

Review Article

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Revolutionizing energy harvesting: A comprehensive review of thermoelectric devices

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Abstract: The necessity for a shift to alternative forms of energy is highlighted by both approaching consequences of climate change and limited availability of fossil fuels. While a large portion of energy required can be generated by solar and wind, a diverse, sustainable energy generation mix is still necessary to meet our energy needs. By capturing otherwise lost heat energy and turning it into valuable electrical energy, thermoelectric can play a significant part in this. Using the Seebeck effect, thermoelectric generators (TEG) have established their capability to transform thermal energy into electrical energy directly. Furthermore, because they do not include chemical compounds, they are silent in operation and can be built on various substrates, including silicon, polymers, and ceramics. Moreover, thermoelectric generators have a long operational lifetime, are position independent, and may be integrated into bulky, flexible devices. However, the low conversion efficiency of TEG has confined their broad application, hampering them to an academic subject. Until now, recent developments in thermoelectric generators and devices are presuming the technology to catch its place among state-of-the-art energy conversion systems. This review presents the commonly used methods for producing thermoelectric modules (TEMs) and the materials currently studied for TEMs in bulk and printed thermoelectric devices.

Keywords: thermoelectric generators, thermoelectric modules, nanoparticles, thermoelectric devices, Seebeck coefficient

1 Introduction

Seebeck made the initial discovery of the thermoelectric phenomena in 1821. By heating the connection between two separate electrical conductors, he demonstrated how to create an electromotive force. It is discovered that the meter registers a minor voltage when the connection between the wires is heated. It has been determined that the size of the thermoelectric voltage is proportional to the difference in temperature between the connections to the meter and the thermocouple junction. The second of the thermoelectric effects was noticed by French watchmaker Peltier 13 years after Seebeck made his discovery. He discovered that depending on the direction of the electric current, passing through a thermocouple results in a minor heating or cooling impact. Since the Joule heating effect always accompanies the Peltier effect, it is very challenging to show the Peltier effect using metallic thermocouples. Showing less heating when the current is transmitted in one way instead of the other is often all that can be done. The Peltier effect may be theoretically illustrated using the setup in Figure 1 by exchanging the meter for a direct current source and mounting a tiny thermometer on the thermocouple junction. The thermoelectric (TE) is explained using the aforementioned phenomena [1–3]. These three phenomena and Joule effect provide the physical foundation for TE transformation procedures [4]. Seebeck observed a magnetic field surrounding the two distinct metal wires in a loop when he warmed up one connection but kept the other at a low temperature (Figure 1(a) and (b)). The formation of the magnetic field due to the temperature difference between two junctions was named thermo-magnetism.

A further explanation was offered by Oersted, whose experiment showed that the magnetic field near the circuit was unaffected by the temperature change. The temperature variation produced a voltage V_{ab} and an electric current in the circuit, which produced the experiment's

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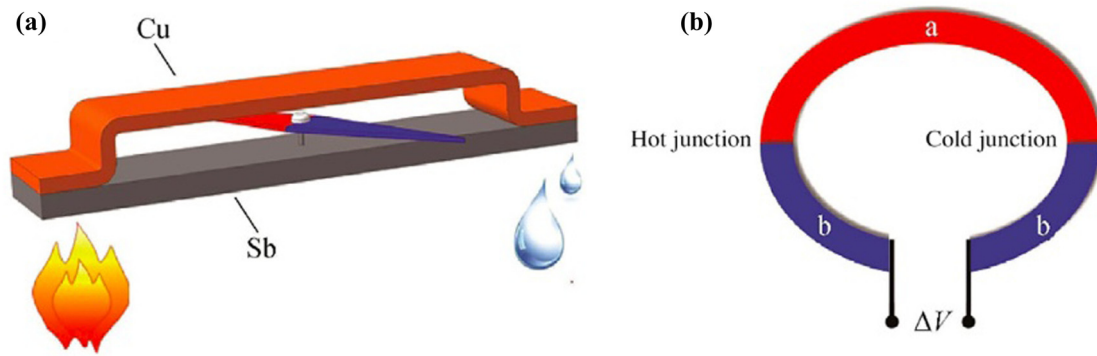


Figure 1: (a) Experimental observation and (b) seebeck effect, reproduced with the permission of Chen *et al.* [5].

magnetism. The formation of electric current due to the temperature difference was named thermoelectricity [6]. Seebeck first identified the phenomenon; since then, it has been referred to as the Seebeck (Sb). The Seebeck effect, which explains the phenomenon of the carrier's directly pumping heat, is the reverse of the Peltier effect. Additional heat will be released or absorbed at these two junctions in addition to the Joule heat created when a current is applied to a circuit of two conductors. Since J. C. A. Peltier, a French physicist, first identified this effect in 1834, it has been named after him.

He connected the Bi and Sb wires and observed how a metal connection caused water droplets to condense when an electric current was sent across the circuit. When the current turned around, the ice disintegrated (Figure 2a). When electric current applied at junction of two different Fermi-level conductors, the electrons will either jump from the high to the low or in the opposite direction across the

interface potential barrier [7]. They will absorb or release energy in the form of heat at the junction (Figure 2b). It was not until 1855 that William Thomson (Lord Kelvin) recognized this issue as being related to the link between the Seebeck and Peltier effects [8]. He reasoned that there must also be a third effect. In addition to the Joule heat, a uniform conductor should experience reversible heat absorption or release as an electric current passes through it. The Thomson effect was introduced in 1867 after this effect had been empirically verified. Several research works take the constant value of Thomson coefficients and establish that the coefficient showed a significant impression on the efficiency of thermoelectric generators (TEGs). Subsequently, Yamashita *et al.* [9] investigated the linear temperature-dependent effect of TE materials on TEG performance. The asymptotic solution to the nonfundamental heat control equation for TE materials was proposed by Sandoz-Rosado *et al.*, [10] and the equations were derived from

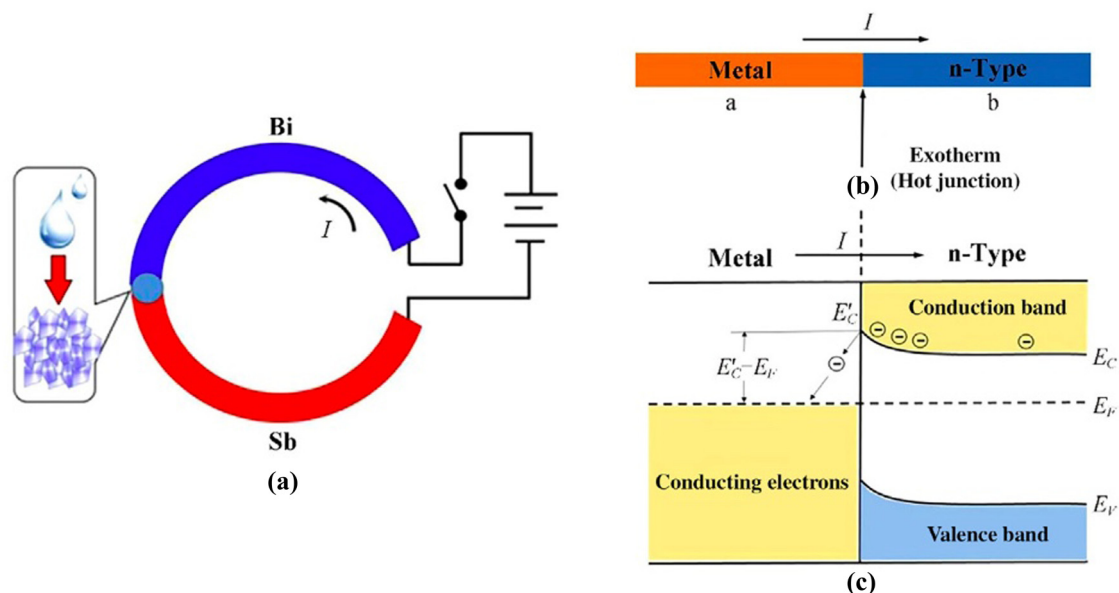


Figure 2: (a–c) The Peltier effect, reproduced with the permission of Chen *et al.* [5].

the temperature-dependent cumulative model incorporating Thomson's effect by Kim *et al.* [11].

2 Thermoelectric devices

Thermoelectric devices are like heat engines. Their efficiency constraint is comparable to the Carnot efficiency [12]. According to the Carnot cycle [13], the energy can be absorbed or dumped from a high- or low-temperature reservoir; it cannot be taken from room temperature. Because of the trade-off between efficiency and power that applies to all heat engines, achieving the Carnot efficiency at finite power is complicated. It is noted that without violating the trade-off relation, the Carnot efficiency at finite power might be reached in the vanishing limit of a system's relaxation times. The TEG characteristic and outputs not only depend on the Carnot efficiency but also on the dimensionless parameter of material called a figure of merit (ZT) [14]. This dimensionless parameter depends on numerous material characteristics such as temperature [15], Seebeck coefficient [16], electrical [17], and thermal conductivity [18]. Modern TE device designs, including traditional TE module design, flexible device design, microdevice design, and applications in various sectors, require high-efficiency levels. TE devices for power generation have received significant attention due to their high reliability, solid-state operation, and stability. When designing thermoelectric power generation devices, it is necessary to consider design principles, fabrication techniques and testing methods, interface optimization, barrier layer design, electrode fabrication, protective coatings, and component and module estimation aspects of TE devices. These TE designs depend on elegant, dimensional designs that guarantee excellent stability and performance [19]. Environmental factors that may harm or improve device performance include oxidation, magnet fields, pressure, and radiation.

Usually, the metal-electrode links n- and p-type TE material to form the TE module (element) [20]. TE devices are created by joining many TE modules in series electrically and in parallel thermally due to the low output voltage of a single TE module [21]. The device is usually assembled by soldering a copper electrode to the ceramic sheet with a thermally conductive surface that acts as the electrical insulating board for both the cold and hot ends. A copper sheet is directly adhered to a ceramic substrate using the copper-cladding technique in a preset pattern [22]. The n- and p-type thermoelectric modules (TEMs) are linked using standard soldering methods to fabricate

a sandwich structure. The wettability of TE can be improved with the help of Nickel, which can be used in solders and soldering surfaces. Solder alloys are essential for the fabrication of integrated circuits and electronic devices. Nickel solders are popular because of their low melting point, superior solderability, and low price [23]. Despite the simplicity and low cost of this preparation procedure, the use of ceramic leads to brittleness and mechanical stress.

For the construction of thermoelectric devices (TEDs), complex electrode preparation techniques such as arc-spraying [24], high-temperature diffusion welding [25], and a combination with Spark plasma sintering [26] have been adopted. The TE device can work at temperatures that are not constrained by the melting point of the solder when using the arc-spraying technique, which shows outstanding thermal stability [27]. A transition or intermediary layer is often formed on the electrode connection surface of TEMs in the interim to facilitate interface bonding. The arc spraying apparatus also has good mechanical stress resistance because the TEMs are fastened and packed in the gap between the frame and electrode. Recently, a one-step method for producing devices was developed that uses hot pressing or SPS to concurrently make n-type and p-type materials and sinter electrodes [28], barrier [29], and TEMs [30]. Using integrated methods, creating low interface contact resistance and high-strength bonding of TEMs and electrodes is straightforward. TEDs are divided into three types: π -type [31], O-type [32], and Y-type [5], based on their diverse designs [33]. The TE gadget of the π -type is the most well-known of them (Figure 3).

The TE modules in π -type TE devices are housed within two efficient heat-conducting electrically insulated ceramic panels [34]. The heat flow is generally directed to a ceramic flat plate, which is appropriate for a flat source of heat's operational environment [35]. Unidirectional heat flow makes it simple to establish uniform heat flow density in TEMs [36], and the best structure design can enhance the TE performance to achieve high efficiency. However, a TE device frequently constrains cold and hot surfaces. The temperature change caused severe thermal stress and impaired the structure's endurance due to the different thermal expansion of other materials [37]. In an O-type TE device, the columnar heat source alternates coaxially with n- and p-type TEMs [26]. The annular insulation material's layer is positioned in the middle of each layer of TE material to establish electrical isolation between the adjacent layers [38]. In addition, metal coil electrodes are used to connect nearby TEMs. In particular, a radial heat flow is observed in the cylindrical heat source. Therefore, a ring-like TE device is suitable for nonflat heat sources. However, the efficient design of electric fields and temperature makes achieving the radial heat flow and current difficult.

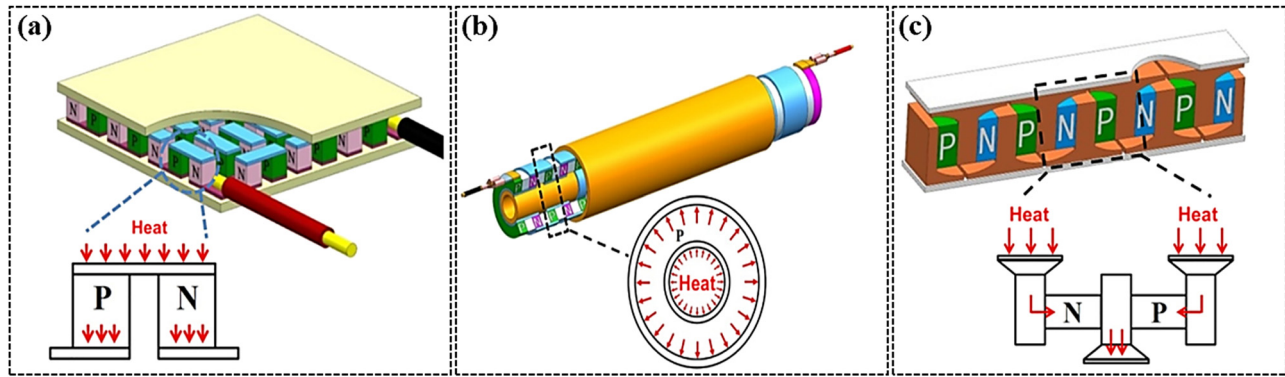


Figure 3: (a) π -shaped TE module, (b) tube-shaped TE module, and (c) Y-shaped TE module, reproduced with the permission of Yubing *et al.* [34].

Compared to a flat device, welding of specifically shaped TEM, metal electrodes, and device integration is much more complex, and the production cost is significantly higher [26].

