

A New Era in Sustainability: Te- and Pb-Free Thermoelectrics

Kivanc Saglik ^{1,2} , Yannan Lu ³ and Xizu Wang ^{2,*} 

¹ School of Materials Science and Engineering, Nanyang Technological University, 50 Nanyang Ave, Block N4.1, Singapore 639798, Singapore; kivanc001@e.ntu.edu.sg

² Institute of Materials Research and Engineering (IMRE), Agency for Science, Technology, and Research (A*STAR), 2 Fusionopolis Way, Singapore 138634, Singapore

³ Manufacturing Research Unit, CSIRO, Research Way, Clayton, VIC 0810, Australia; yannan.lu@csiro.au

* Correspondence: wangxz@a-star.edu.sg

Abstract

Nearly two-thirds of global energy is lost as waste heat. Thermoelectric materials enable the direct conversion of heat into electricity, offering a promising recovery strategy. However, many high-performance systems rely on toxic or scarce elements such as Pb and Te, limiting sustainability. This review compares transport mechanisms in bulk and thin-film thermoelectrics, highlights environmentally benign material alternatives and fabrication routes, and discusses key challenges and future opportunities for sustainable energy applications. To provide a comprehensive overview of tellurium- and Pb-free bulk thermoelectrics, recent developments are summarized in a comprehensive table detailing material composition, peak zT values, temperature differences (ΔK), efficiencies (η), power output (P_{out}) (or maximum power density (P_{max})), and device geometries.

Keywords: tellurium-free thermoelectrics; magnesium antimonide; bulk vs. thin-film thermoelectrics; energy harvesting; waste heat recovery

1. Introduction

Waste heat is an abundant yet untapped energy source, as nearly two-thirds of the energy produced globally is released into the surroundings through industrial and domestic processes [1,2]. Thermoelectric generators (TEGs) offer a viable route to reclaim this dissipated energy by converting thermal gradients directly into electricity. Because TEGs are solid-state devices without moving parts, they provide maintenance-free, silent and reliable power generation across a wide temperature range [3,4]. Consequently, thermoelectric systems have found use in diverse applications including spacecrafts [3], wearable electronics [5,6], automotive systems [7], and the Internet of Things (IoT) [8].

The power conversion efficiency (η) of a thermoelectric generator depends on the leg shape and dimension [9], temperature difference between its hot (T_h) and cold ends (T_c), and the average dimensionless figure of merit (zT) of the materials employed [3]. The figure of merit (zT) is defined as $zT = (S^2\sigma)/\kappa T$, where S , σ , κ , and T represent the Seebeck coefficient, electrical conductivity, thermal conductivity, and absolute temperature, respectively. High zT values are achieved through a combination of a large Seebeck coefficient, high electrical conductivity, and low thermal conductivity, which is intrinsically difficult due to the coupling between electrical and thermal transport [10]. Various strategies such as band engineering [11], nanostructuring [12], and, more recently, high-entropy design have been employed to decouple transport properties and enhance performance. Notably, high-entropy systems such as AgMnGePbSbTe_5 and $\text{AgSnSbSe}_{1.5}\text{Te}_{1.5}$ have demonstrated



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promising thermoelectric properties by exploiting configurational disorder to reduce thermal conductivity [13–15].

Many state-of-the-art thermoelectric materials incorporate heavy metals such as Pb and Te, whose large atomic masses induce mass fluctuations that reduce lattice thermal conductivity and enhance the thermoelectric figure of merit (zT). Similarly, introducing heavy elements successfully reduces the thermal conductivity of various thermoelectric materials through the same mechanism. Bi_2Te_3 - and PbTe-based systems offer a compelling combination of high band degeneracy and intrinsically low thermal conductivity, making them one of the best and most studied thermoelectric systems with commercial applications [16]. However, Bi_2Te_3 - and Pb-based thermoelectric materials, or other systems doped by Pb or Te, raise concerns regarding toxicity, environmental impact, and long-term resource security despite their record efficiencies [17–19]. Tellurium is a critically scarce element (less than 0.001 ppm in Earth's crust), while lead is a known neurotoxin, limiting the scalability and sustainability of such systems [8,17,20,21]. These issues underscore the growing need for thermoelectric materials composed of Earth-abundant, low-toxicity elements that enable large-scale deployment. Unlike scarce and geographically bottlenecked tellurium (Te), elements like silicon (Si), magnesium (Mg), and iron (Fe) feature crustal abundances several orders of magnitude higher, fundamentally mitigating geopolitical supply chain risks. Consequently, shifting to these Earth-abundant matrices removes them from critical raw material risk registers, ensuring long-term resource availability for terawatt-scale energy harvesting. Regarding environmental impact and toxicity, the toxicological profile of lead- and tellurium-free alternatives eliminates the risk of heavy-metal leaching during raw material extraction and operational service. This transition significantly lowers potential ecotoxicity and human toxicity scores, clearing regulatory hurdles that restrict hazardous substances in consumer electronics. From a lifecycle perspective, tellurium-free bulk alloys such as silicides exhibit high mechanical and chemical stability, which minimizes operational degradation and extends device lifespans. Furthermore, their non-hazardous compositions allow for simplified end-of-life processing, bypassing the complex, energy-intensive isolation protocols required to reclaim toxic chalcogenides. Table 1 provides a summary of the toxicity, abundance and scalability of the most common thermoelectric material families. The scalabilities of these systems are evaluated based on toxicity and abundance.

