , changes in  $V$  can be directly used to monitor  $\Delta T$ , even under a small  $\Delta T$  (<2 K, Figures 5E and 5F). Notably, the polarity of  $V$  directly reflects the direction of  $\Delta T$ , with positive and negative gradients producing corresponding positive and negative voltages. Thus, a correlation between  $V$  and flow rate can be established, demonstrating a strong linear relationship (Figure 5G). Moreover, the  $V$  response (positive or negative) allows for direct determination of the sap flow direction (Figure 5H). Additionally, the excellent voltage stability across varying  $\Delta T$  and flow rates further confirms the reliability of this approach for sap flow monitoring (Figures 5F and 5H). Compared with other time-based detection methods, this approach provides higher efficiency and simpler operation.<sup>57,58</sup>

By integrating an F-TEG onto a plant leaf, we demonstrate its capability to power the sap flow monitoring system. In this scenario, the  $\Delta T$  induced by solar radiation can be harnessed to generate electricity. Specifically, we designed a device consisting of 10 series-connected thermoelectric legs, with CNT films and Al<sub>2</sub>O<sub>3</sub>-doped silk fabric (Figure S38) serving as photothermal conversion and radiative cooling layers, respectively.<sup>59</sup> This configuration establishes a  $\Delta T$  across the thermoelectric legs (Figure 5I). Outdoor experiments demonstrate that a maximum  $\Delta T$  of 21.9 K can be achieved during daytime operation, resulting in a  $V_{oc}$  of 31.3 mV (Figure 5J).

Overall, through a proof-of-concept demonstration, we show that integrating thermoelectric sensing with energy-harvesting devices enables simultaneous sap flow monitoring and on-site power generation under controlled conditions. By strategically optimizing monitoring intervals, implementing dormancy-activation schemes for system components, and compensating for environmental temperature fluctuations, this approach

establishes a robust foundation for long-term, stable monitoring and reliable data transmission in practical plant sap flow applications.

## DISCUSSION

In summary, we have successfully developed the facile fabrication of freestanding Ag<sub>1.8</sub>Se/CNT thermoelectric composite films that overcome the long-standing trade-offs among mechanical flexibility, thermoelectric performance, power output, and sustainable manufacturing, through refined structural design. With an ultralow CNT content of 0.9 wt %, the composite achieves an exceptional  $PF$  of 20.9  $\mu\text{W cm}^{-1} \text{K}^{-2}$  at 380 K, outstanding flexibility enduring 20,000 bending cycles at a 2-mm radius, and facile fabrication under a mild processing temperature of 413 K. The resulting flexible generator exhibits a compelling  $P$  of 15.4  $\mu\text{W}$  and a normalized  $P_d$  of 4.63  $\mu\text{W cm}^{-1} \text{K}^{-2}$  at a  $\Delta T$  of 50 K. The remarkable performance of the composite film and the derived devices stems mainly from the optimized Ag/Se molar ratio and the homogeneous co-dispersion of Ag<sub>1.8</sub>Se and CNTs within the matrix, and it is anticipated to be further enhanced through the involvement of microstructural optimization strategies, such as orientation engineering. The assembled device combines high-sensitivity sensing, a continuous and stable self-powered energy supply, and adaptability to diverse surfaces, expanding its utility to flexible electronics, wearable technologies, and non-planar thermal management well beyond the applications demonstrated here. In particular, the excellent biocompatibility of the components creates opportunities for bio-integrated thermoelectric systems operating at the biology-technology interface. Moreover, the capacity to conform to various substrates presents a promising pathway toward multifunctional devices with improved versatility and adaptability. Notably, the structural design strategy presented here is not material-specific and can be applied to other inorganic thermoelectric systems (Figure S39), paving the way for high-performance, flexible, and sustainable electronics in next-generation wearable and IoT technologies.

## METHODS

Detailed methods can be found in the [supplemental information](#).

## RESOURCE AVAILABILITY

### Lead contact

Requests for further information and resources should be directed to and will be fulfilled by the lead contact, Yu Wang ([yuwang87@nju.edu.cn](mailto:yuwang87@nju.edu.cn)).

