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Low Temperature Inorganic Thermoelectric Telluride P-type Materials: A Review

Renewable energy sources based on thermoelectric materials (TE) will be essential for solving the power crises and increasing global demand in the future. These materials have many advantages to use in power generation, cooling and heating and can generate usable electricity from different waste heat sources working at different temperature ranges. Our focus is on p-type low temperature inorganic Thermoelectric Telluride (TET) material, suitable near the room temperature. This review starts with a fundamental understanding of thermoelectric materials, their synthesis methodologies and advance strategies for enhancing the performance. Low temperature chalcogenide-based telluride TE materials play a very essential role and provide foundation along with promises for future applications. Finally, we summaries challenges and future prospective for inorganic TE materials.

Keywords: thermoelectric materials, telluride thermoelectric materials, low temperature thermoelectricity, renewable energy.

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Introduction

Growing global population and rapid development of economy requires larger global demand for sustainable and clean energy sources for human life and industrial production which are highly obstructed by the increasing price of electricity [1]. According to the international energy outlook of 2019 the US energy information administration predicts that energy consumption will rise 50 % from 2018 to 2050. In conventional fossil fuel (coal, natural gas, oil and petroleum) during combustion, just 35 % of the released energy is used effectively and rest is lost as waste heat in the environment [2, 3]. To address these challenge, renewable energy sources such as wind, solar and Thermoelectric (TE) material will be essential for solving the power crises in the future. These materials have the advantages of being noiseless, free of vibration, simple, solid-state device, long life, no moving part, light weight, maintenance free, zero greenhouse gas emission, environment friendly, making them use in power generation, cooling and heating etc. [4-5].

Concept of thermoelectric effect is based on three fundamental principles: the Seebeck effect (discovered in 1821 by German scientist Thomas Johann Seebeck), the Peltier effect (discovered in 1834 by French scientist Jean Charles Athanase Peltier) and the Thomson effect observed in 19th century and consequently their application to thermometry, power generation and refrigeration (cooling electronic devices). The temperature gradient induced electric current is termed as thermoelectricity [6]. Commercial applications are limited due to low efficiency of about 5 % to 20 %, relative cost with conventional power generation and other conversion technologies.

TE performance is based upon conversion efficiency of the material for converting heat into electricity. It depends on the dimensionless key parameter of TE materials termed as figure of merit often written as "ZT" [11]. ZT can be defined as

$$ZT = S^2\sigma T/\kappa$$

Where S is the Seebeck coefficient, T is the absolute temperature, σ is electrical conductivity and κ is thermal conductivity. For an ideal TE material, ZT should be high. For this reason, one must have high S for large thermo-voltage, high σ to reduce the joule heating effect and low κ for large temperature difference. The evolution of high-quality TE materials requires an insight understanding of the electron transport phenomena. To improve energy conversion efficiency various strategies such as optimum carrier concentration, reducing k by nanostructuring and mass defect engineering are suggested [12-13].

Up to now different inorganic TE materials have been studied in detail. Since 1950s and mid-1990s research in the area of TE materials has concentrated on improving TE performance of inorganic materials such as Bi-Te and Pb-Te based alloys [14-16]. In 1950s study of chalcogenide semiconductor especially tellurides, like Bi_2Te_3 and PbTe, chalcogenide glass semiconductor attracted strong interest because of large Seebeck coefficient and small κ and variable σ values.

TE materials are classified into three main categories based on their working temperatures: low temperature 273 K to 500 K (room temperature properties provide a useful baseline), medium temperature 500 K to 900 K & high temperature 900 K to1300 K TE materials.

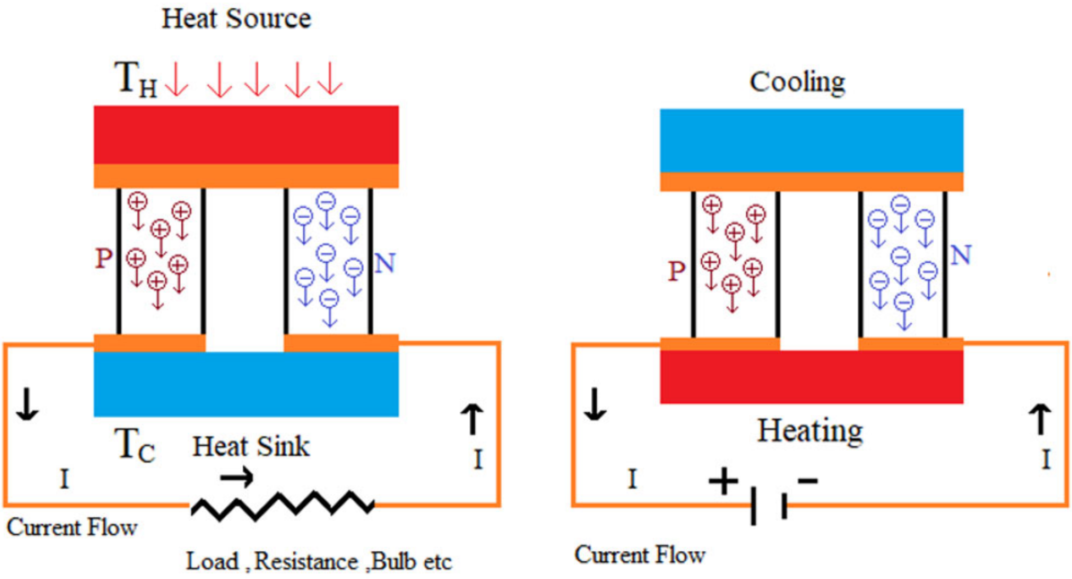

Fig. 1.1. (a) Seebeck Effect (b) Peltier Effect

1.1. Low Temperature Thermoelectric Materials (LTTEM)

The family of TE material those are suitable for working temperature less than 500K exist in this category. It is also called near room temperature TE materials, such as Bismuth based alloy [17], Bismuth chalcogenides (Bi_2Se_3, Bi_2Te_3, Bi_2S_3), Antimony chalcogenides, Sb_2Se_3, Sb_2Te_3, Sb_2S_3.

1.2. Medium Temperature Thermoelectric Materials (MTTEM)

The family of TE material those are suitable for working temperature between 500K to 900K exist in this category. Such as skutterudite TiO_2 [18], Cu_2Se based [19-23], Tin chalcogenide (SnSe, SbTe, SnS) [24-29], Lead chalcogenide (PbTe, PbSe, PbS [30-34], GeTe based [35], Indium chalcogenide (InSe, InTe, InS), Germanium chalcogenide (GeTe, GeSe, GeS) [36-41].

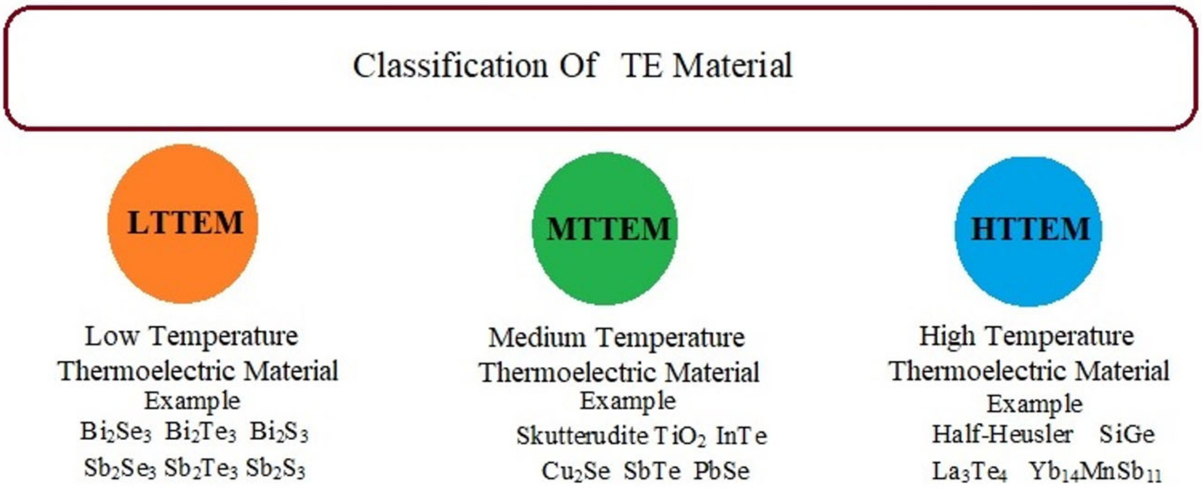

Fig. 1.2. Classification of TE materials

1.3. High Temperature Thermoelectric Materials (HTTEM)

This category involves the family of TE material which are suitable for working at temperatures greater than 900K, such as SiGe, La_3Te_4 [42], $Yb_{14}MnSb_{11}$, half-Heusler [43].