As Te-free alternatives for sustainable thermoelectrics, Mg_3Sb_2 -based systems offer significant promise at room temperature due to their highly degenerate band structure and intrinsically low thermal conductivity [22]. The subsequent sections provide a comprehensive discussion on the utilization of Mg_3Sb_2 and related materials in bulk and thin-film thermoelectric devices. The performance divergence among reported Mg_3Sb_2 -based systems stems from a delicate competition between carrier concentration optimization and grain boundary engineering. Specifically, peak thermoelectric performance is achieved only when excess Mg compensation suppresses intrinsic killer vacancies, while targeted interstitial doping (e.g., with Y or Nb) activates multi-valley band convergence to maximize electrical transport without accelerating lattice thermal conductivity.

Electronic waste can also be directly reused to manufacture thermoelectric materials, merging waste management with energy conversion. For example, Ge- and P-doped silicon recovered from old photovoltaic cells shows a zT of 0.45 at 873 K [23]. This brings important advantages in terms of sustainability because Si is widely available in Earth's crust, it is lightweight, and recycling it from e-waste keeps it out of landfills and reduces environmental electronic waste [24]. Furthermore, tellurium in the used thermoelectric modules could be recycled back into new thermoelectric materials [25]. Current high-performance materials like Bi_2Te_3 and PbTe use large amounts of tellurium. Reclaiming tellurium

from spent devices prevents toxic waste from entering the environment and secures a secondary source for these critical elements, making the entire lifecycle of thermoelectric technology sustainable.

Table 1. A systematic comparison of the thermoelectric figure of merit, operating temperature (K), toxicity, and elemental abundance of thermoelectric materials, used to evaluate the scalability of these systems.

S/N	Material	Peak zT	Operating T (K)	Toxicity	Abundance (ppm)	Scalability	Ref.
1	SnSe	3.1	783	moderate	Sb: 0.2 Se: 0.05	medium	[26]
2	AgSb _{1-x} Yb _x Te ₂	2.4	573	high	Ag: 0.075 Sb: 0.2 Te: 0.001	low	[27]
3	Cu _{0.002} Pb _{0.99} Ge _{0.01} Te	1.4	673	high	Pb: 14 Te: 0.001	low	[28]
4	Ge _{0.89} Bi _{0.05} Sb _{0.04} Te _{0.96} + CuI	1.9	773	high	Ge: 1.5 Te: 0.001	low	[29]
5	Bi _{0.5} Sb _{1.5} Te ₃	1.6	375	high	Bi: 0.025 Sb: 0.2 Te: 0.001	low	[30]
6	Ce _{0.4} Yb _{3.6} Bi _{0.2} Sb _{2.8}	0.4	1273	moderate	Yb: 3.3 Sb: 0.2	medium	[31]
7	Mg _{2.13} In _{0.02} (Si _{0.3} Sn _{0.7}) _{0.98} Sb _{0.02}	1.7	700	low	Mg: 23,000 Si: 272,000 Sn: 2.3	high	[32]
8	Cu ₂ SnS ₃	0.4	650	moderate	Cu: 50 Sn: 2.3 S: 350	medium	[33]
9	MgAg _{0.97} Sb _{0.99}	1.0	550	low	Mg: 23,000 Ag: 0.075 Sb: 0.2	high	[34]
10	Mg ₃ Sb ₂	2.0	425	low	Mg: 23,000 Sb: 0.2	high	[35]

Thin-film thermoelectrics present a compelling opportunity in this regard. Compared with bulk materials, thin films consume substantially less material while offering unique advantages derived from their reduced dimensionality. Such films can be engineered for transparency, flexibility, and tunable thermal or electrical transport, expanding their potential integration into optoelectronic [36] or ferroelectric [37,38] devices alongside traditional thermoelectric applications [38].

This perspective explores recent advances in sustainable thermoelectric materials, emphasizing bulk and thin-film systems and their transport phenomena. It compares the performance and mechanisms of bulk and thin-film thermoelectrics, reviews major developments from the past five years, and discusses the challenges and opportunities for realizing high-performance, environmentally benign thermoelectric technologies.

2. Transport Phenomena in Bulk vs. Thin-Film Thermoelectrics

The nanoscale thickness (<1 µm) of thin films fundamentally alters their electronic band structure compared to bulk counterparts. Band structure calculations (Figure 1) reveal a sharp increase in the density of states (DOS) near the band edge in thin films (Figure 1b)

compared to bulk thermoelectrics (Figure 1a). This band structure modification due to quantum confinement effects enhances the power factor ($S^2\sigma$) of thin films [39].

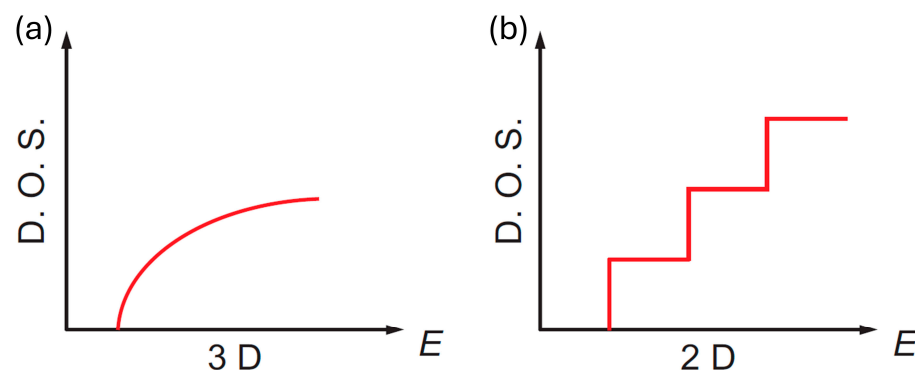


Figure 1. Comparison of electronic band structures of bulk and thin-film thermoelectric materials (Adapted with permission from ref. [40]). (a) Bulk Semiconductor. (b) Quantum Well.

