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Properties and Recent Progress in Oxide-Based Thermoelectric Materials

Chinchu Y.M.[1,*], T. Beena[2]

Abstract

Advanced thermoelectric materials that can be used efficiently for energy conversion have received significant research interest over the last decade because of the mounting demand for sustainable and eco-friendly energy technologies. The main advantage of thermoelectric materials is their capability to directly convert waste heat into electrical energy, which makes them highly promising for the recovery of waste heat from industrial processes, automotive systems, and power generation applications. Oxide-based thermoelectric materials are interesting due to their high abundance, low cost, low mass density, excellent chemical and thermal stability, non-toxicity, and capability to work at higher temperatures in oxidizing conditions, among the different classes of thermoelectric materials. Although they are useful, high thermoelectric efficiency is still hard to attain because of the interdependence of electrical conductivity, Seebeck coefficient, and thermal conductivity. This review gives a thorough overview of the basic properties of oxide-based thermoelectric materials and the methods used to optimize these transport properties to improve the thermoelectric properties. Different synthesis and fabrication methods such as solid-state reaction, sol-gel, polymer solution, pressureless sintering, and spark plasma sintering are discussed, and their effects on the material properties are explored. The developments of the systems that have been extensively studied, such as $Ca_3Co_4O_9$, $SrTiO_3$, and $CaMnO_3$, over the last few years are also summarized, including the effect of element doping and microstructural engineering. In addition, the review identifies the recent progress, current challenges, and future research trends to enhance the figure of merit (ZT) and further extend the application of thermoelectric oxide materials to efficient and sustainable energy harvesting.

Keywords: Thermoelectric, Seebeck coefficient, electrical conductivity, power factor, thermal conductivity

INTRODUCTION

There are several issues with energy use globally. Social and political instability is increasing as a result of the global demand for energy. Over 60% of the energy lost globally is converted to heat. On the other hand, there is a growing demand for greenhouse gases, especially CO_2, in the environment [1, 2]. Similarly, the effects of using fossil fuels on the ecosystem and climate change are becoming more concerning. Challenges to develop alternative technologies that can convert waste heat into electrical energy play a key role in current days [3–5]. This can reduce our dependence on fossil fuels and reduce greenhouse gas emissions. Scientists worldwide are in search of such materials that can help to meet the increasing energy demands with minimal environmental impact [6–8]. In several fields, materials with the ability to directly and reversibly transform thermal energy into electrical energy have sparked attention. Materials that possess these properties are called thermoelectric materials (TE). Home heating, automotive exhaust,

***Author for Correspondence**
Chinchu, Y.M.
E-mail: chinchukayal03@gmail.com

[1]Research Scholar, Department of Physics, Scott Christian College (Autonomous) (affiliated with Manonmaniam Sundaranar University), Nagercoil, Tamil Nadu, India
[2]Department of Physics, Scott Christian College (Autonomous) (affiliated with Manonmaniam Sundaranar University) Nagercoil, Tamil Nadu, India

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and industrial processes can use TE materials [9–15]. High-efficiency TE materials must be developed immediately for waste heat recovery, as this will have significant positive effects on the economy and the environment [16–25]. They are highly applicable for remote applications in the absence of any compressors or pumps, especially in spacecraft. Heat lost in engine coolant or exhaust gas can be used to produce electricity by using TE [26–35].

THERMOELECTRIC MATERIALS

Environmentally friendly energy conversion materials that are compact, highly reliable, pollutant-free, and feasible throughout a broad temperature range are called thermoelectric materials [36–42]. Thermoelectric materials can be used to convert the vast amounts of waste heat produced by industrial processes, vehicle exhaust, and home heating into energy. It is critical to develop high-efficiency thermoelectric materials for waste heat recovery because they have significant positive effects on the economy and the environment [43–54].