TE material can generate usable renewable electricity from different wasted heat sources working at different temperature range. About 70% of the energy produced in the world, is dissipated into environment in the form of waste heat. Waste heat distribution on the basis of temperature [44] is shown in fig. 1.3, in which 63% is dissipated at low temperature.

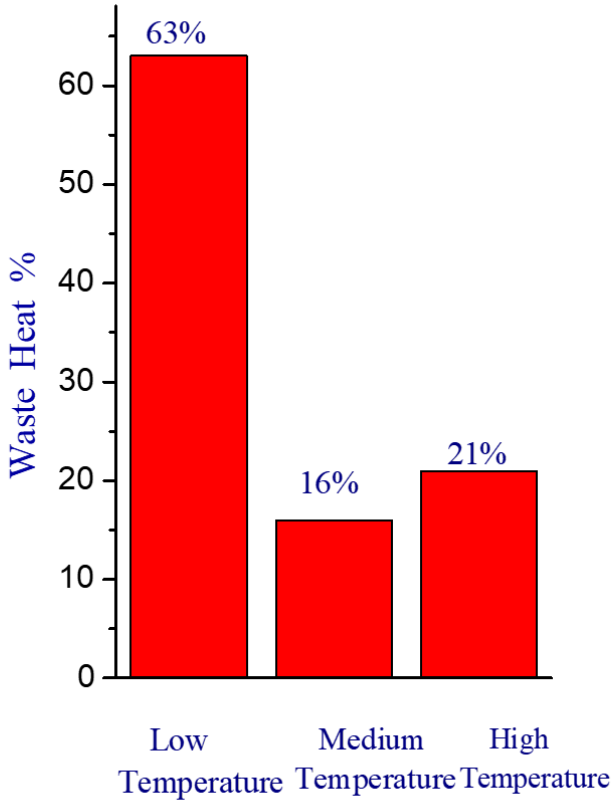

Fig. 1.3. Waste heat distribution on the basis of temperature

Thermoelectricity is the way to convert this waste heat into sustainable and usable energy at low cost. Twenty first century industries consume energy for different manufacturing process plants in which over 60% of total energy loses as waste heat [45, 46] and this waste heat can easily be collected and re-used in industries in the range of less than 573K [47] through conventional energy conversion technologies [48].

Present review, discusses characterization, properties, performance at low temperature and near the room temperature to develop a better understanding for modifying and improving various parameters involved and variation of parameter of wide range of inorganic TE materials in p-type categories of telluride families. Finally, we summaries problems and time ahead prospective for inorganic TE materials. This review also highlights the relationship, similarities and difference of current materials.

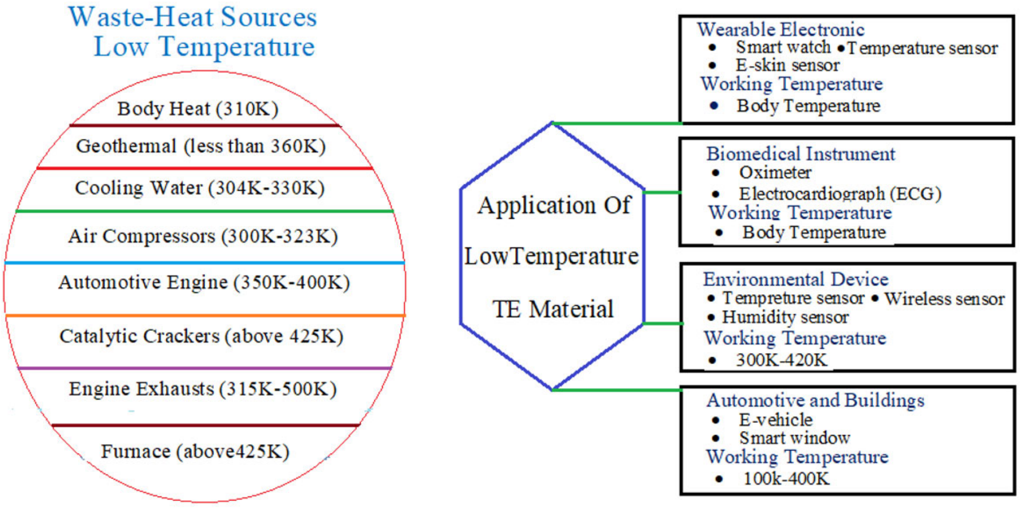

Fig. 1.4. Waste-Heat Sources and application of low temperature TE material

2. P-type Low Temperature Telluride TE Materials: Telluride material such as bismuth telluride Bi_2Te_3 were established as the first commercial TE material and operate at low temperature and peak performance near 380 K due to theses material is most suitable thermoelectric at low temperature. Feature of low temperature dependent telluride p-type early reach very clear peak after which ZT decreases with increasing temperature such property shows operating temperature range of most suitable as thermoelectric. So average ZT can be more important in determining performances at one temperature range Mostly n-type material shows higher ZT value than p-type materials

2.1. Bi Based P-type Low Temperature Telluride TE materials

In 1954 first Proposed that Bi_2Te_3 is low temperature thermoelectric material and crystallizes in to hexahonal layered crystal structure with 5 atomic layers Te^1-Bi-Te^2-Bi-Te^1. It has low melting point of 585 °C (858K) and can operate up to 227 °C (500 K). It is an indirect band gap semiconductor (Eg ~ 0.15), highly degenerate band structure and low effective mass result high Seebeck coefficient. Bulk Bi_2Te_3 prepared by zone melting spinning and SPS process by Tang et al. and they found decrease in κ and increase in σ when rolling speed is increases and maximum Seebeck coefficient (234 μV/K), σ (1100 S/cm), PF (49.43 $\mu W/cm\ K^2$), figure of merit ZT (1.35) and minimum κ (1.09 W/mK) was obtained at room temperature [49]. Poudel et al. synthesizing an alloy composed of Bi and Sb that find this alloying process yields an σ (1250 S/cm), PF (42.78 $\mu W/cm\ K^2$) and ZT is 1.4 [50]. Nanostructuring process for preparation of p-type Bi_2Te_3 system by Xie et al .and result ZT ~ 1.5 and low lattice thermal conductivity $\kappa_{lattice}$ ~.8 W/mK [51]. A p-type $Bi_{0.5}Sb_{1.5}Te_3$ nano grain architecture with grain size 2.-50 nm yields a ZT ~ 1.36 at 87 °C due to which much decrees in κ (~0.9W/mk). Nanocomposite composed of $(Bi_2Te_3)_{0.2}(Sb_2Te_3)_{0.8}$ by ball-Milling distribute with amorphous SiO_2 nanoparticles of ~ 50 nm size were synthesizing by SPS which results ZT of 1.12 and 1.27 at 30 °C (303 K) and 90° C (363 K) and increases power factor due to decrees in thermal conductivity because of scattering of phonon at the phase boundaries and nano particles.

Table 1

Physical and Thermoelectric Properties of Bi based p-type Low Temperature Telluride