Thin-film thermoelectrics enable integration onto flexible substrates for wearable sensors and energy harvesting from irregular surfaces. However, thermal expansion mismatch between the thermoelectric film and substrate often induces cracking [41], limiting high-temperature applications. In contrast, bulk thermoelectrics excel in diverse scenarios: Bi_2Te_3 -based materials dominate low-temperature Peltier cooling ($<300\text{ }^\circ\text{C}$), while PbTe -based systems are preferred for high-temperature power generation ($>500\text{ }^\circ\text{C}$), such as spacecraft applications. Regarding material toxicity concerns, Bi_2Te_3 and PbTe thin films offer greater sustainability than bulk counterparts due to dramatically reduced material consumption [12].

Measurement challenges further distinguish thin-film characterization. Conventional ZEM-3 systems designed for electrical conductivity and Seebeck coefficient testing of bulk samples require specialized adaptation for thin films. Standard thermal diffusivity measurement for bulk materials via laser flash analysis is unsuitable for thin films as well. Alternative techniques like 3ω or time-domain thermoreflectance (TDTR) [42] provide reasonable accuracy but fail at elevated temperatures ($>400\text{ }^\circ\text{C}$). Consequently, thin-film studies typically report power factors or device-level zT values rather than intrinsic material properties [12].

Similarly to the electronic structure, the phonon spectrum also differs markedly between bulk and thin-film thermoelectrics. In bulk materials, grain boundaries, dislocations, and point defects are key scattering centers that effectively suppress phonon transport. When the characteristic dimensions are reduced to the nanometer scale, as in thin films, additional interface scattering at the film–film and film–substrate interfaces further reduces lattice thermal conductivity [39]. Importantly, many strategies developed to lower lattice thermal conductivity in bulk systems, such as alloying, nanostructuring, and defect engineering, can be translated to thin-film architecture as well.

Both bulk (Figure 2a) and thin-film thermoelectric generators (Figure 2b) connect n- and p-type legs electrically in series and thermally in parallel. However, thin-film devices adopt planar architectures where heat flows in-plane to establish the temperature gradient, contrasting the out-of-plane configuration typical of bulk π -shaped TEGs [43].

As a summary, thin films and bulk thermoelectrics have different application areas and various advantages. The advances made in both fields will be elaborated in the next sections.

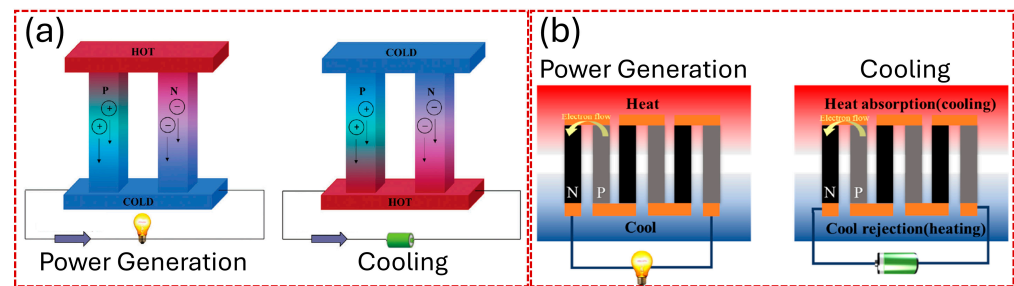


Figure 2. (a) Schematic illustration of bulk thermoelectric generator (left) and bulk thermoelectric cooler (right). (Adapted with permission from ref. [44], under CC BY 4.0 license.) (b) Architecture of thin-film thermoelectric generator (Adapted with permission from ref. [45] under CC BY 4.0 license).

3. Bulk Thermoelectric Materials and Device Optimization

Bulk thermoelectric materials are synthesized via established methods including solid-state reactions, melting, and mechanical alloying (Figure 3) [46]. These materials enable efficient power generation at high temperatures and Peltier cooling at low temperatures.

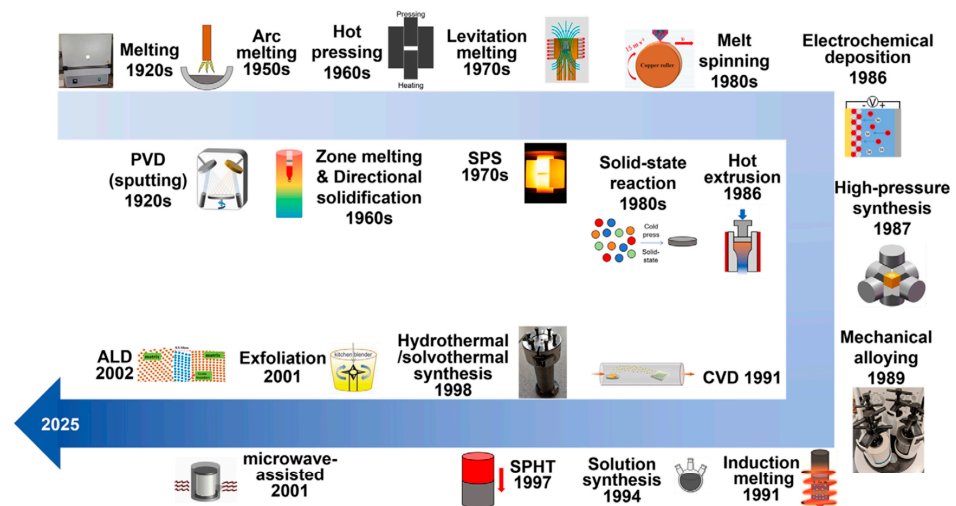


Figure 3. The evolution of synthesis methodologies for thermoelectric materials and films (Adapted with permission from ref. [46] under CC BY 4.0 license).