PROPERTIES OF TE

To carry out such thermoelectric conversion, TE materials should possess a few special qualities. The figure of merit (ZT), which is defined as the conversion efficiency of a TE material, $ZT = \frac{S^2\,\sigma T}{K}$. S, σ, T, and k are the Seebeck coefficient, electrical conductivity, temperature, and thermal conductivity. $S^2\,\sigma$ is the power factor. The relationship between the Seebeck coefficient can be expressed as $S = \frac{8\pi^2 {K_B}^2}{3eh^2}\, m^*\, T\left(\pi/3n\right)^{2/3}$Where k_B, e, h, m^* and n are the Boltzmann constant, carrier charge, Planck's constant, effective mass, and carrier concentration, respectively. Low thermal conductivity and high electrical conductivity are necessary to optimize the thermopower of ZT materials.

Electrical Conductivity

Electrical conductivity is given by $\sigma = ne\mu$. Where μ and n are carrier mobility and carrier concentration. Strongly doped semiconductors with carrier concentrations between 1019 and 1021 cm^3 are good TE materials. One type of carrier should be chosen to guarantee a high Seebeck coefficient; mixing carriers can reduce thermopower. To obtain a single type of carrier, materials with sufficient doping and energy bandgaps that allow for good separation between n-type and p-type are selected. Consequently, semiconductors with an energy bandgap of less than 1 eV that are extensively doped are effective TE materials.

Effective Mass

Another contradiction arises from the effective mass of the charge carriers, as a large effective mass results in limited electrical conductivity and high thermopower. The m^* refers to the density of states effective mass, which increases with a flat, narrow bandgap with density of states at the Fermi surface,$\sigma = \frac{ne^2\tau}{m^*}$. Electrical conductivity results from heavy carriers moving with stronger velocities and, consequently, small mobilities, as m* is linked to n. Effective mass and mobility have a complicated relationship that is influenced by anisotropy, electronic structure, and scattering mechanisms. Effective mass and the dominating charge carriers must be balanced to strike a compromise between high mobility and high effective mass.

Thermal Conductivity

Thermal conductivity in thermoelectric materials comes from two sources

1. Electron and hole transporting heat (k_e)
2. Phonons travelling through the lattices(k_l)

$$k = k_l + k_e$$

$$k_e = LenuT$$

Because k_e is linearly proportional to the electrical conductivity, decreasing k affects the electrical

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conductivity. The lattice thermal conductivity was tuned to change the thermal conductivity because reducing k has little to no improvement in ZT.

PHONON-GLASS-ELECTRON CRYSTAL

As the ZT increases, the complex parameter relationship gets closer. A "phonon-glass-electron crystal (PGEC)" model is a necessary component of high-quality thermoelectric materials. The characteristics of thermoelectric materials are described by this model. An ideal thermoelectric material is said to have both crystal-like electrical characteristics and glass-like thermal conductivity. The lattice thermal conductivities of glasses are poor. The idea of low thermal conductivity arises from the perception that thermal conductivity in glass is a random travel of energy through the lattice rather than quick transit via phonons. However, because they lack the necessary electron crystal characteristics, they perform poorly as thermoelectrics. Thus, crystalline materials that scatter phonons without substantially altering electrical conductivity are good thermoelectric materials. For a thermoelectric material to be capable of such a conversion process, it must possess specific characteristics. The thermoelectric material's conversion efficiency is correlated with the figure of merit (ZT), which is defined as

$$ZT = S^2\sigma T\ k$$

where,
S → Seebeck coefficient (μV/K)
σ → electrical conductivity ($\Omega^{-1}cm^{-1}$)
k → thermal conductivity ($Wm^{-1}\ k^{-1}$)
T → temperature (K)
$S^2\sigma$ → power factor

RESEARCH PROGRESS ON THERMOELECTRIC MATERIALS

The 1950s witnessed remarkable advancements in the field of thermoelectrics. Bi_2Te_3, PbTe, and SnGe bulk thermoelectric materials are examples of the first generation of thermoelectric materials created for room temperature applications. In 1960, solid solution formation and doping control were the main strategies for increasing ZT. Global interest in the field of thermoelectrics was low between 1960 and 1990 [55–59].