	Material	Seebeck coefficient S μV/K	Electrical conductivity σ S/cm	Thermal conductivity κ W/mK	Figure of merit, ZT	Temperature T K	Ref	Year
1	Bi_2Te_3 (ZM)	227	940	1.81	0.8	300K	49	2007
2	Bi_2Te_3 (ZM)	234	650	1.97	0.65	360K	49	2007
3	Bi_2Te_3 (ZM)	225	550	2.5	0.45	420K	49	2007
4	Bi_2Te_3 (SPS)	212	1100	1.09	1.35	300K	49	2007
5	Bi_2Te_3 (SPS)	222	800	1.09	1.3	360K	49	2007
6	Bi_2Te_3 (SPS)	220	620	1.2	1.05	420K	49	2007
7	$Bi_{0.5}Sb_{1.5}Te_3$(NC-BM+HP)	185	1250	1.12	1.2	300K	50	2008
8	$Bi_{0.5}Sb_{1.5}Te_3$(NC-BM+HP)	212	750	1.04	1.4	400K	50	2008
9	$Bi_{0.5}Sb_{1.5}Te_3$(NC+BM+HP)	201	490	1.27	0.8	500K	50	2008
10	Bi_2Te_3(MS+SPS)	225	625	0.8	1.5	390K	51	2010
11	5%Bi_2Te_3-$Ge_{0.87}Pb_{0.13}Te$	130	1087	1.05	0.66	373K	62	2014
12	$Bi_{0.5}Sb_{1.5}Te_3$	241	647	0.33*	1.86	320K	54	2015
13	$Bi_{0.3}Sb_{1.625}In_{0.075}Te_3$	185	890	0.61*	0.75	300K	63	2016
14	$Bi_{0.3}Sb_{1.625}In_{0.075}Te_3$	220	618	1.09	1.4	500K	63	2016
15	$Bi_{0.5}Sb_{1.495}Cu_{0.005}Te_3$	154	1371	0.34*	0.97	300K	64	2016
16	$Bi_{0.5}Sb_{1.495}Cu_{0.005}Te_3$	201	610	0.31*	1.4	450K	64	2016
17	$Bi_{0.05}Sb_{0.1}TeGe_{0.85}$	130	805	0.48*	0.47	300K	65	2017
18	$Sn_{0.97}Bi_{0.03}Te$-3%PbTe	27	4847	1.15*		300K	66	2017
19	$Bi_{0.5}Sb_{1.5}Te_3$	237	600	0.65*	1.24	350K	67	2018
20	$Bi_{0.46}Sb_{1.54}Te_{3.015}Zn_{0.015}$	185	1220	0.52*	1.14	300K	68	2018
21	$Bi_{0.46}Sb_{1.54}Te_{3.015}Zn_{0.015}$	208	850	0.5*	1.4	373K	68	2018
22	$Bi_{0.4}Sb_{1.6}Te_3$	232	778	1.03	1.38	323K	69	2019
23	$(GeTe)_{0.8}(AgBiSe_2)_{0.2}$	242	124	0.28*	0.63	300K	70	2019
24	$(GeTe)_{0.8}(AgBiSe_2)_{0.2}$	279	150	0.32*	1.3	467K	70	2019
25	$BiCuO_{0.88}Te$	185	336	0.59*	0.48	323K	70	2020
26	$Bi_{.04}Sb_{1.6}Te_{3.01}$	195.37	1145.9			300K	55	2021
27	$Bi_2Te_{2.8}Se_{0.2}$ (ZM)	205		1.5	1.05	410K	61	2021
28	$Bi_{0.3}Sb_{1.7}Te_{3.01}$ (MA+YSZ+HP)	227	981	1.17	1.30	300K	56	2022
29	$Bi_{1.99}Pb_{0.01}Te_3$	157	931		0.42	300K	60	2023
30	$BI_{0.50}Sb_{1.50}Te_3$	261		1.32			58	2024
31	$Bi_{0.3}Sb_{1.7}Te_3$ (HP-HE)	197	1260	1.45	1.1	300K	57	2024
32	$Bi_{0.3}Sb_{1.7}Te_3$(ZM)	208	1160	1.56		300K	57	2024
33	$Bi_{0.3}Sb_{1.7}Te_3$ (HP)	199	1050	1.37		300K	57	2024
34	$Bi_{.5}Sb_{1.5}Te_3$+0.10%wt Graphene	190	983	.01186	0.89	300K	59	2024

ZM=Zone Melting, NC= Nano Crystalline, BM=Ball Milling, MS=Melt Spinning * is κ_L

Development of coherent interface between SiC nano inclusion and $Bi_{0.3}Sb_{1.7}Te_3$ matrix enhance Seebeck coefficient, mechanical properties and reduced thermal conductivity due increase in phonon scattering [52]. Doping of rare earth materials (Ce, Y and Sm) in $R_{0.2}Bi_{1.8}Se_{0.3}Te_{2.3}$ achieving a ZT of 1.21 at 140 °C (413 K) for $Y_{0.2}Bi_{1.8}Se_{0.3}Te_{2.7}$. Melt-centrifugation effectively control the microstructure in (Bi, Sb)$_2$ Te_3 with micro scale dislocation array and porous network results superior TE performance than ZM and HP [53]. Liquid phase sintering is used to develop low energy grain boundary with dense dislocation array this result scattering of mid frequency phonon without simultaneously decreases in the charge carrier velocity and record high thermoelectric p type ZT value of 1.86 at 320 K in $(Bi,Sb)_2Te_3$ alloy [54]. For large scale fabrication of p-type $Bi_{0.4}Sb_{1.6}Te_{3.01}$ material with Te self-compensation by Ball milling that result high average ZT value of 1.39 from 30 to 200 °C and peak value 1.55 at 100 °C, low thermal conductivity 0.48 $Wm^{-1}K^{1}$[55]

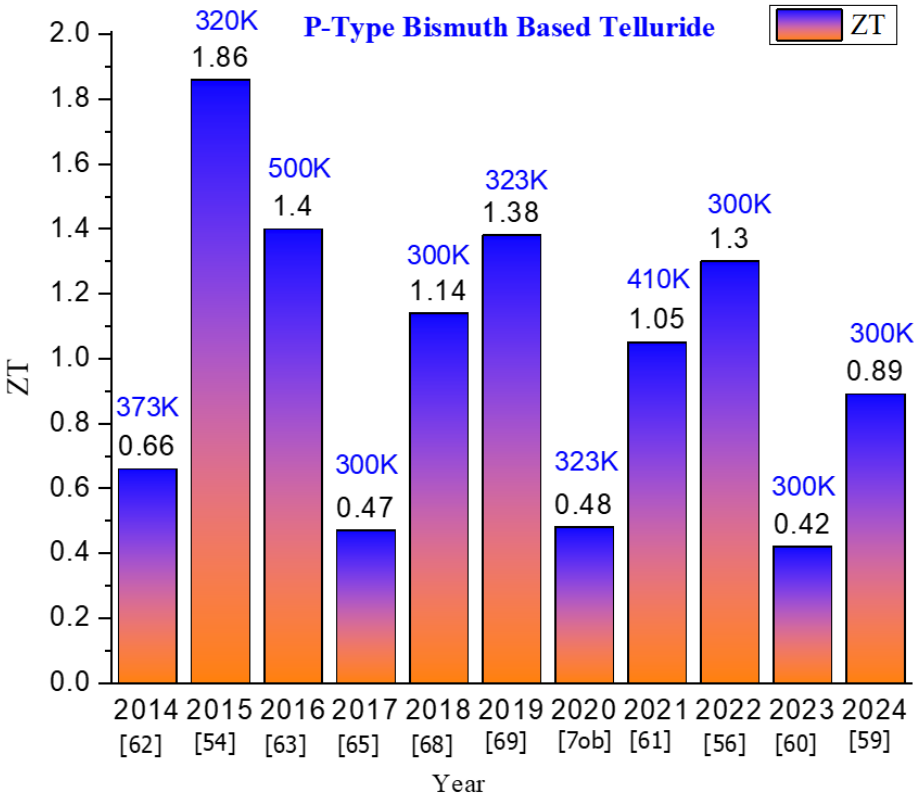

Fig. 2.1. ZT of different Bismuth based p type TET at different temperature and year

TE properties of the $Bi_{0.3}Sb_{1.7}Te_{3.01}$ sintered bulk formed by MA with YSZ milling media followed by HP result high thermoelectric performance at room temperature ($S = 227 \mu V/K$, $\sigma = 9.81 \times 10^4 S/m$, $\kappa = 1.17 W/mK$) [56]. P-type Bi_2Te_3 Samples prepared Hot extrusion (HE) technique can achieve quantitative control over grain size, grain boundaries, inherent point defect and dislocation that regulate thermoelectric performance and gives a low thermal conductivity 0.73 W/mK at 300 K, practical high $ZT \sim 1.1$ at room temperature [57]. $BI_{0.50}Sb_{1.50}Te_3$ alloy prepared by a melting-quenching combined with the SPS process this high Sb doping content enhanced point defect scattering result increase long wave phonon scattering that reduced thermal lattice conductivity [58]. Nanostructured P type $Bi_{0.5}Sb_{1.5}Te_3$ composite obtained by SPS from a powder prepared by melt spinning with addition of 0.10 wt. % of grapheme plates that increase it ZT [59]. Pb doping in Bi_2Te_3 can change carrier type n-type to p-type with high ZT [60].

2.2. Pb Based p-type Low Temperature Telluride TE materials

Te based thermoelectric material PbTe have been used for the past 50 year and shows low κ of 2.3 W/mK and high Seebeck coefficient 500 μV/K at room temperature and its band gap 0.32 eV. Te rich PbTe is a p-type semiconductor**.** PbTe crystalline in the cubic rock salt structure at ambient temperature it has both band converses and resonant level formation strategies is firstly demonstrated in PbTe. resonant level formation in PbTe due to TI doping results in highest ZT ~ 1.5 in 2008 and band merging in $PbTe_{1-x}$: Na results in highest ZT ~ 1.8 in 2011. Another successful approach based on phonon scattering modification namely all scale hierarchical architecture was first proposed in PbTe resulting highest ZT due reducing thermal conductivity. Dopant atom introduce electronic state in PbTe that modifying the electronic density of state and increase Seebeck coefficient from ~ 220 μV/K undoped PbTe to ~ 290 μV/K doped PbTe at 427 °C [71]. More than one element doping in TE materials increase power factor of 16.66 μW/cm K^2, thermal conductivity 1.8 W/mK and ZT (0.27) is attain as for $Pb_{0.91}Cr_{0.009}Te$ co-doped I at 300 K.