In Y-type TED, connecting electrodes contain cylindrical or rectangular n- and p-type TEM. The connection plates of electrodes offer conductive channels for nearby n- and p-type TEM and function as heat-collecting and

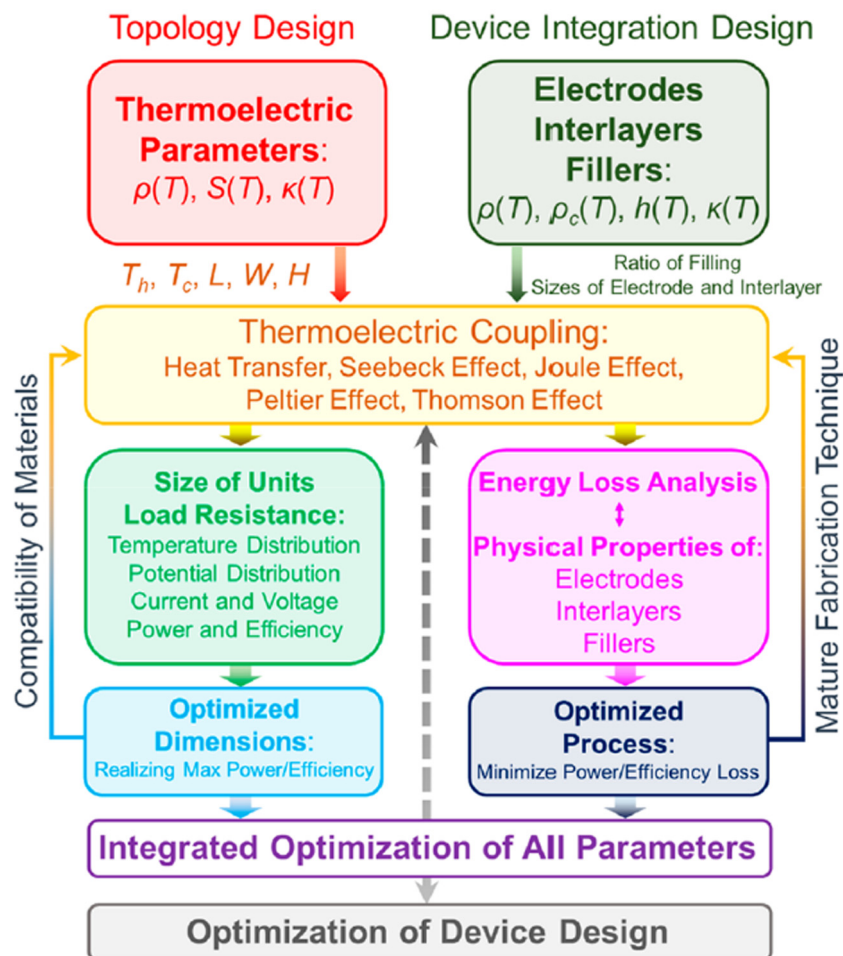


Figure 4: The flowchart description of the basic design criteria for TED, reproduced with the permission of Deepthi [39].

transmission devices [40]. The lateral series thermal expansion of n- and p-type TEM avoids stress concentration due to variations in expansion coefficients and provides better freedom in TEM design parameters [41]. Furthermore, the Y-type construction allows each TE module to be individually optimized structurally [42]. TEM could have a variety of sizes in terms of structure interface and height, which is advantageous in the fabrication of divided structures. The heat and current density nonuniformity flow in Y-shaped TED influenced the efficiency of the TEM materials and shrink device performance [39].

Figure 4 depicts the basic design criteria for TED, which applies to all TED types. The flow diagram shows that the device's topological layout directly impacts output attributes, such as device energy density. As a result, matching the device topologies under real operating conditions is the foundation for optimizing a TE power-generating system's comprehensive indicators [43]. The limits and affecting elements must be thoroughly studied depending on the operating mode of the TED. In the device design phase, it is critical to incorporate mathematical investigation, thermal-electric structure coupling inquiry, finite element numerical modeling, and transient structure analysis [44]. Several interconnected structural parameters must be optimized simultaneously following the device's overall modeling. The efficiency of TED's energy is affected by the TEM performance and the difference in temperature between the two ends of the materials and by their size [45]. When different resistivity of the p- and n-type TEM composes a TE unit, the performance is reduced, which can be countered by adopting two TE "legs." In general, the finite element approach is beneficial for resolving such problems. When a temperature gradient (ΔT) is introduced, the change carriers (electrons for n-type materials and holes for p-type materials) starts moving from the hot side and diffuse to the cold side, this phenomenon is called material's TE energy-harvesting mechanism. An electrostatic potential (ΔV) is consequently created. A solitary n- or p-type TE leg produces an

extremely low electrostatic potential (varying from μV to mV depending upon the situation). TE generators are usually composed of dozens or even hundreds of TE couples to gain high output voltage and power.

Furthermore, a single TEM's ideal working temperature range is frequently small [46]. The efficiency is limited when utilizing a single TEM [47]. Every material has different physical characteristics; therefore, the degree of nonlinearity increases in temperature distribution. In the case of segment TED's, they have many interfaces and complicated topologies, making it challenging to optimize device performance. Another critical component to consider in designing the device is the fill factor (F), which is the ratio of the TEM to the area of the ceramic plate [48]. In general, F must be optimized to maximize efficiency [49]. The link between electrodes and TEM is critical in TED design, particularly for high-temperature electrodes [50]. Because TEDs may remain in service for lengthy durations, interdiffusion and chemical reactions take place at the interfaces of high-temperature electrodes and TEM interfaces [51]. This reduces the efficiency due to increased electrical and thermal resistance. The reasonable remedy for this problem is to place a suitable transition layer between the electrodes and the TEMs, acting as a diffusion barrier [52]. In low temperatures, TED's transition layer's purpose is to increase solder and material's wettability. It also helps reduce thermal and interface resistance. However, its selection and design are much more complicated in the case of high-temperature TEDs [53]. Here is the real example of TEM and TEG in daily life as shown in Figure 5.

3 Nano-particle (Na-Par) preparation methods

In this section, we present some of the commonly used methods for preparing the TEMs.

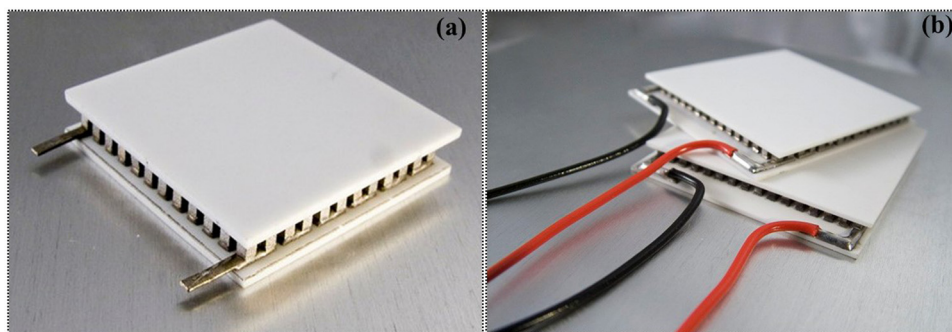


Figure 5: (a) Thermoelectric module and (b) thermoelectric generator.

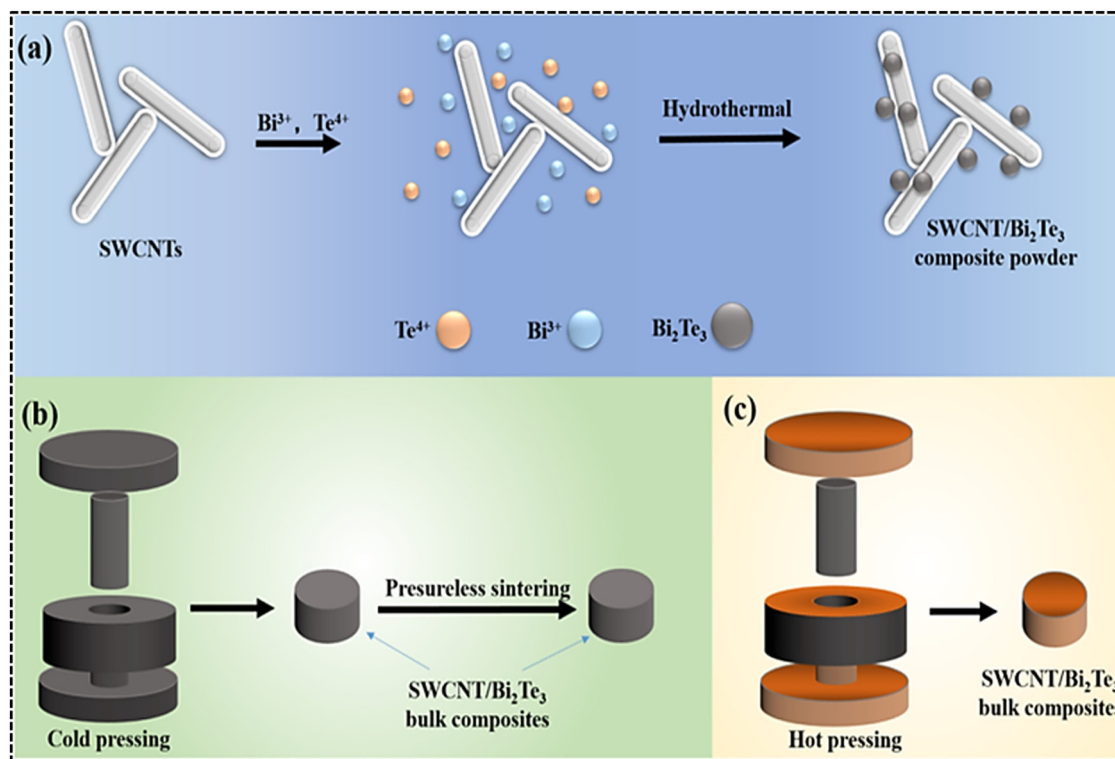


Figure 6: (a) Hydrothermal process, (b) Cold pressing and (c) Hot pressing, reproduced with the permission of Liu *et al.* [79].

3.1 Hydrothermal (HD) method

One of the most popular and efficient ways to create nano-materials (Nan-Mat) with diverse morphologies is HD. This method uses an autoclave to conduct the reaction at high temperatures and pressures while using water or an organic substance as the reactants. The name “hydrothermal process” (HD) refers to the use of water throughout the preparation process [54,55]. Comprehensive discussions on various autoclave types and their purposes have also been reported in the literature [56,57]. Teflon-lined autoclaves can often withstand high temperatures and pressures [58]. Compared to glass and quartz autoclaves, it tolerates alkaline media and greatly resists hydrofluoric acid. A Teflon-lined autoclave is the best container to react under the necessary circumstances. The main element that makes it possible to synthesize different nanostructured inorganic materials during the HD process is precise control [59]. This technique can stimulate hydrolysis by crystal growth, resulting in the self-assembled Nan-Mat in solution. It can also facilitate and speed up the interaction between the reactants. In addition, it is simple to tune the characteristics, morphology, size, and structure of Nan-Mat by adjusting the various parameters, such as temperature, reactant concentration, reaction medium, pressure, pH, reactant concentration, reaction time, as well as the filled capacity of the autoclave. The following steps are part of the HD synthesis process: (i) preparing precursors by

combining soluble metal salt using aqueous solution, (ii) combining the solution into an autoclave to start the reactions at an elevated temperature and pressure, and (iii) finalizing the process by filtering, washing, and then drying [60]. Many Nan-Mat have been studied using the HD technique [61–79].

3.2 Sol-gel method

Currently, numerous methods and techniques are employed to produce nano-materials. However, the sol-gel technique is mainly used for industrial purposes because of its more comprehensive range of applications [80–84]. The industrial-level employment of this technique is due to producing high-quality nanoparticles in large quantities [85–87]. It is also used for metal oxide nanoparticles and mixed oxide composites. The surface and textural characteristics of the materials might be manipulated using this technique. The sol-gel procedure primarily goes through three steps to get the final metal oxide treatments: drying [88], hydrolysis [89], and condensation [90]. The process of creating metal oxide comprises many sequential processes (Figure 6).

First, the matching metal precursor quickly hydrolyzes to generate a metal hydroxide solution, which is immediately condensed to form 3D gels. Depending on the drying method, the resultant gel can be transformed into Xerogel

or Aerogel [91]. Depending on the solvent used, the sol–gel process may be divided into two categories: aqueous sol–gel and nonaqueous sol–gel [92]. The aqueous sol–gel technique uses water as the reaction medium, whereas the nonaqueous sol–gel technique uses organic solvents for reaction media. Figure 7 depicts the chemical route used in the sol–gel process to produce metal oxide nanostructures [93]. The metal precursor and solvent composition in the sol–gel process is essential for creating metal oxide Na-Par. In the aqueous sol–gel method, water solvent provides the oxygen, which is required for the formation of metal oxide. Typically, the metal precursors used in this process include metal acetates and nitrates. However, due to the strong attraction of alkoxides to water, metal alkoxides are often utilized as precursors for forming metal-oxide nanoparticles [94,95]. The aqueous sol–gel approach does come with specific challenges, though. In various situations, the crucial steps – hydrolysis, condensation, and drying – occur concurrently, making it challenging to regulate particle shape and ensure reproducibility of the end procedure during the sol–gel method [96]. The nonaqueous sol–gel technique uses solvents such as ketones, aldehydes, alcohols, or metal precursors to provide the oxygen needed to create metal oxide.

These organic solvents also deliver oxygen while providing a flexible tool for adjusting several essential factors, including the final oxide material's composition, particle size, surface characteristics, and morphology. Although nonaqueous sol–gel approaches are less well known than aqueous sol–gel

approaches, they have demonstrated superior effects on synthesizing nano-oxides compared to aqueous sol–gel process. The generation of metal oxide nanoparticles, a nonaqueous sol–gel pathway, may be separated into two key strategies: surfactant-controlled and solvent-controlled approaches [93]. The hot injection approach uses a surfactant-controlled strategy that directly transforms metal precursor into metal oxide at higher temperatures. Some recent examples are as follows: Bi_2Te_3 is one of the most widely used materials for thermoelectric applications due to its high thermoelectric efficiency. Sol–gel methods can synthesize Bi_2Te_3 nanoparticles or thin films, which can then be assembled into TEMs.

Similarly, the PbTe is another commonly used thermoelectric material, especially in high-temperature applications. Sol–gel techniques can fabricate PbTe nanoparticles or thin films for constructing TEMs. Also, the SiGe alloys exhibit good thermoelectric properties, particularly at elevated temperatures. Sol–gel processes can deposit SiGe thin films or nanoparticles, which can be integrated into TEMs. Sam as copper selenide is an emerging thermoelectric material known for its nontoxicity and earth abundance. This method can synthesize Cu_2Se nanoparticles or thin films, which can be incorporated into TEMs. This technique eliminates particle agglomeration and allows for excellent control over the formation and development of the Na-Par [93].