With ZT ~1, the alloy family was the best commercial material. Two distinct strategies have been developed over the last 20 years to find the next generation of high-performance thermoelectric materials. One is the synthesis and application of low-dimensional thermoelectric material systems, and the other is the discovery and use of novel families of bulk materials with intricate crystal structures. A high ZT of ~2.4 at 300 K was recorded for Bi_2/Sb_2Te_3. However, at high temperatures, these substances become unstable and poisonous. As a result, the focus of the research has somewhat changed to oxides. The high chemical and thermal stability of oxides make it possible to apply a significant temperature gradient across materials in air [60].

Oxides are distinguished by their structural, compositional, and chemical versatility. Oxides are inexpensive and environmentally benign. Metal oxides were not thought to have good TE at first, but studies on transition metal oxides showed promising results because of their strong electronic correlation, narrow bands, which resulted in strong spin and orbital fluctuations, and high degeneracy, which can occur in the d-electrons and is beneficial for enhancing TE.

The most common oxide materials are p-type $Ca_3Co_4O_9$, n-type ZnO, $SrTiO_3$, and $CaMnO_3$. The various manufacturing techniques employed and the potent thermoelectric outcomes attained in these materials in recent years are discussed in this review.

$Ca_3Co_4O_9$ (CCO)

CCO is a p-type semiconducting material well known for its electronic, magnetic, and electro-optic properties. In the family of cobaltites, CCO has gained a special place because of its misfit layered

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structure. Its crystal structure consists of two different CdI_2–type monoclinic structures. The middle layer consists of Ca and O. The other layers contain Ca and O. The substructure has the composition of Ca_2CoO_3 and is usually referred to as a rock salt structure. Various techniques have been used to prepare CCO nanoparticles. The thermoelectric properties also vary depending on the fabrication techniques used and the conditions of annealing and sintering. Various techniques used for the fabrication and their outcomes are given in this article.

Solid-State Reaction Method

The simplest yet effective method for fabricating CCO nanomaterials is the solid-state reaction method. $CaCO_3$ and Co_3O_4 were mixed for 5 min at 400 rpm in an agate ball. The obtained powder was heated at 850°C for 8 h by varying the concentrations (x=1,10,20). Spark plasma sintering was performed. The decomposition of $Ca_3CO_4O_9$ to $Ca_3CO_2O_6$ was studied using thermogravimetric analysis (TGA) analysis, which showed two peaks at 911 and 965°C. All samples were *p*-type conductors with positive Seebeck coefficients. The electrical conductivity also increased with temperature, and a higher ZT was obtained for pure CCO than for the doped one. $CaCO_3$ and Co_3O_4 were mixed and calcined for 20 h at 700°C. The prepared pellets were annealed at 900°C for 20 hours. Again, ground wood powders [1, 1.05, 0.15, 0.25] were added to 1 g of CCO. The samples were sintered at different temperatures of 500°C and 900°C for 6 h and 18 h, respectively. Structural analysis and thermoelectric studies were performed for all samples. Seebeck coefficient varies from 18 μV/k to 144 μV/k for x=0 and 0.15 at 800 K. Among all the samples, CCO [0.15] shows high thermoelectric properties, attaining ZT=1.19 at 800 K

Pressureless Sintering Method

Zirconia grinding media were used to combine $CaCO_3$ and Co_3O_4 with water. It was calcined twice at 900°C for 15 hours after drying. The samples were then cooled to 900°C and annealed at 900°C for 12, 24, and 72 h after being fired in air at 1000–1300°C for 6 h. At 890°C and 940°C, the TGA displayed two peaks, signifying the breakdown into Ca3CO2O6 and CaO at 940°C. The electrical conductivity of the 920°C sample was 56 S/cm at 100°C, and the Seebeck coefficient progressively increased from 130 to 160 μV/k within the measured temperature range of 100–670°C. At 600°C, a ZT of 0.06 was recorded.