### Materials availability

All materials generated in this study will be made available by the [lead contact](#) upon reasonable request.

(G) Correlation between  $V$  of the TE sensor and plant sap flow rate. The error bars represent three independent measurements.

(H) Relationship between TE sensor  $V$ , plant sap flow rate, and flow direction.

(I) A TEG attached to a plant leaf, along with its detailed structural organization, including both a schematic illustration and the corresponding photograph.

(J) Measured  $\Delta T$  and  $V$  of the TEG under daytime exposure.

### Data and code availability

The data and code that support the conclusions of this study are available from the [lead contact](#) upon reasonable request.

### ACKNOWLEDGMENTS

This work was supported by the National Key R&D Program of China (no. 2022YFA1203700 [Y.W.], no. 2022YFA1405000 [Y.-Q.L.], and no. 2021YFA1202000 [Y.W.]); the National Natural Science Foundation of China (no. 62175102 [Y.W.] and no. T2488302 [Y.-Q.L.]); the Natural Science Foundation of Jiangsu Province (no. BK20243067 [Y.-Q.L.]); and the Fundamental Research Funds for the Central Universities (Y.W.).

### AUTHOR CONTRIBUTIONS

Y.W. and G.L. conceived the idea and designed the research. G.L. synthesized and characterized the thermoelectric films and devices. X.-R.G. participated in the preparation and characterization of the PLA film. T.W. and J.W. assisted in material morphology characterization. All authors contributed to the data analysis. G.L., Y.W., G.C., and Y.-Q.L. wrote and revised the manuscript. Y.W., G.C., and Y.-Q.L. supervised the research. All authors approved the final version of the manuscript.

### DECLARATION OF INTERESTS

The authors declare no competing interests.

### DECLARATION OF GENERATIVE AI AND AI-ASSISTED TECHNOLOGIES IN THE WRITING PROCESS

During the preparation of this work, the authors used ChatGPT (OpenAI) to improve the readability and language of the manuscript. After using this tool, the authors reviewed and edited the content as needed and take full responsibility for the content of the publication.

### SUPPLEMENTAL INFORMATION

Supplemental information can be found online at <https://doi.org/10.1016/j.joule.2026.102487>.

