



Ultrafast microstructural reconstruction and 3D percolation networks in tape-casted flexible thermoelectric films via flash Joule heating

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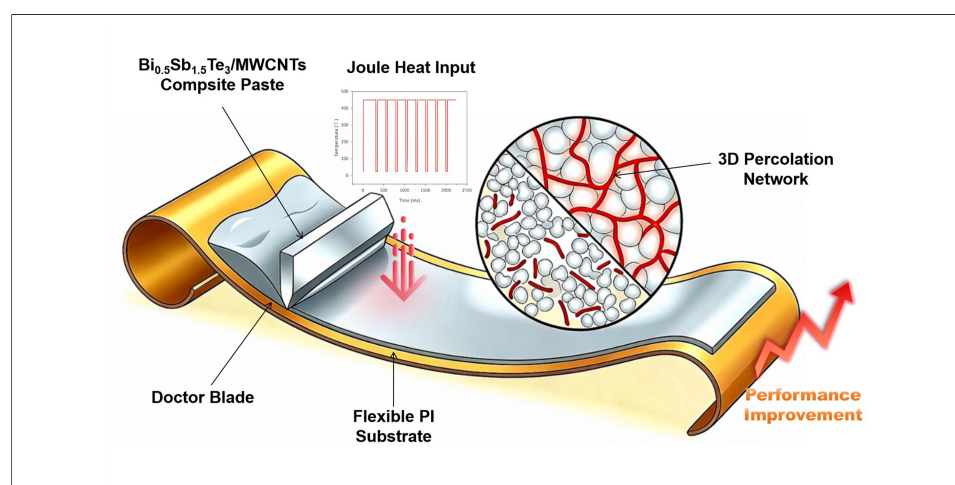
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Abstract

Flexible thermoelectric materials are highly desirable for self-powered wearable electronics, yet scalable fabrication of high-performance films remains a significant challenge. Tape casting offers a low-cost, large-area manufacturing route, but typically yields films with severe porosity and high interfacial resistance. Herein, we report a synergistic strategy combining scalable tape casting with ultrafast pulsed Joule heating to fabricate high-performance flexible $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ and multi-walled carbon nanotubes (MWCNTs) composite films. The millisecond-level pulsed Joule heating is proposed to promote localized Ostwald ripening and reduce micropores without damaging the flexible substrate. Crucially, the incorporation of high-aspect-ratio MWCNTs constructs a robust 3D percolation network that bypasses grain boundary barriers, leading to a remarkable enhancement in electrical conductivity. Meanwhile, the $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ -MWCNTs heterointerfaces are proposed to introduce an energy filtering effect, contributing to the

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well-maintained Seebeck coefficient. The optimized 20 wt% MWCNTs composite film achieves an electrical conductivity of $4,200 \text{ S}\cdot\text{m}^{-1}$ at 300 K. The power factor reaches a peak value of $146 \text{ }\mu\text{W}\cdot\text{m}^{-1}\cdot\text{K}^{-2}$ at 380 K, approximately 387% higher than that of the $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ film without MWCNT addition ($30 \text{ }\mu\text{W}\cdot\text{m}^{-1}\cdot\text{K}^{-2}$). This work demonstrates a highly efficient, cost-effective and scalable paradigm for the microstructural reconstruction of flexible thermoelectric materials.

INTRODUCTION

The rapid evolution of the Internet of Things (IoT) and wearable electronics has stimulated an urgent demand for sustainable, self-contained power sources. Flexible thermoelectric (TE) generators, capable of directly converting low-grade body or environmental heat into electricity without moving parts, have emerged as an ideal energy harvesting solution^[1,2]. Among state-of-the-art TE materials, Bi_2Te_3 -based alloys remain one of the optimal choices near room temperature^[3]. However, traditional bulk Bi_2Te_3 is inherently brittle and rigid, severely restricting its integration into conformable wearable devices^[4].

To achieve mechanical flexibility, thin-film TE materials have been extensively investigated. Currently, high-performance Bi_2Te_3 -based films are predominantly fabricated using vacuum-based techniques, such as magnetron sputtering and pulsed laser deposition (PLD)^[5–7]. While these methods yield dense microstructures and excellent TE performance, their substantial equipment costs, relatively low deposition rates, and strict substrate compatibility constraints can pose challenges for cost-effective, large-scale commercialization^[8]. In contrast, solution-based non-vacuum techniques, particularly doctor-blade coating, offer compelling advantages including low cost, operational simplicity, and superior compatibility with continuous roll-to-roll manufacturing^[9,10]. Despite these merits, doctor-bladed films inherently suffer from poor electrical transport properties. The evaporation of solvents and the presence of residual insulating organic binders create abundant micropores, cracks, and high-resistance grain boundaries, which severely scatter charge carriers.

To overcome transport barriers in tape-casted films, integrating one-dimensional (1D) conductive nanofillers - such as multi-walled carbon nanotubes (MWCNTs)^[11] - into the $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ slurry matrix

has emerged as a promising pathway. MWCNTs bridge isolated grains and establish a 3D conductive percolation network^[12]. However, adding MWCNTs alone cannot fully eliminate insulating binder residues or establish dense interparticle necking without post-deposition thermal treatment^[13]. Conventional isothermal furnace annealing, while somewhat helpful for crystallization, requires prolonged heating cycles. This leads to excessive grain coarsening, compositional volatilization, and, most critically, prolonged high-temperature exposure that may cause structural degradation of flexible polymer substrates [e.g., polyimide (PI)] under sustained annealing conditions^[14,15]. Consequently, developing an ultrafast, localized sintering modality to reconstruct the grain interfaces of tape-casted composite films without damaging the polymer substrates remains a key challenge^[16].

Beyond Bi_2Te_3 -based systems, other promising *p*-type flexible thermoelectric materials have been reported. For instance, MgAgSb -based alloys have recently demonstrated competitive thermoelectric performance near room temperature, with high power factors (PFs) achieved through nanostructuring strategies^[17]. However, these materials typically require more complex synthesis routes and are less amenable to solution-based processing. In contrast, Bi_2Te_3 -based composites processed via tape casting offer a compelling combination of near-room-temperature performance, compositional tunability, and compatibility with scalable wet-process manufacturing^[18], motivating the present study.

In this work, we demonstrate a synergistic microstructural and compositional engineering strategy to boost the TE performance of doctor-bladed $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ /MWCNT composite films. Rather than relying on traditional furnace annealing, we implement millisecond-level flash Joule heating (FJH) as an ultrafast non-equilibrium sintering modality. The FJH technique, originally developed for the rapid synthesis of flash graphene from carbon sources^[19],

delivers intense, localized energy bursts directly to high-resistance contact points^[20]. This rapidly heals interparticle defects and drives localized densification that is proposed to proceed via Ostwald ripening while maintaining a low bulk substrate temperature to preserve PI integrity. Concurrently, the incorporated high-aspect-ratio MWCNTs establish continuous 3D electron “highways” across grain boundaries, while the engineered $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ /MWCNT heterointerfaces are proposed to induce energy-filtering effects that selectively scatter low-energy charge carriers to decouple electrical conductivity (σ) and Seebeck coefficient (S). By co-optimizing the MWCNT loading and FJH processing parameters, we achieved a remarkable PF of $146 \mu\text{W}\cdot\text{m}^{-1}\cdot\text{K}^{-2}$ at 380 K - an approximately 387% enhancement over the MWCNTs-free $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ film. This study provides a scalable processing paradigm for high-performance flexible TE electronics.