Performance of p-type 2% Na doped PbTe by changing HP in which pressure is changing from 70 to 130MPa and sintering time 0.5 to 2 hours results ZT of value .077 and power factor 9.61 μW/cm K^2. Heavily doping in the $PbTe_{1-x}Se_x$ brings out large valley degeneracy through merging of the conduction band [72, 73]. Nano structuring is results a large Seebeck coefficient in PbTe. Kanatzidis et al.gives a complex system with formula MQ-AB composed of dispersed nanoscale inclusion with rock-salt crystal structure. Sono chemical method used to synthesized PbSe-coated PbTe nano partials which enhanced Te properties such as Seebeck coefficient 546 μV/K which is 5 to 10 times larger than uncoated pristine PbTe due to reduction in thermal velocity because of increased scattering of phonon at the PbTe/PbSe interface and small particle size by Sengupta et al. Lead based nanolyers of PbTe have higher thermoelectric properties such as more ZT value than tellurium based nanolayere [74]

Nano crystalline PbSnTeSe HEA (High Entropy Alloy) results good TE properties of the p-type.[75] Nano structured series of p-type $PbSb_xTe_{1-x}$ ($x = 0.1, 0.3, 0.5$) is formed by high energy ball-milling and hot-pressing. [76] Single crystal $Pb_{0.75}Sn_{0.25}Te$<Pb>is formed by the Bridgman method [77]. Different concentration of the doped Pb in the $Tl_{10-x}A_xTe_6$ nanoparticle is synthesized by solid state reaction that results decrees electrical conductivity due the nanoparticle, increase power factor and Seebeck coefficient. Lead Telluride (PbTe) grown by Czochralski technique that result Seebeck coefficient decreasing from 400 μV/K at 450 K to 0μV/K at 550K indicate p-type and after increasing temperature 550 K Seebeck coefficient is –110 μV/K shows n-type thermoelectricity [78].

Table 2

Physical and Thermoelectric Properties of Pb Based P-Type Low Temperature Telluride

	Material	S, μV/K	σ, S/cm	κ, W/mK	ZT	T, K	Ref	Year
1	Tl$_{0.02}$Pb$_{0.98}$Te	130	520	1.8	0.147	300K	71	2008
2	PbTe$_{0.75}$Se$_{0.25}$+2%Na doped	56	1612	2.7	0.056	300K	72	2011
3	PbTe: Na (2%)	62	2500	3.7	0.077	300K	72	2011
4	Na$_{0.02}$Mg$_{0.03}$Pb$_{0.95}$Te 2% Na doped	110	1000	2.6	0.14	300K	73	2011
5	Pb$_{0.96}$Mn$_{0.04}$Te:Na	100	1333	2.2	0.2	300K	79	2012
6	Pb$_{0.98}$Na$_{0.02}$Te-8%SrTe	91	2041	1.7*	0.18	300K	80	2016
7	Pb$_{0.953}$Na$_{0.04}$Ge$_{0.007}$ Te	69	2941	2.15*	0.1	300K	81	2018
8	PbSnTeSe	159.66 (623k)	613.8104	1.18*	.47 (623k)	373K	75	2021
9	PbSb$_{0.1}$Te$_{0.9}$	610	3112.7	1.52	0.12	0.09	76	2021
10	PbSb$_{0.1}$Te$_{0.9}$	523.9	1678.5	1.4			76	2021
11	PbSb$_{0.3}$Te$_{0.7}$	764.20	2628.4	1.5	0.15	160K	76	2021
12	PbSb$_{0.3}$Te$_{0.7}$	646.46	1438.0	1.25	0.11	300K	76	2021
13	PbSb$_{0.5}$Te$_{0.5}$	90.84	1961.1	1.42	0.16	160K	76	2021
14	PbSb$_{0.5}$Te$_{0.5}$	773.10	833.6	1.05	0.13	300K	76	2021
15	PbTe	400(450K)		1.43	0.41	300K	78	2022
16	PbTe	424			0.70	300K	79	2023
17	Pb$_{.75}$Sn$_{0.25}$Te at .0% hyper stoichiometric Pb	36.7	1772.6	2.55	0.09	300K	77	2023
18	Pb$_{.75}$Sn$_{0.25}$Te at .01% hyper stoichiometric Pb	41.1	1549.8	2.25	0.12	300K	77	2023
19	Pb$_{.75}$Sn$_{0.25}$Te at .05% hyper stoichiometric Pb	43.6	1696	2.21	0.15	300K	77	2023
20	Pb$_{.75}$Sn$_{0.25}$Te at .1% hyper stoichiometric Pb	46.8	1494.9	2.30	0.14	300K	77	2023
21	Pb$_{.75}$Sn$_{0.25}$Te at .5% hyper stoichiometric Pb	64.2	1026.3	2.08	0.20	300K	77	2023
22	Pb$_{.75}$Sn$_{0.25}$Te at .1% hyper stoichiometric Pb	74.9	301.3	1.88	0.09	300K	77	2023
23	Sn$_{0.94}$Na$_{0.03}$Pb$_{0.03}$S$_{0.97}$Te$_{0.03}$	208	40	0.3 (525K)	0.38 (525K)	300K	82	2024

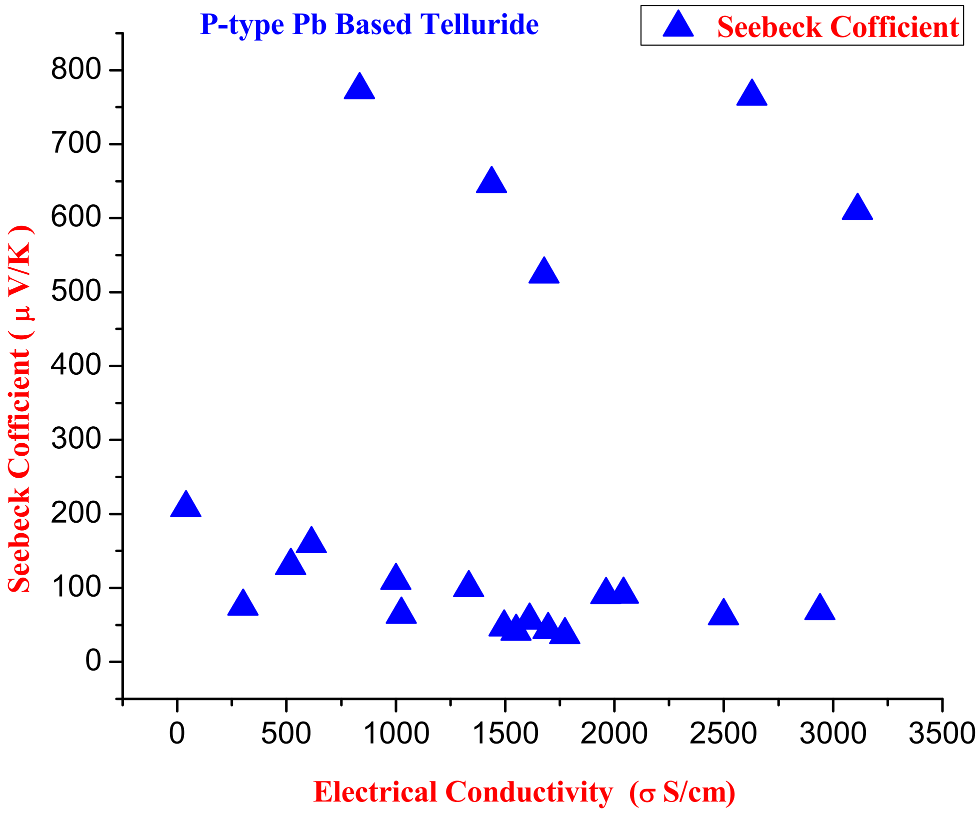

Fig. 2.2. S vs. σ plot of various Pb based P type TET

2.3. Ag Based P-type Low Temperature Telluride TE Materials

In Ag_2Te, AgCuTe are super ionic conductor at room temperature and AgCuTe and Ag_2Te are hexagonal and monoclinic at high temperature and super ionic conduction which result super high electrical conduction making them useful in TE. Se alloying in AgCuTe is effectively suppressing cation vacancies due to stronger AgSe / CuSe bond compare to AgTe/CuTe bonds result decrease thermal conductivity and increase ZT in Se doped AgCuTe[83/170]. Sn doping is used for high hole concentration in Cu_2Te and result increase ZT [84]. $AgSbTe_2$ ternary polycrystalline exhibits exceptional performance at low temperature TE application the effect of different dopant and composition on TE properties are summarized in table the PF increases due to Se doping in $AgSbTe_2$ material [85]. Disorder structure of $AgSbTe_2$ plays an important function in decreases κ [86]

