

Article

Enhanced Thermoelectric Performance of CuInTe₂ via SnTe Incorporation and Microwave Synthesis

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Abstract

Incorporating a secondary phase represents a promising strategy for enhancing the thermoelectric performance of materials. In this study, CuInTe₂ was modified with SnTe powders at various weight fractions (0, 0.5, 1, 2, 4 wt%) and subsequently synthesized via rapid microwave melting. The phase composition and microstructure of the resulting materials were systematically characterized. Structural analyses revealed that the introduction of SnTe induced the incorporation of Sn and Te into the CuInTe₂ lattice, accompanied by a progressive contraction in lattice parameters. Owing to the spontaneous formation of intrinsic Sn vacancies in SnTe and the regulation of carrier concentration, electrical conductivity of up to $2.7 \times 10^4 \text{ Sm}^{-1}$ was achieved, representing a 1.7-fold increase over pristine CuInTe₂. Coupled with a notable reduction in lattice thermal conductivity ($0.9 \text{ Wm}^{-1}\text{K}^{-1}$ at 700 K), a maximum figure of merit of 0.44 was obtained for the CuInTe₂-2 wt% SnTe sample. While the absolute zT value is moderate compared to state-of-the-art CuInTe₂-based materials, this work establishes the feasibility of microwave melting combined with second-phase incorporation as a rapid and energy-efficient synthesis pathway for CuInTe₂ modification. These results demonstrate that the introduction of SnTe is a viable strategy for enhancing the thermoelectric performance of CuInTe₂.

Keywords: thermoelectric material; rapid microwave melting; carrier concentration modulation; defect engineering; CuInTe₂

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1. Introduction

Thermoelectric (TE) technology is considered a compelling pathway for harvesting renewable energy, offering a viable means for waste heat recovery and refrigeration [1–3]. The conversion efficiency of a thermoelectric device is predominantly determined by the dimensionless figure of merit $zT = (\alpha^2 \sigma) T / \kappa$, where α , σ , $\alpha^2 \sigma$, κ , and T are the Seebeck coefficient, electrical conductivity, power factor, thermal conductivity, and absolute temperature, respectively [4,5]. Approaches to enhancing zT can be categorized into $\alpha^2 \sigma$ (power factor) boosting by optimizing the n (carrier concentration) [6,7], or κ_L (lattice thermal conductivity) reducing by defect engineering [8,9]. Although electrical conductivity modulation by carrier concentration tuning has been widely attempted, the

thermal conductivity is invariably compromised due to inherent coupling between electrical conductivity and carrier thermal conductivity. From this perspective, strategies such as carrier pocket engineering [10–12], interfacial effects [13,14], and entropy engineering [15,16], as well as energy-filtering effects [17,18] have garnered significant interest for their capacity to concurrently modulate the intrinsic carrier concentration and enhance phonon scattering. Consequently, the primary focus in thermoelectric research is the discovery and development of materials that achieve substantially higher zT values.

Among the TE materials, the ternary Cu-based *p*-type semiconductor, CuInTe₂, characterized by a comparatively narrow bandgap, has been extensively investigated [19,20]. A preferred approach to improving thermoelectric properties is introducing alien atoms into the matrix to optimize carrier concentration [21]. In addition, adjusting the stoichiometry in CuInTe₂ will generate stress fields and defects, which are recognized to contribute to significant lattice thermal conductivity reduction. For instance, the use of non-stoichiometric CuInTe₂, synthesized by Kim et al., effectively improved the power factor by $\sim 16.1 \mu\text{W cm}^{-1}\text{K}^2$ at 723 K [22]. Through a systematic variation in Cu content in CuInTe₂, Li et al. achieved controlled modulation of Cu vacancies, thereby establishing vacancy engineering as a viable route to improved thermoelectric performance [23]. Luo et al. demonstrated that the heterogeneous matrix/precipitate interfaces, through an in situ oxidation, effectively enhanced the power factor and enhanced phonon scattering, resulting in the improvement in zT [24]. This approach has motivated the present study on CuInTe₂ modified via SnTe incorporation. These innovative strategies have provided renewed momentum for tailoring both the chemical composition and microstructure of thermoelectric materials to achieve enhanced zT values.

In this study, we specifically explore the role of SnTe incorporation in altering the charge and heat-carrier transport within CuInTe₂. Phonons with varied frequencies were expected to be further scattered by multiscale defects—such as point defects, precipitates, and grain boundaries induced by SnTe incorporation—leading to enhanced thermoelectric properties. In parallel with material composition optimization, the synthesis methodology itself represents a critical, but often overlooked, dimension of thermoelectric materials research. Conventional preparation methods for CuInTe₂ and related compounds typically involve long-duration melting (several hours) followed by slow cooling or hot-pressing. In recent years, rapid synthesis techniques—including microwave-assisted methods, levitation melting combined with spark plasma sintering, and hydrothermal approaches—have emerged as alternatives that significantly reduce processing time from hours to minutes while maintaining or even improving material quality. Meanwhile, microwave heating (MH) offers distinct advantages over conventional synthesis methods, including higher energy conversion efficiency and lower processing cost. Although this technique has been successfully employed in the preparation of thermoelectric materials such as BiCuSeO-Cu₂Se and Cu₂Se [25,26], its application to CuInTe₂ remains largely unexplored.

This work aims to address these gaps by establishing microwave melting combined with SnTe incorporation as a rapid and energy-efficient synthesis pathway for CuInTe₂ modification. While our primary objective is not to achieve the highest possible zT value but rather to demonstrate the feasibility of this innovative synthesis approach, our findings reveal a 1.7-fold enhancement in electrical conductivity relative to pristine CuInTe₂, coupled with significant reduction in lattice thermal conductivity due to multiscale defect scattering. We acknowledge that the current peak zT of 0.44 at 675 K is lower than state-of-the-art CuInTe₂-based materials reported in the literature; however, this study provides critical foundational insights for further optimization of carrier concentration and SnTe doping amounts. Our results establish a viable pathway for CuInTe₂ modification via rapid microwave melting, with systematic optimization

expected to yield significantly improved zT values in forthcoming studies. These findings may serve as a valuable reference for guiding the continued development and optimization of CuInTe_2 thermoelectric systems, particularly emphasizing the balance between performance and energy-efficient synthesis.