3.3 Polyol method

Different types of Na-Par with various morphologies have been created using several potential synthetic techniques [97]. Among these, the polyol approach is a flexible liquid-phase technique that uses multivalent alcohols and high boiling to create Na-Par [98]. Polyols can regulate the development of particles in addition to acting as a solvent and reducing agent. The Fievet's group adopted the word polyol for synthesizing metal nanoparticles in 1989, which marked the beginning of the polyol synthetic pathway. This process has been used with various polyols, including diethylene, ethylene, propylene, triethylene, tetraethylene, butylene, and polyethylene glycol. Moreover, Carroll *et al.* synthesized elemental copper and nickel nanoparticles using the polyol method and investigated further with theoretical calculations [99]. However, polyols offer fantastic benefits in several ways. Several reports have been published using polyol methods, such as Grisaru *et al.* who reported the microwave-assisted polyol method was quite impressive for the fabrication of CuInTe_2 and CuInSe_2 nanoparticles. Also, simple and inexpensive metal precursors can be used as

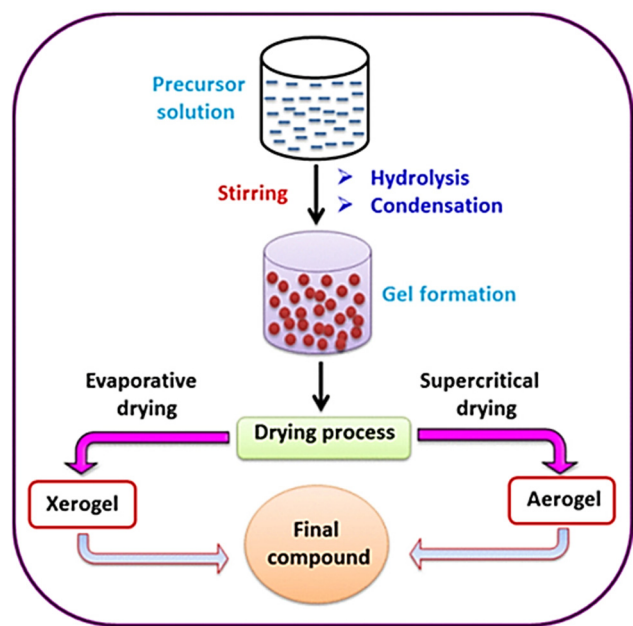


Figure 7: Sol–gel method, reproduced with the permission of Adschiri *et al.* [100].

starting compounds because of polyols' excellent beginning material solubilization properties [101,102]. Riyang *et al.* used the polyol reduction method to synthesize a highly dispersed Ru/SiO₂-ZrO₂ catalyst. The chelating characteristics of the polyol aid in controlling crucial elements of the particle nucleation, growth, and aggregation processes. Another important advantage of polyols is their ability to swiftly reduce metal solutions to produce metal Na-Par at higher temperatures [103]. When choosing a polyol to synthesize Na-Par, the boiling point and reduction potential must be considered.

3.4 Bridgman method

Single crystals may be grown under controlled conditions using the Bridgman method. The crystal formation of choice can be achieved using a regulated temperature gradient [104]. The gradient may be changed by adding various temperature regions to the system or by adjusting the system's overall temperature. This process can produce single crystals in semiconductors and other electrical devices [105,106].

The polycrystalline material is heated over its melting point in an oven using the Bridgman method. A vertical and a horizontal orientation may be used to develop the crystals. Their somewhat differing cooling systems are what distinguish the two approaches. Dynamic vertical single-

crystal development is shown in Figure 8. The approach has historically relied on a dynamic cooling mechanism, with hot and cold zones in the oven. In an ampoule, the melt (Figure 8) is swirled before being transferred through the cooling area of the oven. The nucleation takes place lengthwise in the cold end of the oven, and the single crystal formation begins at the end of the hot zone. To create a homogeneous crystal, cooling during crystallization should be gradual. The cooling rate is determined by the material and the product's requirements. When the crystal is completely formed, the system is cooled down to room temperature. The domain crystalline substance is afterward returned to the melt [105]. In a static cooling setup, the melt does not move as an ampoule's temperature varies over time. The procedure is carried out in a coil-equipped melting oven. Because the process requires constant regulation of the temperature gradient, the oven must support certain temperatures. Ovens can be of many sorts, some providing temperature control in a single or numerous zones. Most melting ovens are appropriate for controlling the temperature in one zone of static cooling systems. There must be several temperature zones in dynamic cooling systems. For instance, because tube ovens allow for numerous temperature zones, they are frequently employed in Bridgman techniques.

3.5 Czochralski method (CZ)

In the CZ process, a small crystal is first inserted into a melt-in crucible, which pulls the seed upward to produce a single crystal [106]. This method of creating nanoparticles approach was established in 1916 by Jan Czochralski. The technique produces numerous single crystals, including Si and Ge [107]. Several researchers have used the CZ approach to create single crystalline SiGe. Kürten *et al.* used a prolonged pulling rate during the CZ development of Si-rich SiGe to create a stable liquid–solid interface. In addition, the gas flow shape was designed to ensure superior temperature stability. As a result, single-crystalline SiGe was produced with a Ge concentration of up to 0.2 [108]. Yonenaga *et al.* have thoroughly investigated Si_{1-x}Ge_x CZ development across the whole range of Ge content and has produced single crystals for $0 < x < 0.1$ and $0.85 < x < 1$ [109–114]. Poly-crystals were formed with an intermediate Ge concentration. On the basis of Tiller's criteria [115], they calculated the critical growth velocity of occurrence for constitutional super-cooling as a function of Ge concentration at a specific temperature gradient. However, poly-crystallization was seen for the intermediate Ge content even when the growth velocity was slower than the critical value [116].

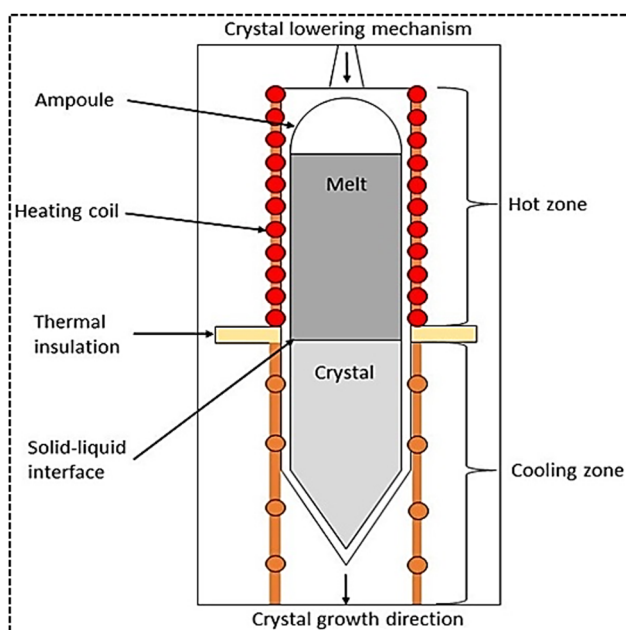


Figure 8: Illustration of the Bridgman method, reproduced with the permission of Mohamed *et al.* [117].

No special attention was paid to the homogeneity of Ge content, and the Si depletion was seen in the pulling direction. Deitch *et al.* [118] proposed a modified CZ technique. They employed pre-grown large-diameter single-crystal Si seeds to speed up cap crystallization and guarantee a single crystal in a large crystal diameter. In actuality, SiGe single crystals with a Ge concentration of up to 0.17 and a diameter of 68 mm were produced. Abrosimov *et al.* employed Si rods for continuous Si feeding to the melt (Figure 9) to prevent Si depletion during the CZ development of SiGe [119]. They have tried to maintain a consistent Ge content in SiGe by regulating the Si rods' movement rate and the rate at which the developed crystal is pulled. Global modeling of the entire chain, including Si dissolution across the solid-liquid interface and the transport in the melt, will be required to optimize the process.

3.6 Conjugated polymers

However, inorganic semiconductor materials have a few intrinsic drawbacks, such as high manufacturing costs, toxicity, poor processability, and scarcity. Furthermore, most high-performance inorganic TE materials work best at temperatures above 200°, so they cannot satisfy the growing need to capture waste heat below 150° [120]. However, there may be answers to the issues with organic semiconductors, which have been disregarded in recent

decades because of their low energy conversion efficiency and potentially poor thermal stability. Conducting polymers, for example, are interesting candidates for organic semiconductors because they can convert low-end thermal energy into usable electricity [121]. This could lead to developing larger-area, more flexible, low-toxicity, and sustainable TEDs [122]. Over the past 10 years, significant efforts have been made to enhance the performance of organic thermoelectric materials. These efforts include the development of novel molecular design strategies and the creating of nanocomposites that combine nanomaterials and conducting polymers [123].

3.7 Graphene

Two-dimensional carbon lattice graphene has superior thermal and electrical characteristics [124]. Researchers' interest in studying the thermoelectric properties of graphene skyrocketed following its discovery as the best conductor of heat and electricity. While graphene's exceptional electrical conductivity can be attributed to its ultrahigh carrier mobility ($15,000 \text{ cm}^2 \cdot \text{V}^{-1} \cdot \text{s}^{-1}$), its exceptionally high thermal conductivity (which can reach up to $2,500\text{--}5,000 \text{ W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$) poses a significant obstacle to its application as a thermoelectric element [125]. Furthermore, generating an effective thermoelectric voltage in graphene is challenging due to its low Seebeck coefficient, commonly $10\text{--}100 \mu\text{V} \cdot \text{K}^{-1}$ [126]. Except for a few recent investigations, all these variables contribute to the extremely low reported ZT values for pristine graphene, which are approximately 10^{-4} . For instance, a record-high ZT of 0.1 has been recorded in suspended graphene nanoribbons with widths of about 40 nm and lengths of about 0.25 microns. In contrast, a typical semiconductor has a respectable ZT of 1–2 [127].

4 Uniqueness of bismuth telluride

In recent years, TEMs have been extensively explored in terms of their characteristics, structure, and TE capabilities with various dopants. Many of these dopants give information about their features. This section examines the dopants utilized in the current and past and their influence on the TE characteristics of novel alloys. Bi_2Te_3 and its companion substances were traditional TEM. Numerous TE cold modules with BiTe components have been extensively used in X-ray and laser detectors. According to theoretical calculations, the 5 Å wide Bi_2Te_3 quantum wire might achieve the

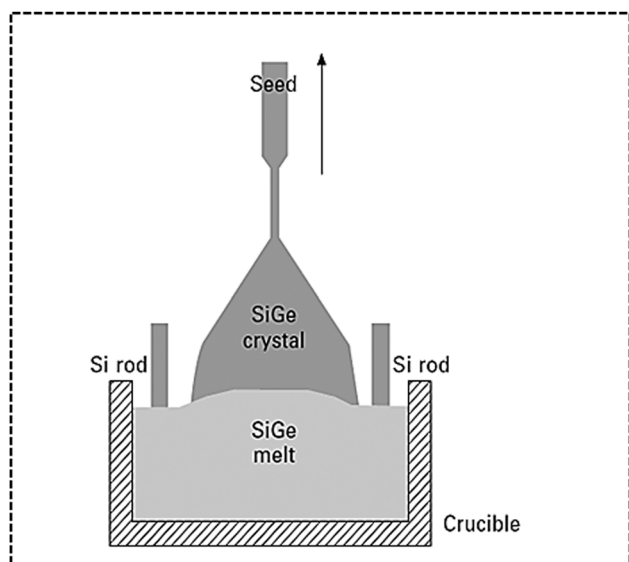


Figure 9: Diagram illustrating the CZ development of a bulk SiGe crystal with Si being continuously added to the melt, reproduced with the permission of Wei *et al.* [120].

maximum ZT value of 14 [128]. ZT might potentially rise by a factor of 13 when Bi_2Te_3 is used in a quantum-well arrangement [129]. Density functional simulations were used to examine the TE characteristics of Bi_2Te_3 nanowires. The ZT value for nanowires with a width of $N = 5$ and a [130] growth direction was 2.3 at 300 K and 2.5 at 350 K [131]. BiTe TEM characteristics varied depending on the preparation techniques used. The more energy used for material fabrication, the greater the efficiency of TE.

From liquid to solid, from low temperature to high temperature, and from straightforward to complex methods, these can affect the production of nanoparticles, and the ZT was affected by these methods [132]. The HD process was used to synthesize nano-powders of Bi_2Te_3 , $\text{R}_{0.2}\text{Bi}_{1.8}\text{Te}_3$ (whereas $R = \text{Sm, Y, Ce}$) [133] and $\text{Bi}_2\text{Te}_{3-a}\text{I}_a$ ($a = 0.0, 0.05, 0.1, 0.2$) [134]. The samples of $\text{Bi}_2\text{Te}_{2.85}\text{Se}_{0.15}:\text{X}_a$ ($\text{X} = \text{I, C, Ag, Ni, and Zn}$) [135], $\text{Bi}_2(\text{Te}_{0.9}\text{Se}_{0.1})_3$ [136], and $\text{Ag}_x\text{Bi}_2\text{Te}_{2.7}\text{Se}_{0.3}$ [137] were prepared using melting and hot pressing in encapsulation. Ajay *et al.* prepares $\text{Bi}_2\text{Te}_{3-x}\text{Se}_x$ samples using the Polyol method [138,139]. Yang prepared N-type Bi_2Te_3 :Pb doped with Pb using radio frequency magnetron sputtering, and Bi_2Te_3 films doped with Pb on SiO_2 substrates at room temperature. Small-size Pb plates were used as the source materials, and silver paste was used to fix Pb plates to the Bi_2Te_3 target so that the doping concentration in the films could be changed [139]. The HD approach was used successfully to produce flower-like nano-powder of $\text{Ti}_x\text{Bi}_{2-x}\text{Te}_{3-x}\text{I}_x$. Then, heat and pressing are used to form bulks from the created nanoparticles [140]. Wu *et al.* prepared $\text{Bi}_{2-x}\text{Zn}_x\text{Te}_3$ single crystals using a modified Bridgman method to study its TE properties [141]. Singh *et al.* prepared $\text{Bi}_{2-x}\text{Gd}_x\text{Te}_3$ samples using the sol-gel method [142]. A KrF excimer laser was used to prepare $\text{Bi}_2\text{Te}_{2.7}\text{Se}_{0.3}$ thin films with a high c -axis texture on fused silicon substrates. A commercially available $\text{Bi}_2\text{Te}_{2.7}\text{Se}_{0.3}$ target was slightly enclosed with a platinum sheet (0.1 mm) along the diameter to form a thin nano-composite coating with platinum inclusions [143]. Hsin prepare polycrystalline $\text{Bi}_{40-y}\text{Ge}_y\text{Te}_{60-x+y}$ alloys using the Bridgman technique for the TE characteristics assessments [144]. CZ technique was used to create single crystals of Bi_2Te_3 . Polycrystalline samples with grapheme (GR) doping were created using a solid-state reaction. The crystals were created using Bi and Te powders. Powders of Bi_2Te_3 were combined with GR, which was then crushed into prill and sintered at 520°C for approximately 600 min in argon [145].