Polymer Solution Method

$Ca[CH_3CO_2].½H_2O$ + $[Co(CH_3CO_2)_2.4H_2O$ were added to distilled H_2O. Polyethyleneimine was added to the above solution. Partial evaporation took place in a rotary evaporator. The powder obtained was thermally heated twice at 750 and 800°C for 12 h and pelletized. The pellets prepared were annealed in air at 900°C and 960°C, indicating the formation of $Ca_3CO_2O_6$. ray diffraction (XRD) studies revealed that $Ca_3CO_4O_9$ was the major phase, accompanied by CaO, $Ca_yCo_{1-y}O$, and $Ca_3CO_2O_6$ for samples annealed for 24 h. For longer annealing times, the number of secondary phases decreased. The electrical resistivity decreased with increasing temperature. The minimum resistivity value at room temperature was 15 mΩcm for the 96 h annealed sample. For the as-prepared sample, the Seebeck coefficient varied from 80 to 550 MvK^{-1} at 400 and 800°C.

Sol-Gel Method

$Ca(NO_3)_2$ and $Co(NO_3).2.6H_2O$ and citric acid were dissolved in distilled water to a metal ion concentration of 0.2–0.3 mol/liter solution was stirred and heated at 70°C for 3 h. The dried gel was ground and calcined at different temperatures from 400 to 800°C, and sintered using spark plasma sintering (SPS) and conventional sintering (CS) techniques. XRD reveals that CCO appears when calcined at 600°C; as the temperature is increased, a pure phase is obtained at 800°C. TGA reveals peaks at 251°C, 377°C, 627°C, and a heavy one at 763°C; a sharp endothermic peak occurs at 956°C. Electrical conductivity at room temperature for the CS sample is 34S/cm and increases at 700°C. Power factors of CS and SPS are noted as $3.51e^{-4}$ and $1.15\ e^{-4}$ W/m^2s at 700°C. ZT values are noted as 0.16 and 0.052 at 700°C.

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DOPING

Bi Doping

Bi-doped CCO was prepared using the chemical sol-gel method. Bi-doped CCO was prepared using the chemical sol-gel method. $Ca(NO_3)_2.4H_2O + Ca(NO_3)_2.6H_2O + Bi\ (NO_3)_2.5H_2O$. Ethylene glycol and polyethylene glycol were used to polymerize the solution. Nitric acid was added, and the mixture was stirred at 353 K; the gel was prepared and calcined at 923 K for 4 h, and the pellets were sintered at 1233 K for 9 h in a furnace with oxygen flow. Power factors of 22.5 and 23.8 were obtained for concentrations of 0.2 and 0.3 of Bi, respectively. ZT of 0.16 at ZT of 0.16 at 823K (550°C) was obtained for Bi-doped with CCO prepared by the spark plasma sintering method.

Lanthanum Doped

$Ca(NO_3)_2.4H_2O + Ca(NO_3)_2.6H_2O + La(NO_3)_2$ were mixed and dissolved in distilled water. The precursors were calcined in a muffle furnace at 650,700, and 750°C for 5 h to produce CCO powders of CCO/LaCCO 650, which revealed a weak signal corresponding to $CaCo_2O_4$.No other phases were observed, confirming the entry of La elements into the lattice of CCO. The power factor exhibited a positive correlation with the temperature. LaCCO 700-900 had a high power factor of 204.25 $\mu Wm^{-1}K^{-2}$ at 600°C, three times higher than CCO. The improvement in K at 950°C is due to density improvement compared with 900°C samples. The ZT reached 0.25 at 600°C.

Silver Doping

Silver-doped CCO was prepared using ball milling and calcined at 1173 K for 24 h in air twice, with an intermediate grinding. The prepared CCO was mixed with different proportions of AgNPS [0, 0.5, 1.0, 1.5, 2.0]. XRD shows that all the samples are single-phase. As AgNPS content grows higher impurity phase of CCO appears in XRD pattern. The conductivities of pure CCO and AgNPS (0.2) were 74 and 85 S/cm, respectively, at 700 K.

ZT of 0.1 at 700 K was obtained.