2. Results and Discussion

The powder XRD patterns of the prepared CIT samples have been depicted based on the tetragonal CuInTe_2 (space group, I-42d, $a = b = 6.18 \text{ \AA}$, $c = 12.39 \text{ \AA}$), as shown in Figure 1. Additionally, weak diffraction peaks corresponding to the cubic SnTe phase (space group, Fm-3m, $a = b = c = 5.99 \text{ \AA}$) are detected within the resolution limit of XRD. The coexistence of CuInTe_2 and residual SnTe -derived features may contribute to the reduction in lattice thermal conductivity through enhanced phonon scattering at lattice distortions and local strain fields. An enlarged view of the diffraction patterns between 47° and 51° is plotted in Figure 1b. A systematic shift in the diffraction peaks toward higher 2θ values is observed with increasing SnTe content, which is primarily attributed to the partial substitution of indium ($1.56 \text{ \AA}$) by tin ($1.45 \text{ \AA}$), as the smaller ionic radius of Sn leads to a decrease in interplanar spacing. This shift corresponds to a progressive contraction in the lattice parameters, which has also been reported in the Cu_2Se and Cu_3SbSe_4 systems [27,28]. Combined with the lattice distortion schematic (Figure 1c) and XRD patterns, it can be inferred that various structural features present within the material may collectively act as potential phonon-scattering centers. Further analysis of the role played by these effects in modulating the electrical and thermal transport properties of CuInTe_2 will be discussed in a later section.

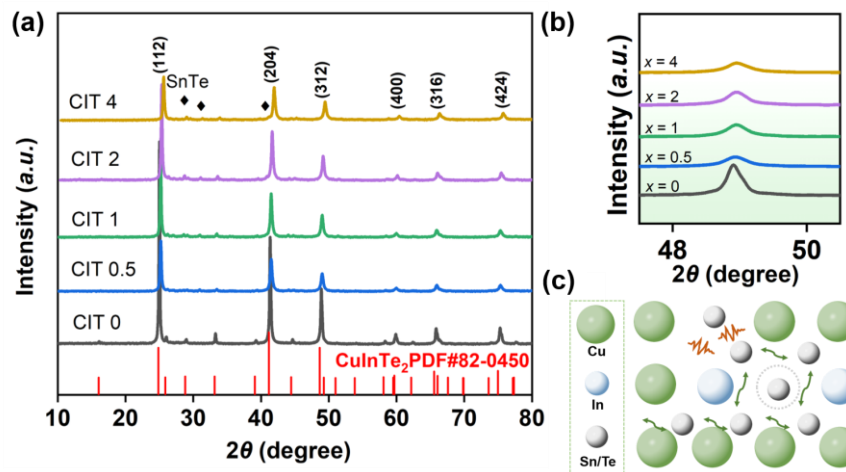


Figure 1. (a) XRD patterns of CIT samples, the black square is labeled as the SnTe phase; (b) the enlarged diffraction peaks of CIT samples; (c) schematic diagram of lattice distortion, schematic diagram of lattice distortion.

To further investigate the fractured surface of CIT samples, SEM analysis was conducted. Densely packed grains were observed, indicating good sinterability and low porosity in the prepared CuInTe_2 . However, a limited number of micro-holes and precipitates, ranging in size from hundreds of nanometers to several microns, were also observed embedded along the grain boundaries of the CuInTe_2 -2 wt% SnTe sample, as marked by the cyan circle in Figure 2. The porosity of the CIT samples was increased upon SnTe incorporation, which is considered beneficial for enhancing phonon scattering and suppressing lattice thermal conductivity. In addition, to further determine the distribution of each constituent element, the CuInTe_2 -4 wt% SnTe sample was analyzed by elemental

mapping. A homogeneous distribution of Sn was confirmed by the EDS analysis presented in Figure 2e, suggesting that Sn derived from SnTe was incorporated into the CuInTe₂ matrix. This can be intuitively interpreted from Figure 2f, where numerous interfaces were provided by residual secondary phases, and pores in this system could lead to significant modifications in the electrical and thermal transport properties. It is well-established that microstructural features such as the size, number, and distribution of pores directly affect the relative density. The density and relative density of the CIT samples fabricated via the microwave method are presented in Table 1. The relative density of all samples exceeded 95%, and the theoretical density was calculated based on the nominal composition of CuInTe₂ with varying SnTe additions. It can be inferred from Table 1 that the relative density of SnTe-modified samples was lower than the pristine phase.

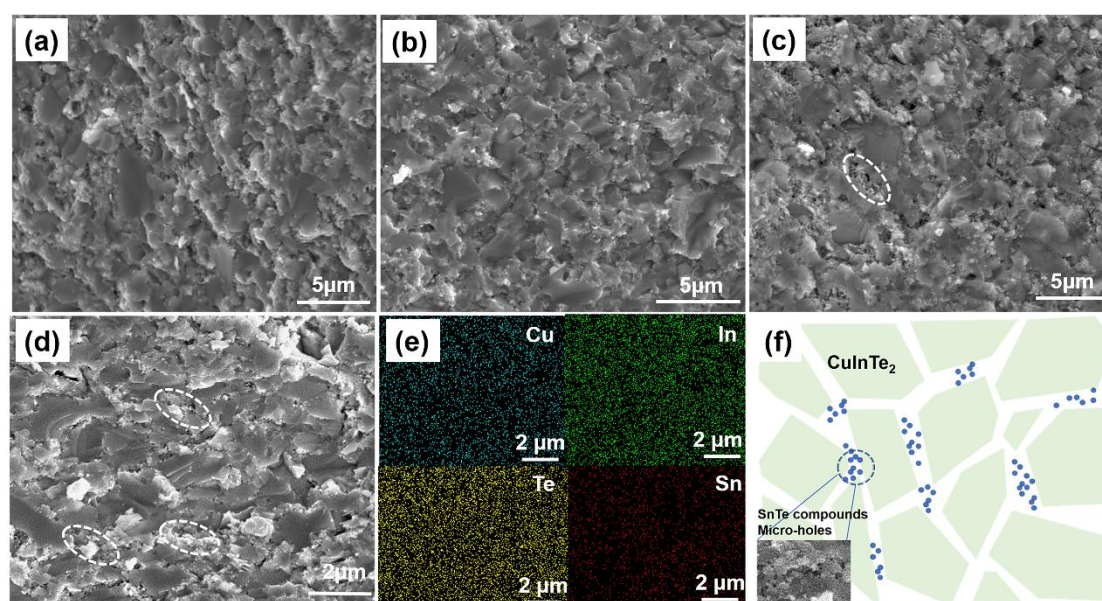


Figure 2. Cross-sectional SEM images of CIT samples. (a) CuInTe₂-0.5 wt% SnTe sample; (b) CuInTe₂-1 wt% SnTe sample; (c) CuInTe₂-2 wt% SnTe sample; (d) CuInTe₂-4 wt% SnTe sample, the white dotted circles clearly show the presence of micrometer-sized holes; (e) EDS elemental mapping for the CuInTe₂-4 wt% SnTe sample; (f) schematic of the phase interface, the blue bullets represent SnTe phase inclusions and micropores

Table 1. The density, relative density, and carrier concentration of CIT samples.