Received: December 23, 2025

Revised: April 1, 2026

Accepted: April 23, 2026

### REFERENCES

- Xiao, Y., and Zhao, L.D. (2020). Seeking new, highly effective thermoelectrics. *Science* 367, 1196–1197. <https://doi.org/10.1126/science.aaz9426>.
- Tang, W., Sun, Q., and Wang, Z.L. (2023). Self-powered sensing in wearable electronics-A paradigm shift technology. *Chem. Rev.* 123, 12105–12134. <https://doi.org/10.1021/acs.chemrev.3c00305>.
- Wan, C., Gu, X., Dang, F., Itoh, T., Wang, Y., Sasaki, H., Kondo, M., Koga, K., Yabuki, K., Snyder, G.J., et al. (2015). Flexible n-type thermoelectric materials by organic intercalation of layered transition metal dichalcogenide TiS<sub>2</sub>. *Nat. Mater.* 14, 622–627. <https://doi.org/10.1038/nmat4251>.
- Yang, Q., Yang, S., Qiu, P., Peng, L., Wei, T.R., Zhang, Z., Shi, X., and Chen, L. (2022). Flexible thermoelectrics based on ductile semiconductors. *Science* 377, 854–858. <https://doi.org/10.1126/science.abq0682>.
- Chen, W., Shi, X.L., Li, M., Liu, T., Mao, Y., Liu, Q., Dargusch, M., Zou, J., Lu, G.Q.M., and Chen, Z.G. (2024). Nanobinders advance screen-printed flexible thermoelectrics. *Science* 386, 1265–1271. <https://doi.org/10.1126/science.ads5868>.
- Mao, J., Liu, Z., Zhou, J., Zhu, H., Zhang, Q., Chen, G., and Ren, Z. (2018). Advances in thermoelectrics. *Adv. Phys.* 67, 69–147. <https://doi.org/10.1080/00018732.2018.1551715>.
- Cao, T., Shi, X.L., and Chen, Z.G. (2023). Advances in the design and assembly of flexible thermoelectric device. *Prog. Mater. Sci.* 137, 101003. <https://doi.org/10.1016/j.pmatsci.2022.101003>.
- Zheng, Z.H., Shi, X.L., Ao, D.W., Liu, W.D., Li, M., Kou, L.Z., Chen, Y.X., Li, F., Wei, M., Liang, G.X., et al. (2023). Harvesting waste heat with flexible Bi<sub>2</sub>Te<sub>3</sub> thermoelectric thin film. *Nat. Sustain.* 6, 180–191. <https://doi.org/10.1038/s41893-022-01003-6>.
- Lu, Y., Zhou, Y., Wang, W., Hu, M., Huang, X., Mao, D., Huang, S., Xie, L., Lin, P., Jiang, B., et al. (2023). Staggered-layer-boosted flexible Bi<sub>2</sub>Te<sub>3</sub> films with high thermoelectric performance. *Nat. Nanotechnol.* 18, 1281–1288. <https://doi.org/10.1038/s41565-023-01457-5>.
- Li, D., Shi, X.L., Zhu, J., Cao, T., Ma, X., Li, M., Han, Z., Feng, Z., Chen, Y., Wang, J., et al. (2024). High-performance flexible p-type Ce-filled Fe<sub>3</sub>CoSb<sub>12</sub> skutterudite thin film for medium-to-high-temperature applications. *Nat. Commun.* 15, 4242. <https://doi.org/10.1038/s41467-024-48677-4>.
- Kim, S.J., Lee, H.E., Choi, H., Kim, Y., We, J.H., Shin, J.S., Lee, K.J., and Cho, B.J. (2016). High-performance flexible thermoelectric power generator using laser multiscanning lift-off process. *ACS Nano* 10, 10851–10857. <https://doi.org/10.1021/acsnano.6b05004>.
- Kim, S.J., We, J.H., and Cho, B.J. (2014). A wearable thermoelectric generator fabricated on a glass fabric. *Energy Environ. Sci.* 7, 1959–1965. <https://doi.org/10.1039/c4ee00242c>.
- Liang, Z., Shu, R., Xu, C., Wang, Y., Shang, H., Mao, J., and Ren, Z. (2025). Substrate-free inorganic-based films for thermoelectric applications. *Adv. Mater.* 37, e2416394. <https://doi.org/10.1002/adma.202416394>.
- Wu, H., Shi, X.L., Mao, Y., Li, M., Liu, W.D., Wang, D.Z., Yin, L.C., Zhu, M., Wang, Y., Duan, J., et al. (2023). Optimized thermoelectric performance and plasticity of ductile semiconductor Ag<sub>2</sub>S<sub>0.5</sub>Se<sub>0.5</sub> via dual-phase engineering. *Adv. Energy Mater.* 13, 2302551. <https://doi.org/10.1002/aenm.202302551>.
- Wu, H., Shi, X., Duan, J., Liu, Q., and Chen, Z.G. (2023). Advances in Ag<sub>2</sub>Se-based thermoelectrics from materials to applications. *Energy Environ. Sci.* 16, 1870–1906. <https://doi.org/10.1039/d3ee00378g>.
- Ding, W., Shen, X., Jin, M., Hu, Y., Chen, Z., Meng, E., Luo, J., Li, W., and Pei, Y. (2024). Robust bendable thermoelectric generators enabled by elasticity strengthening. *Nat. Commun.* 15, 9767. <https://doi.org/10.1038/s41467-024-54084-6>.
- Wu, H., Shi, X.L., Li, M., Gao, H., Liu, W.D., Zhu, M., Yin, L.C., Wang, D.Z., Duan, J., Chen, Z.G., et al. (2025). Sandwich engineering advances ductile thermoelectrics. *Adv. Mater.* 37, e2503020. <https://doi.org/10.1002/adma.202503020>.
- Gao, Z., Yang, S., Ma, Y., Wei, T.R., Chen, X., Zheng, W., Qiu, P., Zeng, X., Chen, L., and Shi, X. (2025). Warm metalworking for plastic manufacturing in brittle semiconductors. *Nat. Mater.* 24, 1538–1544. <https://doi.org/10.1038/s41563-025-02223-9>.
- Chen, Z., Lv, H., Zhang, Q., Wang, H., and Chen, G. (2022). Construction of a cement-rebar nanoarchitecture for a solution-processed and flexible film of a Bi<sub>2</sub>Te<sub>3</sub>/CNT hybrid toward low thermal conductivity and high thermoelectric performance. *Carbon Energy* 4, 115–128. <https://doi.org/10.1002/cey2.161>.
- Jin, Q., Jiang, S., Zhao, Y., Wang, D., Qiu, J., Tang, D.M., Tan, J., Sun, D.M., Hou, P.X., Chen, X.Q., et al. (2019). Flexible layer-structured Bi<sub>2</sub>Te<sub>3</sub> thermoelectric on a carbon nanotube scaffold. *Nat. Mater.* 18, 62–68. <https://doi.org/10.1038/s41563-018-0217-z>.
- Liu, Y.M., Shi, X.L., Wu, T., Wu, H., Mao, Y., Cao, T., Wang, D.Z., Liu, W.D., Li, M., Liu, Q., et al. (2024). Boosting thermoelectric performance of single-walled carbon nanotubes-based films through rational triple treatments. *Nat. Commun.* 15, 3426. <https://doi.org/10.1038/s41467-024-47417-y>.