Thermoelectric generators (TEGs) drive carriers via temperature gradients, allowing for single-leg operation for power generation. However, cooling applications require closed π -shaped circuits with complementary n- and p-type legs driven by applied voltage, emphasizing the critical need for high-performance n-type materials.

Mg-based thermoelectrics offer compelling advantages over Bi_2Te_3 modules, including suppressed bipolar conduction, lower toxicity, and reduced cost [2]. P-type MgAgSb delivers a peak zT of 1.5 at 525 K, driven by its complex crystal structure that scatters phonons effectively and multi-valley electronic structure that boosts the power factor, complemented by exceptional mechanical durability. Its n-type counterpart, Mg_3Sb_2 ($N_v = 6$) [22], reaches $zT > 1$ at 573 K [8], which is ideal for recovering the 50% of waste heat available below this temperature [47]. A single-leg $\text{Mg}_{3.3}(\text{Sb}_{0.3}\text{Bi}_{0.7})_{1.9945}\text{Te}_{0.0055}$ module demonstrated 6.5% conversion efficiency and 64 mW output at $\Delta T = 246$ K [48].

Beyond zT , the filling factor (FF), which is the ratio of the leg cross-sectional area to the substrate area, critically influences device efficiency. The FF can be increased by either (i) increasing the cross-sectional area of legs or (ii) increasing the number of legs by keeping the area of the substrate constant. An optimal FF of 0.64 in $\text{MgAg}_{0.97}\text{Sb}_{0.99}/\text{Mg}_3(\text{Sb},\text{Bi})_2$

couples yielded 8.2% efficiency at $\Delta T = 260$ K, with negligible resistance drops after 10,000 cycles, underscoring geometrical optimization [47].

Despite decent performance, oxidation sensitivity limits Mg-based materials' longevity. The remarkable capacity of the atomic layer deposition (ALD) HfO_2 coating to preserve 93% power output over 65,000 cycles stems from its ultra-dense, pinhole-free morphology, which acts as a robust hermetic barrier. This nanoscale encapsulation effectively suppresses the volatile sublimation of magnesium (Mg) and blocks atmospheric oxidation, thereby preventing compositional drift and structural degradation during prolonged thermal cycling [21]. Similarly, obtaining phase-pure MgAgSb is challenging because of the high vapor pressure of Mg, and its thermoelectric performance is extremely sensitive to slight compositional deviations. As a result, many studies deliberately prepare Ag- and Sb-deficient (or Mg-rich) MgAgSb compositions. To overcome this issue, a two-step ball-milling approach has been introduced: Ag and Mg are milled first, followed by the addition of Sb. This approach modifies the carrier scattering mechanism and enhances weighted mobility. A seven-leg module based on n-type $\text{Mg}_{3.285}\text{Nb}_{0.015}\text{SbBi}_{0.9965}\text{Te}_{0.0035}$ and p-type $\alpha\text{-MgAgSb}$ achieved a power generation efficiency (η) of 12% and a cooling ΔT of 61 K at 300 K [2].

$\text{Mg}_3(\text{Bi,Sb})_2$ has emerged as a sustainable alternative to Bi_2Te_3 -based thermoelectrics, yet its performance is often constrained by electron scattering at grain boundaries. To mitigate this limitation, Chen et al. introduced a tailored heating–annealing approach (Figure 4a) that promotes controlled nucleation followed by extensive grain growth. The resulting ingot exhibited grain sizes of approximately 800 μm to 1 mm, corresponding to nearly two-thirds of the typical transport measurement sample length. The specialized crucible and the coarse-grained ingot are represented in Figure 4b. Such coarse grains significantly enhanced electrical conductivity while preserving a Seebeck coefficient comparable to that of polycrystalline counterparts. Importantly, thermal conductivity remained largely unaffected, reflecting the distinct mean free paths of electrons and phonons. Consequently, coarse-grained $\text{Mg}_3\text{Bi}_{1.25}\text{Sb}_{0.75}$ composition achieved higher Z values than commercial n-type $\text{Bi}_2\text{Te}_{2.7}\text{Se}_{0.3}$, particularly within 375–600 K. A module consisting of n-type $\text{Mg}_{3.15}\text{Sb}_{1.4975}$ and p-type $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ delivered a power conversion efficiency (η) of 5.8% and a cooling ΔT of 68 K at 300 K [22]. These findings underscore the critical role of phase-diagram-guided grain engineering in advancing sustainable thermoelectric materials.

Half-Heuslers combine mechanical strength, thermal stability, and relatively inexpensive constituent elements, making them attractive candidates for thermoelectric power generation [49,50]. Their properties can be tuned to both p- and n-type materials through doping and compositional control, minimizing thermal expansion mismatch between module legs. Their mechanical strength also allows for leg lengths as short as 2 mm, improving power density. For example, an n-type $\text{Hf}_{0.6}\text{Zr}_{0.4}\text{NiSn}_{0.98}\text{Sb}_{0.02}$ and p-type $\text{Hf}_{0.2}\text{Zr}_{0.75}\text{Ti}_{0.05}\text{CoSb}_{0.8}\text{Sn}_{0.2}$ couple achieved a power density of 7.5 W cm^{-2} at $\Delta T = 575$ K. Oxidation studies indicated stable operation below 575 K due to negligible oxidation [51].

