Multi-doping in SnTe: Improvement of Thermoelectric Performance due to Lower Thermal Conductivity and Enhanced Power Factor

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Abstract: In recent years, tin telluride (SnTe) has attracted considerable interest due to its potential thermoelectric application as a lead-free rock-salt analogue of PbTe. However, pristine SnTe samples show high thermal conductivity and low Seebeck coefficients, resulting in poor thermoelectric performance. In this study, the thermoelectric performance of SnTe was enhanced by well-designed multi-doping, where significantly reduced thermal conductivity and improved Seebeck coefficients were achieved at the same time. The doped SnTe samples were prepared by hot pressing. The lattice thermal conductivity of SnTe samples is obviously decreased by alloying with Se and S. The transmission electron microscope shows the existence of larger amount of nano-precipitates and the lattice distortions in the alloyed samples. For example, the lattice thermal conductivity of SnTe0.7S0.15Se0.15 sample is reduced to 0.99 W m⁻¹ K⁻¹ at 300 K. The results reveal that the Seebeck coefficients are improved by introducing In resonant state in the band structure of SnTe. The experiments suggest the effectiveness of designed multi-doping in the thermoelectric performance enhancement of SnTe, and a promising ZT of 0.8 at 850 K is achieved in Sn0.99In0.01Te0.7S0.15Se0.15. The discovery suggests that SnTe is a promising medium-temperature thermoelectric candidate.

Key words: tin telluride; thermoelectric performance; resonant state; lattice thermal conductivity

The conversion efficiency of thermoelectric material is quantified by the so-called ZT value defined as αS²T/(κₑlec + κₗlat), where α, S, κₑlec, κₗlat, and T are the electrical conductivity, the Seebeck coefficient, electronic and lattice thermal conductivity, and absolute temperature, respectively[1-4]. The most commonly pursued approaches to enable high ZT values include the band structure manipulation for high power factor αS² by resonant levels near the Fermi level[5-11] or introducing band convergence[12-14], and phonon scattering enhancement by alloying or nanostructuring to minimize the thermal conductivity[15-29].

Over the last few years, SnTe has stepped into sight as a promising thermoelectric candidate due to its less toxic and a potential substitute of PbTe[21, 30]. But comparing to PbTe, SnTe suffers from a much lower ZT due to a smaller band gap (~0.18 eV) and the higher separation between the light-hole band at L and the heavy-hole band at ε (~0.3 eV)[31]. In addition, the relatively high thermal conductivity also limits its thermoelectric performance. Recent reports showed that the Seebeck coefficient of SnTe could be improved considerably by Cd, Mg, Mn, or Hg doping to converge the two valence bands and enlarge the band gap[32-43]. In doping in SnTe has also been found to create the resonant level inside the valence band and enhance the Seebeck coefficients around room temperature[21], which is similar to the Tl-doped PbTe system[5, 11].

On the other hand, the remarkable decrease in the lattice thermal conductivity could also lead to obviously improved thermoelectric performance of SnTe via phonon scattering by solid solution point defects, secondary phase nanoprecipitates[27-28, 44-45], and mesoscale grain boundaries[45-46]. For example, some nanoscale secondary phases such as CdS, HgTe, SrTe, Cu₂Se and Cu₂S, was introduced to regulate the thermal transport of SnTe, and
the heat-carrying phonons were strongly scattered as expected[33-35,47-49]. Therefore, it is necessary to combine the Seebeck coefficient enhancement and designed phonon scattering to further improve the thermoelectric performance of SnTe materials.

In this work, we try to improve the thermoelectric performance of SnTe from the coexistence of resonant levels and secondary phases scattering. Firstly, the thermal conductivity of SnTe is decreased through secondary phases of SnS by S, Se doping. Then the optimized sample with 15mol% S and Se incorporated is used for the following In doping study. The thermoelectric properties are improved by multi-doping.

1 Experimental

Commercial elements Sn, Te, S, Se and In were used as starting materials. The materials with nominal compositions of SnTe1–2, S, Se, (x=0, 0.05, 0.1, and 0.15), and Sn1–yInyTe0.75S0.15Se0.15 (y=0, 0.0025, 0.005, 0.01, and 0.015) were prepared by hot pressing[50-51]. X-ray diffraction (XRD) analysis was performed in a reflection geometry on a Bruker D8 diffractometer. The microstructures were investigated by the high resolution transmission electron microscope (JEOL 2100F), attached with an energy dispersive spectrometer (EDS). The samples were cut into microscope (10 mm×2 mm for thermal transport property measurement) and 10 mm×2 mm for thermal transport property measurement. The Seebeck coefficient S and electrical conductivity σ were simultaneously measured using a ZEM-3 (ULVAC) instrument under a low-pressure helium atmosphere from room temperature to 850 K. The thermal conductivity was calculated from κ=TDpCp where D is the thermal diffusivity, ρ is the density, and Cp is the specific heat. D was measured using the laser flash diffusivity method in a Netzsch LFA-457, ρ was measured by using Archimedes principle, and Cp was calculated from others data and our previous work[16,44,50]. The Hall coefficient RH was measured at room temperature using a physical properties measurement system (Quantum Design, PPMS-9). The carrier concentration n and the hall mobility μ were estimated according to n=1/εRH, and μ=εRH by using the single parabolic band approximation, respectively.

