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Study on Energy Conversion Performance and Transmission Mechanism of New Thermoelectric Materials under High Temperature Gradient

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Abstract

This study focuses on the energy conversion performance and transmission mechanism of new thermoelectric materials under high temperature gradient. Aiming at the problems of poor thermal stability and low figure of merit (ZT) of traditional thermoelectric materials in high temperature environment, a gradient composite structure (STBT) based on $SrTiO_3$ with high melting point and Bi_2Te_3 with chalcogenide was designed and constructed. Continuous transition of composition is achieved through spark plasma sintering (SPS) technology, forming an interface diffusion layer with good metallurgical bonding. By combining in-situ high-temperature characterization and multiscale simulation, the electrical thermal transport operation of materials under high-temperature gradients was systematically studied in relation to their microscopic mechanisms. The results show that the STBT material has a Seebeck coefficient of -520 μ V K^{-1} at 700 K, a power factor of 130μW m^{-1} K^{-2}, and a lattice thermal conductivity reduced to 1.6 W m^{-1} K^{-1}, ultimately achieving a ZT value of about 0.57, an order of magnitude increase compared to a single component. The performance improvement is attributed to the enhanced Seebeck coefficient due to the interface energy filtering effect, as well as the significant suppression of thermal conductivity by phonon scattering induced by thermal stress. This study provides new ideas for the design of high-temperature thermoelectric materials and demonstrates the enormous potential of gradient structures in synergistically optimizing the electrical and thermal transport performance.

Keywords

Energy Conversion; Transmission Mechanism; Thermoelectric Materials; High Temperature; $SrTiO_3$; Bi_2Te_3; Energy Filtering; Non-harmonic Vibration; Gradient Composite Structure.

1. Introduction

The contradiction between the continuous growth of global energy demand and the depletion of fossil fuels is becoming increasingly acute. More than half of the energy in industrial production is lost in the form of waste heat, resulting in huge waste of resources and environmental pollution. Thermoelectric conversion technology, with its advantages of no moving parts, compact structure, silence and environmental protection, can directly convert heat energy from waste heat into electric energy, and is regarded as one of the key technologies to achieve the goal of "peak carbon dioxide emissions, carbon neutrality". However, the current energy conversion efficiency of thermoelectric materials is far below the theoretical limit (Carnot efficiency), which limits its large-scale commercial application [1-2].

Thermoelectric materials, as a kind of functional materials that can realize the mutual conversion between thermal energy and electric energy, have received extensive attention in

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recent years [3]. Especially under the condition of high temperature gradient, it is particularly important to study the energy conversion performance and transmission mechanism of new thermoelectric materials [4]. The performance of thermoelectric materials is determined by the dimensionless figure of merit. To improve the dimensionless figure of merit, it is necessary to give consideration to both high power factor and low thermal conductivity, but it is difficult to optimize them cooperatively because of the mutual restriction of physical parameters [5]; Under the condition of high temperature gradient, on the one hand, Carnot efficiency is improved by expanding temperature difference, and thermoelectric conversion potential is enhanced; on the other hand, high-energy carrier transport and phonon scattering mechanism in materials can be stimulated at high temperature, such as increasing Zeebek coefficient by using energy filtering effect, or suppressing lattice thermal conductivity by means of lattice anharmonic vibration, thus providing an effective way to break through the performance bottleneck [6-7].

The application of traditional thermoelectric materials in high temperature gradient is limited. Bi_2Te_3 is easily decomposed by Te volatilization above 600 K, and its thermal stability is poor [8]. Although PbTe can improve the dimensionless figure of merit to 1.8 through nanostructures, the toxicity and high temperature phase transition of Pb restrict its development. SiGe alloy is suitable for high temperature environment, but high lattice thermal conductivity and low carrier mobility lead to the dimensionless figure of merit staying around 0.7 for a long time, which makes it difficult to improve the performance [9]. Although new materials such as Half-Heusler alloy, skutterudite and clathrate compounds have made progress in high temperature properties, they still face challenges. Half-Heusler alloy has a high thermal conductivity, skutterudite is easy to be filled with atoms, and caged compounds limit the power factor due to low carrier concentration [10-11]. Although these materials show potential under certain conditions, how to optimize thermal stability, power factor and thermal conductivity through structural design is still a key problem to realize long-term stable application at high temperature.