Addition of Sn atoms to the LAST develop p-type materials known as LASTT its is Ag $Pb_m Sn_n Sb Te_{2+m+n}$ In this transport properties is changed by Pb:Sn ratio and effect of various alloying element summarized in table .TE properties of LASTT ($AgPb_mSn_nSbTe_{2+m+n}$) affected by charge and phonon transport by AgSbTe [87]. After Ag doping and low proportion of Se alloying produced sample $Te_{0.94}Se_{0.03}Ag_{0.02}Sb_{0.01}$ by mechanical alloying combined with SPS that result to enhancement grain boundary scattering and reduce lattice thermal conductivity and boosted power factor [88]. 1-D metal chalcogenide nanowire Ag_2Te suitable for inkjet printing without any additives for good flexible thermoelectric application [89]. Adjustment of Te content in AgCuTe observed remarkable defects which increase phonon scattering, decrease lattice thermal conductivity by over 30% and also increase ZT [90]

Table 3

Physical and Thermoelectric Properties of Ag Based N-Type Low Temperature Telluride

	Material	S μV/K	σ S/cm	κ W/mK	ZT	T K	Ref.	Year
1	$AgSbTe_2$	264	95	0.5	0.4	300K	86	2008
2	$Ag_{0.88}Sb_{1.04}Te_2$	200	161	0.73	0.26	300K	86	2008
3	$Ag_{0.83}Sb_{1.06}Te_2$	195	200	0.7	0.32	300K	86	2008
4	$AgSbSe_{0.02}Te_{1.98}$	270	155	0.58	0.58	300K	85	2010
5	$Ag_{0.5}Pb_6Sn_2Sb_{0.2}Te_{10}$	75	1000	1.4	0.12	300K	85	2010
6	$AgSn_mSbTe_{m+2}$ (m=5)	64	2400	2.18	0.13	300K	87	2012
7	$AgSn_mSbTe_{m+2}$ (m=10)	50	2800	3	0.07	300K	87	2012
8	$AgSbTe_{1.85}Se_{0.15}$	203	179	0.37*	0.53	300K	91	2017
9	AgCuTe	31	1055	0.35*	0.15	300K	83	2018
10	$Sn_{0.83}Ag_{0.03}Mn_{0.17}Te$	46	2669	1.64*	0.05	320K	82	2020
11	$AgSb0._{94}Cd_{0.06}Te_2$	248	220	0.15*	1.5	300K	92	2021
12	$Te_{0.94}Se_{0.03}Ag_{0.02}Sb_{0.01}$	220		1.38	.57 (ave)	300K	88	2022
13	$Ag_{1.9}Te$	80.4	185.8				89	2023
14	$Ag_{2.1}Te$	65.2	523.3				89	2023
15	$AgCuTe_{.925}$	63		0.5 (300K)	1.02 (573K)	320K	90	2024

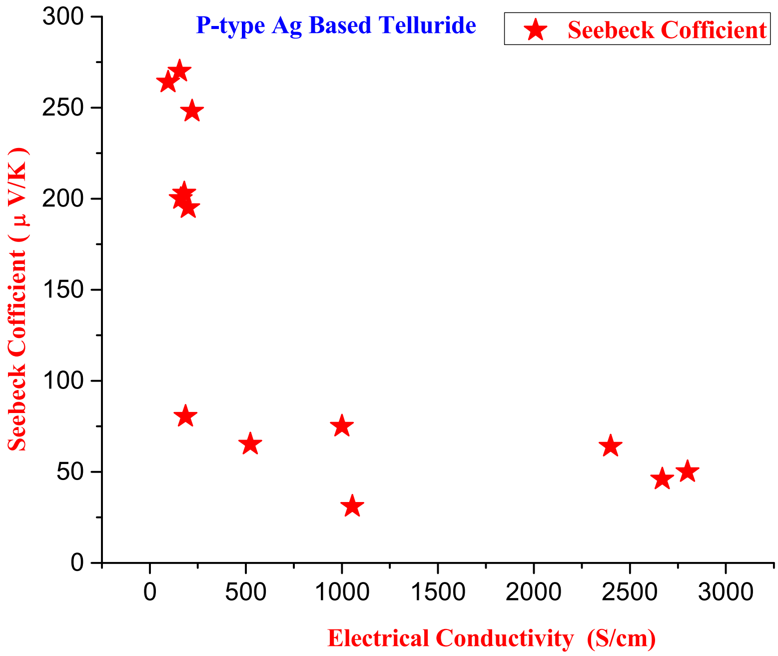

Fig. 2.3. Seebeck coefficient(S) vs Electrical conductivity (σ) plot of various Ag based P type TET

2.4. Sn Based P type Low Temperature Telluride TE Materials

SnTe is crystalline cubic rock salt at ambient condition with narrow band gap semiconductor as result that band gap highly symmetrical, degenerate electronic bands, easily changing electronic structure by alloying and doping. Doping is much effective in inducing resonant level in SnTe [93]. SnTe has rock salt structure the larger band gap between primary and secondary band valence SnTe results small Seebeck coefficient so result poor TE performance. Doping and alloying reduce band gap that improve TE performance such as co doping of In and Cd in SnTe and In and Ag. Due to co doping of Ag and Mn synergistic effect have also been achieved in SnTe. Main problem in SnTe is p-type carrier concentration due to cation vacancies this is reducing hole concentration by self-compensation in SnTe [94]. Furthermore, novel strategies engineering ferroelectric instability in SnTe for High TE performance [95].

First time synthesis of both pristine and Sr doped SnTe by using thermal evaporation technique result enhanced Seebeck coefficient due Sr addition by carrier localization specifically the energy filtering. The bulk tin telluride (SnTe) is synthesized through flame melting while the 2D SnTe is produced via liquid phase exfoliation experimental results shows high ZT ~ 0.17 due to low thermal conductivity for 2D SnTe compared to bulk ZT ~ 0.005 at room temperature [96]. A colloidal hot injection approach for fabricating binary SnTe [97]. Doping and lattice strain engineering, thermal conductivity is decreased by scattering phonon and disrupting heat pathway. Te doped sample maintain a stable power factor between 1.8 and 2.25 µW/cm K^2 over a broad temperature range 300 K to 573 K and Seebeck coefficient 208 µV/K at 300 K to 350 µV/K at 573 K. The cationic Na, Pb and anionic Te doped sample result low thermal conductivity 0.3 W/mK at 525 K and result ZT of 0.38 [98]. High power factor achieved for $Sn_{0.7}Sr_{0.3}Te$ thin film at different post annealing temperature using a simple thermal evaporation. Nanoparticle shaped tin (Sn) and layer shaped palladium (Pd) obtained on the Bi_2Te_3 nano plates via electrode deposition results low thermal conductivity 1.1 W/mK because of nanoparticle shaped make easy the scattering of phonons.

Table 4

Physical and Thermoelectric Properties of Sn Based P-Type Low Temperature Telluride

	Material	S µV/K	σ S/cm	κ W/mK	ZT	T K	Ref	Year
1	$Sn_{0.9975}In_{0.0025}Te$	50	4300	1.61*	0.09	300K	93	2013
2	$Sn_{0.985}In_{0.015}Te_{0.85}Se_{0.15}$	66	1674	1.28*	0.09	300K	99	2014
3	$Sn_{0.94}Mg_{0.09}Te$	35	3126	2.72*		300K	100	2015
4	$Sn_{0.94}Ca_{0.09}Te$	44	5466	1*	0.8	300K	101	2016
5	$Sn_{0.915}Mn_{0.11}In_{0.005}Te$	117	634	1.64*	0.13	300K	102	2017
6	$Sn_{0.57}Sb_{0.13}Ge_{0.3}Te$	67	2004	0.48**	0.16	300K	95	2019
7	$Sn_{0.83}Ag_{0.03}Mn_{0.17}Te$	46	2669	1.64*	0.05	300K	94	2020
8	$Sn_{1.03}Te_{0.85}Se_{0.075}S_{0.075}$ -2%Ag-2%In	90	1387	1.21*	0.1694	300K	94	2020
9	SnTe	30	$2.1x10^4$			300K	97	2021
10	$(SnGe_{0.03}Te)_{0.9}(Ag_{0.5}Bi_{0.5}Se)_{0.1}$	68			0.89 (873K)	300K	103	2022
11	$AgSn_4(Sb_{0.8}Bi_{0.2})\ Te_6$	40	1429	108		300K	104	2023
12	$AgSn_2(Sb_{0.8}Bi_{0.2})\ Te_{3.97}Br_{0.03}$	74	685	1.2		300K	104	2023
13	$AgSn_4(Sb_{0.8}Bi_{0.2})\ Te_{5.97}Br_{0.03}$	57	1443	2.0	0.07	300K	104	2023
14	SnTe 2D nanostructured	280			0.17	300K	96	2024
15	$Sn_{0.94}Na_{0.03}Pb_{0.03}S_{0.97}Te_{0.03}$	208		.3 (525K)	.38 (525K)	300K	98	2024