22. Wang, H., Liu, X., Zhou, Z., Wu, H., Chen, Y., Zhang, B., Wang, G., Zhou, X., and Han, G. (2022). Constructing n-type Ag<sub>2</sub>Se/CNTs composites toward synergistically enhanced thermoelectric and mechanical performance. *Acta Mater.* 223, 117502. <https://doi.org/10.1016/j.actamat.2021.117502>.
23. Zhang, L., Shi, X.L., Shang, H., Gu, H., Chen, W., Li, M., Huang, D., Dong, H., Wang, X., Ding, F., et al. (2025). High-performance Ag<sub>2</sub>Se-based thermoelectrics for wearable electronics. *Nat. Commun.* 16, 5002. <https://doi.org/10.1038/s41467-025-60284-5>.
24. Lv, W., Tang, W., Yu, P., Kainat, J., Zhou, Z., Lan, J.L., Yang, X., and Lin, Y.H. (2025). Polymorphic interface engineering for high-performance Ag<sub>2</sub>Se-based flexible thermoelectric device. *J. Colloid Interface Sci.* 699, 138307. <https://doi.org/10.1016/j.jcis.2025.138307>.
25. Wang, Y., Hong, M., Liu, W.D., Shi, X.L., Xu, S.D., Sun, Q., Gao, H., Lu, S., Zou, J., and Chen, Z.G. (2020). Bi<sub>0.5</sub>Sb<sub>1.5</sub>Te<sub>3</sub>/PEDOT: PSS-based flexible thermoelectric film and device. *Chem. Eng. J.* 397, 125360. <https://doi.org/10.1016/j.cej.2020.125360>.
26. Zhou, H., Zhang, Z., Sun, C., Deng, H., and Fu, Q. (2020). Biomimetic approach to facilitate the high filler content in free-standing and flexible thermoelectric polymer composite films based on PVDF and Ag<sub>2</sub>Se nanowires. *ACS Appl. Mater. Interfaces* 12, 51506–51516. <https://doi.org/10.1021/acsami.0c15414>.
27. Xu, Q., Qu, S., Ming, C., Qiu, P., Yao, Q., Zhu, C., Wei, T.R., He, J., Shi, X., and Chen, L. (2020). Conformal organic-inorganic semiconductor composites for flexible thermoelectrics. *Energy Environ. Sci.* 13, 511–518. <https://doi.org/10.1039/c9ee03776d>.
28. Na, Y., Kim, S., Mallem, S.P.R., Yi, S., Kim, K.T., and Park, K.-I. (2022). Energy harvesting from human body heat using highly flexible thermoelectric generator based on Bi<sub>2</sub>Te<sub>3</sub> particles and polymer composite. *J. Alloys Compd.* 924, 166575. <https://doi.org/10.1016/j.jallcom.2022.166575>.
29. Li, J., Shi, Q., Röhr, J.A., Wu, H., Wu, B., Guo, Y., Zhang, Q., Hou, C., Li, Y., and Wang, H. (2020). Flexible 3D porous MoS<sub>2</sub>/CNTs architectures with ZT of 0.17 at room temperature for wearable thermoelectric applications. *Adv. Funct. Mater.* 30, 2002508. <https://doi.org/10.1002/adfm.202002508>.
30. Wang, H., Lin, X., Han, G., Zhang, B., Chen, Y., Zhang, L., Lu, X., Wang, G., and Zhou, X. (2025). Flexible porous Ag<sub>2</sub>Se films: From freestanding inorganic films to inorganic-network/organic-skeleton thermoelectric generators. *Adv. Funct. Mater.* 35, 2413605. <https://doi.org/10.1002/adfm.202413605>.
31. Liang, L., Wang, M., Wang, X., Peng, P., Liu, Z., Chen, G., and Sun, G. (2022). Initiating a Stretchable, Compressible, and Wearable Thermoelectric Generator by a Spiral Architecture with Ternary Nanocomposites for Efficient Heat Harvesting. *Adv. Funct. Mater.* 32, 2111435. <https://doi.org/10.1002/adfm.202111435>.