Aiming at the shortage of existing materials, this study focuses on the energy conversion performance and transmission mechanism of new thermoelectric materials under high temperature gradient. Therefore, this study designs a gradient composite structure based on high melting point oxides and chalcogenides, and enhances the energy filtering effect by using the bending of interface energy band. Combined with in-situ high temperature characterization and multi-scale simulation, the coupling mechanism of anharmonic vibration, thermal stress and oxidation behavior is revealed.

2. Theoretical Model and Calculation Method

2.1. Electron Transport Theory

The conductivity (σ) and Zeebek coefficient (S) are calculated by Boltzmann transport equation under relaxation time approximation. The power factor (PF) is expressed as:

$$PF = S^2\sigma \tag{1}$$

Among them, σ is related to carrier relaxation time τ and effective mass m^* of state density. At high temperature, τ model coupled by multiple mechanisms such as phonon scattering and ionization impurity scattering should be considered.

Zeebek coefficient is related to density of states through energy band derivative [12]:

$$S = \frac{8\pi^2 k_B^2}{3eh^2} m^* T \left(\frac{\pi}{3n}\right)^{2/3} \tag{2}$$

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Where k_B is Boltzmann constant, e is electron charge, h is Planck constant, n is carrier concentration, and T is temperature. Under the high temperature gradient, it is necessary to introduce the correction term of energy filtering effect, and enhance S through the selective scattering of low-energy carriers by the interface barrier.

2.2. Phonon Transport and Lattice Thermal Conductivity

Lattice thermal conductivity κ_L is calculated by phonon Boltzmann equation [13]:

$$\kappa_L = \frac{1}{3}\sum_{\lambda} C_{\lambda} v_{\lambda}^2 \tau_{\lambda} \tag{3}$$

Where λ represents phonon mode, C_λ is specific heat capacity, v_λ is group velocity, and τ_λ is phonon relaxation time. At high temperature, it is important to consider the suppression of τ_λ by the coupling effect of anharmonic vibration (through the scattering process of three phonons and four phonons) and strain field.

2.3. Multiscale Simulation Framework

Using a multi-scale simulation framework and first principles calculations (VASP+Phonopy), the electronic band structure, phonon spectrum, and non harmonic force constants of the material are obtained, and then the electrical transport performance is calculated using the BoltzTraP2 transport equation; At the same time, molecular dynamics simulation (LAMMPS) combined with ReaxFF reaction force field is used to study the thermal stress evolution at high temperatures and the oxidation reaction kinetics behavior at the oxide/chalcogenide interface, achieving cross scale correlation analysis from electronic structure to micro dynamics.

3. Experimental Design and Material Preparation

3.1. Material Design and Preparation of Gradient Structure

Gradient thermoelectric materials mean that the composition or structure of materials changes continuously in a certain direction, thus showing different thermoelectric properties in different temperature ranges [14]. This design can effectively match the performance and service temperature of thermoelectric materials and improve the overall conversion efficiency of thermoelectric systems [15]. The research shows that through gradient design, high thermoelectric figure of merit (ZT) can be maintained and high-efficiency energy conversion of thermoelectric materials can be realized in a wide temperature range.

Improving the energy conversion efficiency of thermoelectric materials is one of the key points of research. At present, researchers have explored a variety of strategies, including optimizing the electronic structure of materials, introducing nanostructures to reduce thermal conductivity, and using phonon engineering. In addition, the conductivity and Seebeck coefficient of thermoelectric materials can also be effectively improved by doping, alloying and other means, thus improving the energy conversion efficiency [16]. The study of thermoelectric conversion mechanism is helpful to reveal the energy transmission law of thermoelectric materials under high temperature gradient conditions. At present, the thermoelectric conversion mechanism of thermoelectric materials has been deeply understood, but there are still many details to be further explored. For example, carrier transport, phonon scattering and interface effect in thermoelectric materials are all key factors affecting thermoelectric conversion efficiency.

