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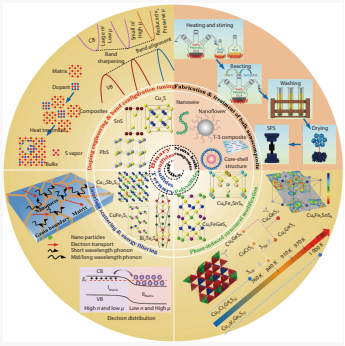
PERSPECTIVE

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Exploring electrical property improvement for thermoelectric sulfides

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Metal sulfides have been the subject of extensive research as a promising candidate in the study of thermoelectric energy transfer due to their eco-friendly abundance and similarities in chemical and structural properties with tellurides and selenides. Their intrinsically low thermal conductivity decouples the intercorrelated thermoelectric parameters that comprise the figure merit of *ZT*. This perspective aims to further advance thermoelectric sulfides application in structural designing, performance optimization and mechanism analysis, providing new insights to explore their electrical properties. The influence of chemical modification, heterogeneous phase mixture, and multi-physical technologies for matrix on electrical transport is used to demonstrate the performance improvement for thermoelectric sulfides. Lastly, key outcomes along with future thermoelectric conversion applications are outlined.



Thermoelectric (TE) technology, namely the direct conversion of heat to electricity and vice versa, can potentially solve environmental problems driven by the increasing global energy demand and the tremendous amount of heat wasted in industrial processes. This sparked interest over the past few decades due to their advantages over conventional energy conversion systems based on vapor-compression cycling, with no vibrations, gas-free emissions, and improved safety.[1] However, most commercial devices are limited by their high cost and low efficiency.[2] The conversion efficiency is defined as the dimensionless figure of merit $ZT = \alpha^2\sigma T/\kappa$, where α, σ, κ and T are Seebeck coefficient, electrical conductivity, thermal conductivity and absolute temperature, respectively. Enhancing ZT requires a trade-off between the inversely related electrical (α and σ) and thermal properties (κ). Modulating the band structure[3] can effectively enhance α and maintain a high σ by engineering the density-of-states (DOS) near the Fermi level. Doping with heavy elementals[4], designing nano-structures, or introducing hetero-phases[5] can enhance phonon scattering while providing a relatively high α.

Despite these advances in optimizing TE performance, they do not provide a comprehensive path to decoupling these intercorrelated TE parameters. Traditional approaches to lowering κ require heavy mass elements (e.g., Pb), which increase the carrier transport barrier (decreasing the carrier mobility μ). Many high-performance TE materials rely on the careful construction of a chemical profile that struggles to maintain structural homogeneity and high-performance stability in large-scale production. These interdependencies and compromises exist even at the fundamental material level, which presents a challenge in comprehensively understanding their physical and chemical properties. Therefore, there is an urgent need for earth-abundant and TE materials with easily regulatable properties that can be synthesized with a facile method.

Metal sulfides are comprised of earth-abundant elements and have generated great interest due to their low cost and unique chemistry. A progressive overview of the structural merits and TE performance-enhanced strategies in sulfur-based bulks and bottom-up composites is shown in Fig. 1. These compounds have a low Debye temperature (T_D) and a large atomic displacement parameter (ADP), due to their complex crystal structures with possible phase transformations, and intrinsically low κ.[6] It is thus essential to manipulate the carrier (electron e^- and hole h^+) transport of TE sulfides, and thereby develop a high power factor ($PF = \alpha^2\sigma$) by controlling the chemical and physical structures at the same spatial scale.