Given the urgent demand for efficient n-type thermoelectric materials, n-type Zr-NiSn Half-Heusler alloys have emerged as strong candidates. Systematic doping studies show that Cu incorporation enhances weighted mobility, electrical conductivity, and consequently the power factor. Notably, the effective mass remains nearly unchanged, allowing the Seebeck coefficient to be largely preserved despite the increase in carrier concentration (n). In addition, Cu atoms introduce extra phonon scattering centers, which suppress lattice thermal conductivity (κ_L) and promote the partial decoupling of electrical and thermal transport. As a result of this synergistic optimization, the composition $\text{ZrNiCu}_{0.02}\text{Sn}$ achieves a maximum zT of 0.81 at 775 K. Beyond transport performance, Cu doping also improves mechanical robustness, as evidenced by increased Vickers hardness and fracture tough-

ness [50]. This combination of enhanced thermoelectric and mechanical properties makes the material particularly attractive for practical thermoelectric generator applications.

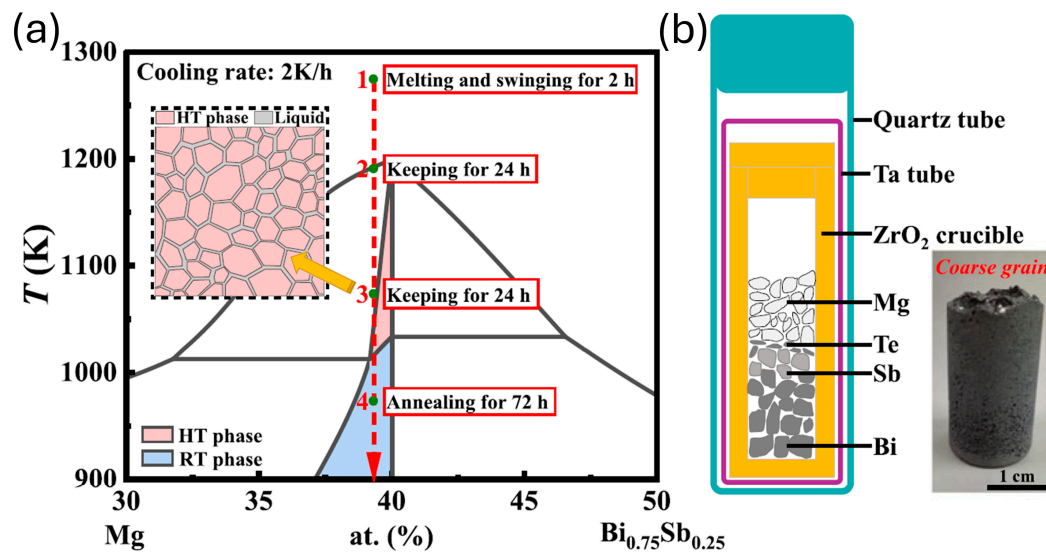


Figure 4. (a) Overview of fabrication process and grain coarsening mechanism of coarse-grained $\text{Mg}_3\text{Bi}_{2-x}\text{Sb}_x$ ($x = 0.5, 0.75$) bulk samples, accompanied by schematic pseudobinary Mg - $\text{Bi}_{0.75}\text{Sb}_{0.25}$ phase diagram. (b) Schematic depiction of crucible configuration used during synthesis process (Adapted with permission from reference [22], under CC BY 4.0 license).

Table 2 summarizes the performance metrics of tellurium-free bulk thermoelectric devices. Reported parameters include the composition and peak zT of the n- and p-type legs, alongside key operational characteristics such as temperature difference (ΔT , K), conversion efficiency (η , %), maximum power density (P_{max} , Wcm^{-2}) or output power (P_{out} , W), leg dimensions, total number of legs, and corresponding references.

Table 2. A summary table for bulk thermoelectric devices: their composition, corresponding peak zT values, applied temperature difference (ΔT , K), efficiency (η , %), maximum power density (P_{max} , Wcm^{-2}) (or output power (P_{out} , W)), number of legs, leg dimensions and reference numbers.