EXPERIMENTAL

Slurry preparation and film post-treatment

The as-synthesized *p*-type bismuth telluride ingot (home-made) was ball-milled into fine powders ($< 45 \mu\text{m}$) for subsequent slurry preparation. The composite slurry was primarily formulated with an active material, a home-made binder (synthesized according to literature^[21], denoted as MF-BPA; MF-BPA is a melamine-formaldehyde resin cross-linked with bisphenol A, which provides high thermal stability and stepwise carbonization behavior) and a conductive agent (multi-walled carbon nanotubes, denoted as MWCNTs). In brief, for the synthesis of MF-BPA, paraformaldehyde (12 equiv.) was dissolved in distilled water or ethanol under base catalysis (NaOH, pH 8.5–9.5) at 60 °C, followed by sequential addition of melamine (2 equiv.) at 80–85 °C for 30–45 min and bisphenol A (3 equiv.) at 80–90 °C for 1.5–2 h. The reaction was neutralized to pH 7.0–7.5 and collected without solvent removal, affording a viscous pale yellow to amber resin solution (50–70 wt% solids) functionalized with hydroxyl, methylene, and ether bridges.

A preliminary 1.5 g slurry was prepared with a mass ratio of $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$:MF-BPA:MWCNTs = 7:2:1. A series of slurries were subsequently prepared by maintaining a constant $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$:MF-BPA mass ratio of 7:2 while varying the MWCNTs doping concentrations (0, 5, 10, 15, and 20 wt%).

Absolute ethanol and ethylene glycol monobutyl ether were utilized as co-solvents, with the latter serving as a retarder to control the solvent evaporation rate. After introducing the BYK-2150 dispersant, the mixture underwent high-energy blending for 3 min to ensure uniform dispersion. The well-dispersed slurry was then coated onto a surface-treated PI substrate (RAYITEK Hi-Tech Film Company, Ltd., Shenzhen, 100 μm thickness) using a doctor blade with a 100 μm clearance, resulting in large $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ /MWCNTs composite films. The thin film was punched into a round shape (12 mm diameter) and subsequently placed in a FJH instrument (model: HTS-7026D, Shenzhen Zhongke Jingyan Technology Co., Ltd., China) operating in pulse mode. Nine pulse operations were performed under a peak temperature of 450 °C, with each pulse width set at 200 ms.

The FJH peak temperature of 450 °C was selected via systematic optimization. Gradient screening experiments with fixed pulse width and pulse count were carried out at four peak temperatures: 350, 400, 450 and 500 °C. Temperatures below 400 °C cannot drive adequate atomic diffusion and binder carbonization, leading to marginal enhancement in electrical conductivity. Partial decomposition of the PI substrate occurred at 500 °C. Consequently, 450 °C achieved the best trade-off between microstructural reconstruction and substrate integrity. The 200 ms pulse width was optimized to maximize energy input into the film and reduce bulk heat accumulation on the substrate.

Characterization

The crystallographic structures and phase compositions of the $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ powders, pristine films, and $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ /MWCNTs composite films were characterized by X-ray diffraction (XRD) using an X'Pert Powder diffractometer (Malvern Panalytical, Netherlands) equipped with Cu K α radiation ($\lambda = 0.154 \text{ nm}$). Molecular vibrational information and the structural defect states of the incorporated MWCNTs were analyzed by Raman spectroscopy (Invia Reflex, Renishaw, UK, 532 nm laser excitation). The elemental distribution of Bi, Sb, Te, and C within the samples was characterized using energy-dispersive X-ray spectroscopy (EDS). Thermogravimetric analysis (TGA) was performed (TA Instruments Q500, USA) to evaluate the thermal decompo-

sition behavior of the binder systems. The rheological properties of the slurries were measured (Anton Paar MCR 302, Austria) to assess their suitability for doctor-blade coating. Cross-sectional and surface morphologies were examined by scanning electron microscopy (SEM; Hitachi SU-8000, Japan).

Thermoelectric transport property measurements

Thermoelectric properties, including the S and σ , were simultaneously measured using an MRS-3 system (Wuhan JouleYacht Technology Co., Ltd., China) under low vacuum from 300 to 400 K. S was determined via the dynamic method, where linear regression of pulse-induced ΔV vs. ΔT curves was employed to eliminate parasitic thermopower and instrumental drift. Electrical conductivity was measured using the standard direct current (DC) four-probe technique to exclude contact and lead resistances. The PF was calculated as $S^2\sigma$ to evaluate the overall transport performance of the $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ -based composite films. Room-temperature Hall effect measurements, including Hall coefficient (R_H), sheet/bulk resistivity (ρ), carrier concentration (p) and Hall mobility ($\mu_H = \frac{R_H}{\rho}$), were performed on an Accent HL5500 Hall System (Nanometrics, USA) with the Van der Pauw configuration. The four-contact Van der Pauw geometry eliminates strict requirements for sample shape.

RESULTS AND DISCUSSION

Fabrication process and baseline characterization

Figure 1 presents a detailed schematic of the $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ -based slurry and film preparation workflow. $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ powder serves as the thermoelectric matrix material, MF-BPA is introduced as a novel composite binder, and MWCNTs are added as a conductive network. These components are precisely proportioned in a mixed solvent of ethanol and 2-butoxyethanol. The mechanical shearing and collision action generated by ball milling achieves fine particle size reduction and uniform dispersion of the solid particles. The resulting homogeneous slurry system provides a solid material foundation for subsequent high-quality film casting or screen printing. The lower part of Figure 1 illustrates the FJH for the post-treatment of thin-film samples. The punched film pellets (12 mm diameter) are fixed in a custom metal fixture and placed under vacuum to

prevent oxidation. The system employs a temperature pulse mode, performing 9 cyclic pulse heating operations at 450 °C with a fixed pulse width of 200 ms (heating duration for every single pulse, left bottom of Figure 1). Compared to conventional time-consuming tube furnace annealing, this millisecond-level ultrafast thermal shock process offers significant advantages in microstructural control. It can rapidly eliminate residual stress and partial organic volatiles within the film layer, while the extremely high heating and cooling rates effectively suppress excessive grain growth of $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ crystallites. This thermal processing mechanism, while improving material densification, may retain abundant nanoscale grain boundaries, which are expected to strongly scatter low-frequency phonons and further reduce lattice thermal conductivity. Notably, the peak temperature of the FJH process can be flexibly adjusted by tuning pulse voltage, pulse width, and the number of pulses. This tunability enables the process to be adapted to different thermoelectric material systems, binder formulations, and substrates with varying thermal tolerance, extending its applicability to a broader range of flexible electronic material systems.