32. Sun, L., Ao, D.W., Hwang, J., Liu, Q., Cao, E.S., and Sun, B. (2024). Realizing high thermoelectric performance flexible free-standing PEDOT:PSS/Bi<sub>0.5</sub>Sb<sub>1.5</sub>Te<sub>3</sub> composite films for power generation. *Rare Met.* 43, 5985–5993. <https://doi.org/10.1007/s12598-024-02860-0>.
33. Li, J., Liu, Y., Wang, Z., Chen, L., and Cai, K. (2023). Ultra-flexible self-supporting Ag<sub>2</sub>Se/nylon composite films for wearable thermoelectric devices. *Compos. B Eng.* 265, 110946. <https://doi.org/10.1016/j.compositesb.2023.110946>.
34. Cao, T., Shi, X.L., Hu, B., Yang, Q., Lyu, W.Y., Sun, S., Yin, L.C., Liu, Q.Y., Chen, W., Wang, X., et al. (2025). Advancing Ag<sub>2</sub>Se thin-film thermoelectrics via selenization-driven anisotropy control. *Nat. Commun.* 16, 1555. <https://doi.org/10.1038/s41467-025-56671-7>.
35. Hu, Q.X., Liu, W.D., Zhang, L., Gao, H., Wang, D.Z., Wu, T., Shi, X.L., Li, M., Liu, Q.F., Yang, Y.L., et al. (2024). Carrier separation boosts thermoelectric performance of flexible n-type Ag<sub>2</sub>Se-based films. *Adv. Energy Mater.* 14, 2401890. <https://doi.org/10.1002/aenm.202401890>.
36. Chen, Y.X., Shi, X.L., Zhang, J.Z., Nisar, M., Zha, Z.Z., Zhong, Z.N., Li, F., Liang, G.X., Luo, J.T., Li, M., et al. (2024). Deviceization of high-performance and flexible Ag<sub>2</sub>Se films for electronic skin and servo rotation angle control. *Nat. Commun.* 15, 8356. <https://doi.org/10.1038/s41467-024-52680-0>.
37. Li, Y., Lou, Q., Yang, J., Cai, K., Liu, Y., Lu, Y., Qiu, Y., Lu, Y., Wang, Z., Wu, M., et al. (2022). Exceptionally high power factor Ag<sub>2</sub>Se/Se/polypyrrole composite films for flexible thermoelectric generators. *Adv. Funct. Mater.* 32, 2106902. <https://doi.org/10.1002/adfm.202106902>.
38. Yang, D., Shi, X.L., Li, M., Nisar, M., Mansoor, A., Chen, S., Chen, Y., Li, F., Ma, H., Liang, G.X., et al. (2024). Flexible power generators by Ag<sub>2</sub>Se thin films with record-high thermoelectric performance. *Nat. Commun.* 15, 923. <https://doi.org/10.1038/s41467-024-45092-7>.
39. Hou, S., Liu, Y., Yin, L., Chen, C., Wu, Z., Wang, J., Luo, Y., Xue, W., Liu, X., Zhang, Q., et al. (2021). High performance wearable thermoelectric generators using Ag<sub>2</sub>Se films with large carrier mobility. *Nano Energy* 87, 106223. <https://doi.org/10.1016/j.nanoen.2021.106223>.
40. Hou, S., Liu, Y., Luo, Y., Wang, X., Yin, L., Sun, X., Wu, Z., Wang, J., Li, M., Chen, Z., et al. (2022). High-performance, thin-film thermoelectric generator with self-healing ability for body-heat harvesting. *Cell Rep. Phys. Sci.* 3, 101146. <https://doi.org/10.1016/j.xcrp.2022.101146>.
41. Lei, Y., Qi, R., Chen, M., Chen, H., Xing, C., Sui, F., Gu, L., He, W., Zhang, Y., Baba, T., et al. (2022). Microstructurally tailored thin β-Ag<sub>2</sub>Se films toward commercial flexible thermoelectrics. *Adv. Mater.* 34, e2104786. <https://doi.org/10.1002/adma.202104786>.