Composition (n/p)	Peak zT	ΔT (K)	η (%)	$P_{\text{max}}/P_{\text{out}}$	# Legs	Dimensions (mm^3)	Ref.
Mg_3Sb_2	2.0	425	12.6	-	1	$3.8 \times 3.8 \times 6.0$	[35]
$\text{Mg}_3\text{Sb}_2/\text{MgAgSb}$	-	276	7.1	-	2	$3.8 \times 3.8 \times 6.0$	[35]
$\text{Mg}_{3.2}\text{Ag}_{0.01}\text{Sb}_{1.5}\text{Bi}_{0.49}\text{Te}_{0.01}$	1.64	400	11	-	1	-	[52]
$\text{Mg}_{3.2}\text{Ag}_{0.01}\text{Sb}_{1.5}\text{Bi}_{0.49}\text{Te}_{0.01}/\text{Mg}_{0.99}\text{Cu}_{0.01}\text{Ag}_{0.97}\text{Sb}_{0.99}$	-	277	7.9	-	2	$3 \times 3 \times 5.1$	[52]
$\text{Mg}_{3.2}\text{Sb}_{1.5}\text{Bi}_{0.49}\text{Te}_{0.01}\text{Cu}_{0.01}/\text{MgAgSb}$	1.9/-	420	7.3	0.51 Wcm^{-2}	2	-	[53]
$\text{Mg}_{1.98}\text{Ag}_{0.02}\text{ZnSb}_2/\text{Mg}_{3.2}\text{SbBi}_{0.996}\text{Se}_{0.004}$	0.45/-	380	7.5	0.23 Wcm^{-2}	2	$3 \times 3 \times 5.7$	[54]
$\text{Mg}_3(\text{Sb,Bi})_2/\text{Ce}_{0.9}\text{Fe}_3\text{CoSb}_{12}$	-	440	11	0.4 W	2	$3 \times 3 \times 6/2 \times 2 \times 6$	[55]
$\text{Mg}_{3.16}\text{Mo}_{0.04}\text{Sb}_{1.3}\text{Bi}_{0.49}\text{Te}_{0.01}$	1.84	420	12	1.46 Wcm^{-2}	1	$4 \times 4 \times 6$	[56]
$\text{Mg}_{3.2}\text{SbBi}_{0.99}\text{Te}_{0.01}/\text{Mg}_{1.594}\text{Yb}_{0.2}\text{Na}_{0.006}\text{Zn}_{1.2}\text{Sb}_2$	-/0.64	276	5.5	-	2	$2 \times 2 \times 4.6/2.8 \times 1.8 \times 4.6$	[57]
$\text{MgAg}_{0.97}\text{Sb}_{0.99}$	1.0	250	6.5	-	1	$2 \times 2 \times 6.5$	[34]
$\text{Mg}_{3.3}\text{Bi}_{1.498}\text{Sb}_{0.5}\text{Te}_{0.002}/\text{MgAg}_{0.97}\text{Sb}_{0.99}$	0.9/1.0	250	7.0	-	2	$2 \times 2 \times 6.5$	[34]
$\text{CuFeSe}_2/\text{Cu}_3\text{SbS}_4$	0.35/0.32	433	4.6	$3 \mu\text{Wcm}^{-2}$	14	1: 7 mm, 2r: 9 mm	[58]

Table 2. Cont.

Composition (n/p)	Peak zT	ΔT (K)	η (%)	P_{max}/P_{out}	# Legs	Dimensions (mm ³)	Ref.
Mg _{2.06} Si _{0.3} Sn _{0.665} Bi _{0.035} / Mg _{1.98} Li _{0.03} Si _{0.3} Sn _{0.7}	1.3/ 0.3	375	3.6	0.9 Wcm ^{−2}	4	3.4 × 3.04 × 4.05/ 4.5 × 4.5 × 3.96	[59]
Mg _{2.06} Si _{0.3} Sn _{0.665} Bi _{0.035} / α -MgAg _{0.97} Sb _{0.995}	1.0/ 1.2	275	6.4	0.8 W	4	0.3 × 0.3 × 0.3/ 0.3 × 0.6 × 0.4	[60]
Mg _{2.13} In _{0.02} (Si _{0.3} Sn _{0.7}) _{0.98} Sb _{0.02} / MgAg _{0.97} Sb _{0.99}	1.7/-	300	7.3	0.35 W	4	2.4 × 2.4 × ()/4 × 4 × ()	[32]

Thermal expansion studies further show that inserting an Ag layer between Cu electrodes and thermoelectric legs reduces cracking. Using this approach, the internal resistance of an Mg₂Si_{0.6}Sn_{0.4}/MnSi_{1.77} couple decreased, yielding a power density of 0.95 W cm^{−2} at $\Delta T = 400$ K [61]. Non-cuboid thermoelectric legs have been shown to enhance waste heat harvesting, and additive manufacturing enables complex geometries [9]. Thimont et al. fabricated layered Ge-doped Mn₂Si_y (1.73 < y < 1.77) structures with different orientations. Samples with layers perpendicular to the pressing direction showed higher density, highlighting binder effects. The power factor was orientation-dependent, indicating that this method can tailor anisotropy to optimize thermoelectric performance [62].

Silicides are promising mid-temperature materials because they are cheap, scalable, and mechanically robust. However, synthesizing Mg₂Si is tricky because magnesium reacts easily with nitrogen (N₂). This usually forces researchers to use argon-filled gloveboxes for synthesis, even though N₂ gas is much cheaper than argon. To address this, Solco et al. combined high-energy ball milling with self-propagating high-temperature synthesis (SHS) to prepare Mg₂Si in a N₂ atmosphere. XRD and SEM-EDS analysis confirmed phase purity without Mg₃N₂ formation. The material exhibited a zT around 0.3 at 700 K, increasing to 1.0 upon Zn, Sn, and Bi doping. Beyond increasing carrier concentration, doping also introduced mass-strain fluctuations that enhanced phonon scattering [63]. This approach improves synthesis sustainability and could be extended to other systems. While bulk thermoelectric systems excel at large-scale, high-thermal-throughput waste heat recovery, their rigidity, heavy mass, and geometric constraints limit their integration into microelectronics and flexible wearable sensors. Consequently, thin-film thermoelectrics are critically needed to enable low-profile, high-power-density micro-cooling and localized energy harvesting where spatial footprints are severely restricted.

4. Advances in Thin-Film Thermoelectrics

Flexible thermoelectric generators (FTEGs) are promising for low-grade waste heat recovery in industrial systems [64] and wearable electronics such as e-skins, smart watches, ear buds or smart rings [20,65]. Thin films can be synthesized by magnetron [66,67] or radio-frequency (RF) sputtering [42], spin coating, pulsed laser deposition (PLD) [68], chemical vapor deposition (CVD) [69], metal–organic vapor phase epitaxy (MOVPE), and atomic layer deposition (ALD). Sputtering is widely used for flexible devices [70], while MOVPE enables precise compositional control (e.g., oxygen in oxides) [69]. On the other hand, PLD operates at low oxygen pressures, which may result in oxygen vacancies. ALD provides angstrom-level thickness control and is suitable for alloys, semiconductors and metals. These methods allow for the tuning of gas partial pressure, morphology, grain size, and thickness to optimize performance [42,71,72].