Composition	Density (g/cm ³)	Theoretical Density (g/cm ³)	Relative Density (%)	<i>n</i> (cm ⁻³)
CIT 0	5.970	6.092	98.0	6.7×10^{17}
SnTe	6.322	6.464	97.8	1.4×10^{20}
CIT 0.5	5.919	6.076	97.4	7.5×10^{17}
CIT 1	5.880	6.077	96.7	2.5×10^{18}
CIT 2	5.878	6.081	96.6	6.4×10^{18}
CIT 4	5.867	6.088	96.4	9.6×10^{18}

The electrical properties of CIT samples obtained by the microwave method are shown in Figure 3. The electrical conductivity (σ) and electrical conductivity gradient (k) of CIT samples are displayed in Figure 3a,b, respectively. It can be seen that the electrical conductivity for all samples exhibited a monotonic increase as the temperature was elevated. For a given temperature region, the σ_{ave} of samples increased substantially with SnTe content, rising from $7 \times 10^3 \text{ Sm}^{-1}$ to $2.7 \times 10^4 \text{ Sm}^{-1}$, representing a 170% enhancement

over that of pristine CuInTe₂. It can be observed that the σ of the CuInTe₂-2 wt% SnTe sample and the CuInTe₂-4 wt% SnTe sample remained nearly constant at 750 K, and was 142% higher than that of the pristine phase. In order to quantify the manipulation of electric performance in CuInTe₂ after SnTe addition, the relationship between the slope of σ and the added amount of SnTe at different temperatures were summarized, as shown Figure 3b. For the $x \leq 2$ composites, the value of k increased with the SnTe addition, which was attributed to the high $n \sim 10^{20} \text{ cm}^{-3}$ (Table 1) owing to the spontaneous formation of intrinsic Sn vacancies. For the $x = 4$ sample, the increasing trend of k was slowed down after 475 K. This observation suggests that the interaction between the Cu and SnTe might potentially contribute to the variation in electrical conductivity, although the exact mechanism remains to be further investigated. It is speculated that an ionic exchange process could possibly influence the overall carrier contribution to electrical properties within the temperature range, [29,30] but definitive confirmation requires additional characterization such as XPS analysis. Consequently, with increasing x , a diminution trend of the k for CuInTe₂ is observed. Figure 3c depicts the energy-band model of the SnTe-modified CuInTe₂ system [31]. The significant enhancement in electrical conductivity can be primarily attributed to the substantial increase in hole carrier concentration (from $6.7 \times 10^{17} \text{ cm}^{-3}$ for CIT 0 to $9.6 \times 10^{18} \text{ cm}^{-3}$ for CIT 4, as listed in Table 1), which arises from the spontaneous formation of intrinsic Sn vacancies (V_{Sn}) within the incorporated SnTe. As illustrated in Figure 3c, the introduction of SnTe establishes a type-I (straddling) band alignment at the SnTe/CuInTe₂ hetero-interfaces due to the narrower band gap (0.20 eV) and higher-lying valence band maximum (VBM) of SnTe compared to CuInTe₂ (1.06 eV), driving hole injection from SnTe into the CuInTe₂ matrix and elevating the overall carrier concentration while creating potential barriers at the interfaces. These potential barriers simultaneously enable the energy-filtering effect that preferentially scatters low-energy carriers while allowing high-energy carriers to pass through, enhancing the Seebeck coefficient without significantly compromising electrical conductivity. Furthermore, Sn incorporation and the resulting lattice contraction alter the band curvature near the Fermi level, with calculations based on the single parabolic band (SPB) model indicating an increase in the density-of-states effective mass (m_e) from $0.04 m_e$ (CIT 0) to $0.08 m_e$ (CIT 2). Therefore, the superior electrical transport properties result from a synergistic effect combining enhanced carrier concentration from Sn vacancies, carrier energy filtering at hetero-interfaces, and moderately increased density-of-states effective mass due to band structure modulation, which decouples the traditional inverse relationship between electrical conductivity and the Seebeck coefficient and enables the observed significant improvement in the power factor. The carrier concentration values were extracted from Hall effect measurements using the van der Pauw method. The Hall coefficient R_H was measured, and the carrier concentration n was calculated using the relation $n = 1/eR_H$, where e represents the elementary charge. The temperature-dependent Seebeck coefficient for the CIT samples is plotted in Figure 3d. The positive values of the Seebeck coefficient indicated that holes were the dominant charge carriers across all compositions. Moreover, the Seebeck coefficient decreases with increasing SnTe content, which is attributed to the enhanced carrier concentration. Additionally, the effective mass was calculated to be $0.04 m_e$ and $0.08 m_e$ for CIT0.5 and CIT2, where m_e represents the free electron mass. These observed variations suggest that SnTe incorporation influences both the carrier concentration and the effective mass, as summarized in Figure 3e. Within the framework of a single parabolic band (SPB) model, assuming acoustic phonon scattering, the effective mass of charge carriers (m^*) was derived using the following expressions:

$$m^* = \frac{1}{2k_B T} \left(\frac{\hbar^3}{4\pi F_{\frac{1}{2}}(\eta)} \right)^{\frac{2}{3}} \quad (1)$$

where the k_B , $\hbar$, η , and e are the Boltzmann constant, reduced Planck constant, reduced Fermi energy, and elementary charge, respectively.