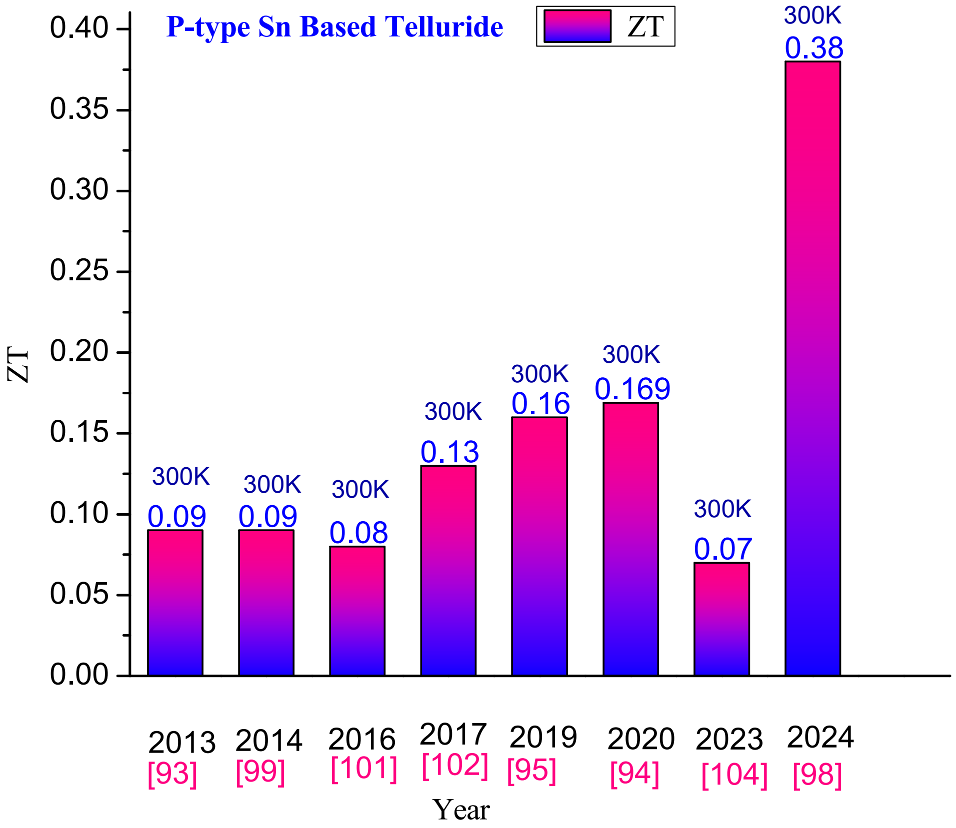

Fig. 2.4. ZT of various Sn based P type TET at different temperature and year

2.5. Cu Based P-type Low Temperature Telluride TE Materials: Cu_2Te is transition metal-based telluride it is super ionic conductor which support high electrical conduction. Cu dopant has been increasing carrier concentration which result high ZT. Sn doping is used to increase large hole concentration of Cu_2Te [105]. The Cu related telluride $CuGaTe_2$ and $CuInTe_2$ both have high σ and κ thus the lowering κ by inclusion and point defect, the inclusion of Nano phase in Cu_2se in $CuGaTe_2$ that reduce thermal conductivity, ZnS nanoscale hetero structures [106] and In_2O_3 nano inclusions [107] have been incorporated in $CuInTe_2$ to lower κ. Alloying of Ag in to $CuInTe_2$ lower κ_L by forming weak Ag-Te bond [108], multicomponent alloying Ag and In in $CuGaTe_2$ also lowering lattice thermal conductivity and increases ZT. Disordering of Cu atom with increasing S (Sulphur) content results in stronger phonon scattering in sample $Cu_2Te_{0.2}S_{0.8}$, and reduce thermal conductivity [109]. Doping of both the Se and Te material with Zn results decrees in carrier concentration and electrical conductivity [110]. The copper telluride thin film with 1:1, Cu: Te ratio gives the highest PF due to carrier filtering at the interface between the two phases [111]. Ball milling method used for synthesize Bi doped Cu_7Te_4/Sb_2Te_3 nanocomposite sample results high power factor to be 788.28 W/mK^2 at 310 K.

Table 5

Physical and Thermoelectric Properties of Cu Based P-Type Low Temperature Telluride

	Material	S μV/K	σ S/cm	κ W/mK	ZT	T K	Ref	Year
1	$Cu_{0.75}Ag_{0.2}InTe_2$	234	69	1.86	0.07	300K	112	2014
2	$CuInTe_{1.99}Sb_{0.01}$+1 %ZnO	104	610	3.6*		300K	107	2016
3	$CuInTe_2$+6%ZnS	157	312	3.54*		300K	106	2017
4	$Cu_{1.9}Sn_{0.1}Te$	38	2638	1.22	0.1	320K	105	2018
5	Cu_2Te	15	4818	6.1		320K	105	2018
6	$Cu_{0.89}Ag_{0.2}In_{0.91}Te_2$	446	19	2.68		300K	108	2020
7	$Cu_4ZnSn_2Se_7$	80	265	3.0	.20 (630K)	300K	110	2021
8	$Cu_4ZnSn_2Te_7$	21	3100	4.6	.05 (575K)	300K	110	2021
9	$Cu_5Sn_2Se_7$	16	8800	6.7	.10 (630K)	300K	110	2021
10	$Cu_5Sn_2Te_7$	9	9200	10	.009 (575K)	300K	110	2021
11	$Cu_{0.5}Te$	346	20.74			300K	111	2022
12	CuTe	40.8	4632			300K	111	2022
13	Cu_2Te	53.8	178.71			300K	111	2022
14	$Cu_2Te_{0.2}S_{0.8}$	338		.29		300K	109	2023
15	5at%Cu- Bi_2Te_3 (brain ice sol)	155		1.396	0.17	300K	113	2024

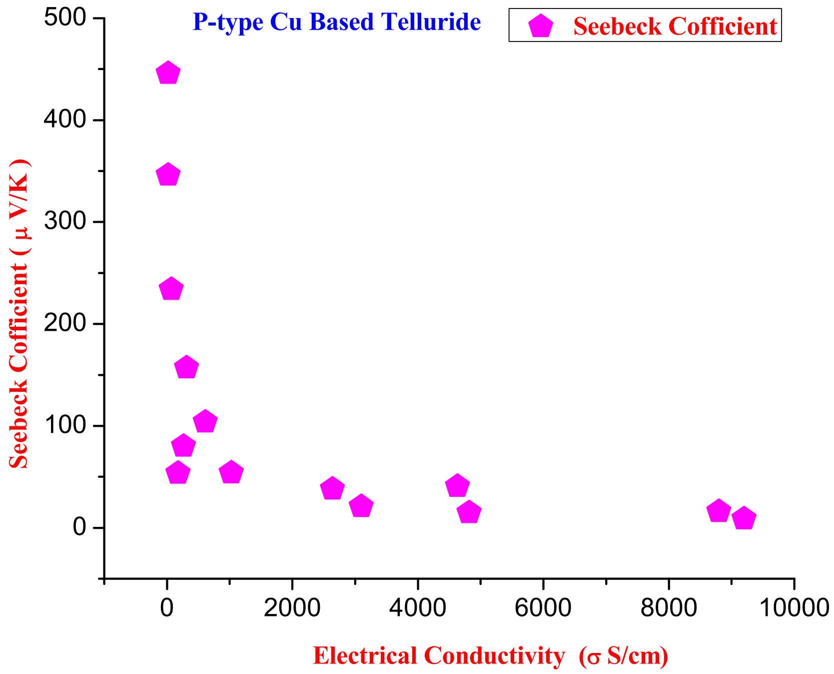

Fig. 2.5. S vs σ plot of various Cu based p-type TET

3. TE Materials Synthesis Methodologies

3.1. Melt Growth (MG) Alloying

TE bulk materials developed by mixing more than one element in desired stoichiometric ratio, after that heating of sealed quartz ampules at temperature 500–2000 °C. After melting and heating the desired structure is prepared by annealing, HP or SPS etc. The processing temperature changes for material after doping or alloying such as PbTe, Bi_2Te_3 etc.

3.2. Mechanical Alloying

Mechanical alloying is commonly used to produce nano/microstructure TE material with reduced thermal conductivity in this method powder of element are mixed in precise proportion and grind in BM and then processed by HP or SPS, Cu_2Se formed by ball milling and HP [114].