42. Zheng, Z.H., Li, Y.L., Niu, J.Y., Wei, M., Zhang, D.L., Zhong, Y., Nisar, M., Abbas, A., Chen, S., Li, F., et al. (2022). Significantly (001)-textured Ag<sub>2</sub>Se thin films with excellent thermoelectric performance for flexible power applications. *J. Mater. Chem. A* 10, 21603–21610. <https://doi.org/10.1039/d2ta06356e>.
43. Lu, Y., Qiu, Y., Cai, K., Li, X., Gao, M., Jiang, C., and He, J. (2020). Ultrahigh performance PEDOT/Ag<sub>2</sub>Se/CuAgSe composite film for wearable thermoelectric power generators. *Mater. Today Phys.* 14, 100223. <https://doi.org/10.1016/j.mtphys.2020.100223>.
44. Lu, Y., Qiu, Y., Cai, K., Ding, Y., Wang, M., Jiang, C., Yao, Q., Huang, C., Chen, L., and He, J. (2020). Correction: Ultrahigh power factor and flexible silver selenide-based composite film for thermoelectric devices. *Energy Environ. Sci.* 13, 1287–1288. <https://doi.org/10.1039/d0ee90012e>.
45. Mao, D., Zhou, Y., Yu, Y., Wang, Y., Han, M., Meng, Q., Lu, Y., Feng, J., Kong, M., Yang, H., et al. (2024). Scalable and sustainable manufacturing of twin boundary-enhanced flexible Bi<sub>0.4</sub>Sb<sub>1.6</sub>Te<sub>3</sub> films with high thermoelectric performance. *Joule* 8, 3313–3323. <https://doi.org/10.1016/j.joule.2024.08.009>.
46. Li, X., Lu, Y., Cai, K., Gao, M., Li, Y., Wang, Z., Wu, M., Wei, P., Zhao, W., Du, Y., et al. (2022). Exceptional power factor of flexible Ag/Ag<sub>2</sub>Se thermoelectric composite films. *Chem. Eng. J.* 434, 134739. <https://doi.org/10.1016/j.cej.2022.134739>.
47. Liang, J., Wang, T., Qiu, P., Yang, S., Ming, C., Chen, H., Song, Q., Zhao, K., Wei, T.R., Ren, D., et al. (2019). Flexible thermoelectrics: From silver chalcogenides to full-inorganic devices. *Energy Environ. Sci.* 12, 2983–2990. <https://doi.org/10.1039/c9ee01777a>.
48. Liu, Y., Zhang, Q., Huang, A., Zhang, K., Wan, S., Chen, H., Fu, Y., Zuo, W., Wang, Y., Cao, X., et al. (2024). Fully inkjet-printed Ag<sub>2</sub>Se flexible thermoelectric devices for sustainable power generation. *Nat. Commun.* 15, 2141. <https://doi.org/10.1038/s41467-024-46183-1>.
49. Ding, Y., Qiu, Y., Cai, K., Yao, Q., Chen, S., Chen, L., and He, J. (2019). High performance n-type Ag<sub>2</sub>Se film on nylon membrane for flexible thermoelectric power generator. *Nat. Commun.* 10, 841. <https://doi.org/10.1038/s41467-019-08835-5>.
50. Chen, H., Shao, C., Huang, S., Gao, Z., Huang, H., Pan, Z., Zhao, K., Qiu, P., Wei, T.R., and Shi, X. (2024). High-entropy cubic pseudo-ternary Ag<sub>2</sub>(S, Se, Te) materials with excellent ductility and thermoelectric performance. *Adv. Energy Mater.* 14, 2303473. <https://doi.org/10.1002/aenm.202303473>.
51. Kumar, S., Battabyal, M., Sethupathi, K., and Satapathy, D.K. (2024). High-performance printed Ag<sub>2</sub>Se/PI flexible thermoelectric film for