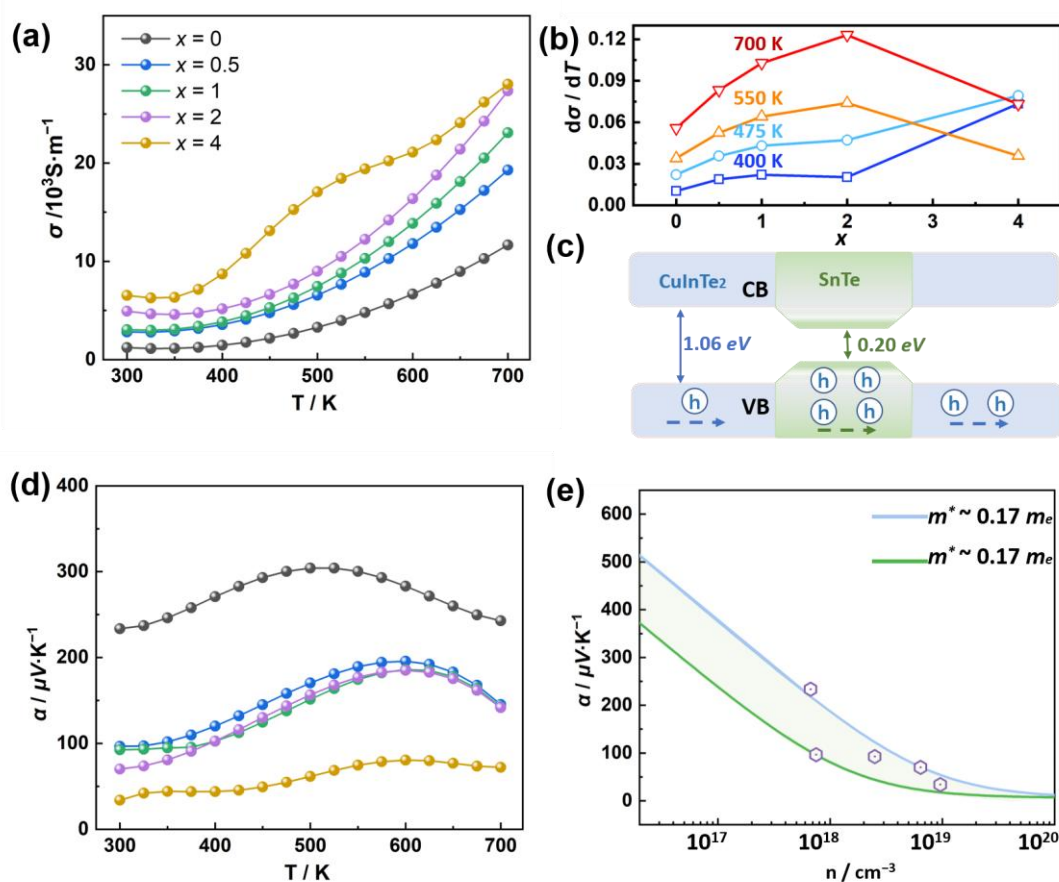


Figure 3. (a) Electrical conductivity of CIT samples; (b) electrical conductivity gradient; (c) schematic diagram of the energy band of SnTe/CuInTe₂ composite material, where h represents the hole; (d) Seebeck coefficient; (e) carrier-concentration-dependent Seebeck coefficient.

The total thermal conductivity (κ_{tot}) of CIT samples is shown in Figure 4a. Owing to strong phonon–phonon interactions, which are typical for crystalline materials, the κ_{tot} was observed to decreased rapidly with the increasing temperature [32]. Additionally, the total thermal conductivity measured for the $x = 2$ sample was found to be substantially lower than that of the pristine composition. To further elucidate the composition dependence of thermal conductivity, we extended our measurements to include additional samples with 1.5% and 2.5% SnTe inclusion. As clearly demonstrated in Figure 4a, the thermal conductivity exhibits a non-monotonic variation with respect to SnTe content. The lattice conductivity decreases progressively from the pristine sample to the $x = 1.5\%$ sample, reaches a minimum at $x = 2$ wt%, and then shows a noticeable increase for both the $x = 2.5\%$ and $x = 4\%$ samples. This non-monotonic behavior can be rationalized by considering the competing mechanisms governing phonon scattering in the SnTe-modified CuInTe₂ system. The electronic thermal conductivity (Figure 4d) was calculated using $\kappa_c = L\sigma T$. To avoid underestimation of the lattice thermal conductivity contribution, the Lorentz constant (L) obtained from the single parabolic band (SPB) model was adopted, as shown in Figure 4b. It can be seen that the trend of electronic thermal conductivity of CIT samples was consistent with the electrical conductivity. With increasing SnTe content, the κ_L of CIT samples decreased and reached $2.3 \text{ Wm}^{-1}\text{K}^{-1}$ at 300 K in the CuInTe₂-4 wt% SnTe sample (Figure 4c), which can be partly attributed to

enhanced phonon scattering at lattice distortions and local strain fields induced by SnTe incorporation.

It is speculated that the presence of Sn-related defects and residual structural features enhanced phonon scattering across the entire measured temperature range. To further elucidate the origin of low thermal conductivity, the structural defect parameters u (anion positional parameter) and η (tetragonal distortion parameter) of samples were calculated. These parameters are derived from the Abrahams and Bernstein formulas and characterize the degree of structural distortion in the chalcopyrite crystal lattice:

$$\mu = \frac{1}{2} - \frac{1}{4\sqrt{2}} \left(\frac{c^2}{a^2} - 2 \right)^{\frac{1}{2}} \quad (2)$$

$$\eta = \frac{c}{2a} \quad (3)$$

The tetragonal distortion parameter η (Equation (1)) quantifies the elongation or compression of the anion tetrahedron along the c -axis relative to the a -axis for an ideal zinc-blend derivative structure with a perfectly cubic unit cell, $\eta = 1$. Deviation from unity indicates tetragonal distortion of the lattice. The anion positional parameter u (Equation (2)) defines the position of the chalcogen atom (Te) along the body diagonal of the tetrahedron. In an ideal chalcopyrite structure, the theoretical value is $u = 0.25$. Deviations from this ideal value ($|\Delta u| = |u - 0.25|$) reflect anion displacement due to differences in cation–anion bond strengths and local strain fields. The lattice constants, which are critical for structural defect characterization, are listed in Table 2. The crystal structure and lattice parameters were determined by X-ray diffraction (XRD) analysis. The lattice constants were calculated using Bragg's law ($n\lambda = 2d \sin \theta$), where λ is the X-ray wavelength (Cu $K\alpha$, $\lambda = 1.5406 \text{ \AA}$), θ is the diffraction angle, and d is the interplanar spacing. For the cubic crystal structure, the lattice constant (a) was calculated using the relationship $a = \lambda \sqrt{h^2 + k^2 + l^2} / (2 \sin \theta)$, where (h , k , and l) are the Miller indices of the diffraction peaks. The lattice constant values reported in Table 2 were obtained by averaging calculations from multiple diffraction peaks to ensure accuracy. To estimate the experimental uncertainty, each sample was measured three times. The relative standard deviation was approximately $\pm 2\%$. As presented in Table 2, the lattice constants of the CIT samples decreased progressively with SnTe incorporation, indicating lattice contraction induced by the substitution of Sn ions into the CuInTe_2 crystal structure. The value of $|\Delta u|$ was increased from 0.0008 to 0.0052, indicating that the lattice distortion was significantly altered by the introduction of SnTe. It was proposed that the slight lattice parameter variation originated from internal lattice mismatch within the CIT samples, which was, in turn, expected to play a role in determining the thermal transport properties of the SnTe-modified CuInTe_2 system [33]. At lower SnTe concentrations ($\leq 2 \text{ wt\%}$), the primary scattering mechanism is point-defect scattering induced by the substitutional incorporation of Sn ions into the CuInTe_2 lattice. This results in significant lattice distortion, as evidenced by the increased anion positional parameter deviation $|\Delta u|$ (from 0.0008 in pristine to 0.0052 at $x = 2$), which strongly suppresses phonon propagation. However, as the SnTe concentration exceeds 2 wt%, additional microstructural features emerge that may alter the thermal transport landscape.