Ba, La and Ag Doped

Citrate sol-gel synthesis followed by spark plasma sintering was used to prepare $Ca_{2.9}M_{0.1}Co_4O_9$ [M=Ca, Ba, La, Ag]. Ca, Ba, La, Ag, and Co were dissolved in an aqueous solution of citric acid. Ethylene glycol was added, and the solution was heated under continuous stirring at 358 K to form a precursor gel. The gel was dried at 393 K in air for 24 h and then heated at 1073 K for 8 h. Activation energy.

$SrTiO_3$

In subsequent years, studies based on $SrTiO_3$ materials have expanded in a wide range of fields. A high power factor of 1.8 $Wm^{-1}K^{-1}$ was achieved for heavily La-doped single-crystal strontium titanate (STO), which increased the study of the TE properties of STO. The electronic transport in $SrTiO_3$ can be tuned over a wide range of physical properties. The electrical conductivity of n-type STO materials can be achieved over a very broad range. Effective doping helps in achieving high ZT.

Pr Doped $SrTiO_3$

$SrCO_3$ was mixed with Pr_2O_3 and TiO_2 and formed into pellets, which were calcined at 1300–1400 °C. The pellets were ground, wrapped in graphite foil, and sintered using SPS. Power factors of 1.3 $Wm^{-1}K^{-1}$ and 2 $Wm^{-1}K^{-1}$ are observed at 500°C and 100°C.

La-Doped $SrTiO_3$

La_2O_3 was mixed with TiO_2 and $SrCO_3$ using planetary ball milling for 60 min and calcined at 1100°C for 7 h in an alumina crucible. The pellets were sintered at 1500°C. La-doped $SrTiO_3$ was prepared using SPS at 925, 1100, and 1200°C. A high ZT of 0.2 was reported for the sample sintered and annealed at 500°C. This is due to the high conductivity from 750 Scm^{-1} to 400 Scm^{-1}.

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Nb-Doped $SrTiO_3$

Nb-doped $SrTiO_3$ is prepared by taking $SrCO_3$, La_2Co_3, and TiO_2 as starting materials, mixed using an attrition mill for 60 min. calcined at 1373 K for 3 h. Pellets were prepared using 200 MPa. Sintering in air/N_2. ($Sr_{1-3x/2}$ La_x TiO_3) ZT was calculated for varying x. The highest ZT of 0.41 was calculated for x = 0.125 - 0.175.

Titanium Nitrate and Y

Y-doped $SrTiO_3$ is prepared by calcination of strontium carbonate + yttrium oxide + titanium oxide. Y-doped $SrTiO_3$ and titanium nitride (TIN) powders are mixed. Pelletized under 47 MPa and sintered at 1573 K for 2 h; ZT of 0.26 at 1073 K.

La, Dy

La_2 + $SrCO_3$ +TiO_2 +Dy_2O_3 is mixed, ball-milled for 12 h, dried, pressed, pelletized, and calcined at 1350°C for 6 h in air. The pellets were crushed into fine powders, pelletized again, and sintered at 1460°C for 4 h to obtain $La_{1-x}Dy_{0.1+x}Sr_{0.8}TiO_3$ [x= 0.02, 0.05, 0.08, 0.10]. The highest power factor of 1.9 e^3 was obtained at 303 K.

Rare Earth

The multiple dopant method was also used to enhance the thermoelectric properties of $SrTiO_3$. Rare earth elements Y, La, Sm, Gd, and Dy were doped simultaneously into $SrTiO_3$.

$SrTiO_3$ is mixed with TiO_2, Ti, Y_2O_3, La_2O_3, Sm_2O_3, Gd_2O_3 and Dy_2O_3. All were mixed and made into pellets, then sintered at 80 MPa and 1873 K for 4 h. Thermoelectric studies were conducted on $Sr_{1-X}RxTiO_3$ by varying the concentrations from 0.05 to 0.2. A ZT of $3.8 * 10^{-4}$ k^{-1} was obtained for $Sr_{0.9}Dy_{0.1}TiO_3$ at 573 K.