Bi_2Te_3 beads were used to create the Bi_2Te_3 powder and was then processed using zirconia balls in an inert environment to prevent oxidizing the particles' surfaces. A diluted HCl solution was used to rinse the ball-milled powder to remove any oxidized coatings on the surface. Based on the methods previously reported [146], with a few changes, the Bi_2Te_3 nano-wires were synthesized. A modified version of Hummers' approach, as described in earlier

published publications [147], was used to create GR oxidized nano-sheets. The produced GO sheets had several micrometers dimensions and a 4–5 nm thickness. The combination was first ultrasonicated for 1 h after being suspended in 500 mL of deionized water containing GR oxidized and Bi_2Te_3 powder. After that, a suitable amount of $\text{N}_2\text{H}_2\text{-H}_2\text{O}$ was added to limit the dispersion. This combination was cooked at 368 K for 2 h while being stirred. The finished item was filtered, cleaned with deionized water, and dried for 24 h at 333 K. To create the GR/ Bi_2Te_3 -powder composites, the finished product was ground into an adequate powder, put into an iron mold, and hard-pressed under high pressure [148]. The composite specimen of Bi_2Te_3 :G was made by incorporating 0.05% by weight of G into the Bi_2Te_3 powder [149]. These powders were well combined with agate mortar and pestle before being compressed into pellets. The pressure was applied to room-temperature air to compact the powder. The samples were held in storage for 5–7 min. Similarly Bi_2Te_3 :C composite sample was created by combining Bi_2Te_3 sample and graphite powder [150]. Qijiang *et al.*'s [108] study shows no notable impure phases such as tellurium, bismuth, or cerium are observed. The lattice parameters $\text{R}_{0.2}\text{Bi}_{1.8}\text{Te}_3$ are all significantly more significant than the Bi_2Te_3 . The minor shift in the crystal characteristic can be due to the small changes in the radius of the atoms. SEM revealed that their morphologies change greatly when nano-powders are doped with rare earth elements. A specimen free of rare earth elements displays a flower-like appearance. Nano-sheets contract when Ce, Y, and Sm are introduced. It suggests that the doping of rare earth elements has a substantial impact on the shape of the powders. It is unknown why rare earth element doping may drastically alter morphology [151]. The presence of rare earth metals alters the bonds' robustness and significantly affects their growth [108].

Doping with rare earth elements boosts electron concentrations and lowers electrical resistance. The electrical resistivity of $\text{R}_{0.2}\text{Bi}_{1.8}\text{Te}_3$ is, nevertheless, more significant than that of un-doped Bi_2Te_3 [152]. There is an inverse relation between electrical resistivity and carrier mobility and concentration; hence, differences in carrier numbers will affect variations in electrical resistivity. Decreased carrier mobility will result in greater electrical resistivity – two plausible explanations for reduced carrier mobility of doped samples. First, substituting rare-earth elements can decrease the carrier mobility by amplifying alloy scattering. Second, chemical bonding can have a significant impact on carrier movement. Covalent bonds are generally more favorable for carrier movement than ionic components [153]. The fraction of the ionic part of the bond of two

elements may be determined using the conventional Pauling empirical equation and electronegativity of bonding atoms [154]. Temperature affects the Sb coefficients. The specimens display n-type conduction because of negative Sb coefficients in the investigated temperature [155]. The intrinsic excitation temperatures of $\text{R}_{0.2}\text{Bi}_{1.8}\text{Te}_3$ was 480 K, indicating similar intrinsic excitation temperatures (approximately 500 K) and band gap to binary Bi_2Te_3 . The thermal conductivity was decreased by substituting Ce, Sm, and Y [156]. The most effective dopant was Ce onto Bi_2Te_3 for reducing thermal conductivity. As a result, the ZT value of $\text{Ce}_{0.2}\text{Bi}_{1.8}\text{Te}_3$ reached to 1.29 at 398 K, higher than Bi_2Te_3 -based alloys but greater than zone-melting ingots [157–160] (Figure 10 and Table 1).

Wu *et al.* [134] explored I-doped $\text{Bi}_2\text{Te}_{3-x}\text{I}_x$ nano-powders. The elemental diffraction scattering (EDS) investigation was done by gathering spectra from over 15 nano-crystals. The EDS and X-ray diffraction (XRD) measurements reveal that I was successfully doped onto Bi_2Te_3 . The lattice characteristics of $\text{Bi}_2\text{Te}_{3-x}\text{I}_x$ compounds derived by Rietveld refinement were all shorter than the Bi_2Te_3 lattice values (JCPDS15-0863 card). This is mostly due to I's smaller atomic radius (0.133 nm) compared to Te's (0.137 nm) [161]. The Nan-Mat showed flower-like morphology ($x < 0.1$) [162–165] and exhibits thick layers that are less than 100 nm in thickness. These morphologies were destroyed when the concentration of I was increased. The sheets of such morphologies became significantly smaller, so I doping does not prompt large nano-sheets. As the temperature climbed, the electrical resistivity of each sample increased, behaving as a degenerate semiconductor. The I-doped nanoparticle specimens from $\text{Bi}_2\text{Te}_{2.9}\text{I}_{0.1}$ displayed electrical resistivity that was smaller than that of un-doped Bi_2Te_3 sample but

Table 1: Lattice parameters of the prepared powders [138]

Sample	Bi_2Te_3	$\text{Ce}_{0.2}\text{Bi}_{1.8}\text{Te}_3$	$\text{Y}_{0.2}\text{Bi}_{1.8}\text{Te}_3$	$\text{Sm}_{0.2}\text{Bi}_{1.8}\text{Te}_3$
$a(\text{\AA})$	4.382	4.387	4.382	4.386
$c(\text{\AA})$	30.485	30.495	30.492	30.496

higher than those of $\text{Bi}_2\text{Te}_{2.95}\text{I}_{0.05}$ and $\text{Bi}_2\text{Te}_{2.8}\text{I}_{0.2}$ samples. It implies that the optimal level of I-doping is 0.1. Indicating that the specimens show n-type conduction, all samples had *–ive* Sb coefficients throughout the observed temperature range. A donor dopant is something like I replacing Te. After a maximum value was reached, continued temperature rise caused a reduction in Sb coefficients.

When n and p kinds of carriers are present, the bipolar effect may occur [166]. The specimen's power factor values are smaller than the un-doped sample's. This is the result of the Sb coefficient and electrical resistivity working together. I-doping reduces the Sb coefficient while increasing electrical resistance. However, in contrast to an un-doped sample, the power factor of the $\text{Bi}_2\text{Te}_{2.9}\text{I}_{0.1}$ the sample is almost unaltered. The thermal conductivity variance between I-doped and un-doped samples is nearly identical. Only the $\text{Bi}_2\text{Te}_{2.9}\text{I}_{0.1}$ sample's value is marginally less than the amount of the un-doped sample. The Wiedemann–Franz equation for near-degenerate or degenerate semiconductors was used to calculate the electronic impact on thermal conductivity [167]. The lattice thermal conductivity has not changed much except for the value of the doped samples ($x = 0.1$) before 498 K. Three potential causes exist for the I-doping samples' continued reduced thermal conductivity. First, just like the un-doped Bi_2Te_3 , the doped nanopowders have a flower-like shape. As a result, some minor grains are amid the bigger ones in the doped bulk, which is ideal for lowering thermal conductivity. Second, the I-doping would not significantly raise the phonon frequency since the respective atomic masses of I (126.9 g) and Te (127.6 g) are so near. Third, changing Te to I will result in additional alloying, which might surge phonon scattering, reducing thermal conductivity. The $\text{Bi}_2\text{Te}_{2.9}\text{I}_{0.1}$ sample, which has a more excellent ZT value over the observed temperature span, can be attributed to a reduction in thermal conductivity and electrical resistivity. In contrast to commercial zone melting ingots, the ZT value of $\text{Bi}_2\text{Te}_{2.9}\text{I}_{0.1}$ sample reaches a value of 1.1 at 448 K [168]. The ZT value of $\text{Bi}_2\text{Te}_{2.9}\text{I}_{0.1}$ is also relatively high for an n-type TE material, especially considering that n-type alloys based on Bi_2Te_3 have recently seen significant improvements in ZT values above 1 [169,170] (Figure 11 and Table 2).

Wu *et al.* [135] investigated ways to increase TE performance by manipulating the carrier concentration by doping. This work used encapsulated melting and hot pressing to create solid solutions of $\text{Bi}_2\text{Te}_{2.85}\text{Se}_{0.15}$:D = I, Ni, Ag, Zn, and

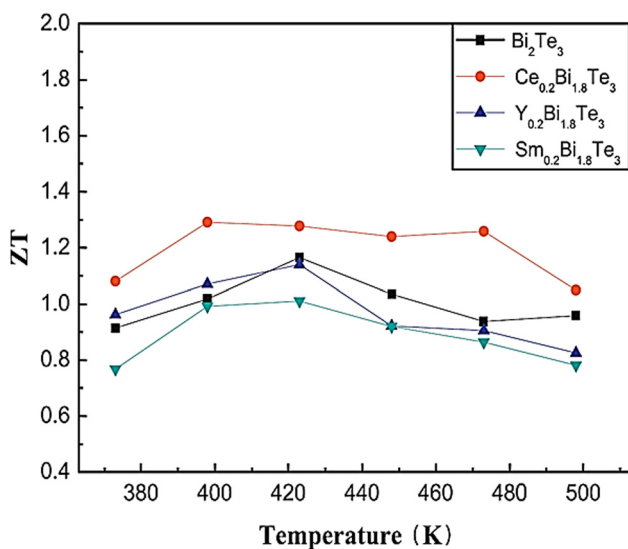


Figure 10: The ZT of $\text{R}_{0.2}\text{Bi}_{1.8}\text{Te}_3$ bulk samples versus reproduced with the permission of [153].

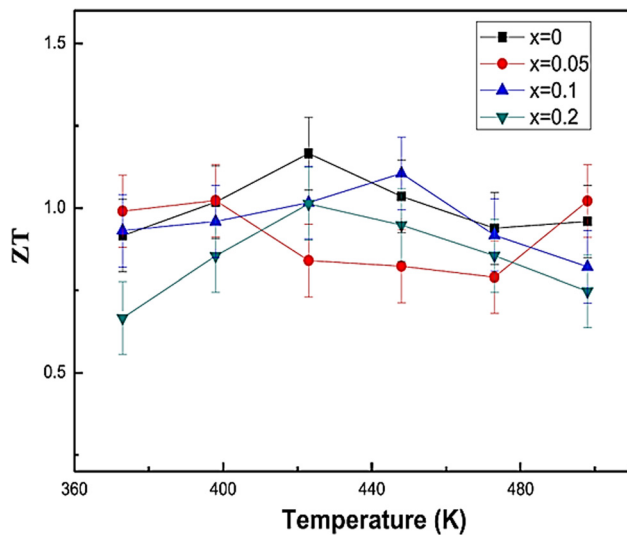


Figure 11: The ZT of $\text{Bi}_2\text{Te}_{3-x}\text{I}_x$ bulk samples versus temperature, reproduced with the permission of Wu et al. [134].

Cu. Table 3 lists the electrical transport characteristics and the greatest figure of merit of $\text{Bi}_2\text{Te}_{2.85}\text{Se}_{0.15}$:Dopants. Halogen element (I, occupies the Te site), which generates electrons, can increase the carrier concentration and decrease mobility. Though the transition elements and other dopants have taken up residence at the Bi site, the reduction in carrier concentration was observed, and mobility was enhanced. The phases and TE characteristics of the samples with the highest ZT for each doping element are listed below.

Table 2: Lattice parameters of as-synthesized powders [2]

Sample	Bi_2Te_3	$\text{Bi}_2\text{Te}_{2.95}\text{I}_{0.05}$	$\text{Bi}_2\text{Te}_{2.9}\text{I}_{0.1}$	$\text{Bi}_2\text{Te}_{2.8}\text{I}_{0.2}$
$a(\text{\AA})$	4.381	4.377	4.373	4.370
$c(\text{\AA})$	30.481	30.435	30.406	30.364

Hot-pressed $\text{Bi}_2\text{Te}_{2.85}\text{Se}_{0.15}$:Dopant XRD patterns were consistent with the literature, and a secondary phase was not discernible. Successful preparation of solid $\text{Bi}_2\text{Te}_{2.85}\text{Se}_{0.15}$:Dopant solutions with excellent crystallinity was achieved. The compositions and lattice constants for a few samples are listed in Table 3. The dopant's quantity was too small; hence, it did not impact diffraction patterns and the constant value of the lattice. The as-synthesized samples were electrical conductivities marginally reduced as the temperature rose, indicating degenerate semiconductor behavior. By acting as an electron donor, I doping boosted electrical conductivity. However, the electrical conductivities of the other dopants were similar to or lower than those of $\text{Bi}_2\text{Te}_{2.85}\text{Se}_{0.15}$, acting as electron acceptors. Samples showed negative Sb coefficients at each of the temperatures used to test them, which is consistent with the Hall coefficients' negative sign and indicates n-type semiconductor properties. The reduction in carrier concentration enhanced absolute values of the Sb coefficient for copper and Argon doping $\text{Bi}_2\text{Te}_{2.85}\text{Se}_{0.15}$ while not affect I-, Ni-, and Zn-doped $\text{Bi}_2\text{Te}_{2.85}\text{Se}_{0.15}$ (Table 4).