It should be noted that the reduction in thermal conductivity may result from multiple mechanisms, including porosity, SnTe inclusions, grain boundaries, and Sn incorporation into the CuInTe_2 lattice. The present observations suggest that there exists an optimal SnTe concentration ($\sim 2 \text{ wt\%}$) at which the phonon-scattering efficiency is maximized, balancing between sufficient point-defect density and minimal adverse effects from secondary phase formation. This optimal composition represents a trade-off between the beneficial effect of enhancing point-defect scattering and the potential

detrimental impact of excess SnTe on the overall microstructure and thermal conductivity. However, the relative contribution of each factor requires quantitative analysis that is beyond the scope of this study. We plan to conduct systematic investigations in our future work to characterize the microstructure and quantitatively evaluate the contribution of each scattering mechanism to the overall thermal conductivity.

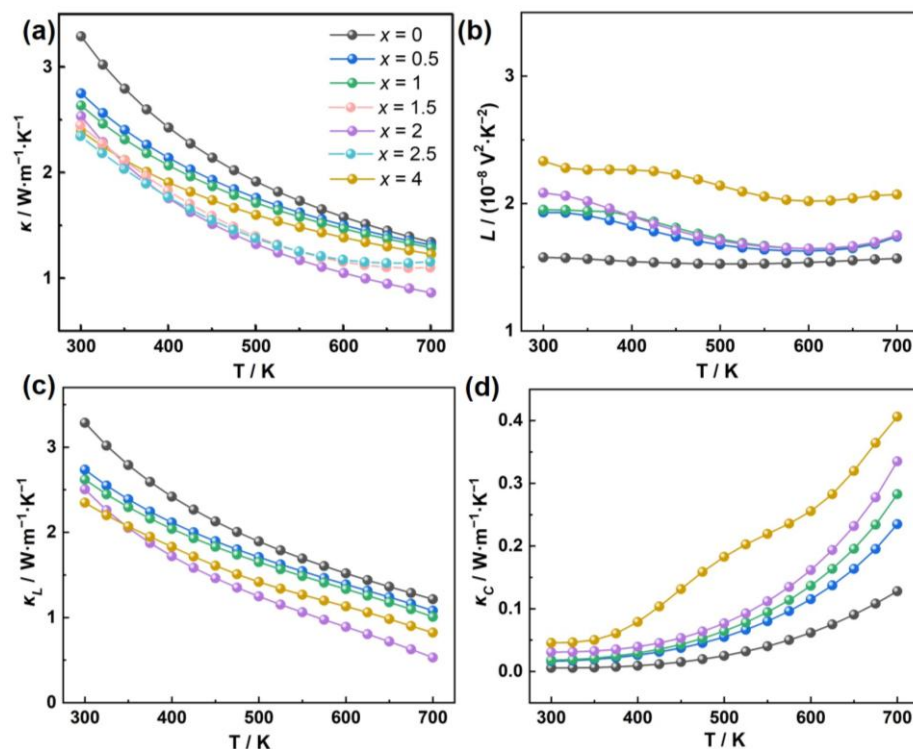


Figure 4. Thermal performance of CIT samples. (a) Thermal conductivity. (b) Lorentz constant; (c) lattice thermal conductivity; (d) carrier thermal conductivity.

Table 2. The structural defect parameters of CIT samples.

Composition	$a/\text{\AA}$	$c/\text{\AA}$	u	$ \Delta u $ $= u - 0.25 $	η	$ \Delta \eta $ $= \eta - 1 $
CIT 0	6.1852	12.3910	0.2492	0.0008	1.0017	0.0017
SnTe	5.99	5.99				
CIT 0.5	6.1501	12.3992	0.2460	0.0040	1.0080	0.0080
CIT 1	6.1334	12.3910	0.2450	0.0050	1.0101	0.0101
CIT 2	6.1323	12.3929	0.2448	0.0052	1.0105	0.0105
CIT 4	6.1852	12.3910	0.2492	0.0008	1.0017	0.0017

The thermoelectric performance of the CIT samples was evaluated by the dimensionless figure of merit, zT , with its temperature-dependent values shown in Figure 5a. The zT of CIT samples was observed to increase with the temperature. Benefiting from the combination of enhanced electrical performance and reduced lattice thermal conductivity, a peak zT value of 0.44 at 675 K was achieved for the CuInTe₂-2 wt% SnTe sample, representing a 1.4-fold enhancement compared to the pristine matrix. The average zT values of CIT samples are shown in Figure 5b. The average zT was calculated by integrating the temperature-dependent zT curve over the measured temperature range and dividing by the temperature interval. The average zT value for CuInTe₂-2 wt% SnTe sample was higher than that of the pure CuInTe₂. Furthermore, a comparison of the temperature-dependent zT values for several reported *p*-type CuInTe₂ materials with excellent thermoelectric performance is summarized in Figure 5c [28,34–37]. In addition,

pure CuInTe_2 can achieve zT values of up to 1.18 at 850 K through specific preparation techniques involving Cu-vacancy engineering [38]. More recent studies have reported even higher zT values: $\text{Cu}_{0.992}\text{InTe}_2$ achieved $zT \approx 0.9$ at 873 K with precision copper vacancy regulation, and CuInTe_2 -based alloys with chemical composition modulation achieved $zT = 1.51$ at 838 K [23,39]. Our work adopted a rapid microwave melting method combined with SnTe inclusion, which represents an initial exploratory. Nevertheless, we recognize that the current zT value of 0.44 at 675 K is lower than state-of-the-art CuInTe_2 -based materials reported in the literature. Future work will focus on further optimizing the Cu-deficiency level, carrier concentration, and SnTe doping amount to enhance the thermoelectric performance.