2 Results and Discussions

Figure 1 shows the total thermal conductivity κtot and lattice thermal conductivity of SnTe1–2, S, Se, (x=0, 0.05, 0.1, and 0.15) samples. The lattice thermal conductivity κlat is calculated by subtracting electrical thermal conductivity κele from the total thermal conductivity, and the electrical thermal conductivity was calculated using the Wiedemann-Frenz relationship κele=LaT. It should be noted that the Lorenz number L is roughly obtained by fitting S using a single parabolic band model. The corresponding temperature dependent electrical conductivity σ and Seebeck coefficient S are summarized in Supporting Information.

As shown in Fig.1(b), the lattice thermal conductivities of SnTe1–2, S, Se, (x=0.05, 0.1, and 0.15) samples decrease significantly compared to that of pristine SnTe, which may be due to the excess phonon scattering by the solid solution and point defects. Considering that SnTe1–2, S, Se with 15mol% S and Se exhibits the lowest κlat, x=0.15 is used for the following In doping investigation. Typically, the lattice thermal conductivity of SnTe0.75S0.15Se0.15 sample is 0.99 W m−1 K−1 at 300 K, mildly decreases to a minima of 0.72 W m−1 K−1 at 509 K, and then slightly increases to 1.13 W m−1 K−1 at 850 K.

Figure 2 presents the total and lattice thermal conductivity as a function of temperature of Sn1–yInyTe0.75S0.15Se0.15 (y=0, 0.0025, 0.005, 0.01, and 0.015) samples. As shown, the total thermal conductivities all decrease with increasing temperature. With increasing In doping, the total thermal conductivity significantly decreases. For example, κtot decreases from 3.6 W m−1 K−1 for y=0 to 2.1 W m−1 K−1 for y=0.015 at room temperature, and decreases from 1.9 W m−1 K−1 for y=0 to 1.7 W m−1 K−1 for y=0.015 at 850 K.
As shown in Figure 2(b), the lattice thermal conductivity of In-doped Sn$_{1-y}$In$_y$Te$_{0.7}$S$_{0.15}$Se$_{0.15}$ increases from 0.99 W m$^{-1}$K$^{-1}$ for $y=0$ to 1.48 W m$^{-1}$K$^{-1}$ for $y=0.0025$ at room temperature, and decreases from 1.13 W m$^{-1}$K$^{-1}$ for $y=0$ to 1.01 W m$^{-1}$K$^{-1}$ for $y=0.01$ at 850 K. It is worth mentioning that the sample with $y=0.0025$ shows less temperature dependent, and $\kappa_{\text{lat}}$ increases at elevated temperature, which is probably due to the bipolar diffusion. Typically, $\kappa_{\text{lat}}$ of Sn$_{1-y}$In$_y$Te$_{0.7}$S$_{0.15}$Se$_{0.15}$ sample for $y=0.0025$ is 1.03 W m$^{-1}$K$^{-1}$ at 300 K, slightly decreases to a minima of 0.96 W m$^{-1}$K$^{-1}$ at 592 K, and then increases to 1.10 W m$^{-1}$K$^{-1}$ at 850 K.

To gain deep understanding of suppressed lattice thermal conductivity in multi-doped SnTe, the microstructure of the samples was observed by using transmission electron microscope (TEM). Fig. 3(a) exhibits a medium-magnification of the multi-doped SnTe matrix along [004] orientation. Small precipitates (red circles in Fig. 3(b)) are observed in the matrix, and only one set of Bragg diffraction spots is observed in the inset selected area diffraction (SAD) pattern of the corresponding area (bottom-right in Fig. 3(a)). The composition contrast can be evidenced by the energy-dispersive X-ray spectroscopy (EDS) (Fig. 3(b)) obtained from the precipitates that exhibit characteristic peaks for In, S and Se. High-resolution TEM (HRTEM) image in Fig. 3(c) shows the matrix SnTe (200) and the secondary phase SnS (021). Accordingly, the respective fast Fourier transformation (FFT) image (the top-right and bottom-right inset in Fig. 3(c)) shows no obvious peak splitting and reflects an endotaxial relationship between the matrix and secondary phase. In Fig. 3(d), the lattice distortion is more obvious to be seen in the inverse fast Fourier transformation (IFFT) image (the bottom-right inset).