For example, adjusting N₂(g) pressure during the growth of n-type ZnSnN₂ modified the band structure and effective mass, increasing the power factor, although device performance was limited by p-type NiO formation [66]. Comparisons between thermal ALD and plasma-enhanced ALD (PEALD) show that PEALD films exhibit higher roughness

and carrier concentration and improved crystallinity due to plasma assistance. Transport properties improved with film growth temperature (up to 400 °C) and cycle number. However, electrical and thermal measurements were performed on films of different thicknesses because thermal diffusivity measurements require relatively thick films. As a result, the zT values of the materials were not calculated [73].

Film thickness critically influences leg resistance and output power in thin-film thermoelectric devices. Using metal-induced layer exchange, films up to 1000 nm thick can be achieved, enabling the fabrication of both n- and p-type SiGe thin films with optimized electrical performance. SiGe-based flexible thermoelectrics are particularly attractive because they consist of group IV elements that are abundant and nontoxic. Al-induced SiGe legs exhibited comparable power factors for both n- and p-type films, suggesting balanced carrier transport and stable device operation. The corresponding flexible module delivered an output power of approximately 1 μW at room temperature. These results highlight the strong influence of fabrication strategy, particularly thickness control and layer exchange growth, on the performance of thin-film thermoelectrics [67].

Oxide thin films are attractive due to their chemical and thermal stability. ZnO, a transparent n-type material, has been doped with Al, Ga, In and Sb to enhance its power factor [68]. Ga-doped ZnO prepared by sputtering showed reduced lattice thermal conductivity via impurity scattering. Therefore, a device with 10 n-type legs produced 230 nW at $\Delta T = 138\text{ K}$ [42]. In contrast, PLD-grown ZnO exhibited a modified conduction band with higher effective mass, slightly reducing the power factor but remaining competitive [68]. These results highlight the strong influence of fabrication methods on performance. When evaluating the transition from laboratory fabrication to industrial technology transfer, processing scalability and defect density become paramount. While pulsed laser deposition (PLD) offers excellent stoichiometric fidelity for complex multi-element oxides on a small scale, it suffers from a restricted ablation plume size and the inherent ejection of particulate agglomerates (droplets), which compromises large-area device uniformity. Conversely, despite challenges regarding the preferential re-sputtering of lighter elements, RF/DC magnetron sputtering remains the most industrially viable route for high-throughput, large-area thin-film thermoelectrics. Industrial scaling mitigates stoichiometry drift through the deployment of over-stoichiometric targets or multi-target co-sputtering configuration systems, successfully leveraging sputtering's superior film uniformity, step coverage, and compatibility with continuous roll-to-roll or batch manufacturing workflows.

SrTiO_3 is a perovskite thermoelectric material characterized by an intrinsically high Seebeck coefficient but relatively low electrical conductivity, which limits its practical performance. To overcome this limitation, Baki et al. synthesized SrTiO_3 using MOVPE, achieving high crystallinity together with precise dopant control. The resulting La-doped SrTiO_3 films exhibited enhanced electrical conductivity, primarily attributed to the reduced concentration of oxygen vacancies enabled by the controlled growth environment. As an n-type oxide, SrTiO_3 allows for effective donor doping through the substitution of Sr^{2+} with La^{3+} , which increases carrier concentration. However, despite the improved conductivity, the power factor of the La-doped films was lower than the values reported for samples grown by molecular beam epitaxy (MBE). This reduction is attributed to the possible formation of SrO interfacial layers, which enhance charge carrier scattering and reduce mobility [69]. Overall, these findings highlight the strong influence of growth conditions on thermoelectric transport properties, demonstrating that synthesis route and microstructural control are as critical as compositional optimization.

Systematic doping studies on ZnO (Mg-, Al-, and Ga-doped) revealed that electrical conductivity increases as electronegativity differences decrease and orbital overlap increases, leading to high power factors. The results indicate that even though electrical

conductivity and the Seebeck coefficient depend on defects and dopant characteristics, power factors are less sensitive to these factors. Therefore, Ga-doped ZnO achieved a power factor of $148 \mu\text{Wm}^{-1}\text{K}^{-2}$ at 573 K [74]. This study shows the importance of electronegativity in the performance of thermoelectrics.

Mg_3Sb_2 - and Mg_3Bi_2 -based compounds [20] are promising alternatives to Bi_2Te_3 -based flexible thermoelectric generators (FTEGs) due to their layered [75], anisotropic structures, which help decouple electronic and thermal transport and enable high band degeneracy and a $zT > 1$ [20]. A full FTEG using n- and p-type Mg_3Bi_2 legs delivered 0.17 nW and $1.67 \mu\text{Wcm}^{-2}$ at $\Delta T = 2.5$ K [20]. By adjusting the Bi content during sputtering, the same material could be made either n-type or p-type, allowing the module to use a single material for both legs and reducing stress from thermal expansion differences. The low bandgap of 0.2 eV limits wider temperature operation due to bipolar conduction. A related module using p-type $\text{Mg}_3\text{Sb}_{1.1}\text{Bi}_{0.9}$ and n-type Ag_2Se achieved an output power of 9.96 nW and a power density of $77.38 \mu\text{Wcm}^{-2}$ at $\Delta T = 10$ K.