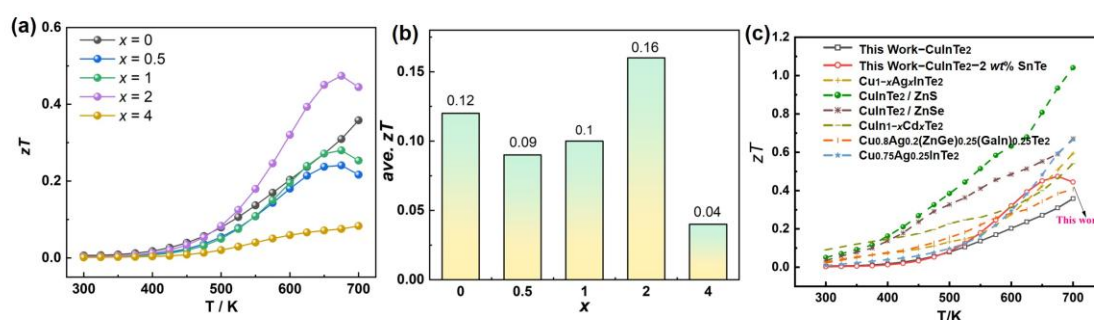


Figure 5. (a) zT of CIT samples as a function of temperature. (b) The average zT of CIT samples. (c) The comparison of reported zT for CuInTe_2 compound (continuous thin line).

3. Materials and Methods

The stoichiometric CuInTe_2 and SnTe compounds were fabricated via a cold pressing–microwave melting method. High-purity copper, indium, and tellurium powders (Aladdin, $\geq 99.99\%$) were first homogenized by ball milling, and then the mixture was pressed into bulk precursors. The pressed CuInTe_2 was placed into a carbon crucible and encapsulated in an evacuated silica ampoule. Microwave heating of the encapsulated sample was conducted for 10 min, after which the ampoule was slowly cooled. Following the same procedure, crystalline SnTe ingots of high quality were synthesized from appropriately proportioned mixtures of Sn (99.99%) and Te (99.999%) powders. The CuInTe_2 and SnTe powders were mixed according to the desired SnTe weight fractions ($x = 0, 0.5, 1, 2, 4$ wt%), and then ball milled for 12 h at a rotational speed of 300 rpm using a planetary ball mill. Isopropanol was used as the milling medium to prevent oxidation, and the slurry was dried at 80 °C under vacuum for 24 h after milling. The resulting composite powders were then densified by rapid hot pressing (RHP) at 673 K for 30 min under an axial compressive stress of 60 MPa, yielding disc-shaped specimens with dimensions of $\phi 12 \times 1.5$ mm. Ultimately, the high-density bulk CuInTe_2 - x wt% SnTe ($x = 0, 0.5, 1, 2, 4$) sample thermoelectric materials were obtained, denoted as CIT 0, CIT 0.5, CIT 1, CIT 2, and CIT 4, respectively.

The relative density (d) of the CIT specimens was determined using the Archimedes method. Phase identification was conducted by X-ray diffraction (XRD) with $\text{Cu K}\alpha$ radiation, over a 2θ range of 10 – 80° at a scanning rate of $4^\circ/\text{min}$. Under a low-pressure ($\sim 10^2$ Pa) helium atmosphere, electrical performance testing of the samples was conducted using a Seebeck coefficient/resistance determination system (LSR-3, Linseis, Germany) from 300 to 750 K to determine the electrical conductivity and Seebeck coefficient. All the block samples used for testing were ground and polished into disc-shaped samples with a diameter of ~ 12.7 mm and a thickness of ~ 1.3 mm. Moreover, the thermal conductivity of the disc-shaped samples was determined using a flash laser thermal conductivity meter

(LFA1000, Linseis, Germany) in a flowing argon atmosphere within a temperature range consistent with the electrical properties. The thermal conductivity was derived using the standard formula $\kappa = \lambda \rho C_p$, where λ , ρ , and C_p are the thermal diffusivity, density, and specific heat, respectively. Thermal diffusivity data were collected employing a laser flash diffusivity instrument. The inherent error associated with this ultrasonic method was quantified as 1%, arising predominantly from the measurement accuracy of sample thickness. At ambient temperature, the electrical transport performance parameters, including hole carrier concentration and carrier mobility, were measured using the temperature-variable hall effect testing system (CH-100, Beijing, China) in a reversible magnetic field of 0.5 T.

4. Conclusions

An effective strategy for improving the electrical properties of *p*-type CuInTe₂ is proposed, wherein SnTe incorporation is employed to achieve enhanced thermoelectric performance. Electrical conductivity of up to 7000 Sm^{−1} at room temperature was achieved due to the increased carrier concentration upon 4 wt% SnTe addition. The introduction of lattice distortion and micro-holes into CuInTe₂ efficiently enhanced phonon scattering across a wide spectrum of mean free paths. The κ_L of the CIT sample decreased from 3.4 Wm^{−1}K^{−1} to 2.3 Wm^{−1}K^{−1} at room temperature, while the electrical properties remained undegraded. As a result, an average *zT* of 0.16 was attained in *p*-type CuInTe₂-based compounds. This work demonstrates an alternative strategy for simultaneously optimizing carrier concentration and thermal conductivity, providing a novel route to enhancing the thermoelectric performance of copper-based ternary compounds.

Author Contributions: Conceptualization, L.B., Y.W., W.W., and L.S.; validation, L.B., W.Z., X.L., and D.Z.; formal analysis, L.B. and D.Z.; data curation, L.B.; writing—original draft preparation, L.B. and D.Z.; writing—review and editing, L.B. and D.Z.; funding acquisition, L.B., W.Z., X.L., and D.Z. All authors have read and agreed to the published version of the manuscript.

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Data Availability Statement: The original contributions presented in this study are included in the article. Further inquiries can be directed to the corresponding author.