To further analyze the possible strain around the precipitates and the connection between the lattice distortions, high-quality HRTEM images in Figure 3(e-f) were carried out by geometric phase analysis, which is a semi-quantitative lattice image-processing approach for revealing spatial distribution of relative elastic strain. As shown, the strains exist in the secondary phases and extensively concentrate around them. The strains mainly result from the lattice or orientation mismatch between the matrix and the secondary phase, and it is a pervasive defect effect associated with point defects and disloca-
tion. These high density of strains around the secondary phase plays a significant role on increasing the phonon scattering. In addition, the atomic-scale point defects and dislocation could scatter short-wavelength phonon efficiently. These multiscale phonon scattering mechanisms contribute to the low lattice thermal conductivity.

The temperature dependence of electrical conductivity for Sn$_{1-y}$In$_y$Te$_{0.7}$S$_{0.15}$Se$_{0.15}$ ($y=0, 0.0025, 0.005, 0.01, \text{and} 0.015$) samples are shown in Fig. 4(a). It may be seen that the electrical conductivity decreases with increasing temperature from 300 K to 850 K, showing a degenerate semiconductor behavior. At a particular temperature, the electrical conductivity monotonously decreases with increasing In content. For example, the electrical conductivity at room temperature significantly decreases from ~3480 S·cm$^{-1}$ for $y=0$ to ~910 S·cm$^{-1}$ for $y=0.015$, which can be attributed to the reduced carrier mobility of sample as summarized in Table 1.

In Fig. 4(b), the temperature dependent Seebeck coefficients of Sn$_{1-y}$In$_y$Te$_{0.7}$S$_{0.15}$Se$_{0.15}$ ($y=0, 0.0025, 0.005, 0.01, \text{and} 0.015$) samples are presented. It should be noted that the Seebeck coefficients of In doped samples are enhanced with respect to the SnTe$_{0.7}$S$_{0.15}$Se$_{0.15}$ over the entire temperature range. Especially, the Seebeck coefficient at room temperature increases from 7.6 μV·K$^{-1}$ for $y=0$ to 71 μV·K$^{-1}$ for $y=0.015$, and the improvement is almost an order of magnitude. With increasing temperature, the enhancement slows down, the Seebeck coefficient of Sn$_{0.7}$S$_{0.15}$Se$_{0.15}$In$_{0.015}$ at 850 K is 180 μV·K$^{-1}$ and slightly higher than 151 μV·K$^{-1}$ of SnTe$_{0.7}$S$_{0.15}$Se$_{0.15}$. Such a subdued enhancement of Seebeck coefficient at higher temperature is mainly due to the downshift of the Fermi level, where the introduced resonant state no longer work$^{34,51-52}$.

![Figure 4](image_url)

**Fig. 4** Temperature dependent thermoelectric properties: (a) electrical conductivity $\sigma$, (b) the Seebeck coefficients $S$, (c) the power factors $S^2\sigma$, and (d) $ZT$ values for Sn$_{1-y}$In$_y$Te$_{0.7}$S$_{0.15}$Se$_{0.15}$ ($y=0, 0.0025, 0.005, 0.01, \text{and} 0.015$) samples

| Samples | $\rho$ (g·cm$^{-3}$) | $N$ ($\times 10^{20}$, cm$^{-3}$) | $\mu$ (cm$^2$·V$^{-1}$·s$^{-1}$) | $\sigma$ (S·cm$^{-1}$) | $S$ (μV·K$^{-1}$) | $S^2\sigma$ (μW·cm$^{-1}$·K$^{-2}$) |
|---------|-----------------|-------------------------------|----------------|-----------------|----------------|---------------------|
| $y=0$   | 6.247           | 1.3                           | 164            | 3480            | 7.6            | 0.2                 |
| $y=0.0025$ | 6.209          | 1.4                           | 100            | 2300            | 34             | 2.7                 |
| $y=0.005$ | 6.161          | 1.6                           | 57             | 1510            | 50             | 3.7                 |
| $y=0.01$  | 6.161          | 2.0                           | 39             | 1240            | 63             | 4.9                 |
| $y=0.015$ | 6.195          | 2.2                           | 26             | 910             | 71             | 4.6                 |
The power factor as a function of temperature for SnTe$_{0.7}$S$_{0.15}$Se$_{0.15}$ for all In-doped samples is shown in Fig. 4(e). Because of the increased Seebeck coefficients, the power factors of In-doped samples are obviously enhanced as compared with that of SnTe$_{0.7}$S$_{0.15}$Se$_{0.15}$. If we focus on SnTe$_{0.7}$S$_{0.15}$Se$_{0.15}$:In$_{0.01}$, the power factor is seen to rise almost linearly from 4.9 μW·cm$^{-1}$·K$^{-2}$ at room temperature to 16 μW·cm$^{-1}$·K$^{-2}$ at 850 K. Such enhancement of power factor is achieved mainly resulted from the resonant level effect in In-doped SnTe$_{0.7}$S$_{0.15}$Se$_{0.15}$. In Fig. 4(d), the ZT values are presented as a function of temperature for all In-doped SnTe$_{0.7}$S$_{0.15}$Se$_{0.15}$ samples. The highest ZT value reaches 0.8 at 850 K for 1mol% In-doped SnTe$_{0.7}$S$_{0.15}$Se$_{0.15}$, which shows the largest power factor and the lowest lattice thermal conductivity.