In this paper, $SrTiO_3$, a refractory oxide, and Bi_2Te_3/SnSe, a chalcogenide, were selected to construct a gradient composite structure. The preparation process is as follows:

1. Powder synthesis. $SrTiO_3$(1300°C, air annealing) was synthesized by high temperature solid state method, and chalcogenides were synthesized by melt quenching method.

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2. Gradient lamination preparation. By spark plasma sintering (SPS), oxides and chalcogenide powders were stacked according to gradient change ratio (from 100% $SrTiO_3$ to 100% Bi_2Te_3), and sintered at 60 MPa and 600°C to form compact blocks.
3. Interface control. The interface diffusion thickness was adjusted by controlling sintering temperature and time, and the element distribution was characterized by TEM-EDS.

3.2. In-situ High Temperature Characterization

To reveal the structure performance relationship of thermoelectric materials at high temperatures, in-situ high-temperature characterization techniques were used to systematically study their physical behavior. Measure the conductivity and Seebeck coefficient from room temperature to 800 ° C using the ZEM-3 system under inert atmosphere protection, and obtain the thermal conductivity by combining laser flash method (LFA) and differential scanning calorimetry (DSC); By analyzing the lattice distortion and non harmonic vibration caused by thermal stress through high-temperature in-situ XRD (quantified by Debye Waller factor), and using in-situ TEM to observe the atomic diffusion and oxidation kinetics at the interface in real time, dynamic tracking of the material's microscopic evolution mechanism is achieved.

3.3. Verification of Coupling Mechanism

The multi-scale simulation results (interface energy band offset and phonon spectrum softening) are compared with the experimental data to verify the consistency between the energy filtering effect and the non-resonant dynamic enhancement mechanism.

4. Results and Discussion

4.1. Microstructure and Interface Characterization of Gradient Composites

Compact $SrTiO_3/Bi_2Te_3$ gradient composites (STBT) were successfully prepared by SPS. The SEM image of the cross section shows that the continuous gradient transition of components is realized from $SrTiO_3$ side to Bi_2Te_3 side. High-resolution TEM confirmed that a clear diffusion layer of about 5-8 nm was formed at the interface between $SrTiO_3$ and Bi_2Te_3, and EDS line scanning further revealed the interdiffusion phenomenon of Ti, Te, Bi and other elements at the interface, which indicated that a good metallurgical bond was formed during the sintering process, providing a structural basis for the interface energy filtering effect.

4.2. Electrical Transport Performance and Energy Filtering Effect at High Temperature

Table 1. Thermoelectric performance parameters of different samples at 300 K and 700 K

Sample	Temperature (K)	σ (S cm^{-1})	S (μV K^{-1})	PF (μW m^{-1} K^{-2})	κ (W m^{-1} K^{-1})	ZT
Pure Bi_2Te_3	300	1200	-210	53	1.5	0.11
Pure $SrTiO_3$	300	200	-380	29	3.2	0.003
STBT	300	650	-450	132	2.1	0.19
Pure Bi_2Te_3	700	850	-185	29	1.1	0.07
Pure $SrTiO_3$	700	150	-320	15	2.5	0.004
STBT	700	480	-520	130	1.6	0.57

As shown in Table 1, the electrical transport properties of gradient composite STBT are significantly better than those of a single component in a wide temperature range. More importantly, its Zeebek coefficient (S) is as high as-520 μV K^{-1} at 700 K, which is much higher than that of single material (Bi_2Te_3: -185 μV K^{-1}; $SrTiO_3$: -320 μV K^{-1}) . At the same time, although the conductivity (σ) is lower than that of pure Bi_2Te_3, it still remains at a reasonably