$CaMnO_3$

Compared with chalcogenides and skutterudites, metal oxides are good candidates for thermoelectric applications. Due to their high thermal and chemical stability at high temperature in air, easy manufacture and low cost. However, ionic bonding in metal oxides usually leads to strong scattering of electrons by optical phonons, which results in low carrier mobility and generally poor thermoelectric performance. Therefore, metal oxides have been avoided in the search for good thermoelectric materials. In 1997, $Na_xCo_2O_4$ showed unexpected thermoelectric properties that made thermoelectric metal oxides highly attractive. Perovskite families have also been explored and considered as thermoelectric oxides. Among them, $SrTiO_3$ is a good TE material, and its TE properties and variations with dopants are mentioned below.

Solid-State Reaction Method

La^{3+}, Y^{3+}, and Ce^{4+} were added to replace Ca^+ in $CaMnO_3$. Multi-dopants were used to yield high efficiency by using a simple solid-state reaction method with $CaCo_3$, MnO_2, La_2O_3, CeO_2, and Y_2O_3 as mixing agents. They were mixed individually and calcined in air at 1273 K for almost 12 h. They were pelletized and sintered at 1573K (1300°C) for 24 h to obtain $Ca_{1-x}La_xMnO_3$, $Ca_{1-x}Ya_xMnO_3$ and $Ca_{1-x}Ce_xMnO_3$. Thermoelectric results were obtained by varying the concentrations. The minimum resistivity was noted for x=0.12 for La- and Y-doped and for x=0.06 for Ce-doped. The largest Seebeck coefficient of 500 to 600 μVk^{-1} was noted for $CaMnO_3$. A large ZT is obtained for $Ca_{0.9}La_{0.1}MnO_3$ at 1000 K.

SAMARIUM DOPED $CaMnO_3$:

Sm-doped $CaMnO_3$ was prepared using the sol-gel method. $Ca_{0.1}Sm_{0.1}MnO_3$ shows a high-power factor of 3.11 $\mu Wm^{-1}K^2$ at 800°C.

Bi- and Nb-Doped

Bismuth- and niobium-doped $CaMnO_3$ was prepared using the solid-state reaction method by taking $CaCO_3$ and MnO_2 as the starting materials and grinding them at 1373 K. The prepared powders were

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mixed with Nb_2O_5 and Bi_2O_3 and calcined at 1373 K. The powders were pelletized and sintered at 1573 K for 12 hrs. $Ca_{1-x}Bi_xMn_{1-y}Nb_yO_3$ was prepared. The maximum power factor was obtained for x=y=0.04 and ZT~ 0.1 at 873 K.

Bi-Doped

By using the planetary ball milling method in an ethanol medium for 5 h, $CaCo_3$, MnO_2, and Bi_2O_3 were calcined at 117 K for 10 h and again annealed at 1273 K for 10 h. The resultant powder was then again ball-milled and pelletized. The pellets were sintered at 1423 K for 10 h, and the results were studied by varying the concentrations [0, 0.02, 0.03, 0.04, 0.06 and 0.10]. ZT of 0.25 is observed at 973 K for x=0.03, and a high power factor of 4×10^{-4} is noted.

CONCLUSION

The global demand for sustainable energy and environmental concerns regarding the use of fossil fuels have imposed an increasing need for the advancement of alternative energy conversion technologies, including thermoelectrics. To improve the performance of thermoelectric materials, we have established in the past years an arsenal of technical tools and methods to achieve low lattice thermal conductivity. There is little overlap between lattice thermal conductivity reduction and power factor improvement; hence, we should pay closer attention to enhancement approaches with dual purposes. In this review, we have discussed various preparation techniques used and the power factors and ZT values obtained. In conclusion, thermoelectric research is an exciting field. It will also provide students of the next generation with invaluable skill sets, including intimate knowledge of phonon and electron scattering. We feel that this review article is very timely and necessary for understanding the power factor of thermoelectric materials and hope that readers will find it very useful in their future research.

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