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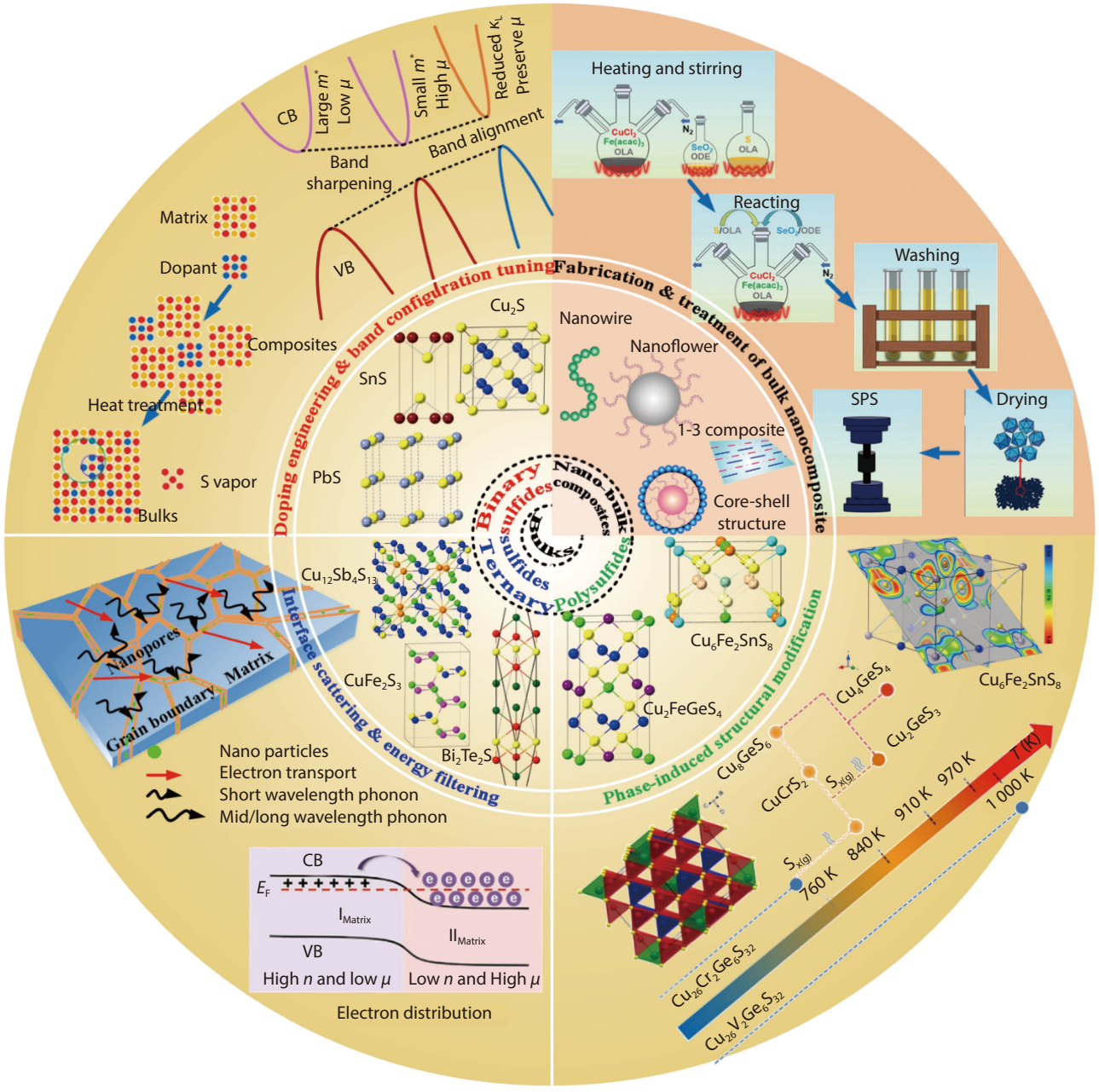


Fig. 1 Progressive overview of structural merits and performance-enhanced strategies in sulfur-based bulks and bottom-up composite.[7-12] Copyright 2019, American Chemical Society; Copyright 2019, Elsevier Ltd.; Copyright 2018, Elsevier Ltd.; Copyright 2017, The Authors; Copyright 2021, Wiley-VCH GmbH; Copyright 2019, American Chemical Society.

The peak *PF* in p- and n-type TE sulfides are shown in Fig. 2a, as previously reported in the literature. These are two strategies for improving the *PF*, which are further discussed herein. (1) adjusting the independent crystal structure and/or microstructure design. (2) tuning the composition, e.g., chemical modification, heterogeneous phase mixture, and multiphysical modulation, in relaton to the carrier transport property.