When I resides at the Te-site, the electron is produced, increasing the carrier concentration and, thus, the electrical conductivity [171]. This led to an improvement in

Table 3: Electrical transport characteristics and figure of merit of $\text{Bi}_2\text{Te}_{2.85}\text{Se}_{0.15}$:Dopant [135]

Samples	Hall coefficient (cm^3C^{-1})	Mobility ($\text{cm}^2\text{V}^{-1}\text{s}^{-1}$)	Carrier Conc. (cm^{-3})	ZT _{max}
$\text{Bi}_2\text{Te}_{2.85}\text{Se}_{0.15}$	-0.085	92.58	7.37×10^{19}	0.57@99.85°C
$\text{Bi}_2\text{Te}_{2.85}\text{Se}_{0.15}\text{I}_{0.0025}$	-0.076	85.67	9.38×10^{19}	0.76@99.85°C
$\text{Bi}_2\text{Te}_{2.85}\text{Se}_{0.15}\text{I}_{0.005}$	-0.058	92.93	1.07×10^{20}	0.90@149.85°C
$\text{Bi}_2\text{Te}_{2.85}\text{Se}_{0.15}\text{I}_{0.01}$	-0.046	81.84	1.36×10^{20}	0.77@199.85°C
$\text{Bi}_2\text{Te}_{2.85}\text{Se}_{0.15}\text{I}_{0.015}$	-0.029	62.21	2.14×10^{20}	0.71@199.85°C
$\text{Bi}_2\text{Te}_{2.85}\text{Se}_{0.15}\text{I}_{0.03}$	-0.017	42.65	3.63×10^{20}	0.60@259.85°C
$\text{Bi}_2\text{Te}_{2.85}\text{Se}_{0.15}\text{I}_{0.045}$	-0.015	48.60	4.22×10^{20}	0.47@249.85°C
$\text{Bi}_2\text{Te}_{2.85}\text{Se}_{0.15}\text{Cu}_{0.045}$	-0.135	123.94	4.62×10^{19}	0.58@149.85°C
$\text{Bi}_2\text{Te}_{2.85}\text{Se}_{0.15}\text{Cu}_{0.0075}$	-0.131	120.60	4.78×10^{19}	0.64@99.85°C
$\text{Bi}_2\text{Te}_{2.85}\text{Se}_{0.15}\text{Cu}_{0.01}$	-0.175	144.48	3.56×10^{19}	0.76@49.85°C
$\text{Bi}_2\text{Te}_{2.85}\text{Se}_{0.15}\text{Cu}_{0.015}$	-0.190	135.18	3.28×10^{19}	0.71@49.85°C
$\text{Bi}_2\text{Te}_{2.85}\text{Se}_{0.15}\text{Ag}_{0.005}$	-0.176	142.44	3.54×10^{19}	0.65@99.85°C
$\text{Bi}_2\text{Te}_{2.85}\text{Se}_{0.15}\text{Ag}_{0.01}$	-0.171	143.59	3.65×10^{19}	0.75@99.85°C
$\text{Bi}_2\text{Te}_{2.85}\text{Se}_{0.15}\text{Ag}_{0.015}$	-0.213	123.44	2.93×10^{19}	0.68@49.85°C
$\text{Bi}_2\text{Te}_{2.85}\text{Se}_{0.15}\text{Ag}_{0.02}$	-0.245	136.12	2.54×10^{19}	0.68@49.85°C
$\text{Bi}_2\text{Te}_{2.85}\text{Se}_{0.15}\text{Ag}_{0.03}$	-0.320	102.97	1.95×10^{19}	0.40@49.85°C
$\text{Bi}_2\text{Te}_{2.85}\text{Se}_{0.15}\text{Ni}_{0.005}$	-0.083	90.68	7.56×10^{19}	0.56@99.85°C
$\text{Bi}_2\text{Te}_{2.85}\text{Se}_{0.15}\text{Ni}_{0.01}$	-0.279	118.41	2.23×10^{19}	0.32@49.85°C
$\text{Bi}_2\text{Te}_{2.85}\text{Se}_{0.15}\text{Zn}_{0.005}$	-0.084	98.37	7.39×10^{19}	0.57@99.85°C
$\text{Bi}_2\text{Te}_{2.85}\text{Se}_{0.15}\text{Zn}_{0.01}$	-0.176	113.25	3.56×10^{19}	0.51@49.85°C

Table 4: Lattice parameters of the prepared powders [135]

Sample	$a(\text{\AA})$	$c(\text{\AA})$
$\text{Bi}_2\text{Te}_{2.85}\text{Se}_{0.15}$	4.438	3.042
$\text{Bi}_2\text{Te}_{2.85}\text{Se}_{0.15}\text{:I}_{0.005}$	4.437	3.048
$\text{Bi}_2\text{Te}_{2.85}\text{Se}_{0.15}\text{:Cu}_{0.01}$	4.437	3.040
$\text{Bi}_2\text{Te}_{2.85}\text{Se}_{0.15}\text{:Ag}_{0.01}$	4.437	3.040
$\text{Bi}_2\text{Te}_{2.85}\text{Se}_{0.15}\text{:Ni}_{0.005}$	4.438	3.035
$\text{Bi}_2\text{Te}_{2.85}\text{Se}_{0.15}\text{:Zn}_{0.005}$	4.437	3.039

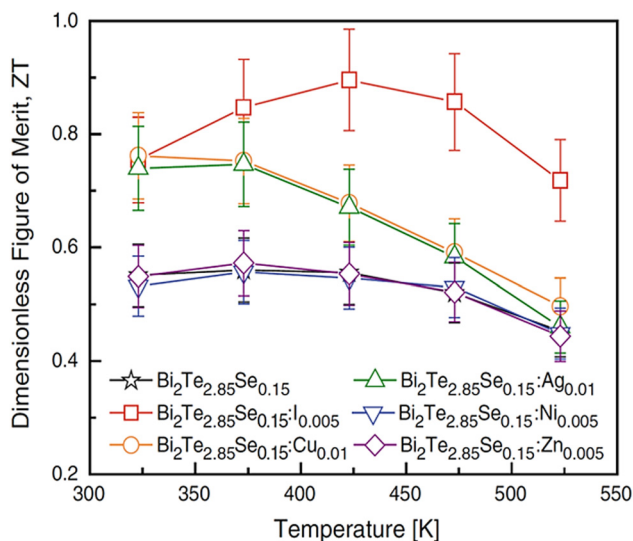
the PF value for the I-doped solution. In contrast, a hole is created when Cu and Ag atoms replace the Bi site, which lowers carrier concentration [172,173]. The Cu doping prevents the development of BiSe antisite flaws, which prevents the free electron concentration from rising. The enhanced power factor from I, Cu, and Ag doping improved the ZT value, having ZT value 0.90 at 423 K for $\text{Bi}_2\text{Te}_{2.85}\text{Se}_{0.15}\text{:Cu}_{0.005}$ peak and 0.76 at approximately 49.85°C for $\text{Bi}_2\text{Te}_{2.85}\text{Se}_{0.15}\text{:Cu}_{0.01}$ was obtained. The improved carrier concentration should increase the ZT value according to the operating temperature (Figure 12).

To improve TE ambient temperature efficiency of n-type polycrystalline, $\text{Bi}_2\text{Te}_{2.85}\text{Se}_{0.15}$ alloys were investigated by Lee *et al.* [137], who reported an integrated optimization approach of Ag doping with hot compression. Bismuth-telluride-based alloys have long been the finest commercial TEM for solid-state refrigeration near ambient temperature. Both traditional zone-melted p- and n-type Bi_2Te_3 -based alloys display $ZT \sim 1$ near the 22°C ambient temperature. However, because of their weak mechanical qualities, the

construction of devices often fails. In comparison to zone-melted equivalents, which have received a lot of focus recently, polycrystalline Bi_2Te_3 -based alloys prepared by metallurgical processing methods offer better mechanical properties and a lower lattice contribution. To attain a high peak $ZT > 1.2$ and room temperature $ZT > 1.0$ in p-type (Bi, Sb) $_2\text{Te}_3$ alloys, nano-structuring methodologies, including the “bottom-up” approach [174–180] and the “top-down” approach [181–183], have been utilized. The lowered ZT may be due to significantly reduced carrier mobility μH following nano-structuring in n-type $\text{Bi}_2(\text{Te, Se})_3$ alloys. Recent years have seen the effective application of the heat deformation (HD) method, which has been widely used in metallic alloys [184,185], to increase the μH of polycrystalline n-type Bi_2Te_3 -based alloys. By repressing the $\text{Bi}_2\text{Te}_{2.7}\text{Se}_{0.3}$ alloy, Hou *et al.* [186] found enhancement in the peak of ZT from 0.85 to 1.04 at 400K. Yan *et al.* got a ZT 1.06 at 400 K in the repressed $\text{Cu}_{0.01}\text{Bi}_2\text{Te}_{2.7}\text{Se}_{0.3}$ sample [187]. Lee *et al.* [137] studied typically $\text{Ag}_x\text{Bi}_2\text{Te}_{2.7}\text{Se}_{0.3}$ samples. After heated deformation, it is clear that the (00l) diffraction intensity rises in the HD samples, showing a favored alignment. The orientation factor F of the (00l) plane is determined using the Lotgering technique [188] to explore the texture degree further. By combining Ag doping and heat distortion, the service temperature of n-type polycrystalline Bi_2Te_3 -based alloys is lowered from around 177–22°C. The power factor increases, but the electrical thermal conductivity close to 22°C decreases due to Ag doping’s optimized carrier concentration. Due to shaping, the hot deformation procedure makes the carrier more mobile. The doubly hot, deformed $\text{Ag}_{0.011}\text{Bi}_2\text{Te}_{2.7}\text{Se}_{0.3}$, an alloy ore exhibits a ZT value of approximately 1.0 at 26.85°C and, and a peak ZT value of 1.1 at 76.85°C, which is encouraging for actual usage in refrigeration near ambient temperature (Figure 13).

Wu *et al.* [138] studied how $\text{Bi}_2\text{Te}_{3-x}\text{Se}_x$ nano-platelet composites made using the polyol technique have improved TE properties. Although the computed lattice parameters nearly adhere to Vegard’s rule, two-mode behavior for $\text{Bi}_2\text{Te}_{3-x}\text{Se}_x$ nano-platelet are demonstrated by a break in shifting the high-frequency phonon modes. SPS pellet composites exhibit semiconducting behavior for intermediate Se compositions and metallic conduction for Bi_2Te_3 nano-platelet composites in terms of electrical resistivity. In all fabricated nano-platelet composites, the thermal conductivity is substantially lower than the bulk numbers and is mainly caused by scattering from microstructural grain boundaries. In compared to Bi_2Te_3 nano-platelet composites, the TE power of S (259 $\text{V}\cdot\text{K}^{-1}$) and ZT is 0.54, approximately four times higher for $\text{Bi}_2\text{Te}_{2.7}\text{Se}_{0.3}$ SPS pellets (Figure 14).

Tentatively, this improvement in TE properties in nano-platelet composites is accredited to extensive grain

**Figure 12:** Figure of merit temperature dependency, reproduced with the permission of Wu *et al.* [135].

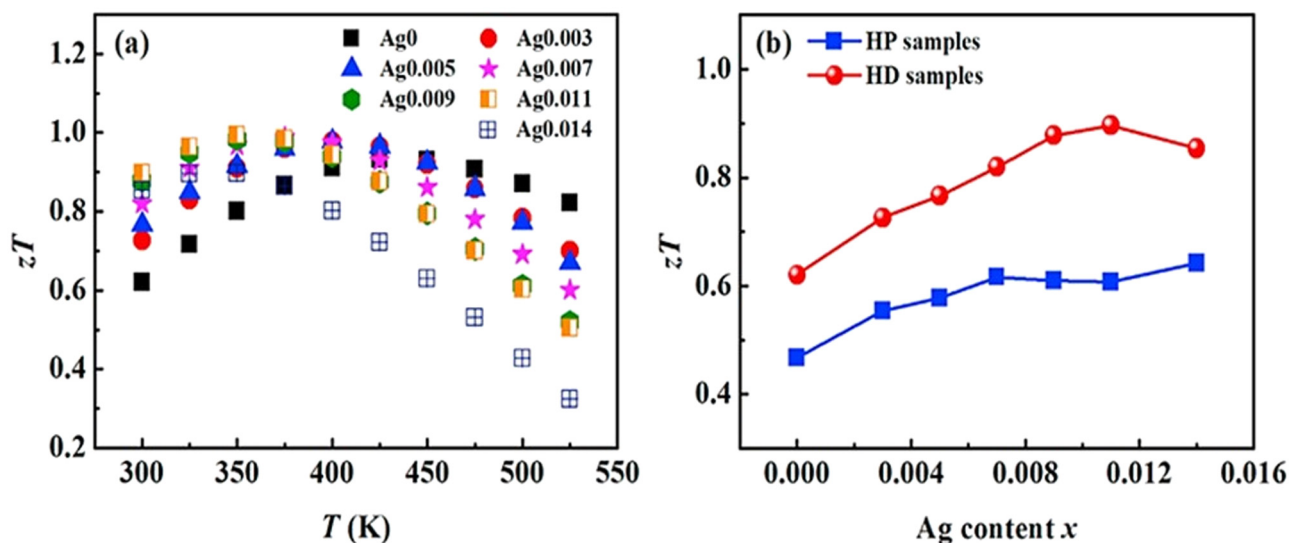


Figure 13: (a) The figures of merit (ZT) of $\text{Ag}_x\text{Bi}_2\text{Te}_{2.7}\text{Se}_{0.3}$ samples versus temperature. (b) ZT versus Ag content, reproduced with the permission of [137].

boundaries in aligned nano-composites that filter low-energy electrons. In a study by Soni *et al.* [139], Pb-doped nano-crystalline n-type Bi_2Te_3 thin films were deposited by radiofrequency magnetron sputtering. The element Pb is suitable for adjusting the carrier concentration of bulk Bi_2Te_3 and doping Pb atoms into the crystal lattice of the material increases the concentration of holes [189,190]. Because typical n-type Bi_2Te_3 films have a high carrier concentration, ZT enhancement is constrained [191–194]. Pb doping reduced the carrier concentration of n-type Bi_2Te_3 films and increase TE characteristics of the Bi_2Te_3 film.

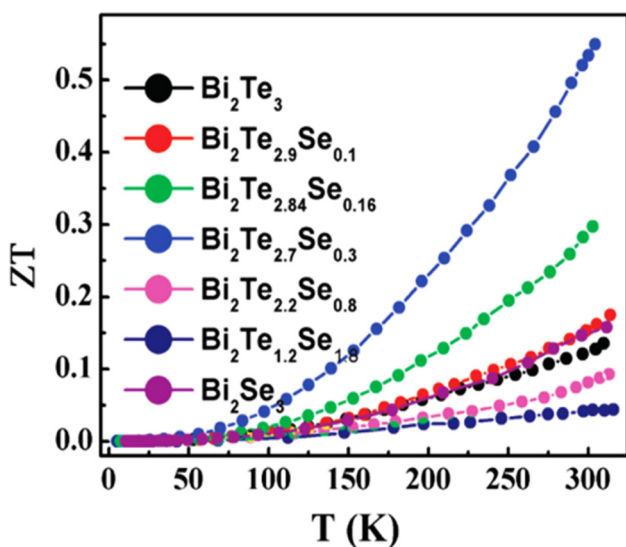


Figure 14: The ZT of $\text{Bi}_2\text{Te}_{3-x}\text{Se}_x$ samples with temperature, reproduced with the permission of Wu *et al.* [138].