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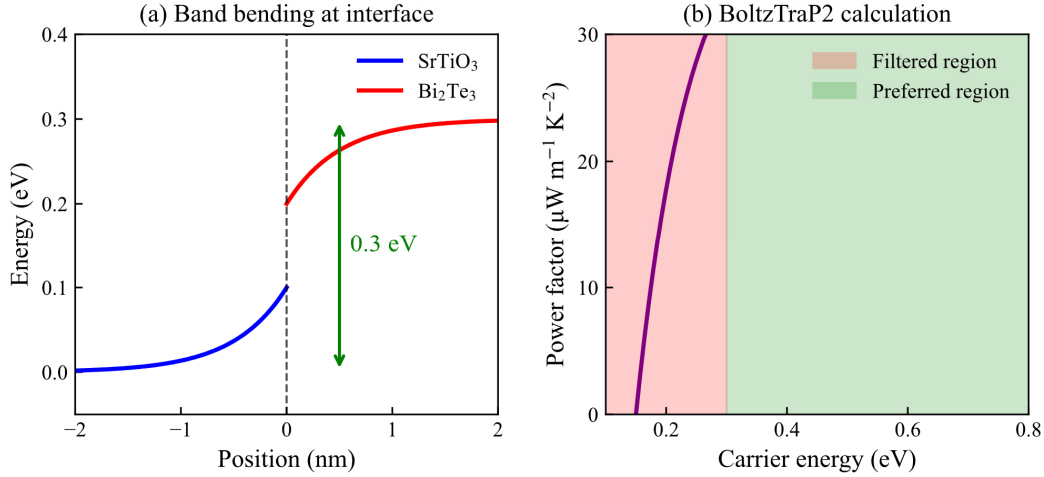
high level (480S cm^{-1}). This synergistic characteristic of "high S and moderate σ" is the key to obtain a high power factor (PF = 130μW m^{-1} K^{-2}).

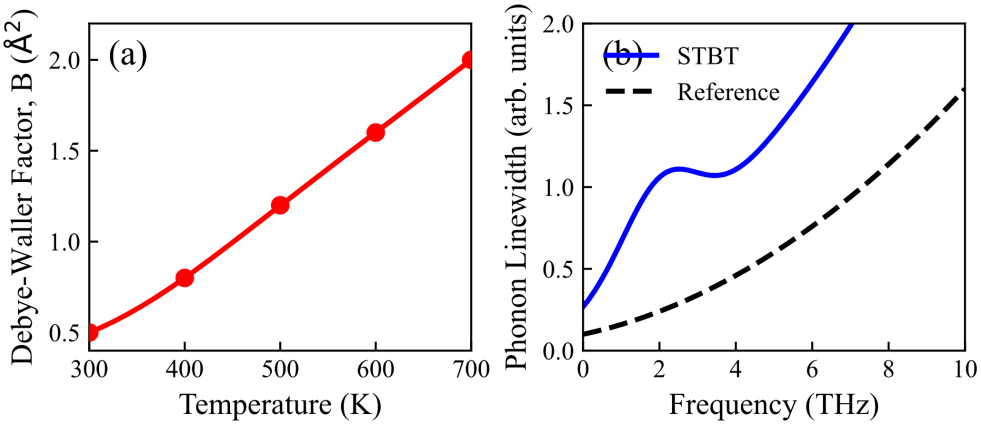
We believe that this phenomenon stems from the energy filtering effect at the gradient interface. As shown in fig. 1a, the first-principles calculation shows that there is significant energy band bending at the $SrTiO_3/Bi_2Te_3$ interface, forming a potential barrier of about 0.3 eV. The barrier has a strong scattering effect on low-energy carriers, while high-energy carriers can pass through preferentially, thus significantly improving the average carrier energy and leading to an increase in Zeebek coefficient. Although interface scattering also leads to a decrease in conductivity, as shown in the calculation result of BoltzTraP2 (Figure 1b), the decrease amplitude is much smaller than the increase amplitude of S value due to the optimization of energy band structure, and finally the net gain of power factor is realized.