Optimization of the carrier concentration (n_{e^-} and n_{h+}) is usually the first step in maximizing *ZT* for the given TE materials. Electrical transport in TE sulfides is coupled such that a low *n* leads to a high α and low σ, making the optimization of *n* necessary to achieve a high *PF*. Except for materials, such as the mineral sulfides of tetrahedrites[52], colusites[53], and pavonites[54], tuning the composition is straightforward in most systems. Introducing foreign elements can generate defects in matrixes, such as point defects and interstitial, which usually increases n_{h+} at canion positions or n_{e^-} at anion positions (heterovalent doping). The biggest challenge is modifying the intrinsic properties, which are governed by the crystal itself, i.e., the band structure and carrier effective mass (m^*). For example, He et al.[22] reported the temperature-dependent evolution and interplay of three separate electronic bands in $SnS_{1-x}Se_x$ crystals, involving two-band convergence, diver-

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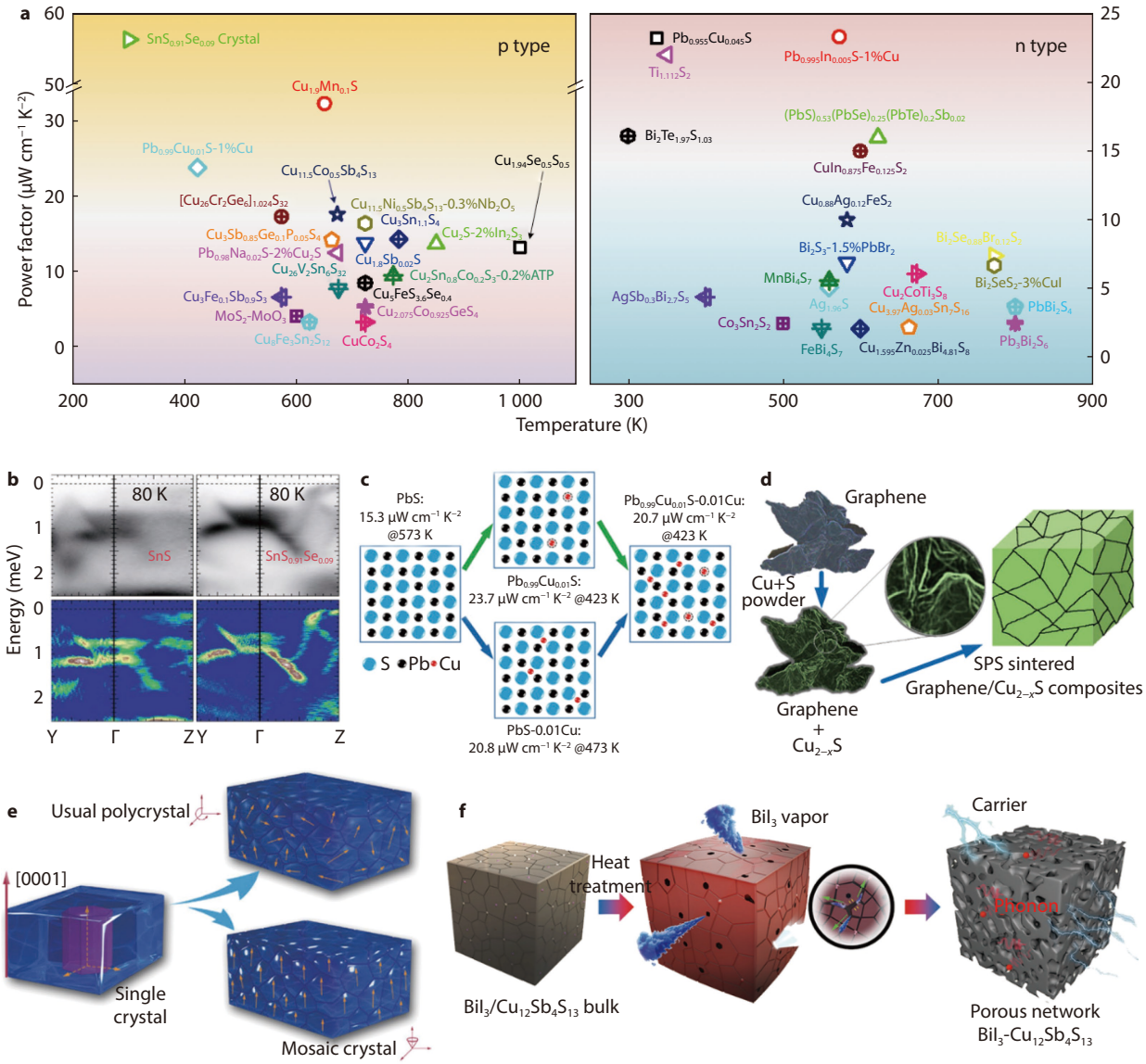