3.3. Zone Melting

Zone melting can directly prepared bulk polycrystalline without further sintering thus keep away from inhomogeneity, volatilization at high temperature.

3.4. Bridgman and Czochralski Methods

Both methods are used to synthesize single crystalline material direct solidification by converting melt from hot zone to the cold zone such as $Ba_8Au_{5.33}Ge_{40.67}$ by Bridgman process with SPS. Czochralski method is used for highly efficient and high purity preparation of single crystalline materials.

3.5. High Pressure Preparation

High pressure preparation method (GPa-5GPa) used to avoid the loos of volatile raw material via sublimation by decreasing reaction temperature and shortening reaction

3.6. Additive Manufacturing (3D printing)

Additive manufacturing can be used to construct to complex geometry and layered structure. The additive manufacturing includes photo polymerization, powder bed fusion, material jetting techniques and extrusion-based methods.

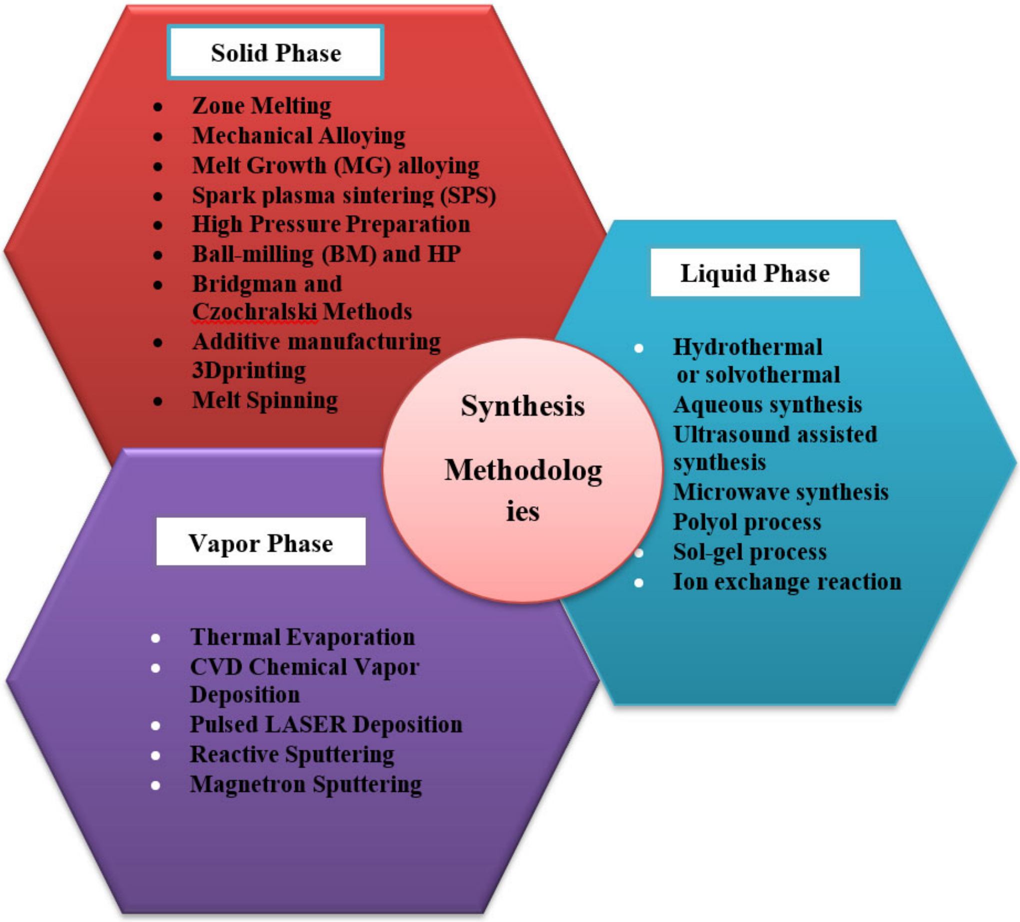

Fig. 3.1. Various synthesis methodologie

3.7. Arc Melting or Levitation

Arc melting or Levitation is an instant melting process for preparation of alloy system such as HH alloy, intermetallic and large temperature material and further processed by SPS or HP such as $Hf_{1-X}Zr_XNiSn_{1-Y}Sb_Y$ formed by Levitation and SPS.

3.8. Spark Plasma Sintering 'SPS'

Spark plasma sintering has the outstanding superiority of attaining a high densification in a short time with limited grain growth .SPS employ a very large current going through the sample to heat it directly such as in case of $Cu_{15}As_{30}Te_{55}$, $Ge_{1-x}Ga_xTe$ TE material.

3.9. Ball-Milling (BM) and Hot Pressing (HP) Method

HP is used to develop PbSe, SiGe TE material. Ball milling and HP combination is powerful method to prepare bulk TE material such as amorphous dense bulk materials 80 $GeTe_2$-20 Ga_2Te_3 at room temperature was obtained with fine microstructure and large electrical conductivity.

3.10. Melt Spinning (MS) Method

Melt spinning is useful method for fast molten solution cooling. In this thin stream of molten glass is flow on spinning wheel and melting heat is quickly transferred to the wheel followed by rotation such technique generates a microstructure with reduced thermal conductivity. Goncalves et al. prepared $Cu_{30}AS_{15}Te_{55}$ by using MS technique.

3.11. Hydrothermal or Solvothermal

Currently material with nanodimension such as nanotube, nanoparticle, nano box, hallow nanospheres and nanodendrites prepared by Hydrothermal and Solvothermal. Synthesis starts in aqueous /non-aqueous solution in closed stainless steel autoclaves at high temperature and pressure such as PbTe nano powder, and Bi_2Te_3

3.12. Aqueous Synthesis

Aqueous synthesis is used for the large-scale production of TE material at room temperature and atmospheric pressure such as PbTe gray powder is obtained by this method.

3.13. Microwave Synthesis

Microwave assisted synthesis are allow in solution and permit by the friction and collision between polar molecules under alternating electromagnetic field. In solid state microwave, a precursor is vacuum sealed quartz ampoule and it is exposed by microwave radiation in a microwave oven.

3.14. Polyol Process

Polyol process is used for preparation of PbTe, Bi_2Te_3 and Cu_7Te_5 by microwave irradiation. The selected precursor (acetate of Pb, Ni, and Cu etc.) is dissolved in the ethylene glycol (EG) in which an elemental powder of Te is added. A solid-state moisture assisted reaction takes place under microwave irradiation result nano crystalline form.

3.15. Sol-Gel Process

For preparing metal oxide, chalcogenide and semiconductor sol gel method is versatile technique such as the formation of of Bi_2T_3 by the sol gel process.

3.16. Ultrasound Assisted Synthesis

In sono chemistry synthesis in which inorganic particle via chemical reaction under strong ultrasound radiation between 20 kHz and 15 KHz is formed.

3.17. Ion Exchange Reaction

Ion exchange reaction is mostly used for synthesis of semiconductor nano crystal where by guest cations are substitute with host atom cations within the crystal lattice such as Ag_2Se thin film.

3.18. Chemical Vapor Deposition 'CVD'

CVD is used for the fabrication of thin film from the gaseous phase through reaction of volatile precursors on the substrate surface such as growth of nanowire arrays in TE micro generators

3.19. Thermal Evaporation

Thermal evaporation is used to fabrication of thin film of Ag_2Se with variable thickness by applying a heat source to evaporate bulk polycrystalline Ag_2Se.

3.20. Pulsed LASER Deposition

Pulse LASER deposition is performed by focusing a high-energy pulsed beam on a solid target material this method used for non-epitaxial growth of TE films with desired phase and composition.

3.21. Reactive Sputtering

Reactive sputtering in which compound film are fabricated on substrates by introducing a reactive gas precursor

3.22. Magnetron Sputtering

Magnetron sputtering in which magnetically confined plasma collides with an electrode or target in order to eject atom for fabrication on the substrate that is also used for series of very good performance TE films at room temperature for application such as wearable device

4. Advance Strategies for Enhancing the Performance of Thermoelectric Material

For designing advance Thermoelectric material various strategies as followings

4.1. Doping and Co-Doping

Various doping element are used as dopant in improving the TE performance due to different mechanism. Generally, the Seebeck coefficient is directly related to concentration n, effective mass m* and T so that concentration is important factor to set peak temperature and maximum power factor. Suitable doping element can enhance carrier concentration by modifying electronic band structure and also increase number of densities of state and Fermi level is shifted near the conduction band that promote transport of electron that increase S, ZT and electrical conductivity.so doping improve the performance of TE materials. Uniform doping reducing carrier mobility and a heavily doped semiconductor shows a high PF

The merging of the valence band and conduction band results in a simultaneously increase in power factor, S and electrical conductivity because of doping result the electronic band to converges in the bulk material with high valley degeneracy. The electronic band in $PbTe_{1-x}Se_x$ doped with 2%Na converges electronic band width increase valley degeneracy and the results of this mechanism a ZT = 1.8 at 527 °C is obtained [115] Both σ and S are increase simultaneously in a host semiconductor embedded with anti-resonance nanoparticles .If the band gap is significantly greater than K_BT ($E_g > 10\ K_BT$) then width of band gap and Fermi level near below conduction band results significantly increasing the thermo power of material and no role of carrier concentration and charge transport for example PbTe a maximum ZT is obtained at 327 °C ($E_g \sim 0.36$ eV).