Conflicts of Interest: The author Yongpeng Wang were employed by Binzhou Special Equipment Inspection and Research Institute company (Binzhou, China). The author Shilai Zeng were employed by Shandong Huanbang Electronic Technology Co., Ltd. Company (Linyi, China). The remaining authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

References

1. Zhao, P.; Xue, W.; Zhang, Y.; Zhi, S.; Ma, X.; Qiu, J.; Zhang, T.; Ye, S.; Mu, H.; Cheng, J.; et al. Plasticity in single-crystalline Mg_3Bi_2 thermoelectric material. *Nature* **2024**, *631*, 777–782.
2. Yu, J.; Liu, X.; Hu, H.; Jiang, Y.; Zhuang, H.-L.; Li, H.; Su, B.; Li, J.-W.; Han, Z.; Wang, Z.; et al. Ultralow thermal conductivity and high zT of Cu_2Se -based thermoelectric materials mediated by TiO_2 nanoclusters. *Joule* **2024**, *8*, 2652–2666.
3. Zhang, Z.; Ming, C.; Song, Q.; Wang, L.; Chen, H.; Liao, J.; Wang, C.; Chen, L.; Bai, S. Grain boundary modulation improved thermal stability of high thermoelectric performance $\text{Mg}_3(\text{Sb,Bi})_2$ -based compounds. *Acta Mater.* **2025**, *287*, 120806.
4. Yu, J.; Hu, H.; Jiang, Y.; Zhuang, H.-L.; Thong, H.-C.; Su, B.; Li, J.-W.; Han, Z.; Li, H.; Pei, J.; et al. Interface-enhanced high-temperature thermoelectricity in $\text{Cu}_{1.99}\text{Se/B}_4\text{C}$ composites with synergistically improved mechanical strength. *Adv. Energy Mater.* **2024**, *14*, 2303942.
5. Zhang, C.; Jin, K.; Dong, H.; Xu, W.; Xu, P.; Yan, Z.; Zhao, W.; Xu, B.; Fu, L. Synergistic enhancement of thermoelectric performance of n -type PbTe by resonant level and single-atom-layer vacancies. *Nano Energy* **2024**, *126*, 109615.
6. Xiong, Q.; Han, G.; Wang, G.; Lu, X.; Zhou, X. The doping strategies for modulation of transport properties in thermoelectric materials. *Adv. Funct. Mater.* **2024**, *34*, 2411304.
7. Wei, S.; Ji, Z.; Li, W.; Zheng, S.; Wang, Q.; Li, Z.; Yu, L.; Luo, S.; Song, W. Enhanced thermoelectric properties of Cu_3SbSe_4 -based materials by synergistic modulation of carrier concentration and phonon scattering. *J. Mater.* **2024**, *10*, 339–347.
8. Wang, D.; Liu, W.; Mao, Y.; Li, S.; Yin, L.; Wu, H.; Li, M.; Wang, Y.; Shi, X.; Yang, X.; et al. Decoupling carrier-phonon scattering boosts the thermoelectric performance of n -Type GeTe -based materials. *J. Am. Chem. Soc.* **2024**, *146*, 1681–1689.
9. Li, L.; Zhai, W.; Wang, C.; Chen, Y.; Li, S.; Fan, P.; Cheng, Z.; Yang, G.; Wang, J.; Mao, Y. Maximizing phonon scattering efficiency by Cu_2Se alloying in AgCuTe thermoelectric materials. *J. Mater. Chem. A* **2022**, *10*, 6701–6712.
10. Chang, C.; Wu, M.; He, D.; Pei, Y.; Wu, C.-F.; Wu, X.; Yu, H.; Zhu, F.; Wang, K.; Chen, Y. 3D charge and 2D Phonon Transports Leading to High Out-of-plane zT in n -Type SnSe Crystals. *Science* **2018**, *360*, 778–783.
11. Ozen, M.; Yahyaoglu, M.; Candolfi, C.; Veremchuk, I.; Kaiser, F.; Burkhardt, U.; Snyder, G.J.; Grin, Y.; Aydemir, U. Enhanced thermoelectric performance in $\text{Mg}_{3+x}\text{Sb}_{1.5}\text{Bi}_{0.49}\text{Te}_{0.01}$ via engineering microstructure through melt-centrifugation. *J. Mater. Chem. A* **2021**, *9*, 1733–1742.
12. Park, J.; Xia, Y.; Ozoliņš, V.; Jain, A. Optimal band structure for thermoelectrics with realistic scattering and bands. *NPJ Comput. Mater.* **2021**, *7*, 43.
13. Li, P.; Shi, J.; Wu, X.; Li, J.; Hu, L.; Liu, F.; Li, Y.; Ao, W.; Zhang, C. Interfacial engineering of solution-processed Bi_2Te_3 -based thermoelectric nanocomposites via graphene addition and liquid-phase-sintering process. *Chem. Eng. J.* **2022**, *440*, 135882.
14. Ahmad, M.; Agarwal, K.; Munoz, S.G.; Ghosh, A.; Kodan, N.; Kolosov, O.V.; Mehta, B.R. Engineering interfacial effects in electron and phonon transport of $\text{Sb}_2\text{Te}_3/\text{MoS}_2$ multilayer for thermoelectric zT above 2.0. *Adv. Funct. Mater.* **2022**, *32*, 2206384.
15. Deng, Q.; Wen, J.; An, X.; Xie, Y.; Luo, J.; Zhao, Z.; Sa, B.; Dong, H.; Ang, R. Thermoelectric cooling and low-temperature power generation in n -type PbTe enabled by band flattening and entropy engineering. *Chem. Eng. J.* **2025**, *506*, 160319.
16. Ma, Z.; Xu, T.; Li, W.; Cheng, Y.; Li, J.; Zhang, D.; Jiang, Q.; Luo, Y.; Yang, J. High entropy semiconductor AgMnGeSbTe_4 with desirable thermoelectric performance. *Adv. Funct. Mater.* **2021**, *31*, 2103197.
17. Pakdel, A.; Guo, Q.; Nicolosi, V.; Mori, T. Enhanced thermoelectric performance of $\text{Bi-Sb-Te/Sb}_2\text{O}_3$ nanocomposites by energy filtering effect. *J. Mater. Chem. A* **2018**, *6*, 21341–21349.
18. Zhou, Z.; Yang, J.; Jiang, Q.; Zhang, D.; Xin, J.; Li, X.; Ren, Y.; He, X. Thermoelectric performance of SnTe with ZnO carrier compensation, energy filtering, and multiscale phonon scattering. *J. Am. Ceram. Soc.* **2017**, *100*, 5723–5730.
19. Wei, J.; Yu, W.; Li, Z.; Hu, J.; Wang, T.; Yu, Y.; Guo, X. Pressure-mediated band engineering and cation ordering for enhanced thermoelectric performance in CuInTe_2 chalcopyrites. *Chem. Eng. J.* **2025**, *521*, 166531.
20. Zhang, T.; Ning, S.; Zhang, T.; Qi, N.; Su, X.; Tang, X.; Chen, Z. Stepwise vacancy manipulation for optimized carrier concentration and blocked phonon transport realizing record high figure of merit zT in CuInTe_2 . *Adv. Funct. Mater.* **2025**, *35*, 2419984.
21. Meschke, V.; Pak, A.; Adamczyk, J.M.; Gomes, L.C.; Baumann, S.; Rome, G.; Porter, C.; Ciesielski, K.; Ertekin, E.; Toberer, E. Amphoteric doping and thermoelectric transport in the $\text{CuInTe}_2\text{-ZnTe}$ solid solution. *J. Mater. Chem. C* **2025**, *13*, 8792–8801.
22. Kim, H.; Kihoi, S.K.; Lee, H.S. Point defects control in non-stoichiometric CuInTe_2 compounds and its corresponding effects on the microstructure and thermoelectric properties. *J. Alloys Compd.* **2021**, *869*, 159381.
23. Li, S.; Xie, C.; Liu, K.; Liu, J.; Mei, J.; Ding, G.; Cui, J.; Xu, W.; Cao, C.; Zhang, Q.; et al. Precision engineering of copper vacancies in CuInTe_2 for synergistically enhanced thermoelectric performance. *J. Mater. Chem. A* **2025**, *13*, 27691–27700.