Fig. 2 **a** Peak *PF* values in sulfur-based TE materials. There are two breakdowns: p-type and n-type binary copper[13-16], lead[17-21], tin[22], silver[23], molybdenum[24], bismuth[25] and titanium[26] sulfides, ternary Cu-[9, 11, 27-36], Co-[37] and Bi-based[38-43] sulfides, and polysulfides.[44-48] TE properties have been unfolded, and the current maximum value is 56.3 μW cm^{-1} K^{-2} at 300 K in p-type $SnS_{0.91}Se_{0.09}$ crystal.[22] **b** Electronic band structures for $SnS_{1-x}Se_x$ (x = 0, 0.09).[22] Copyright 2019, The Authors, AAAS. **c** Roadmaps toward high *PF* in p-type PbS compounds via compositing Cu.[19] Copyright 2021, Wiley-VCH GmbH. **d** Schematic illustration of the process for fabricating graphene/$Cu_{2-x}S$ composites.[49] Copyright 2018, Elsevier Ltd. **e** Structural characteristics among a single crystal, usual polycrystal and mosaic crystal.[50] Copyright 2015, Wiley-VCH Verlag GmbH & Co. kGaA, Weinheim. **f** The progress of porous network-structured $Cu_{12}Sb_4S_{13}$.[51] Copyright 2021, Wiley-VCH GmbH.

gence, and crossing (Fig. 2b). These outstanding behaviors were promoted through the substitution of S with Se, allowing for the successful optimization of m^* and μ. Sn, Mn and Sn/Mn doping[15] can directly and effectively enhance the *PF* by increasing n_{h+} in p-type $Cu_{2-x}S$. The presence of Cu ions[18] allowed for σ to be improved by increasing n_{e-} while simultaneously maintaining a large μ, which translates to a peak *PF* of ~23 μW cm^{-1} K^{-2} in n-type PbS alloys. As another approach, resonant doping, having similar electronic configurations as the host atoms, factors the dopant energy level alignment with the host energy level to form two extended states These states then resonate to form more energy states resulting in a sharp increase in the density of states (DOS). The presence of resonant levels enhances α while only slightly reducing σ to improve the overall *PF*.

Based on the matrix crystal structure, the mixture or composite of TE sulfides with homogeneous metal chalcogenides or heterogeneous phases creates a unique composite structure that generates an energy filtering effect that increases the *PF*. To obtain high-performance p-type PbS, Cu substitutions and interstitials[19] effectively improve μ by lowering m^* and n_{e-} at low temperatures (300-450 K) and further optimizing temperature-dependent n_{e-} at high temperatures (450-823 K). Reducing both m^* and n_{e-} shifted the peak *PF* to the low temperature range (Fig. 2c), leading to an ultrahigh *PF* of ~23 μW cm^{-1} K^{-2} at 423 K. For n-type PbS, the cooperative ef-

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fects[17] of the In resonance level and Cu dynamic doping also improved electronic transport and led to the same high *PF*. The introduction of a certain concentration of 3D graphene into the $Cu_{2-x}S$ matrix provided a high, reproducible *PF* (Fig. 2d). Furthermore, the microstructural characterizations revealed that the foreign atomic and nanoscale defects can both create strain field and mass fluctuations that effectively intensify the phonon scattering effect and reduces κ.