Organic thin films, especially PEDOT:PSS, are attractive for sustainable FTEGs due to their abundance, low weight, biocompatibility, and tunable conductivity [71], though they suffer from cracking and modest conductivity. H_2SO_4 -treated PEDOT:PSS films transferred onto paper and PET maintained electrical conductivity and the Seebeck coefficient after 10,000 bending cycles and showed humidity resistance [76]. Annealing under controlled temperature and humidity further optimized the balance between conductivity and the Seebeck coefficient, achieving a power factor of $103.9 \mu\text{Wm}^{-1}\text{K}^{-2}$; prolonged annealing induced color changes consistent with dedoping [71]. Thin films can achieve high localized efficiencies and exceptional areal power densities due to steep temperature gradients across microscale thicknesses, but their total volumetric power output is strictly limited by low active material mass. Conversely, bulk architectures leverage substantial structural volume to maximize total thermal throughput, making them superior for macroscale power delivery. Physical vapor deposition methods like magnetron sputtering offer superior large-area uniformity and excellent industrial scalability, but they suffer from the preferential re-sputtering of lighter elements that alters film stoichiometry. On the other hand, techniques such as chemical vapor deposition (CVD) and atomic layer deposition (ALD) provide unparalleled conformal step coverage and atomic-scale thickness control, though they are frequently constrained by slower deposition rates and toxic precursor hazards.

5. Challenges and Future Outlook

Although thermoelectric materials provide a sustainable route for converting waste heat into electricity, their efficiencies remain well below those of other power generation technologies [77]. Solar and nuclear systems correspond to effective zT values above 4, a level not yet achieved experimentally. The highest reported zT values include 3.1 at 783 K for SnSe [26] and 2.6 at 573 K for AgSbTe_2 [78]. Thus, significant room remains for developing high-performance, environmentally benign materials.

One promising direction is the integration of machine learning. The synthesis and characterization of thermoelectrics are time- and resource-intensive, whereas machine learning can predict new materials, transport properties, and device performance by identifying correlations within existing datasets [79]. This approach can narrow down optimal compositions and reduce intensive trial-and-error experimentation, improving research sustainability. Databases on borides [44], carbides, and silicides [49] could serve as starting points for data-driven material discovery.

Thin films offer additional opportunities through controlled layering and texturing, enabling alignment along high-performance crystallographic directions. Bulk materials

such as CuSbSe_2 , $\text{Ca}_3\text{Ca}_4\text{O}_9$, and BiCuOSe demonstrate improved performance when textured [80], and similar strategies can be applied to anisotropic thin films.

The accurate measurement of thin-film thermal conductivity remains challenging. As an alternative comparison metric, the electronic quality factor (B_E) can be used, as it depends only on the Seebeck coefficient and electrical conductivity and isolates the electronic contribution by excluding lattice thermal conductivity [81]. Accelerating the discovery of high-performance, tellurium-free thermoelectrics increasingly relies on integrating high-throughput density functional theory (DFT) band structure modeling with AI-driven material discovery workflows. This closed-loop approach enables the rapid screening of vast compositional spaces to identify optimal electronic band convergence and defect-induced phonon scattering centers for maximized ZT optimization.

Finally, maintaining a stable temperature gradient is particularly difficult in thin-film devices. Future efforts should therefore focus not only on improving transport properties but also on effective thermal isolation strategies while preserving device performance and lightweight design.

6. Conclusions

Thermoelectricity represents a promising route toward sustainable energy generation. Nevertheless, many state-of-the-art thermoelectric materials rely on toxic or scarce elements such as Pb and Te, which limit their long-term sustainability. Alternative material systems, including Mg_3Sb_2 -based compounds, Heusler alloys, and oxide thermoelectrics, offer competitive performance while relying on more environmentally benign compositions. The embedded carbon footprint of fabrication remains a key trade-off, as high-temperature mechanical alloying and spark plasma sintering (SPS) require intensive primary energy inputs. However, lifecycle carbon offset modeling indicates that the net emissions avoided by harvesting waste heat over a typical ten-year module operational lifespan fully amortize this initial manufacturing carbon penalty.

In addition, thin-film thermoelectrics reduce the absolute consumption of critical elements, further enhancing sustainability. This review compared the transport properties of bulk and thin-film systems and summarized recent advances in both fields. The materials and strategies discussed here provide viable pathways toward more sustainable thermoelectric generators, contributing to the broader goal of green and accessible energy technologies. Present ML applications in thermoelectrics are primarily limited by sparse, fragmented, and inconsistently reported experimental datasets that hinder the training of robust predictive models. Future research must prioritize the development of automated, standardized high-throughput synthesis platforms that generate high-fidelity, interoperable data to move beyond simple property prediction toward inverse design and material discovery.

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Abbreviations

The following abbreviations are used in this manuscript:

ALD	Atomic Layer Deposition
CVD	Chemical Vapor Deposition
DOS	Density of States
FF	Filling Factor
FTEG	Flexible Thermoelectric Generator
IoT	Internet of Things
MBE	Molecular Beam Epitaxy
MOVPE	Metal–Organic Vapor Phase Epitaxy
PEALD	Plasma-Enhanced Atomic Layer Deposition
PEDOT: PSS	Poly(3,4-ethylenedioxythiophene):Polystyrene Sulfonate
PLD	Pulsed Laser Deposition
RF	Radio-Frequency
TDTR	Time-Domain Thermorefectance
TEG	Thermoelectric Generator

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