The decomposition characteristics of the binder directly affect the purity and charge carrier transport network of the thermoelectric thin film after annealing. The TGA curves in Figure 2A compare the thermal decomposition behavior of three binder systems [sodium carboxymethyl cellulose (CMC), poly(vinylidene fluoride) (PVDF), and MF-BPA]. The results show that conventional binders CMC and PVDF undergo dramatic mass loss between approximately 100 °C and 200 °C, with almost no residual carbon retention. In contrast, the MF-BPA adopted in this study exhibits outstanding high-temperature stability and stepwise decomposition characteristics, retaining approximately 25% residual mass at 500 °C^[21]. This high thermal stability of MF-BPA means that during the rapid Joule heating process, it will not instantaneously gasify and cause film layer structural collapse or macroscopic pore formation. Instead, its moderate carbonization residue may form an *in situ* continuous conductive carbon skeleton between $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ particles, synergistically optimizing the interfacial charge transport channels of the composite material together with the carbon nanotubes^[22].

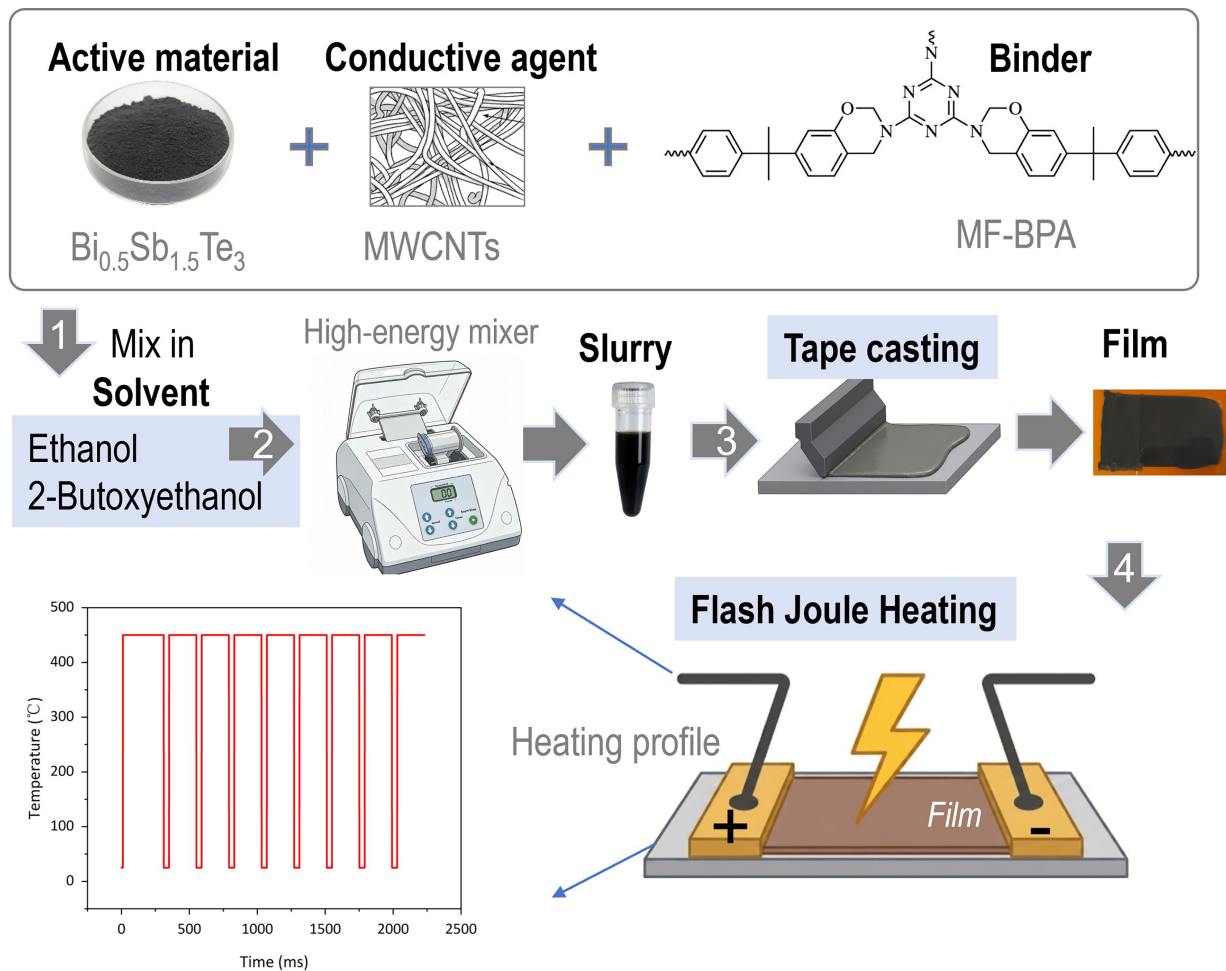


Figure 1. Fabrication workflow of Bi_2Te_3 -based thermoelectric composite films, including the key processes of slurry preparation, tape casting, and flash Joule heating (with a heat profile figure on the bottom left).

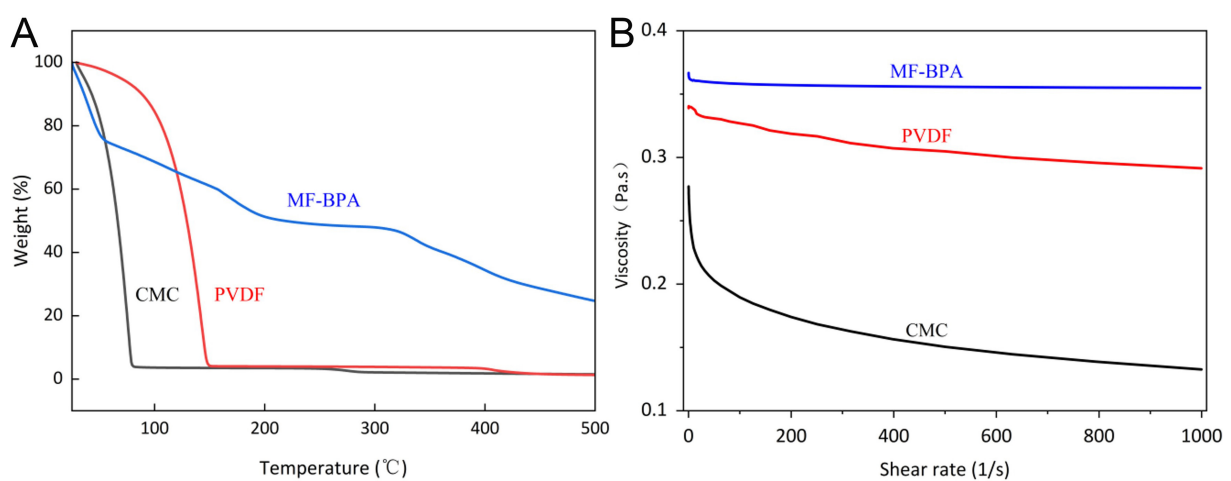


Figure 2. (A) TGA curves comparing three different binders (MF-BPA, PVDF, CMC); (B) Rheological flow curves showing viscosity versus shear rate for slurry systems containing different binders. CMC: Sodium carboxymethyl cellulose; PVDF: poly(vinylidene fluoride); MF-BPA: melamine-formaldehyde resin cross-linked with bisphenol A; TGA: thermogravimetric analysis.