- powering wearable electronics. *ACS Appl. Mater. Interfaces* **16**, 40848–40857. <https://doi.org/10.1021/acsami.4c05537>.
52. Lu, Y., Han, X., Wei, P., Liu, Y., Wang, Z., Zuo, X., Zhao, W., and Cai, K. (2024). Nanoengineering approach toward ultrahigh power factor Ag<sub>2</sub>Se/polyvinylpyrrolidone composite film for flexible thermoelectric generator. *Chem. Eng. J.* **485**, 149793. <https://doi.org/10.1016/j.cej.2024.149793>.
53. Zhang, M., Li, J., Liu, Y., Liu, Y., Huang, C., Liu, R., Wei, P., Zhao, W., Xie, W., Weidenkaff, A., et al. (2025). Screen printing high-performance free-standing Ag<sub>2</sub>Se/carbon composite film for flexible thermoelectric converters. *Nano Energy* **138**, 110836. <https://doi.org/10.1016/j.nanoen.2025.110836>.
54. Gao, J., Miao, L., Lai, H., Zhu, S., Peng, Y., Wang, X., Koumoto, K., and Cai, H. (2020). Thermoelectric flexible silver selenide films: Compositional and length optimization. *iScience* **23**, 100753. <https://doi.org/10.1016/j.isci.2019.100753>.
55. Qin, J., Du, Y., Meng, Q., Chen, S., Hong, M., and Ke, Q. (2025). Versatile polymer-coated Ag<sub>2</sub>Se thermoelectric materials and devices for multi-scenario applications developed by direct-ink printing. *Nat. Commun.* **16**, 8497. <https://doi.org/10.1038/s41467-025-63390-6>.
56. Qi, Y., Xu, C., Zhang, Z., Zhang, Q., Xu, Z., Zhao, X., Zhao, Y., Cui, C., and Liu, W. (2024). Wet environment-induced adhesion and softening of coenzyme-based polymer elastic patch for treating periodontitis. *Bioact. Mater.* **35**, 259–273. <https://doi.org/10.1016/j.bioactmat.2024.02.002>.
57. Chai, Y., Chen, C., Luo, X., Zhan, S., Kim, J., Luo, J., Wang, X., Hu, Z., Ying, Y., and Liu, X. (2021). Cohabiting plant-wearable sensor in situ monitors water transport in plant. *Adv. Sci. (Weinh)* **8**, 2003642. <https://doi.org/10.1002/advs.202003642>.
58. Wang, S., Chai, Y., Sa, H., Ye, W., Wang, Q., Zou, Y., Luo, X., Xie, L., and Liu, X. (2024). Sunflower-like self-sustainable plant-wearable sensing probe. *Sci. Adv.* **10**, eads1136. <https://doi.org/10.1126/sciadv.ads1136>.
59. Zhu, B., Li, W., Zhang, Q., Li, D., Liu, X., Wang, Y., Xu, N., Wu, Z., Li, J., Li, X., et al. (2021). Subambient daytime radiative cooling textile based on nanoprocessed silk. *Nat. Nanotechnol.* **16**, 1342–1348. <https://doi.org/10.1038/s41565-021-00987-0>.