Pb doping allows for precise control of the Bi_2Te_3 films' carrier levels, which have a compelling effect on the transport properties of thin film. The Sb coefficient of Bi_2Te_3 films are highest, and thermal conductivity was lowest at Pb doping concentration. The PF values of 2.50 and 2.15 $\text{mW}\cdot\text{K}^{-2}\cdot\text{m}^{-1}$ were achieved at 200°C, approximately for unannealed and annealed films doped with 0.38% Pb, respectively. The results showed that Pb doping was a workable method for improving the TE characteristics of n-type Bi_2Te_3 films. A potential material for TE micro devices is the Pb-doped Bi_2Te_3 sheet [140]. The HD process was successfully used by Wu *et al.* [141] to create $\text{Tl}_x\text{Bi}_{2-x}\text{Te}_{3-x}\text{I}_x$ nano-powders that were similar to flowers. After that, heat pressing was used to compress the created nanoparticles into bulks. The bulk samples of $\text{Tl}_x\text{Bi}_{2-x}\text{Te}_{3-x}\text{I}_x$ were examined, and their TE characteristics were reviewed. The outcomes show that the effects of Tl doping were greater than those of I doping onto Bi_2Te_3 on electrical resistivity and Sb coefficients of Bi_2Te_3 . The co-doped bulks' PF was consequently lower than the bulk Bi_2Te_3 . The $\text{Tl}_x\text{Bi}_{2-x}\text{Te}_{3-x}\text{I}_x$ bulk samples' thermal conductivities continue to be lower. The results showed that at 398 K, the ZT value of the doped bulk $\text{Tl}_{0.1}\text{Bi}_{1.9}\text{Te}_{2.9}\text{I}_{0.1}$ reaches the value of 1.1 [140] (Figure 15).

New groups of surface materials that have time reversal protection by time-reversal symmetry are named topological insulators [195]. Consequently, the delocalized surface state is unchanged by nonmagnetic dopants and defects. The unusual features of TIs, such as the anomalous quantum Hall effect [195–203], hold great promise for the development of quantum computing. It was demonstrated using ARPS in Bi_2Te_3 and $\text{Bi}_2\text{Te}_3\text{Tl}_3$ that quantum magneto-transport phenomena, like weak anti-localization [204–206], Aharonov-Bohm oscillations [207], and quantum conductance fluctuations

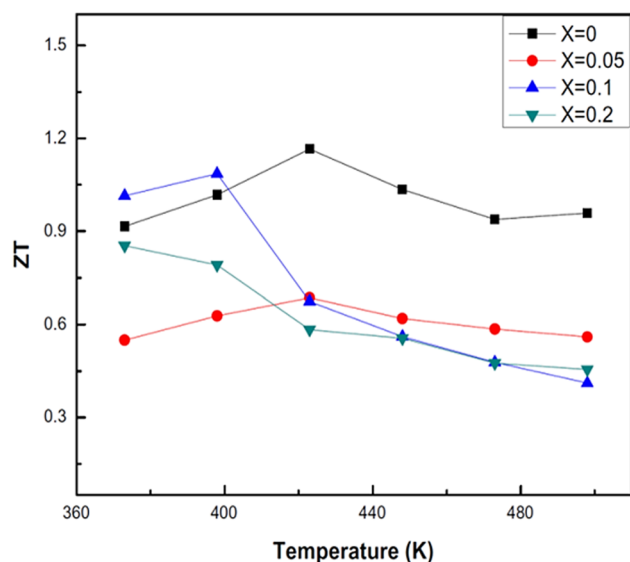


Figure 15: The ZT of $\text{Tl}_x\text{Bi}_{2-x}\text{Te}_{3-x}\text{I}_x$ samples versus temperature, reproduced with the permission of Zhou *et al.* [140].

[208], are associated with topological surface states. Furthermore, strong TE properties in these materials are crucial because TE devices are considered a viable source of power generation and energy conservation. In these devices, n- and p-type materials are coupled in series. $\text{Bi}_{2-x}\text{Zn}_x\text{Te}_3$ single crystals that were created utilizing the modified Bridgman technique were investigated by Singh *et al.* [142]. He studied Zn-doped Bi_2Te_3 topological insulator's electrical resistivity, TE power, magneto-transport, and magnetism. Higher Zn content improves electrical conductivity, and calculated carrier mobility from Hall effect approaches, a phenomenal value of $7.2 \times 10^3 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$. In high-mobility specimens, a significant positive magnetoresistance was seen. It is notably shown that higher Zn-doped Bi_2Te_3 samples break the link between electrical conductivity and the Sb coefficient, significantly improving the TE power factor. Singh *et al.* [142] investigated the synthesis of bismuth telluride doped with the rare earth element gadolinium in two distinct configurations, 0.0 and 0.1. XRD was employed for structural investigation, which covered crystal structure, phase purity, crystallite size, and lattice characteristics. The rhombohedral structure of Bi_2Te_3 was revealed by the structural investigation. N-type $\text{Bi}_2\text{Te}_{2.7}\text{Se}_{0.3}$ thin films containing Pt nano-inclusions were effectively made using pulsed laser deposition, according to Arif *et al.* and Sun *et al.* [143,144].

The grain borders of these semiconductor matrices include embedded Pt nano-inclusions. The PF of a nano-composite thin film may be significantly increased by adding Pt nano-inclusions thanks to the energy-filtering outcome. At the same time, the thermal conductivity was decreased via scattering long-wavelength phonons. The PF

reaches $3.5 \times 10^{-3} \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-2}$ at approximately 22°C with a ZT value of 1.17 thanks to the Pt content optimization. This preparation technique helps the production of anisotropic materials for TE and other renewable energy solutions, as well as the nano-engineering of textures and semiconductors. By gathering information on phase equilibrium from various thermally equilibrated ternary alloys and the three constituent binary systems, Wu *et al.* [145] studied the 523 K isothermal section of the Bi-Ge-Te system (Figure 16). In particular, the ternary system emphasizes the compositional homogeneity, phase relations, and thermal stability within the range restricted by BiTe , GeTe , and Te phases. At 523 K, the thermal stability of four ternary compounds, $\text{Bi}_2\text{Ge}_3\text{Te}_6$, $\text{Bi}_{1.5}\text{Ge}_{1.5}\text{Te}_5$, $\text{Bi}_2\text{Ge}_1\text{Te}_4$, and $\text{Bi}_4\text{Ge}_1\text{Te}_7$, is about along the $\text{Bi}_2\text{Te}_3 - \text{GeTe}$. When the nominal compositions fulfill specific requirements, $\text{Bi}_{40-y}\text{Ge}_x\text{Te}_{60-x+y}$ alloys ($x = 0 \rightarrow 12.5$, $y = 0 \rightarrow 10$) were formed by Bridgman technique, and massive carrier transits from p-type holes for an un-doped stoichiometric sample to n-type electrons. Te must be less than 60.0 or 60.0 at.% and Ge/Bi greater than or equal to 0.2.

Among the alloy p-type (P2), the peak value of ZT is at 0.9 at 325 K, and in the case of n-type (N5), the alloy peak value of ZT is at 0.45 at 525 K, which traces in a $\text{Bi}_2\text{Te}_3 + \text{Bi}_2\text{Ge}_1\text{Te}_4 + \text{Bi}_4\text{Ge}_1\text{Te}_7$ three-phase region (Figures 17 and 18).

The recently established one-atom thick graphite, graphene (GR) [209], is crucial for understanding physics and real-world applications. Due to its extremely high mobility, it showed high electrical conductivity at approximately 22°C . A small amount of GR inserted into the Bi_2Te_3 matrix may be able to play a significant role in inhibiting phonon

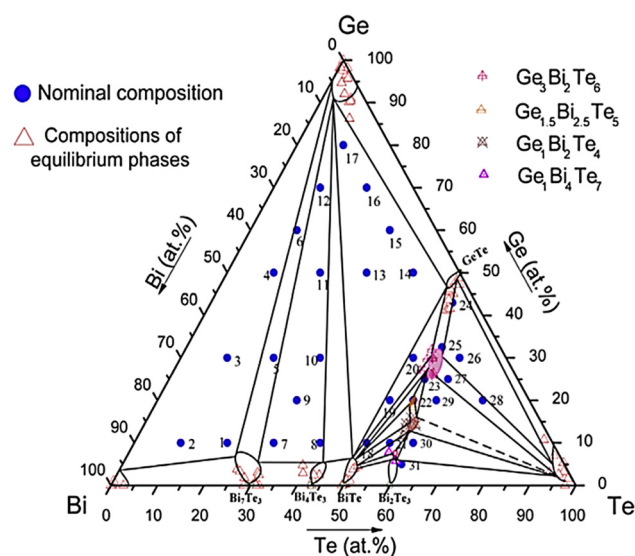


Figure 16: Ternary Bi-Ge-Te system, reproduced with the permission of Wu *et al.* [145].

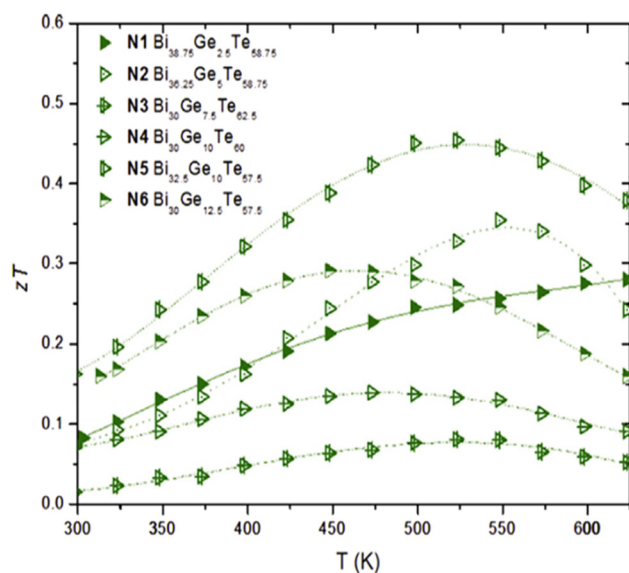


Figure 17: The ZT of $\text{Bi}_{40-y}\text{Ge}_x\text{Te}_{60+x+y}$ with temperature, reproduced with the permission of Wu *et al.* [145].

transport while retaining the excellent electrical conductivity of the Bi_2Te_3 lattice. In this research, both single crystals of Bi_2Te_3 and Bi_2Te_3 doped with GR have had their electrical structure and TE characteristics examined. Wu *et al.* [145] used CZ technique to create single crystals of Bi_2Te_3 . Bi_2Te_3 polycrystalline samples with GR doping were created utilizing a solid-state reaction. The crystals were made using Bi and Te powders. The thermal power is approximately $190 \text{ V}\cdot\text{K}^{-1}$, same as for Bi_2Te_3 [210]; resistivity was approximately $2.2 \text{ m}\Omega\cdot\text{cm}$ and reduces monotonically with declining

temperature for the Bi_2Te_3 crystals. The thermal conductivity of Bi_2Te_3 crystals is approximately $25 \text{ mW}\cdot\text{cm}^{-1}\cdot\text{K}^{-1}$ and gradually rises with a declining temperature, then decreases for 80 K. At room temperature, ZT is around 0.3, which is consistent with what has been observed for ordinary Bi_2Te_3 (Figure 19).

Li *et al.* [146] examined the TE characteristics of two series of GR/ Bi_2Te_3 composites that were made using a straightforward wet-chemical synthesis approach and sintering procedure. By ultrasonically dispersing a well-dispersed solution and vacuum filtering it, the GR sheets were evenly spread through the Bi_2Te_3 matrices. The most significant values were reached with a GR loading of 0.5 wt%. The power factors of the two types of GR/ Bi_2Te_3 composites primarily rose with the growing GR content. They subsequently fell as the amount of GR in the mixture grew. Instead of the lower Sb coefficient values brought on by the reduction in n, this phenomenon was linked to the higher Sb coefficient values of the samples. A little addition of GR (up to 3 weight percent) was found to lower the thermal conductivity value because the GR surfaces produced many boundaries with the Bi_2Te_3 matrices, which served as efficient phonon scattering centers (Figure 20).

The GR/ Bi_2Te_3 -NW composite (1 wt%) had a maximum ZT value of 0.4 at approximately 26.85°C , which is higher than the Bi_2Te_3 NWs and GR/ Bi_2Te_3 -powder composites due to the enhanced power factor and lower k values. So, to summarize, our work has shown the advantages of integrating 1D Bi_2Te_3 NWs with GR as a possible means of developing highly effective TEMs (Figure 21).

Li *et al.* [146] found that the impact of monolayer GR's (G) presence on the conductivities of Bi_2Te_3 at the nano-

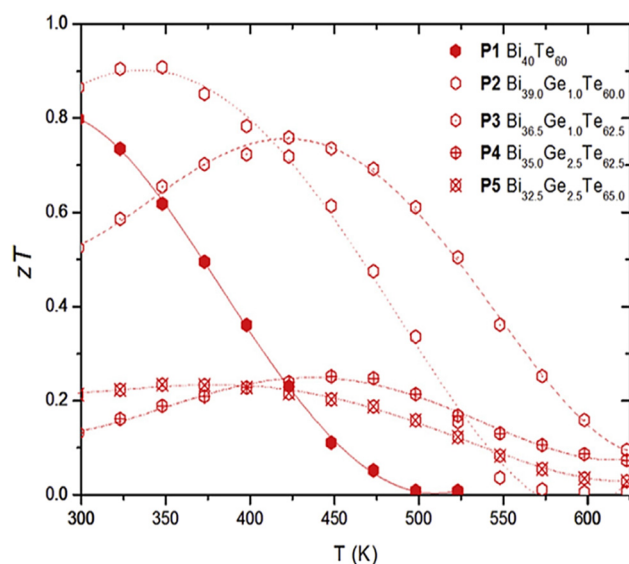


Figure 18: The ZT of $\text{Bi}_{40-y}\text{Ge}_x\text{Te}_{60+x+y}$ with temperature, reproduced with the permission of Wu *et al.* [145].

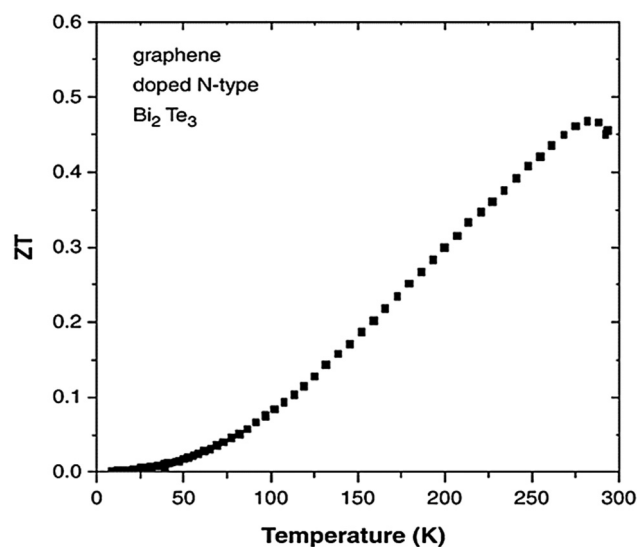


Figure 19: The ZT of Bi_2Te_3 :Gr samples, reproduced with the permission of [146].