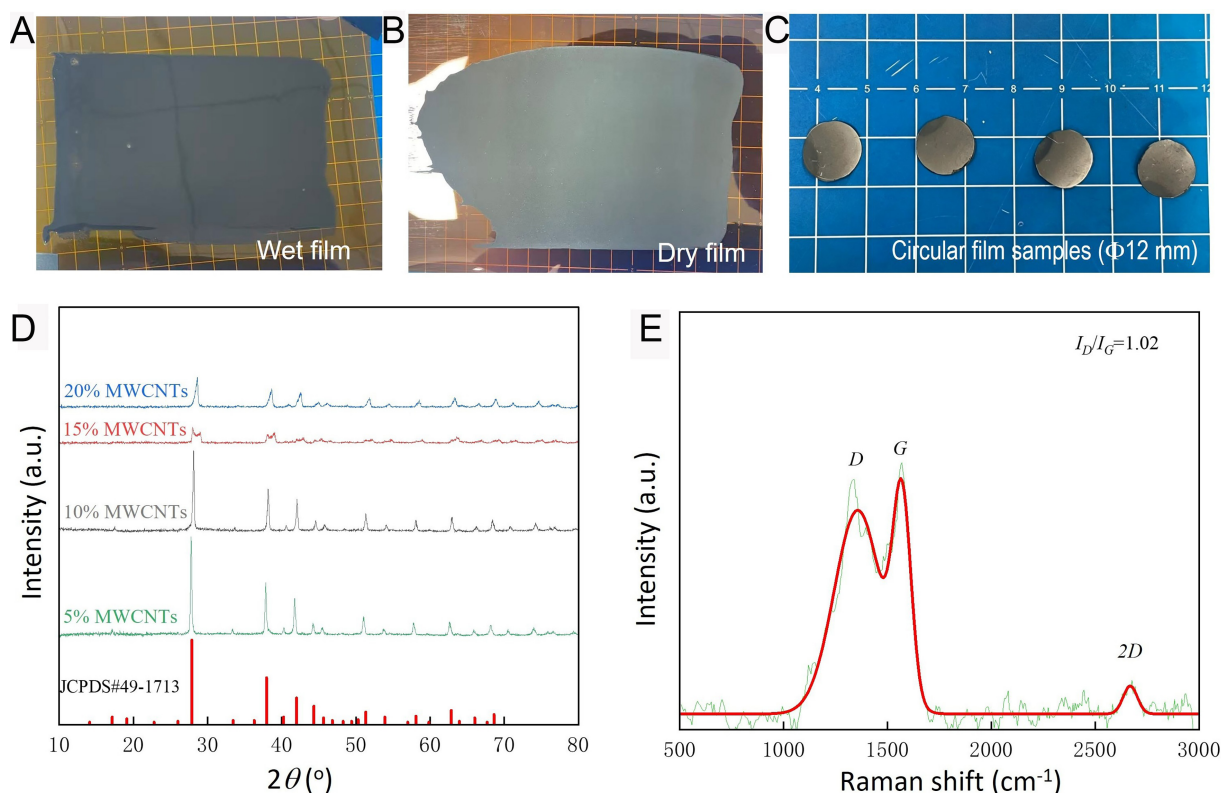


Figure 3. (A) Photograph of the composite wet film on a flexible PI substrate prepared by tape casting; (B) Photograph of the solid film after drying; (C) Punched films (12 mm diameter) for performance testing after the FJH treatment; (D) XRD patterns of composite materials with different MWCNTs mass fractions (5–20 wt%), all characteristic peaks matching the standard card (JCPDS#49-1713); (E) Raman spectrum of the $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ /20 wt% MWCNTs composite film. PI: Polyimide; XRD: X-ray diffraction; MWCNTs: multi-walled carbon nanotubes.

The rheological behavior of the slurry is a core parameter determining film quality and printing resolution. Figure 2B shows the viscosity versus shear rate curves (flow curves) for different binder systems. The MF-BPA system not only exhibits the highest apparent viscosity (stable at approximately 0.35 Pa·s at low shear rates) but also demonstrates typical and stable shear-thinning (non-Newtonian) fluid characteristics. In comparison, the CMC system has extremely low viscosity that drops sharply with increasing shear rate. The superior rheological properties demonstrate that MF-BPA is an ideal choice for preparing heavy metal compound slurries. Its high viscosity effectively suppresses gravitational sedimentation of high-density $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ powder during storage, ensuring excellent suspension stability, while the shear-thinning behavior ensures good flowability and printability of the slurry when external shear force is applied (such as tape casting or screen printing), thereby guaranteeing the surface flatness and internal uniformity of the thermoelectric film^[23].

Cross-sectional morphology and microstructural organization

Figure 3A and B shows the macroscopic morphology of the composite slurry before and after film formation on a flexible PI substrate via doctor blade coating. The wet film surface in Figure 3A is smooth and continuous, indicating that the slurry possesses excellent flowability and wettability. The solid thick film after drying in Figure 3B shows no obvious macroscopic cracks or curling delamination, confirming that the slurry system has excellent film-forming capability and structural adhesion. Figure 3C shows the circular film samples (12 mm diameter) punched after drying and subjected to FJH treatment. This process demonstrates the excellent macroscopic processability of the composite material - capable of meeting customized requirements for components of different geometric dimensions, laying a reliable foundation for subsequent thermoelectric property measurements.

Figure 3D presents the XRD patterns of composite materials with different MWCNTs mass fractions (5