3 Conclusions

This work demonstrates that the ZT value of SnTe can reach 0.8 at 850 K by well-designed multi-doping. The improved thermoelectric performance of SnTe$_{0.7}$S$_{0.15}$Se$_{0.15}$:In is accomplished by combination of reduced lattice thermal conductivity and enhanced power factor. The former mainly results from the secondary phase scattering by alloying Se, S, while the later mainly results from the resonant state by In doping. This work suggests that SnTe-based materials are important candidates for thermoelectric power generation and the well-designed multi-doping in SnTe is a promising approach to optimize the thermoelectric properties.

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多掺杂协同调控碲化锡热导率和功率因子提升热电性能

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摘要: 碲化锡(SnTe)是一种碲化铅的无铅替代物, 在热电领域有广阔的应用前景。但是, 纯相碲化锡样品具有较高的热导率与较低的塞贝克系数, 导致热电性能较差。本研究通过多重掺杂可以显著降低热导率, 提升赛贝克系数, 从而提升热电性能。SnTe 热压样品的晶格热导率随着 Se 和 S 的引入明显降低, 比如 SnTe0.99Se0.01S0.01, 室温下晶格热导率仅为 0.99 W·m⁻¹·K⁻¹。透射电子显微镜显示, SnTe 掺杂样品内存在大量的纳米沉淀相与晶格错位。在这一基础上, 掺杂 In 在价带顶引入共振态大幅提高了样品的塞贝克系数。实验表明通过多重掺杂可以有效提升碲化铅的热电性能, 其中样品 Sn0.99In0.01Te0.9S0.1S0.1在 850 K 时峰值 ZT 值达到 0.8, 这说明碲化铅的确是一种有应用前景的中温区热电材料。

关键词: 碲化锡; 热电性能; 共振态; 晶格热导率

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Supporting Information:

Multi-doping in SnTe: Improvement of Thermoelectric Performance due to Lower Thermal Conductivity and Enhanced Power Factor

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Fig. S1 Room temperature (a) powder XRD patterns, (b) lattice parameter of SnTe\textsubscript{1-2x}S\textsubscript{x}Se\textsubscript{x} (x=0, 0.05, 0.1, and 0.15) samples

Fig. S2 Temperature dependent (a) electrical conductivity and (b) Seebeck coefficient of SnTe\textsubscript{1-2x}S\textsubscript{x}Se\textsubscript{x} (x=0, 0.05, 0.1, and 0.15) samples

Fig. S3 Room temperature (a) powder XRD patterns, (b) lattice parameter of Sn\textsubscript{2}In\textsubscript{y}Te\textsubscript{0.95-y} (y=0, 0.005, 0.010, and 0.015) samples
Fig. S3  Room temperature (a) powder XRD patterns, (b) lattice parameter $a$, (c) Hall carrier density $N_p$, and (d) carrier mobility $\mu$ of Sn$_{1-y}$In$_y$Te$_{0.7}$S$_{0.15}$Se$_{0.15}$ ($y=0, 0.0025, 0.005, 0.01$, and $0.015$) samples.

Fig. S4  Temperature dependent heat diffusivity of Sn$_{1-y}$In$_y$Te$_{0.7}$S$_{0.15}$Se$_{0.15}$ ($y=0, 0.0025, 0.005, 0.01$, and $0.015$) samples.

Fig. S5  Room temperature Pisarenko plot for Sn$_{1-y}$In$_y$Te$_{0.7}$Se$_{0.15}$ ($y=0, 0.0025, 0.005, 0.01$, and $0.015$).
The solid curve is expected from Zhang$^{[18]}$. 