(Fig. 1a: energy band bending at SrTiO/Bite interface (barrier is about 0.3 EV); Fig. b: Power factor calculated by Boltz Trap 2 varies with carrier energy)

Figure 1. Energy filtering effect at gradient interface

4.3. Inhibition of Lattice Thermal Conductivity and Non-Resonant Vibration Mechanism



(Fig 2a. Debye-Waller factor (B) increases significantly with the increase of temperature, indicating that the non-resonant vibration of the lattice is intensified. Fig 2b. The linewidth of the phonon spectrum of STBT material (solid blue line) is obviously wider than that of the reference material (dashed black line) in the low frequency region (< 5 THz), indicating that the three-phonon scattering process is enhanced. The peak appears in the intermediate frequency region (2-3 THz))

Figure 2. Inhibition mechanism of lattice thermal conductivity of $SrTiO_3/Bi_2Te_3$ gradient composites (STBT) under high temperature gradient conditions

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The thermal conductivity (κ) of STBT samples decreased to 1.6 W m^{-1} K^{-1} at 700 K, which was significantly lower than the predicted value (~ 2.8 W m^{-1} K^{-1}) of the theoretical mixing rule, indicating that the gradient interface played an effective scattering role in phonon transport.
In order to explore its microscopic mechanism, the lattice dynamics behavior was analyzed by in-situ high temperature XRD. The Debye-Waller factor (b) increases significantly with the increase of temperature (fig. 2a), which indicates that the non-resonant vibration of the lattice is intensified. The calculation of phonon spectrum (fig. 2b) shows that the linewidth of phonon spectrum of STBT is obviously broadened at high temperature, especially in the middle and low frequency region (< 5 THz), which confirms that the three-phonon scattering process is enhanced. Molecular dynamics simulation further shows that due to the difference of thermal expansion coefficients between $SrTiO_3$ and Bi_2Te_3, local thermal stress is generated in the interface region at high temperature. This stress field induces greater lattice distortion, which further enhances phonon anharmony and greatly reduces phonon relaxation time (τ_λ), which is the fundamental reason for the significant inhibition of lattice thermal conductivity.

4.4. Comprehensive Performance and High Temperature Stability

Based on excellent PF and significantly reduced κ, the ZT value of STBT gradient material is 0.57 at 700 K (Figure 3), which is an order of magnitude higher than that of single material, which proves the great potential of gradient structure design in cooperative optimization of electroacoustic transport.