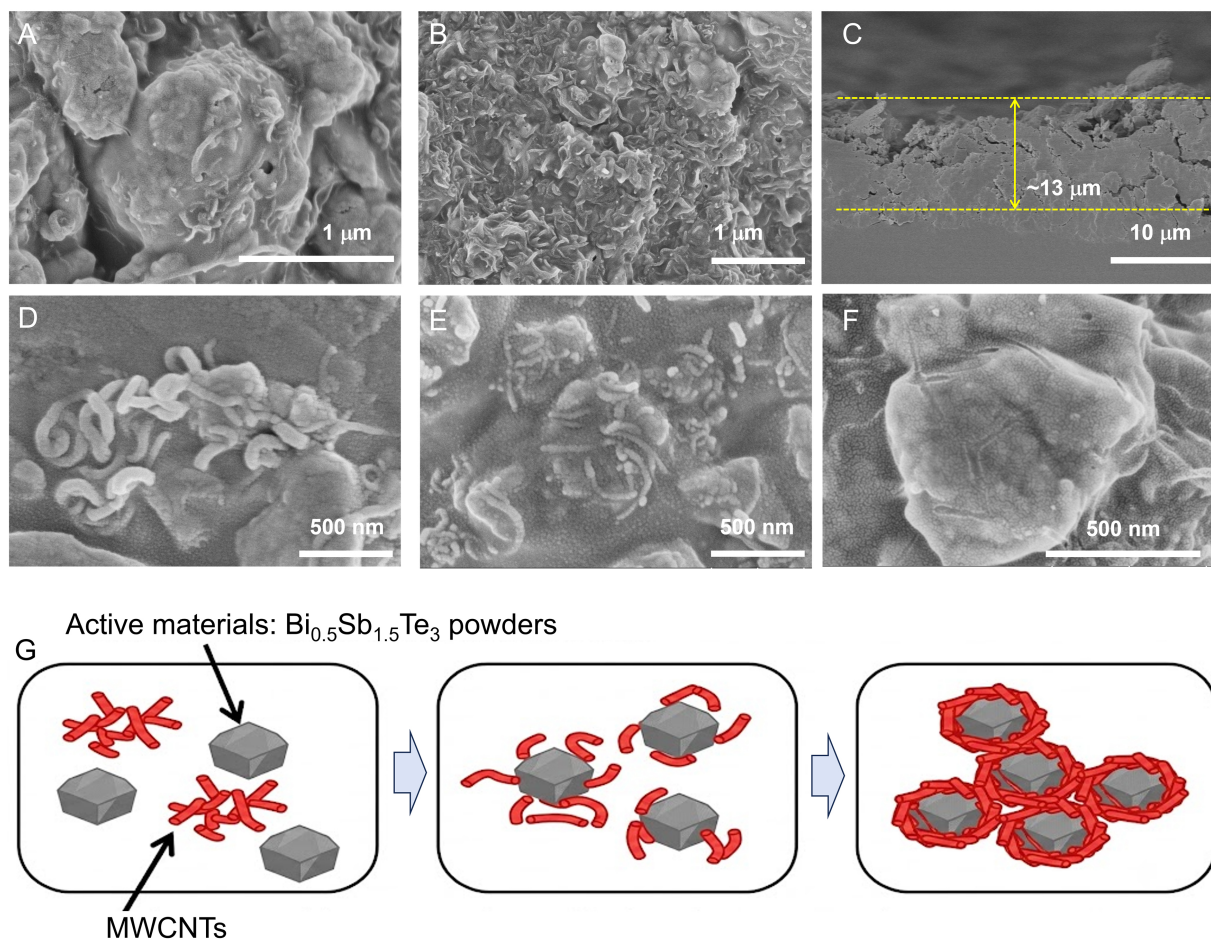


Figure 4. SEM surface morphologies of the composite film with (A) 10 wt% MWCNTs and (B) 20 wt% MWCNTs; (C) Cross-sectional SEM image of the composite film with 20 wt% MWCNTs; (D) SEM image of the composite material before sintering, showing MWCNTs in a loose, free random entanglement state around matrix particles; (E) Morphology at the early stage of Joule heating pulse annealing, with local thermodynamic driving and binder carbonization shrinkage causing MWCNTs to gradually adhere to matrix particle surfaces; (F) Surface morphologies of the composite material after sintering completion, with high-density MWCNTs tightly wrapping $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ particles; (G) Physical model schematic of interface reconstruction and 3D interconnected network formation between $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ particles and MWCNTs driven by the thermal field. SEM: Scanning electron microscopy; MWCNTs: multi-walled carbon nanotubes.

to 20 wt%). The results show that the characteristic diffraction peaks of all composite samples are in high agreement with the standard card of $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ (JCPDS#49-1713, matrix phase), with no obvious impurity peaks observed. This indicates that the introduction of MWCNTs did not alter the intrinsic crystal structure of the thermoelectric matrix, and the two form a physical composite as the primary interaction, avoiding the occurrence of harmful interfacial side reactions, thereby preserving the superior charge carrier transport characteristics of the matrix material.

Figure 3E shows the Raman spectrum of the composite material, clearly displaying the D band ($\sim 1,350 \text{ cm}^{-1}$) and G band ($\sim 1,580 \text{ cm}^{-1}$) characteris-

tic of carbon materials. The calculated intensity ratio I_D/I_G is approximately 1.02, indicating a certain number of structural defects on the MWCNT surfaces. The spectrum also presents a 2D band at $\sim 2,700 \text{ cm}^{-1}$, which together with the D and G bands constitutes the fingerprint features of sp^2 -hybridized carbon materials^[24]. The coexistence of the strong G peak and the broad multi-layer 2D peak confirms the multi-walled nature of the incorporated carbon nanotubes, ruling out the possibility of pure amorphous carbon or single-layer graphene^[25].

Figure 4A and B shows surface-view SEM images of composite thermoelectric materials with 10 and 20 wt% MWCNTs additions, respectively. These two sets of images intuitively reveal the significant evolu-

tion of the internal microstructure and heterogeneous interface of the composite material as the second-phase filler concentration increases. At 10 wt% addition, MWCNTs present a uniformly dispersed state, with the tubular structure sparsely but continuously penetrating and bridging on the surface and grain boundaries of micron-scale $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ matrix particles. When the MWCNTs concentration increases to 20 wt%, the microstructure undergoes dramatic changes. The high-density carbon nanotubes form a highly entangled complex network like a “bird’s nest”, almost completely encapsulating and covering the $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ matrix particles. Figure 4C confirms that through the specific fabrication process (slurry coating combined with annealing), a flexible thermoelectric thick film with a specific thickness ($\sim 13\ \mu\text{m}$) and good uniformity over a larger macroscopic range was successfully obtained.

It is particularly noteworthy that ultrafast Joule heating appears to be the main mechanism driving pore reduction and dense particle fusion, because it creates localized, transient thermal fields that accelerate neck formation and densification^[26], while MWCNT loading mainly affects particle packing/dispersion and can even agglomerate at higher concentrations^[27]. Unlike traditional long-time isothermal annealing, millisecond-level pulsed Joule heating can generate intense non-equilibrium thermal gradients inside the film. This instantaneous local high-energy burst (450 °C, 200 ms pulse width per heating cycle) rapidly activates atomic diffusion on particle surfaces, which is hypothesized to initiate a local Ostwald ripening-like mass transport process^[28]. Driven by such atomic migration, partial micron-scale pores generated during tape casting may be filled, and obvious necking and interfacial welding can form between discrete $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ particles, enabling fast reconstruction of the film microstructure. Together with undecomposed MF-BPA and MWCNTs, a structurally robust and electrically continuous three-dimensional composite network is formed. More critically, since thermal energy is injected instantaneously in the form of extremely short pulses, the global heat cannot accumulate significantly toward the underlying substrate, thereby substantially reducing thermal exposure to the underlying flexible PI substrate and preserving its structural integrity while achieving

densification of the thermoelectric active layer^[29].

The corresponding cross-scale microstructural morphology [Figure 4D–F], the distribution of MWCNTs in the $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ matrix undergoes a significant interface reconstruction process driven by sintering thermodynamics. Before sintering [Figure 4D], due to the spatial obstruction of solvents and undecomposed binders, MWCNTs are mainly loosely distributed around the matrix particles in a free or locally randomly entangled state, with weak interfacial bonding. With the instantaneous injection of Joule heating pulses [Figure 4E], the binder (MF-BPA) undergoes *in situ* carbonization and volume shrinkage, accompanied by particle rearrangement driven by local thermal stress, causing the nanotubes to gradually adhere to the matrix particle surfaces. After sintering is complete [Figure 4F], MWCNTs tightly and uniformly wrap the $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ particle surfaces, significantly reducing the original physical gaps, and successfully constructing a continuous and highly dense three-dimensional interconnected conductive network inside the composite system [as shown in the physical model in Figure 4G].

Thermoelectric transport properties

In the composite material system of MWCNTs and $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$, MWCNTs, as a highly conductive 1D nanomaterial, significantly alter the thermoelectric transport properties of the matrix upon introduction. On one hand, MWCNTs interconnect within the $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ matrix to form an effective 3D conductive network, greatly reducing the overall electrical resistance of the material and significantly improving electrical conductivity with increasing doping amount^[30]. On the other hand, the heterogeneous interfaces formed between MWCNTs and $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ are proposed to have an energy filtering effect on charge carriers, potentially capable of selectively scattering low-energy charge carriers, thereby contributing to an enhancement of the S ^[31].