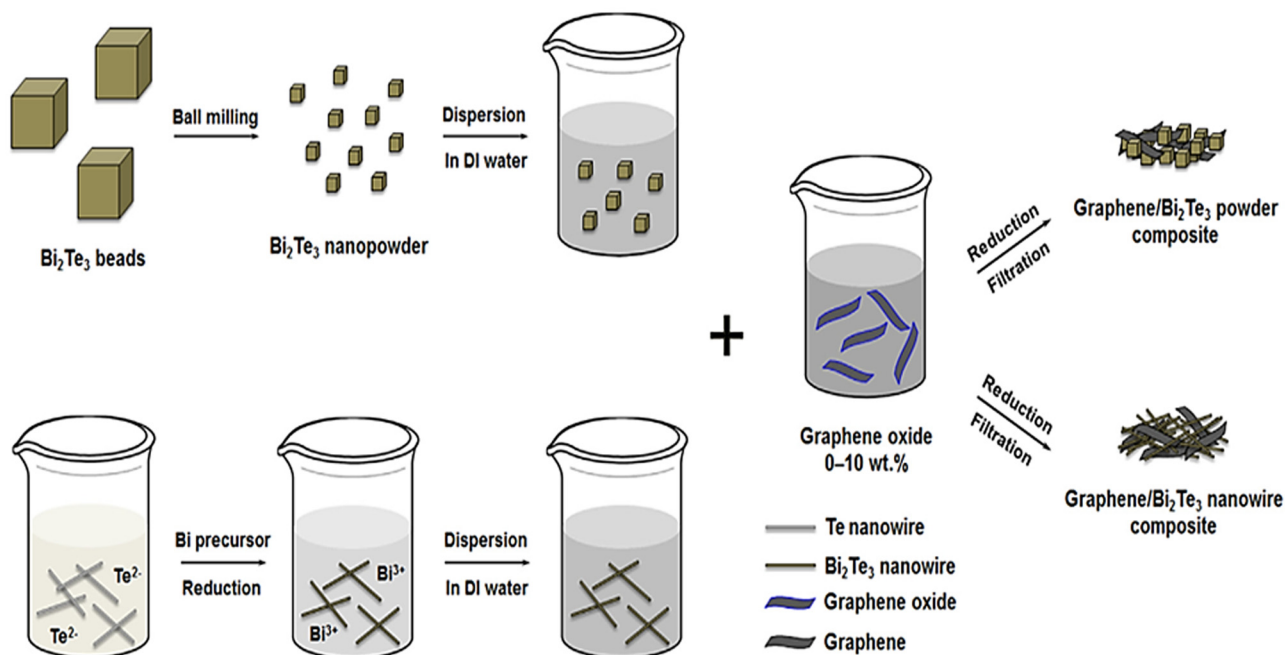


Figure 20: Diagram of the two separate GR/Bi₂Te₃ composites' production process, reproduced with the permission of Li *et al.* [146].

scale. Studies using conductive atomic force microscopy demonstrate that the addition of G causes electrical conductivity at Bi₂Te₃:G contacts to decrease. The thermal conductivity and work function values of Bi₂Te₃ composites are different from those of the Bi₂Te₃ sample, as shown by the Kelvin probe and scanning thermal images. By testing the TE characteristics of Bi₂Te₃ and composites (temperature varies from 300 to 480 K), the reduction in thermal conductivity was further supported. The Bi₂Te₃:G

composite sample's simultaneous rise in PF and reduction of thermal conductivity resulted in an improved ZT = 0.92 value. Due to the huge boundary area and two-dimensional behavior of G, the enhanced scattering of phonon and imperfect impact on electron transport are ascribed to the rise in ZT value. To establish the function of 2D materials in enhancing the TE properties, this work sheds light

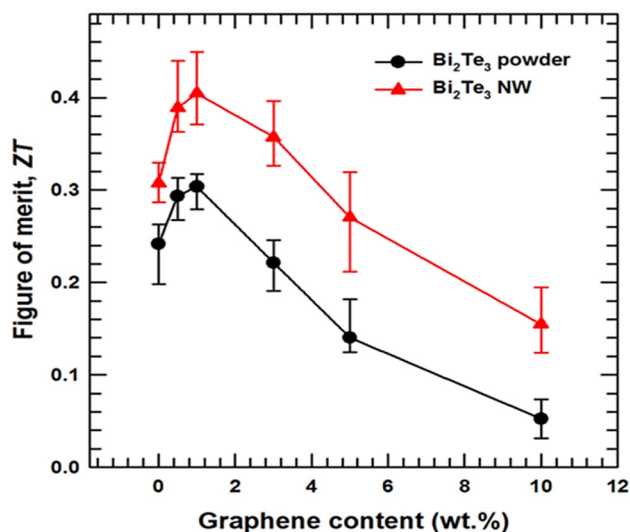


Figure 21: The ZT of Bi₂Te₃:Gr samples versus varied GR content, reproduced with the permission of Li *et al.* [146].

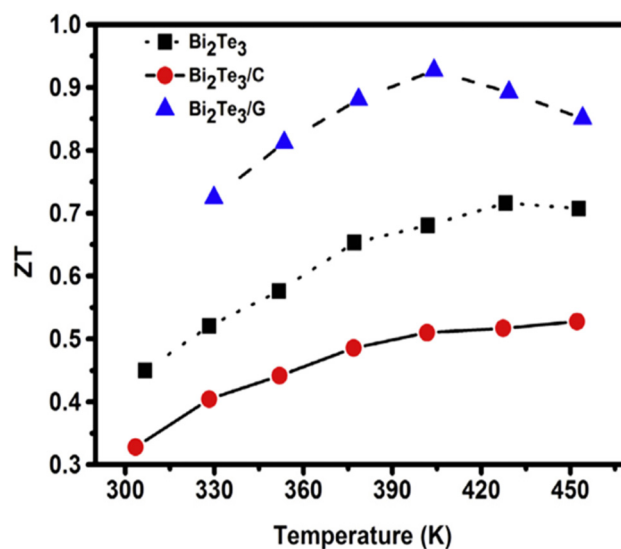


Figure 22: The ZT of Bi₂Te₃, Bi₂Te₃/C, and Bi₂Te₃/G samples versus varied temperatures, reproduced with the permission of Ju and Kim [147].

on a novel approach for straight nano-scale measurements of nano-composite samples (Figure 22).

5 Printable thermoelectric materials

Printing provides a possible means of producing TEMs at a cheaper cost and permits the creation of generators specifically designed to satisfy the needs of waste heat sources. The printing of TEMs has recently increased in terms of TE material and printing techniques, as well as performance with TEMs that are made commercially. According to Figure 23, at the proper temperatures, a ZT of 1 can produce efficiencies at the same level as those of other renewable technologies [211]. While renewable energy sources like wind and solar power are widely used in countries aiming to achieve carbon neutrality, TEs are only used in specialized fields like watches, space [212], or Peltier coolers [130]. It has been crucial from a technological standpoint to optimize the shape of TEGs composed of inorganic materials to increase their efficiency. The thermal-to-electrical conversion efficiencies are still far below the maximum Carnot efficiency, even if the TE characteristics of inorganic materials and the efficiency of their associated TEG have significantly increased. In addition, the conventional inorganic TE materials previously discussed have processability problems and are costly due to the presence of hazardous and rare

elements. This may be somewhat explained by considering the high cost of producing TE devices, which goes toward the \$0.80 price tag for TE generators [213,214] as opposed to \$0.089 and \$0.084 for photovoltaic and wind turbines, respectively (price is per kW·h) [215].

The cost of producing TEG must be decreased, as must the price per kW·h of energy harvested if TEs are widely used. Reducing the price of producing TEMs is one technique to lower the cost of heat-to-electricity conversion. To create TEMs for modern marketable devices, spark plasma sintering, hot pressing, or a mixture of the two processes was used. These processes need expensive equipment, high temperatures, high pressures, and lengthy manufacturing periods. Contrarily, printing can be done in any environment, at high manufacturing rates, and with inexpensive tools. In recent years, printed thermoelectric has made substantial progress. Screen, inkjet, dispenser, and, more recently, 3D/pseudo-3D printing are some methods that have been researched (Figure 24). While not strictly categorized as a printing method, spin coating is frequently used in laboratories to measure the viability of ink mixtures before applying these inks in multiple printing methods. The center of a substrate that is stationary or rotating slowly has the ink formulation used on it. The substrate is programmed to turn rapidly once the ink has been put on it. Several spin velocities may be employed with different acceleration rates. When the procedure is followed correctly, consistent films are produced. This method's drawbacks include the need for a flat substrate, and only thin films are made. Traditionally used to create commercially printed items like posters and textiles, screen printing (Figure 23b) has recently gained new applications in the manufacture of printed circuit boards (PCBs) and other electrical goods [216–220]. Screen-printed electrodes are frequently employed in many industrial applications, including displays, radio frequency identification, solar cells, and sensors.

In traditional screen printing, a patterned stencil is affixed to the mesh, and a woven or stainless-steel mesh screen mask is used. Photolithography is used to create this stencil using a photo emulsion layer. An electrode design is deposited on the substrate through a stencil aperture, and conductive ink dispersed on the screen mask is squeezed into the woven or stainless-steel mesh. Ink is transferred to a substrate during the screen printing process, except for regions of the web that are impervious because of a hindering stencil. To fill the open holes on the screen with ink, a squeegee or blade is passed over it. After that, an opposite stroke lets the screen briefly trace the substrate along the contact line. As a result, the screen retracts when the blade has passed, causing the ink to moisten the substrate

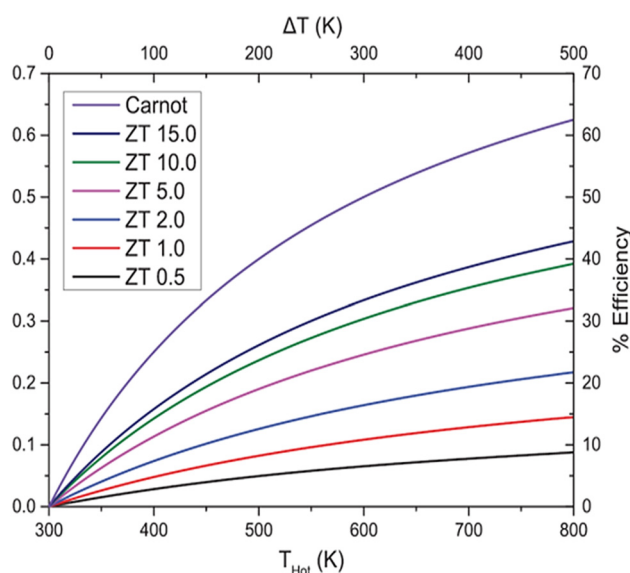


Figure 23: The ZT of different technologies, reproduced with the permission of Department for Business Energy & Industrial Strategy, Electricity Generation Costs [221].

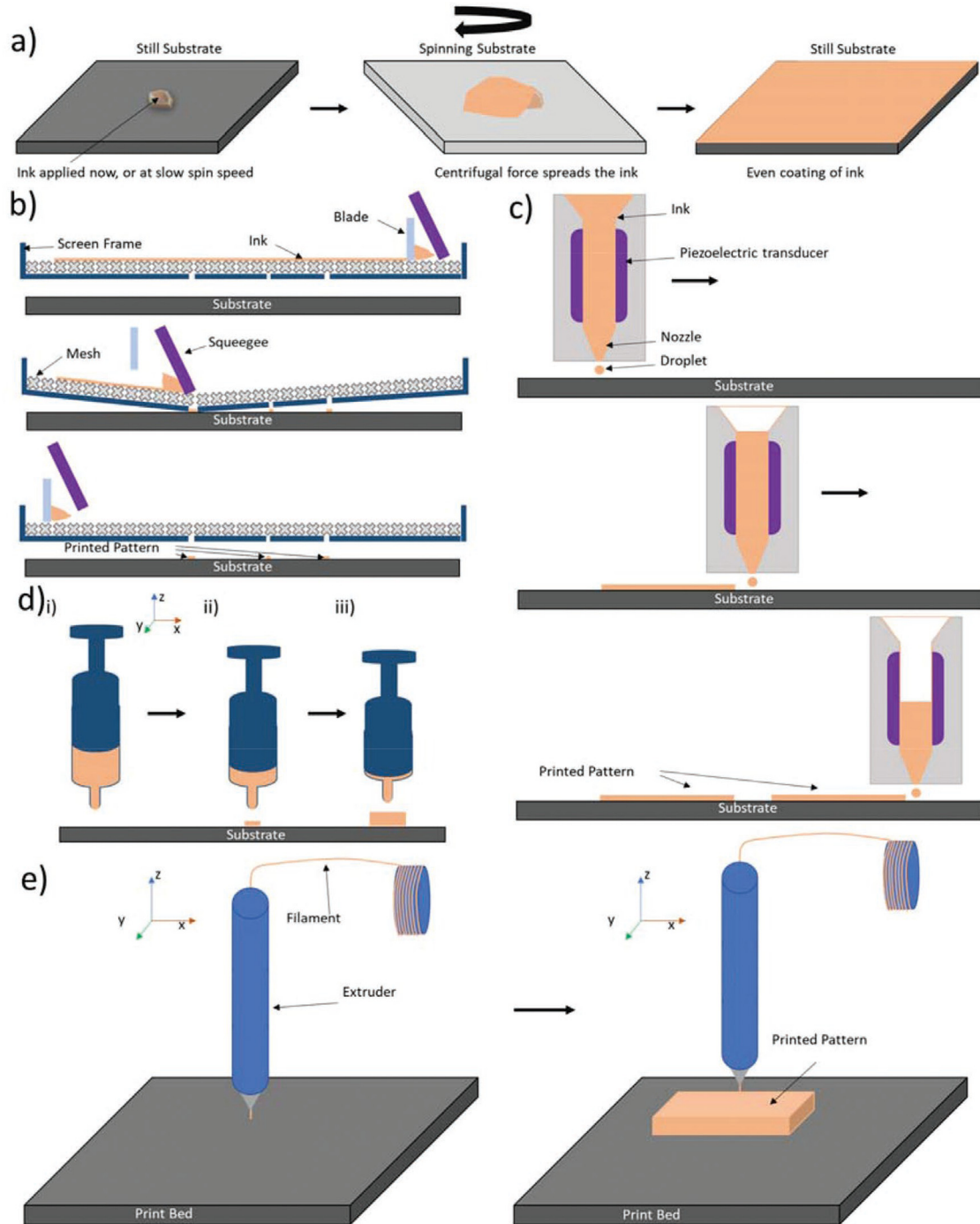


Figure 24: (a) Spin coating, (b) screen, (c) inkjet, (d) dispenser printing, and (e) fuse deposition modeling, reproduced with the permission of Department for Business Energy & Industrial Strategy, Electricity Generation Costs [221].

and be drawn out through mesh apertures. Various widths can be printed from a few microns to several hundred microns. Any level solid substrate can be utilized, and typical examples include polyimide for thermoelectric applications and aluminum, glass, and other common materials. Another constraint is the drying method, which might cause

contraction and surface irregularity, which may result in errors or imperfections in the ultimate product. If complicated pieces are required, the restriction on feature size may cause problems in the top product. Even though the inks can have various viscosities, more excellent viscosity inks are more commonly employed than those used in other printing

procedures [222,223]. Due to oxidation of the surface of films and the fact that the TE particles are not in liquid form, films made using this technique often exhibit reduced electrical conductivity, making electrical paths more challenging to cross [224,225]. The inkjet printing process entails the construction of an image on a substrate drop by drop while being electronically operated. One of the most promising techniques for the simple, low-cost production of high-resolution functional materials is inkjet printing technology. The liquid droplet transfer of ink material to a substrate is crucial to inkjet printing. As a result, there is no need for mechanical contact between the print head and the substrate [226]. The most common kind is drop-on-demand (DOD), with continuous inkjet printing as an alternative.