Microstructural investigations revealed that alloying homomorphic crystallites into the TE matrix triggeed a unique reaction chemistry that created nanoscale sublattices, mesoscale derivatives, and complex structures with atomic-scale defects and micro-scale boundaries. As a result, electrical transport was strongly enhanced, which led to a high *PF*. Using the concept of nanoscale mosaicity, Cu_2S-Cu_2Te bulk nanomaterials[50] were well constructed and achieved a high *PF* of 8 μW cm^{-1} K^{-2} at 1000 K. Unlike the random orientations in ordinary polycrystals or nanocrystals, the mosaic crystal exhibited a nearly identical orientation, causing the bulk to appears like a single crystal with low angle boundaries from a macroscale point of view (Fig. 2e). Electrons are freely transferred along with the frame of quasi-single crystals, resulting in exceptional electronic transport. Similarly, a porous, network structured tetrahedrite material[51] was formed using a unique BiI_3 sublimation technique, accompanied by changes in the hierarchical structures (Fig. 2f). The addition of additives (0.7 vol%) resulted in a ~84% increase in the *PF* at room temperature due to the suddenly enhanced μ.

Metal sulfides continue to surprise us with their unique chemical and structural properties and interesting carrier and phonon transport mechanisms. Numerous advancements have been made with both p- and n-type sulfur-based materials, making them robust candidates for application in the mid to high temperature ranges. However, further study of sulfides in high-impact TE materials and commercialization is necessary.

(1) Sulfur-based TE technology is inhibited by its limited stability under specific operating conditions The electrical power produced by TE devices must be constant over time for practical applications. Instances of instability in these devices include the complete decomposition of the crystal, slight deterioration of the physical properties, and changes to the carrier and/or phonon transport mechanism. Besides thermal cycling and electric field-induced failure, TE devices also encounter mechanical challenges such as compression and shear stress. To handle such conditions, the material needs to possess high mechanical stability.

(2) Designing hierarchical nanostructures is beneficial to achieving a low κ and high *ZT* but directly reduces the *PF* due to the low μ. By tailoring special nano/micro-structures to improve the *PF* in bulk nanocomposites, the manipulation of electron and phonon transport, i.e., controlling grain size, optimizing chemical compositions, and tuning the crystal distortion energy and system entropy, can be greatly improved.

(3) Binary TE sulfides have attracted considerable attention in recent years. Phonon-liquid electron-crystal (PLEC) materials, have a high potential for commercialization, while polysulfide is less competitive in both performance and potential application. They are usually derived from a distorted symmetric crystal structure with a complex cation-S framework (e.g., $[MoS_4]Cu_6$; multiple atomic interactions, ion-covalent and metal-like), exhibiting a relatively low *PF* of < 17 μW cm^{-1} K^{-2}. Therefore, manipulating the interactions between interstitial ions and the conductive intrinsic network can effectively tune electronic transport and achieve a high *PF*, paving the way for high-*ZT* polysulfide TE materials.

(4) Regardless of the low *PF*, most polysulfide materials exhibit hole conductive behaviors (p-type semiconductors). The corresponding peak *PF*s are often located at 650-800 K, which is higher than that of n-type materials. It is necessary to shift the peak *PF* of p-type materials to lower temperatures (< 700 K) to match n-type materials, improving the electrical performance of n-type parts, *PF* > 10 μW cm^{-1} K^{-2}, is another option.

(5) Considering the toxicity of Pb, although PbS-based materials present both high TE performance and good chemical stability, it is vital to prevent human health risk and environmental pollution during the fabrication processes.

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■ CONFLICT OF INTEREST

The authors declare no conflict of interest.

■ AUTHOR CONTRIBUTIONS

F.-H. Sun and H.Z. Li cowrote the manuscript. J. Tan painted the figures. J. Pei revised the manuscript. X. Wang formatted the manuscript. All authors discussed and commented on the manuscript.

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