Further quantitative analysis shows that the $I_D/I_G = 1.02$ of the MWCNTs indicates that there are relatively abundant structural defects on their tube wall surfaces. This is particularly critical for thermoelectric thin films prepared by wet-process techniques such as doctor-blade coating. On one hand, appropriate surface defects significantly

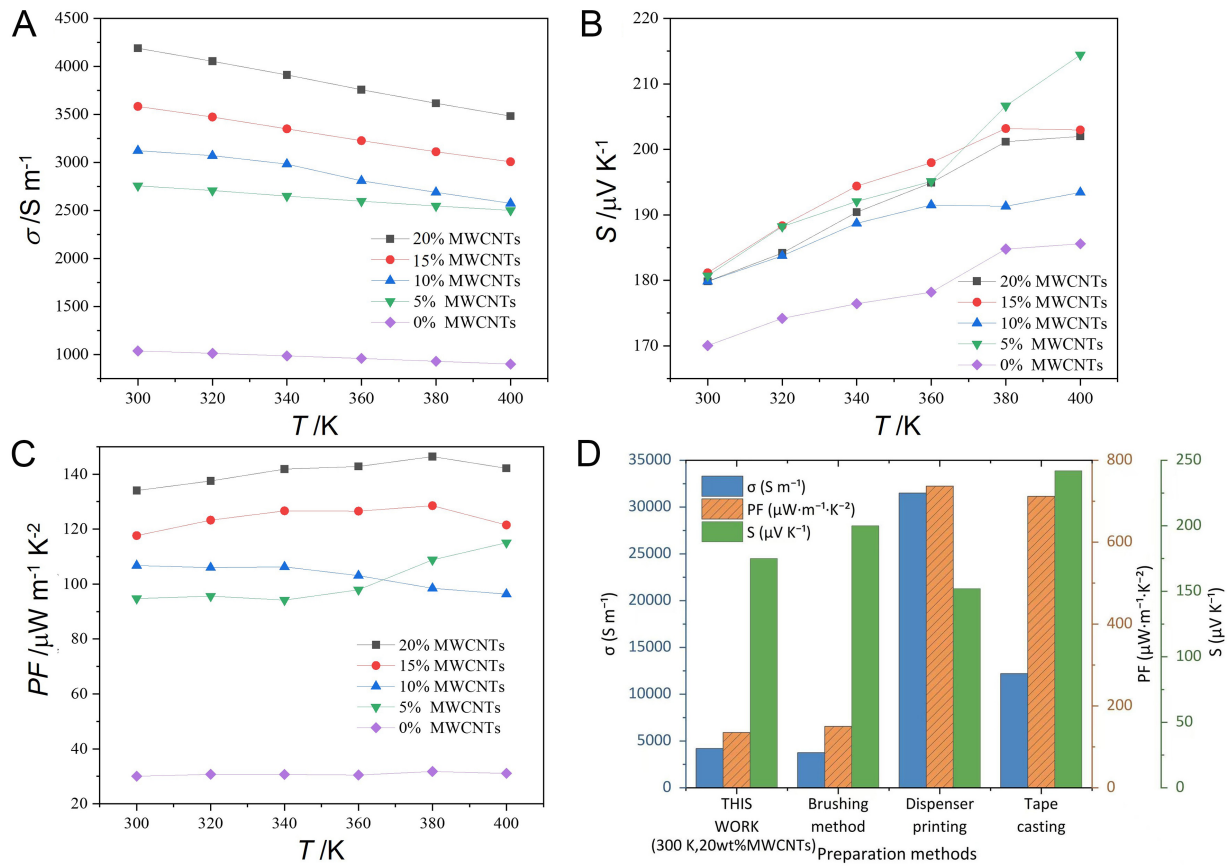


Figure 5. (A) σ , (B) S and (C) PF as a function of temperature (300–400 K) for films with different MWCNTs doping levels after FJH treatment; (D) Comparison of room-temperature thermoelectric performance between this work (doctor-blade coating combined with ultrafast FJH treatment) and other typical thick-film fabrication processes (brush coating, dispenser printing, tape casting) reported in the literature^[32–34]. S : Seebeck coefficient; σ : electrical conductivity; PF : power factor; MWCNTs: multi-walled carbon nanotubes; FJH: flash Joule heating.

improve the dispersibility and rheological properties of carbon nanotubes in the slurry, ensuring microstructural uniformity and macroscopic mechanical flexibility of the doctor-bladed film. On the other hand, the defect sites on MWCNTs surfaces can form tight interfacial coupling with the Bi_{0.5}Sb_{1.5}Te₃ matrix, constructing many heterogeneous interfaces. These interfaces not only act as strong phonon scattering centers to effectively reduce lattice thermal conductivity but also build continuous high-speed charge carrier transport channels inside the film by optimizing the interphase contact potential, thereby achieving synergistic regulation of composite film electrothermal transport.

As shown in Figure 5A, in terms of σ , all samples exhibit a monotonically decreasing trend with increasing temperature. At 300 K, as the MWCNTs

doping amount increases from 0% to 20%, σ gradually increases from 1,000 S m⁻¹ for the pristine Bi_{0.5}Sb_{1.5}Te₃ film (without MWCNTs) to 4,200 S m⁻¹ (20 wt% MWCNTs). When the temperature rises to 400 K, σ generally decreases but still maintains the gradient order of 20 wt% MWCNTs > 15% > 10% > 5% > 0%, with the 20 wt% MWCNTs sample still reaching 3,500 S m⁻¹. This decreasing behavior originates from the reduction in carrier mobility due to enhanced phonon scattering with increasing temperature in semiconductors. The significant increase in σ with MWCNTs doping amount is the basic requirement for electrical transport. However, the Hall mobility of the MWCNT-free pristine film is extremely low at 1.227 cm² V⁻¹ s⁻¹; this low value is mainly limited by intense charge carrier scattering originating from abundant grain boundaries and micropores inside the tape-casted film.

The large leap in electrical conductivity of the composite film in Figure 5A is the inevitable result of the combined action of the 3D carbon nanotube percolation network and the Joule heating interface fusion effects^[28]. For untreated doctor-bladed films, the dense original grain boundaries and organic insulating residues inside constitute extremely high potential barriers, producing strong scattering of charge carriers. However, the local high-temperature melting effect induced by ultrafast pulsed Joule heating greatly improves this transport bottleneck^[28]. On one hand, the instantaneous thermal field effectively clears the insulating potential barriers on particle surfaces; on the other hand, atomic rearrangement and fusion at grain boundaries significantly reduce the interfacial scattering resistance for charge carriers crossing particle boundaries. Therefore, the Joule heating-driven matrix grain boundary optimization, complemented by the cross-grain boundary conductive “highways” built by MWCNTs, fundamentally removes the limitation of extremely low carrier mobility in solution-processed films^[28], enabling the 20 wt% doped composite material to exhibit an outstanding macroscopic electrical conductivity of up to $4,200 \text{ S}\cdot\text{m}^{-1}$.