24. Luo, Y.; Yang, J.; Jiang, Q.; Li, W.; Zhang, D.; Zhou, Z.; Cheng, Y.; Ren, Y.; He, X. Progressive regulation of electrical and thermal transport properties to high-performance CuInTe₂ thermoelectric materials. *Adv. Energy Mater.* **2016**, *6*, 1600007.
25. Lei, Y.; Yong, C.; Qiu, J.; Li, Y.; Xu, L.; Ye, F.; Zhang, S.; Wang, D.; Ni, G. Optimization of thermoelectric performance in BiCuSeO-based materials by constructing Cu₂Se composite interface. *Ceram. Int.* **2025**, *51*, 11331–11337.
26. Bo, L.; Zhang, R.; Zhao, H.; Hou, Y.; Wang, X.; Zhu, J.; Zhao, L.; Zuo, M.; Zhao, D. Achieving high thermoelectric properties of Cu₂Se via lattice softening and phonon scattering mechanism. *ACS Appl. Energy Mater.* **2022**, *5*, 6453–6461.
27. Zhao, K.; Guan, M.; Qiu, P.; Blichfeld, A.B.; Eikeland, E.; Zhu, C.; Ren, D.; Xu, F.; Iversen, B.B.; Shi, X. Thermoelectric Properties of Cu₂Se_{1-x}Te_x Solid Solutions. *J. Mater. Chem. A* **2018**, *6*, 6977–6986.
28. Wang, C.; Ma, Q.; Xue, H.; Wang, Q.; Luo, P.; Yang, J.; Zhang, W.; Luo, J. Tetrahedral distortion and thermoelectric performance of the Ag-substituted CuInTe₂ chalcopyrite compound. *ACS Appl. Energy Mater.* **2020**, *3*, 11015–11023.
29. Olvera, A.; Moroz, N.; Sahoo, P.; Ren, P.; Bailey, T.; Page, A.; Uher, C.; Poudeu, P. Partial indium solubility induces chemical stability and colossal thermoelectric figure of merit in Cu₂Se. *Energy Environ. Sci.* **2017**, *10*, 1668–1676.
30. Hwang, J.; Lee, M.; Yu, B.-K.; Han, M.-K.; Kim, W.; Kim, J.; Al Orabi, R.A.R.; Wang, H.; Acharya, S.; Kim, J. Enhancement of thermoelectric performance in a non-toxic CuInTe₂/SnTe coated grain nanocomposite. *J. Mater. Chem. A* **2021**, *9*, 14851–14858.
31. Tan, G.; Zhao, L.-D.; Shi, F.; Doak, J.W.; Lo, S.-H.; Sun, H.; Wolverton, C.; Dravid, V.P.; Uher, C.; Kanatzidis, M.G. High thermoelectric performance of p-type SnTe via a synergistic band engineering and nanostructuring approach. *J. Am. Chem. Soc.* **2014**, *136*, 7006–7017.
32. Li, X.; Li, D.; Xin, H.; Zhang, J.; Song, C.; Qin, X. Effects of bismuth doping on the thermoelectric properties of Cu₃SbSe₄ at moderate temperatures. *J. Alloys Compd.* **2013**, *561*, 105–108.
33. Zhang, D.; Wang, J.; Zhang, L.; Lei, J.; Ma, Z.; Wang, C.; Guan, W.; Cheng, Z.; Wang, Y. High thermoelectric performance of Bi_{0.46}Sb_{1.54}Te₃-SnTe: Synergistic modulation of electrical and thermal transport by the introduction of thermoelectric hetero nano region. *ACS Appl. Mater. Interfaces* **2019**, *11*, 36658–36665.
34. Xia, Z.; Wang, G.; Zhou, X.; Wen, W. Substitution defect enhancing thermoelectric properties in CuInTe₂. *Mater. Res. Bull.* **2018**, *101*, 184–189.
35. Luo, Y.; Jiang, Q.; Yang, J.; Li, W.; Zhang, D.; Zhou, Z.; Cheng, Y.; Ren, Y.; He, X.; Li, X. Simultaneous regulation of electrical and thermal transport properties in CuInTe₂ by directly incorporating excess ZnX (X = S, Se). *Nano Energy* **2017**, *32*, 80–87.
36. Cheng, N.; Liu, R.; Bai, S.; Shi, X.; Chen, L. Enhanced thermoelectric performance in Cd doped CuInTe₂ compounds. *J. Appl. Phys.* **2014**, *115*, 163705.
37. Cai, J.; Yang, J.; Liu, G.; Wang, H.; Shi, F.; Tan, X.; Ge, Z.; Jiang, J. Ultralow thermal conductivity and improved zT of CuInTe₂ by high-entropy structure design. *Mater. Today Phys.* **2021**, *18*, 100394.
38. Janicek, P.; Kucek, V.; Kasparova, J.; Plechacek, T.; Cernoskova, E.; Benes, L.; Munzar, M.; Drasar, C. Study of Indium Non-stoichiometry in CuInTe₂ and Its Effects on the Thermoelectric Properties. *J. Electron. Mater.* **2019**, *48*, 2112–2119.
39. Qu, L.; Luo, Y.; Li, C.; Du, Z.; Li, X.; Cui, J. Chemical Composition Modulation Realizing Remarkable Improvement of Thermoelectric Performance in CuInTe₂-Based Alloy. *ACS Appl. Mater. Interfaces* **2024**, *16*, 23596–23605.

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