To achieve increasing thermo power and same time decreasing the κ_L by suitable alloying of element that create mass fluctuation and better complex band structure that results disordered scattering of phonon leads to decrease the κ (negligible in case of electron because of large velocity and larger wavelength than phonon) and enhance TE performance. Increase in thermo power by energy filtering effect in a homogeneous material. At the grain- grain interface charge carrier encounters a potential barrier that crosses this barrier shows stronger non equilibrium energy distribution with high energy particle and result of this is increase the S because of carrier with low energy and higher Fermi energy cancel to each other therefore increasing the thermo power by removal of low energy carrier

Doping of element (Li, K and Na) on polycrystalline SnSe improve the performance of TE and ZT increases because of dopants increase electrical conductivity and carrier concentration. N type PbTe based material shows excellent TE material by doping of Bi in PbTe nanotube and highest ZT. First time ZT value greeter than 2 Excellent by K doped $PbTe_{0.7}S_{0.3}$ that result ZT value 2.2 at 923K [116]

4.2. Alloying

Alloying element alters the properties of TE materials that increase PF and decreasing mostly notice in HH alloy, Skutterudites and Clathrates. In HH alloy the electronic structure is changed due to alloying of element that create atomic disorder at an atomic size due to development of atomic fluctuation or strain effect which reduce κ_L. Low κ is obtained within a single material can be controlled with strategies such as Phonon Glass Electron Crystal (PGEC) is particularly in Skutterudites in which guest atom in a matrix of complex material system act as scattering center that results reduce κ and enhance TE performance

4.3. Nano Structuring

Reducing structural dimension of TE materials to the nano scale range by nano structuring for improving ZT Nano structuring reduces thermal conductivity by the scattering of phonon from nanostructure surface or interface effective in nanowire. quantum dot, nanoparticles super lattice and nano thin film system. Presence of interface in nanostructured material result in reducing lattice thermal conductivity and increase ZT such as nano crystalline material prepared by BM shows a ZT ~ 1.4 at 127 ºC [117]. Effect of annealing increases the nanoscopic in homogeneities within the grain that result increases phonon scattering and decreases the value of κ [117] Power factor cannot be affected by nano structuring.

Nano structured material such as 2-D TE nano nanomaterial (quantum wells) ,1D TE material (nanowire, nanotube, nanoribbons and nano mesh),0D quantum dot all these 2D, 1D, 0D called LD used for new TE device nonostructuring can increase the electronic density of state near the Fermi level via quantum confinement due to this ZT value in some nanostructure above 3 N type $Bi_2(Te, Se)_3$ compound in the form of nanowire arrangement by using thermal co evaporation method results ZT ~ 1.01 at 300 K [118]

4.4. Nanocomposite

Nano composite material and nano composite strategies are much used in TE device due to much advantage compared with bulk and nanostructured materials that control phonon and electron transport such as HH alloy, skutterudites and Si-Ge system. Compositional fluctuations at nano scale due to spinodal decomposition in the both composition of the mixed phase system share the same lattice leading to spatial modulation of the local composition at the Nano scale. This spatial modulation is established coherently in the embedded nanoparticles within the matrix there by forming nanostructured TE material. PbS-Ag Nano composite form ohmic contact with bulk semiconductor, it enhances the TE properties with improved ZT ~ 1.7 at 850 K [14]

4.5. Nanoinclusion

Nano inclusion is used due to their cost and easily processing material. Dispersed secondary nanoparticles in the nanostructure host material are referred to as nano inclusion. Nano inclusion is the much fruitful due to reduction in κ_L via phonon scattering from nano inclusion. Nano inclusion is important for increasing the thermo power because of the scattering of phonon or electron at hetero boundaries that an increase in thermo power from –155 to –177μV/K is observed with an increase in the ZrO_2 particle content from 0 to 9 vol%. Nano inclusion results reduce thermal conductivity, increase electrical conductivity and S.

4.6. Superlattice

Periodic layer of nanometer thickness of two or more are called super lattice it homo structure (fabricate by single material) and hetero structure (fabricate by two or more material) is synthesized by molecular beam epitaxial sputtering, metal organic CVD etc. Ultra-thin film nanostructure is fabricated by thermal evaporation, electro deposition pulse laser deposition, CVD etc.

Fig. 4.1. Advance Strategies for enhancing the performance of Thermoelectric Material

QDSL means Quantum Dot Super Lattice composed of PbTe / $PbSe_{0.98}Te_{0.02}$ doping with Bi on BaF_2 substrate with ZT value of 1.6 and 3.5 at 300 K and 570 K. The fabrication of multiple quanta well Bi_2Te_3/Sb_2Te_3 super lattice with ZT ~ 2.4 without change power factor.2D super lattice consisting of PbTe quantum walls and $Pb_{1-X}Eu_XTe$ barrier which significantly improve the TE performance. various supper lattice in which TE performance enhance due to decrease of thermal conductivity for example PbTe/PbSe Ge/SiGe, Si/Ge, Bi/Sb and Bi_2Te_3/Sb_3Te_3 [119]. Firstly, idea of supper lattice was given by Venkatasubramanian to improve the value of ZT by decreasing the κ_L in case of p type Bi_2Te_3 / Sb_3Te_3 super lattice by controlling the phonon transmission and electron in the super lattice so result high ZT value 2.4

5. Conclusions and Future Perspectives

Based on the comparative analysis presented in this review, the development of thermoelectric (TE) materials has progressed significantly, with notable achievements such as the discovery and optimization of Sn-based tellurides. Compounds like Pb{1-x}SnxTe have demonstrated high efficiency in both n- and p-type configurations across wide temperature ranges, underscoring their importance in mid-temperature TE applications. Nevertheless, stabilizing p-type carriers in chalcogenide systems remains a challenge, limiting the widespread realization of high-performance devices.

For clarity, TE materials can be classified according to their operating temperature ranges:

Temperature Range (K)	n-type Materials	p-type Materials
300–600 (Low)	Bi_2Te_3	Bi_2Te_3
600–900 (Mid)	PbTe-based compounds	GeTe-based compounds

Within these temperature regimes, performance optimization has relied on several strategies, including band convergence, defect engineering, nanostructuring, and alloy design. While these approaches have yielded incremental improvements, achieving ZT > 2 in low-temperature chalcogenide-based materials remains an open challenge. Addressing this will be critical for enabling scalable TE devices in refrigeration and energy harvesting applications over the next decade.

In summary, Sn-based tellurides and PbTe/GeTe systems represent major milestones in mid-temperature TE research, but further innovation is required to overcome intrinsic limitations in low-temperature materials. Advancing both low- and mid-temperature TE materials will be essential for scalable energy solutions, ranging from refrigeration to waste heat recovery, thereby ensuring that thermoelectrics contribute meaningfully to sustainable energy technologies.

Conflict of Interest: On behalf of all authors, the corresponding author states that there is no conflict of interest.

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Низькотемпературний неорганічний термоелектричний телуридний матеріал p-типу: огляд

Відновлювані джерела енергії, такі як термоелектричні матеріали, будуть важливими для вирішення енергетичних криз та зростання глобального попиту в майбутньому. Ці матеріали мають багато переваг для використання у виробництві електроенергії, охолодженні та опаленні, і можуть генерувати корисну електроенергію з різних джерел відпрацьованого тепла, що працюють у різних температурних діапазонах. Наша увага зосереджена на низькотемпературному неорганічному термоелектричному телуриді (TET) p-типу, придатному для роботи за температури, близької до кімнатної. Цей огляд починається з фундаментального розуміння термоелектричних матеріалів, методологій їх синтезу та передових стратегій підвищення характеристик. Низькотемпературні телуридні TE матеріали на основі халькогенідів відіграють дуже важливу роль і створюють основу для майбутніх застосувань, а також пропонують перспективи. Нарешті, ми підсумовуємо проблеми та майбутні перспективи неорганічних TE матеріалів.

Ключові слова: термоелектричні матеріали, телуридні термоелектричні матеріали, низькотемпературна термоелектрика, відновлювана енергія.