In the composite system, the significant improvement in overall electrical conductivity under conditions of severely limited intrinsic carrier mobility is fundamentally due to the highly conductive MWCNTs building a three-dimensional continuous percolation network inside the matrix. By virtue of their large aspect ratio and excellent intrinsic electrical conductivity, MWCNTs establish direct physical contact and low-resistance charge transport channels between adjacent matrix particles. When the MWCNTs addition reaches the percolation threshold, the dominant transport path of charge carriers changes, enabling direct transport along the carbon nanotube network, thereby effectively bypassing the high-resistance matrix grain boundary regions. The construction of this continuous microscopic conductive network fundamentally overcomes the limitation of film grain boundary potential barriers on carrier transport, ultimately achieving a large improvement in the macroscopic electrical conductivity of the composite material^[30,35,36].

The S in Figure 5B shows a monotonically increasing trend with increasing temperature. At 300 K, the

S values of all doped samples are approximately $170\text{--}180 \text{ }\mu\text{V/K}$, reaching a maximum of $214 \text{ }\mu\text{V/K}$ at 400 K (5 wt% MWCNTs). Appropriate MWCNTs doping ($\sim 5 \text{ wt\%}$) may improve the S value by a proposed energy filtering effect of MWCNTs- $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ heterogeneous interfaces, increasing the proportion of high-energy charge carriers, thereby optimizing the S value^[37]. Excessive addition (15 and 20 wt%) leads to a slight decrease in S due to the bipolar effect caused by excessively high carrier concentration, partially offsetting this advantage^[38].

Figure 5C shows PF variation versus temperature for samples with different MWCNTs filling contents. Incorporation of MWCNTs significantly enhances the base material's PF; at the same test temperature, sample PF overall increases with rising MWCNTs filling ratio. For 20% and 15% MWCNTs filled samples, PF first rises then falls as temperature increases, both peaking around 380 K. For 10% MWCNTs filled sample, PF stays stable at 300–340 K, then decreases when temperature exceeds 340 K; for 5% MWCNTs filled sample, PF drops slightly at 300–340 K, then continuously increases when temperature exceeds 340 K; the pure base material without MWCNTs has generally low PF, with no obvious variation in 300–400 K.

The optimal PF of $146 \text{ }\mu\text{W}\cdot\text{m}^{-1}\cdot\text{K}^{-2}$ (380 K, $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ /20 wt% MWCNTs) in this study significantly outperforms many reported non-vacuum wet-process coated $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ films^[39]. For most unoptimized formulations without specialized high-energy post-treatment, their PFs are typically below $50 \text{ }\mu\text{W}\cdot\text{m}^{-1}\cdot\text{K}^{-2}$ ^[40,41], while some optimized printed samples can exceed this value. This demonstrates the enormous progress of the 3D conductive network construction strategy in overcoming grain boundary potential barriers. Compared to common flexible organic-inorganic composites such as PEDOT:PSS/MWCNT systems (PF typically $50\text{--}130 \text{ }\mu\text{W}\cdot\text{m}^{-1}\cdot\text{K}^{-2}$), and highly ordered $(\text{Bi,Sb})_2\text{Te}_3$ /single-wall CNT composite films prepared by *in situ* crystal growth (PF up to $\sim 1,680 \text{ }\mu\text{W}\cdot\text{m}^{-1}\cdot\text{K}^{-2}$)^[42], or traditional high-vacuum magnetron sputtered films and hot-pressed bulk alloys (PF up to $2,000\text{--}4,000 \text{ }\mu\text{W}\cdot\text{m}^{-1}\cdot\text{K}^{-2}$)^[43], the slurry wet-process technique represented by doctor-blade coating has become the core development direction in the flexible thermoelectric field due to its superior scalability

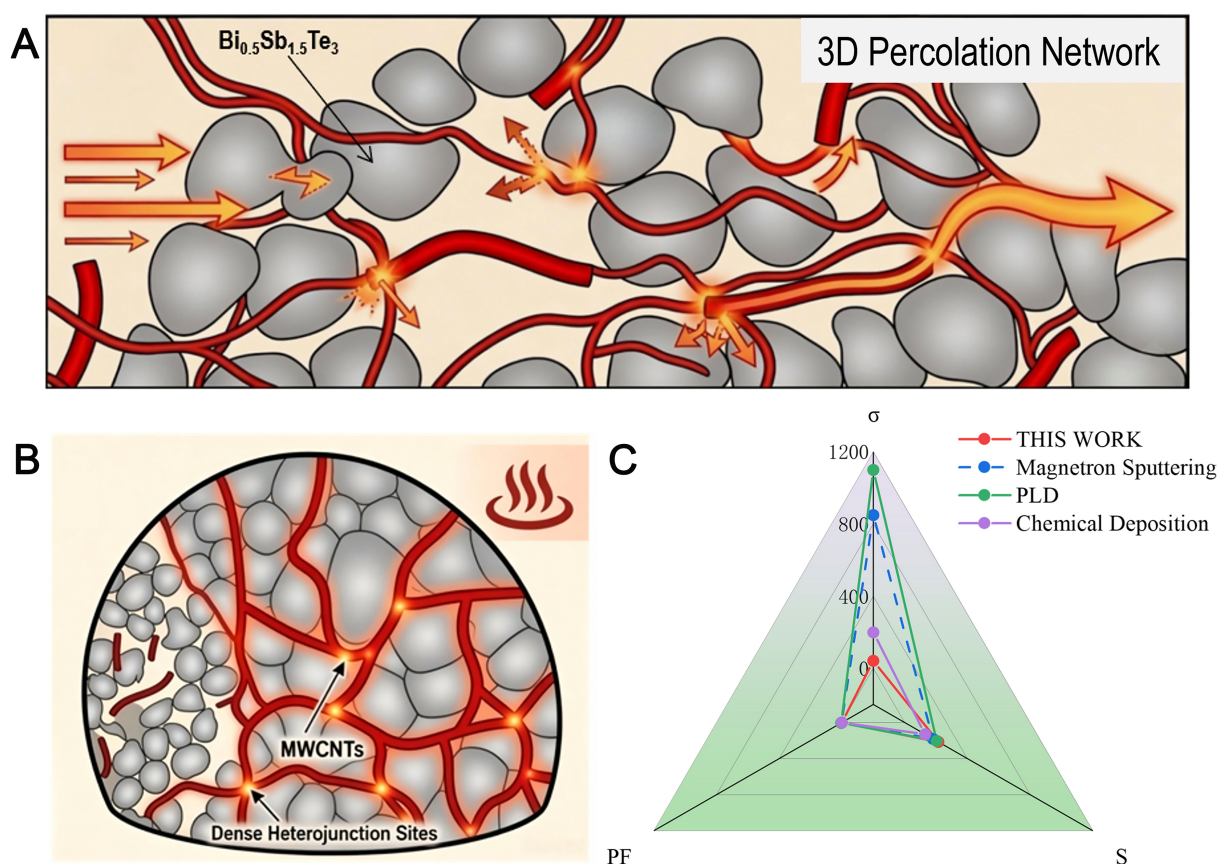


Figure 6. (A) Schematic diagram of the 3D Percolation Network constructed by MWCNTs in $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ -MWCNTs composite film; (B) Schematic illustration showing densely distributed heterojunction sites within the composite; (C) Radar plot comparing the thermoelectric performance of thin films prepared by various fabrication methods. MWCNTs: multi-walled carbon nanotubes; PLD: pulsed laser deposition.

and extremely low manufacturing cost^[44]. The introduction of MWCNTs may not only exert an energy filtering effect but also act as a 1D “nano-skeleton” endowing the film with excellent crack resistance and bending flexibility^[45]. Systematic quantitative evaluation of bendability and long-term mechanical reliability will be further performed in our follow-up work. This design concept of sacrificing partial intrinsic electrical properties in exchange for large-area conformal adhesion capability effectively reduces interfacial contact thermal resistance at the system level, providing an extremely valuable engineering reference for the continuous manufacturing of next-generation low-cost flexible thermoelectric components^[46].