A voltage-activated piezoelectric diaphragm pushes ink into a chamber in a DOD system. When the diaphragm contracts due to a voltage change, there is a higher pressure inside the diaphragm than in the printing chamber. This pressure difference forces ink into the chamber. To print more ink on the subsequent initiation of the piezoelectric diaphragm, the capacity of the diaphragm diminishes when the voltage is reversed to let in air. There is minimal ink waste since ink droplets are only created when necessary. However, if the ink particle size is close to the nozzle diameter in inkjet printing, the printer head is prone to blockage and disfigurement [227]. If a bigger spout diameter is used, the image quality might be decreased [228]. Thus, research into these variables is essential to the development of functional material inkjet printing. Some applications require creating narrow, smooth structures with various geometries, such as integrated circuits and optical waveguides. Increasing printing resolution is a complex undertaking that necessitates an additional technological advancement related to the resource-intensive substrate pretreatment.

5.1 Printed TEDs

The n- and p-type components are linked in series in alternating patterns in the fundamental construction of TEDs such that the voltage created by each component sums up [229]. The output demand and the application determine how many elements are needed. Kim made a flexible TE generator on glass textiles using the screen-printing method. They bent the device to study its adaptability and showed that it maintained a constant efficiency even after 120 iterations [230]. With eight n-type Bi_2Te_3 and p-type Sb_2Te_3 thermocouples, they were able to fabricate printed films with a high power density at the temperature of 50 K and an open-circuit voltage of 0.09 V. They emphasized

how lightweight their product was. In addition, the device's output power on glass fabric was more significant than it was on Al_2O_3 substrates because the fabric has a small thermal resistance. Rösch *et al.* created a TE generator using screen printing, folding, and TE inks of PEDOT nanowires for the p-type and TiS_2 for n-type material for legs. They achieved a $47.8 \text{ W}\cdot\text{cm}^{-2}$ power output at a 30 K temperature differential [231].

The item has 254 p and 253 n-type legs with an area of approximately 44.2854 mm^2 for each leg, arranged in a checkerboard design with a 39 cross-13 matrix. In their research, they employed a folding technique that allowed for a minimal spacing among the TE legs, or effectively the substrate thickness of 0.06 m, to act as an insulating barrier between the TE components. They attained a high thermocouple density of around 190 cm^2 , which led to a high open-circuit voltage. Sb_2Te_3 and Bi_2Te_3 nanoparticles were used together with the screen-printing technique by Rösch *et al.* [232] to create a TEG, which was then infiltrated with PEDOT:PSS in DMSO at a 5 vol% concentration. DMSO aids in raising the materials. They showed that because printed n- and p-type TE legs have distinct thermal and electrical characteristics, it affects how well the TEDs work. When n- and p-type TE's cross-sectional area ratio was 3, they attained a peak output power density of $1.2 \times 10^3 \text{ W}\cdot\text{cm}^{-2}$ at 50°C ΔT with a hot surface temperature of approximately 59.85°C . Here, the flexible substrate for the printed TEG was a polyimide film with seven thermocouples, and the electrode was a screen-printed Ag film. With the help of body heat, their 15 TE couple-equipped device produced 0.0121 V voltage. In a $5 \text{ cm} \times 5 \text{ cm}$ screen-printed TEG, We *et al.* [233] attained a peak output power of 0.2 kW at a ΔT of 43°C .

To create the TEG, they combined a hybrid Bi_2Te_3 -polyimide ink with an ionic liquid. The hybrid $\text{Bi}_2\text{Te}_3/\text{PVA}$ composite was chemically treated by Kato *et al.* [234] to create a screen-printed TEG with 10 stripes, each 2 mm and 25 mm wide. The displayed power output of TEG is up to $9 \text{ W}\cdot\text{cm}^{-2}$ at a ΔT of 46°C . When there was a 50 K temperature differential between a printed TEG's hot and cold sides, Pires *et al.* [235] showed a power density of $2.7 \times 10^3 \text{ W}\cdot\text{cm}^{-2}$. The TEG was created using $\text{Bi}_2\text{Te}_{2.7}\text{Se}_{0.3}$ films that were aerosol jet printed. They also performed experiments using tellurium-based nano solder and ball-milled $\text{Bi}_{0.4}\text{Sb}_{1.6}\text{Te}_3$ NPs. With ΔT of 80°C , the screen-printed device with four legs provided a significantly enhanced power density of $1.88 \times 10^3 \text{ W}\cdot\text{cm}^{-2}$. To create their device, Saeidi *et al.* also employed Bi_2Te_3 ink with a 12-weight percent Te-based nano-solder [236]. It had screen printing and was $10 \text{ mm} \times 5 \text{ mm} \times 166 \text{ m}$. At a ΔT of 105°C , they got a maximum output power density of $3.26 \times 10^3 \text{ W}\cdot\text{cm}^{-2}$. To create a

printed TEG, Varghese *et al.* [237] combined reduced-graphene oxide (RGO) nano-sheets with Bi_2Te_3 nano-plates as ink. When the relative concentration of RGO in the ink was 1%, the greatest power production of 1.72 W at ΔT 20°C was observed. Any increase or decrease in RGO had a negative impact on power output. The power output of a printed TEG at a temperature differential of 20°C with a power density of $75 \text{ W}\cdot\text{cm}^{-2}$ was reported by Ding *et al.* [238], who created TEGs at a millimeter size. On a polyimide substrate, they used dispenser printing to realize a TEG using Sb_2Te_3 and Bi_2Te_3 inks. They used n-type Bi_2Te_3 -epoxy composite films with 1% Se and 62 single-legged printed TEG prototypes to produce a greater output of $130 \text{ W}\cdot\text{cm}^{-2}$ at the same ΔT [239].

Madan *et al.* [240] created an n-type Bi_2Te_3 ink and through screen-printing techniques made a TEG. With a ΔT 60°C, a power output density of $4,100 \text{ W}\cdot\text{cm}^{-2}$ was obtained. The TEG comprised 0.5 nm n-type legs with a cross-sectional area of 0.02 mm^2 . When 0.02 A of applied current was used, Varghese *et al.* [241] developed a cooling device and found a peak ΔT of 1.4°C. They produced a 6.2°C differential between the hot and cold sides when the current was raised to 60 mA. The highest result was 17.9°C at 180 A current. They discover that the Joule heating effect brought on by the device's internal resistance significantly impacts the temperature differential between the two

different temperature sides. The Peltier effect predominates when the applied current is small, and the cold side temperature drops due to the joule and Fourier effect, however, caused more heat to be produced when a high current was supplied. As a result, the cold side's temperature began to rise, and no net temperature of the cold surface was seen. Table 5 presents some of the TEMs and schemes used for printing.

6 Summary and perspectives

Despite significant progress in printed TEMs over the past 10 years, the effectiveness of printed TEDs still falls short of the capabilities of the materials they are made from. High electrical and thermal contact resistances primarily cause low conversion efficiency at the device level. In addition, most TE inks fall short in printability and mechanical properties, even though a few reports demonstrate good results [242,243]. One of the main problems is simultaneously improving the printed materials' mechanical and TE characteristics. Another obstacle to overcome is to make the preparation process simpler. Although its morphology is poor, solid-state processing at high temperatures enables a well-controlled chemical composition [244]. In addition,

Table 5: Summary of the power outputs of printed TEG based on Te

Materials	Printing method	Substrate	Thickness (mm)	Power (μW)	Power density ($\text{mW}\cdot\text{cm}^{-2}$)	Cure T (K)
$\text{Sb}_2\text{Te}_3/\text{Bi}_{1.8}\text{Te}_{3.2}$	Screen	Kapton	0.067	0.048	180	523
$\text{Sb}_2\text{Te}_3/\text{Bi}_2\text{Te}_3$	Screen	Glass Fabric	0.5	—	—	803
$\text{Sb}_2\text{Te}_3/\text{BiTe}$ (epoxy)	Screen	Polyimide	0.078	0.44	180	523
$\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3/\text{Bi}_2\text{Te}_{2.75}\text{Se}_{0.25}$ with epoxy	Dispenser	Free standing	4.4	0.126	—	—
$\text{BiSbTe}/\text{BiSeTe}$	Screen	a-Si quartz/ SiO_2	0.65	76,480	—	—
$\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3/\text{Bi}$ with epoxy	Dispenser	Glass	—	130	720	523
$\text{Sb}_2\text{Te}_3/\text{Bi}_2\text{Te}_3$ (epoxy)	Dispenser	Polyimide	0.12	0.85	—	473
$\text{Sb}_2\text{Te}_3/\text{Bi}_2\text{Te}_3$ with epoxy	Dispenser	Polyimide/glass	0.2	10.5	—	523
Bi_2Te_3 (PVA)	Shadow Mask	PET	0.265	—	120	353
Bi_2Te_3 with epoxy	Dispenser	Polyimide	0.1–0.12	25	720	623
BiTeSe with aтерpineol and Disperbyk – 110	Screen	Polyimide	0.1	0.004	45	703
$\text{Sb}_{1.5}\text{Bi}_{0.5}\text{Te}_3/\text{Bi}_2\text{Te}_{2.7}\text{Se}_{0.3}$	Inkjet	Glass/Polyimide	150 layers	341	673	673
$\text{Bi}_{0.55}\text{Sb}_{1.45}\text{Te}_3/\text{Bi}_2\text{Te}_{2.7}$ with Sb_2Te_4	3D	N/A	0.35	2.8	723	723
$\text{Bi}_{0.4}\text{Sb}_{1.6}\text{Te}_3/\text{Bi}_2\text{Te}_{2.7}\text{Se}_{0.3}$ with Sb_2Te_3	3D	N/A	1.5–2.0	1,620	723	723
Se doped Bi_2Te_3 with epoxy	Dispenser	Polyimide	0.2	1.6	720	523
$\text{Pb}_{0.98}\text{Na}_{0.02}\text{Te}/\text{Pb}_{0.98}\text{Sb}_{0.02}\text{Te}$	3D	N/A	2	216,300	3,150	373–1,023
$\text{Bi}_{0.4}\text{Sb}_{1.6}\text{Te}_3/\text{Bi}_2\text{Te}_{2.7}\text{Se}_{0.3}$	Laser	$\text{Bi}_{0.4}\text{Sb}_{1.6}\text{Te}_3$	1.6	1,450,000	1,440	673
$\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3/\text{Bi}_2\text{Te}_3$	Inkjet	Polyimide	0.0015	0.127	723	723
$\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3/\text{Bi}$ with epoxy	Dispenser	Glass	—	130	720	523

higher temperatures cause the polymer substrates to break down. To increase the conversion efficiency of printed TED's, further in-depth research on device designs, electrical contacts, and thermal contacts is needed. New types of high-efficiency printed TEMs may result from further study. Using fresh, efficient manufacturing techniques might enhance the performance and robustness of the TEDs.

In the past few years, it has been discovered that the photonic curing [245] procedure is an exceptional sintering method that enables excellent efficiency and mechanical flexibility. The procedure minimizes the possibility of oxidation because it only needs a few multiples of 10^{-3} s printed TEM's sintering. The shape of the TEDs also affects the conversion efficiency since it regulates the devices' electrical and thermal impedance [246]. Another crucial area of research is the impact of shape on device efficiency. One of the potential production techniques for ink creation is the solution-based wet chemistry approach, which enhances the ink's surface chemistry [247]. Significant findings from TE inks [248,249] and printed systems [250–254] made with various concentrations and recipes have been published recently. However, further study on the subject is required to overcome the challenges. While Te helps to improve TE characteristics, some of the commonly used dopants are very harmful to the environment. For the bulk manufacture of the TEDs, substitutes like p-type and n-type [255] might be created. Given the potential for printed TEs in various applications, the field will likely see much exploration in the coming years.

To make the TE technology feasible and affordable, a shift in thinking from traditional bulk to printable TEs has been widely noted in recent years. In addition, bulk TEs do not provide shape conformity, which is necessary for numerous applications on irregular surfaces. Shape-conformable TEDs might be produced at a reasonable cost thanks to the printed TEs. To begin with, organic polymers have been the focus of efforts to transform them into powerful printed TEMs. Their low TE efficiency, however, restricts their use despite their good printing ability. Inorganic-based printed TEs have recently made advancements, as detailed in this article. Inorganic materials that can be printed provide a way around these restrictions. We have outlined the basic principles of TEs, the composition of TE ink, and current developments in inorganic and hybrid printed TE materials. Inorganic TEM materials have been the focus of most studies on printed TEs. However, other chalcogenides have also demonstrated promising abilities. However, improving TE efficiency by hybridizing organic and inorganic materials could be fruitful. There are several reasons for the competitiveness of TEDs compared with other renewable energy resources, including efficiency in

waste heat recovery, low maintenance and long lifespan, scalability and flexibility, reliability and durability, low environment impact, diversified energy sources, technological innovation, and cost reduction. These findings might open the door for the production of low-cost TEDs.

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Data availability statement: The datasets generated and/or analyzed during the current study are available from the corresponding author on reasonable request.

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