Figure 5D provides a lateral comparison of this work (doctor-blade coating combined with ultrafast Joule heating pulse annealing) with other typical thick-film fabrication processes (such as brush coating, dis-

penser printing, and tape casting) in the literature in terms of thermoelectric performance parameters. Objectively speaking, since this work employs millisecond-level ultrafast thermal processing at 450 °C, the crystallinity and absolute electrical conductivity of the film are inevitably lower than those of screen-printed or tape-cast thick films that have undergone high-temperature sintering for several hours. At room temperature, the optimized film in this work delivers a competitive electrical conductivity of $4,200 \text{ S}\cdot\text{m}^{-1}$, with a S of $\sim 180 \mu\text{V/K}$ and showing comparable performance among solution-processed flexible thick films. This performance gap is mainly attributed to the “double-edged sword” effect of the ultrafast heating thermodynamic process. Although millisecond-level Joule heating pulses effectively suppress excessive grain growth and maintain high-density heterogeneous interfaces (favorable for maintaining high S and reducing thermal conductivity), they also lead to the inability to achieve high

densification inside the film (as shown by the hierarchical porous structure in the cross-sectional SEM images). The large number of micron-scale pores and amorphous carbonization residues at interfaces constitute strong electron scattering sources, greatly limiting the further improvement of macroscopic carrier mobility.

Comprehensive performance comparison shows that, relying on the constructed 3D percolation network and the energy filtering effect induced by heterogeneous interfaces [Figure 6A and B], this work not only successfully maintains an excellent S , even superior to some traditional chemical deposition and magnetron sputtering processes - S , but also demonstrates advantages in manufacturing cost, process compatibility, and large-area scalable manufacturing that physical vapor deposition techniques cannot match. However, limited by the inevitable grain boundary residues, amorphous phases, and carrier mobility losses caused by interfacial contact resistance in the composite system, the current σ and overall PF still have a certain order-of-magnitude gap compared to extreme processes such as PLD, as shown in Figure 6C^[47-49]. Future research may focus on exploring advanced post-processing techniques (such as flash sintering) to achieve deeper healing of grain boundary defects, while combining heterogeneous band engineering for fine matching and decoupling regulation of carrier concentration, to further narrow the performance gap with vacuum deposition techniques and ultimately realize the continuous manufacturing of truly high-performance, low-cost flexible thermoelectric components. Furthermore, future work should address the integration of electrode layers and other device components to realize fully functional thermoelectric generators, as the scalable fabrication of complete devices remains an important direction for practical applications^[50].

CONCLUSIONS

In this work, we proposed a synergistic strategy combining low-cost tape casting with ultrafast FJH technology, and successfully fabricated high-performance flexible $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ /MWCNTs composite thermoelectric films with a three-dimensional continuous percolation network. By selecting an MF-BPA binder with high thermal stability and applying instantaneous pulsed Joule heating (200 ms

pulse width, 450 °C), efficient local heat injection was achieved. This work elucidates the synergistic regulation mechanism of microstructural evolution and MWCNTs doping in thermoelectric transport.

On one hand, the highly conductive MWCNTs with a large aspect ratio, interwoven with the *in situ* thermally decomposed carbon skeleton of MF-BPA, construct a robust 3D percolation network inside the matrix, fundamentally bypassing the high-resistance potential barriers composed of insulating residues and original grain boundaries, enabling macroscopic electrical conductivity to achieve a large leap. On the other hand, the densely distributed $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ /MWCNTs heterogeneous interfaces are proposed to induce an energy filtering effect, selectively scattering low-energy charge carriers, effectively maintaining an excellent S while electrical conductivity increases markedly.

Benefiting from the synergistic effect of structural densification and conductive network construction, when the MWCNTs doping amount reaches the optimal ratio of 20 wt%, the composite film exhibits outstanding macroscopic thermoelectric performance. At 300 K, its electrical conductivity soars to 4,200 $\text{S}\cdot\text{m}^{-1}$, approximately 4.2 times that of the MWCNTs-free $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ film. At 380 K, a maximum PF of 146 $\mu\text{W}\cdot\text{m}^{-1}\cdot\text{K}^{-2}$ is achieved, representing an approximately 387% enhancement relative to the MWCNTs-free $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ film. This work not only provides profound physical insights into the complex charge transport mechanisms in flexible thermoelectric materials, but also opens up an entirely new technological paradigm for the scalable non-vacuum manufacturing of next-generation wearable electronic devices and self-powered systems at extremely low cost and with outstanding process scalability.

DECLARATIONS

Authors' contributions

Conceptualization, investigation, methodology, visualization, validation, writing - original draft: Chen, Z.

Investigation, methodology, formal analysis, writing - review & editing: Li, H.

Investigation, methodology, validation: Zhang, X.; Li, M.; Qin, J.; He, H.

Resources, supervision, project administration,

writing - review & editing: Zheng, P.; Zheng, Y.
Resources, data curation: Cheng, X.
Data curation, visualization, supervision: Yemata, T.
A.

Availability of data and materials

The original contributions presented in this study are included in the article. Further inquiries can be directed to the corresponding authors.

AI and AI-assisted tools statement

During the preparation of this manuscript, the AI tools Nano Banana Pro (powered by Gemini 3 Pro Image, version 1.0, released 2026-05-28) and PowerPoint (Microsoft 365 Family, version 2603, released 2026-03-24) were used to create and modify schematic illustrations in [Figures 1, 4G, 6A and B](#). In [Figure 1](#), the schematic illustrations of $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$, MWCNTs, the high-energy mixer, tape-casting equipment and flash Joule-heating device highlighted by green boxes were created using Nano Banana Pro. In [Figure 4G](#), the physical model schematic of interface reconstruction and 3D interconnected network formation between $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ particles and MWCNTs driven by the thermal field was created using PowerPoint and modified with Nano Banana Pro. [Figure 6A and B](#) was created using Nano Banana Pro. The tools did not influence the study design, data collection, analysis, interpretation, or the scientific content of the work. All authors take full responsibility for the accuracy, integrity, and final content of the manuscript.

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Conflicts of interest

Dr. Zheng, Y. is an editorial board member of the journal *Advanced Energy Conversion*, but was not involved in any steps of editorial processing, notably including reviewer selection, manuscript handling, and decision-making, while the other authors have declared that they have no conflicts of interest.

Ethical approval and consent to participate

Not applicable.

Consent for publication

Not applicable.

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