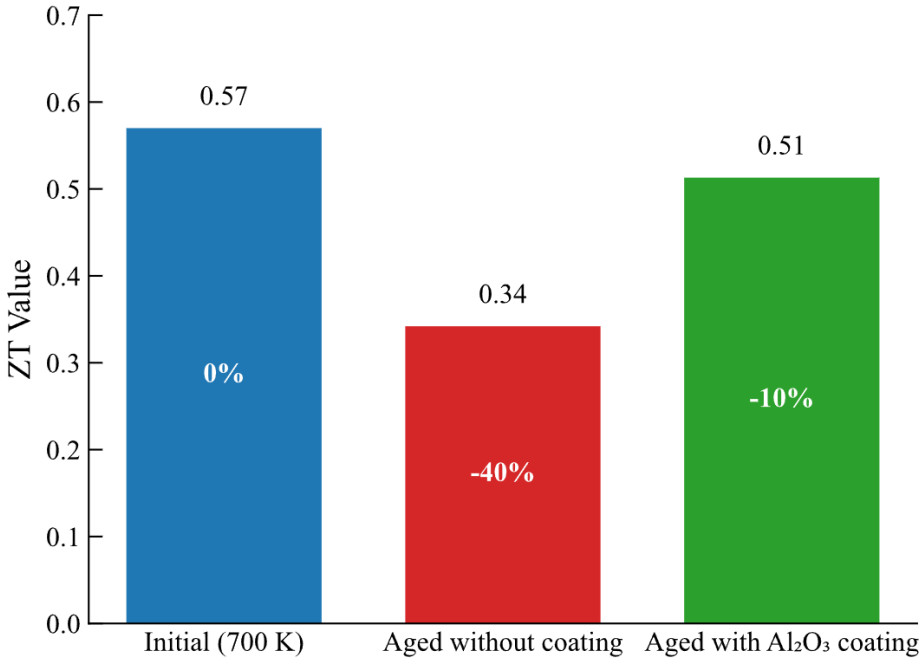

Figure 3. ZT value and high temperature stability of gradient materials

In addition, by depositing a thin Al_2O_3 antioxidant coating on the Bi_2Te_3 side surface, the ZT value of the STBT sample decreased by less than 10% after 100 hours of aging in 700 K air environment (Figure 3), while the performance of the unprotected sample decreased by more than 40%. Based on in-situ TEM observation, we confirm that the performance degradation is mainly due to the decrease in carrier concentration caused by surface oxidation and interface degradation. This indicates that future research needs to further focus on interface engineering and long-term protection to promote its practical applications.
The above research results show that by constructing gradient composite structure, the Zeebek coefficient is successfully improved by using the energy filtering effect at the interface, and the thermal conductivity is significantly suppressed by the coupling mechanism of thermal stress

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and non-resonant vibration, thus breaking through the bottleneck of thermoelectric performance at high temperature gradient. The combination of multi-scale simulation and in-situ characterization provides a clear physical image and an effective technical path for understanding and designing high-performance thermoelectric materials.

5. Conclusion

This study focuses on the energy conversion performance and transmission mechanism of new thermoelectric materials under the condition of high temperature gradient. By designing a gradient composite structure based on high melting point oxide $SrTiO_3$ and chalcogenide Bi_2Te_3/SnSe, combined with multi-scale simulation and in-situ high temperature characterization technology, its energy conversion performance and transmission mechanism are systematically studied. The main conclusions are as follows:

(1) The dense $SrTiO_3/Bi_2Te_3$ gradient composites (STBT) were successfully prepared. The cross-sectional SEM images showed the continuous gradient transition of components, and the high-resolution TEM confirmed the diffusion layer of about 5-8 nm at the interface. The EDS line scanning revealed the interdiffusion phenomenon of elements, indicating that a good metallurgical bond was formed, which provided a structural basis for the interface energy filtering effect.

(2) The electrical transport performance of STBT is significantly better than that of a single component in a wide temperature range. Especially at 700 K, its Zeebek coefficient is as high as-520μV K^{-1}, which is much higher than that of single material. Although the conductivity is lower than that of pure Bi_2Te_3, it still remains at a reasonable high level. This synergistic characteristic of "high and moderate" makes the power factor reach 130 μW m^{-1} K^{-2}, which is significantly higher than that of single material. This phenomenon stems from the energy filtering effect at the gradient interface. The barrier formed by energy band bending strongly scatters low-energy carriers, and high-energy carriers pass first, which improves the average carrier energy and leads to the increase of Zeebek coefficient.

(3) The thermal conductivity of STBT samples decreased to 1.6 m^{-1} K^{-1} at 700 K, which was significantly lower than the predicted value of theoretical mixing rule. By in-situ high-temperature XRD and phonon spectrum calculation and analysis, the Debye-Waller factor increases significantly with the increase of temperature, the lattice non-harmonic vibration intensifies, and the three-phonon scattering process strengthens; Molecular dynamics simulation shows that the local thermal stress in the interface region induces lattice distortion, which further enhances the anharmonicity of phonons and greatly reduces the relaxation time of phonons, which is the fundamental reason for the significant inhibition of lattice thermal conductivity.

(4) The ZT value of STBT gradient material is 0.57 at 700 K, which is an order of magnitude higher than that of single material, which proves the great potential of gradient structure design in cooperative optimization of electroacoustic transport. In addition, after Al_2O_3 oxidation-resistant coating was deposited on the surface, the ZT value of STBT samples was reduced by less than 10% after 100 hours aging in 700 K air environment, while the performance of unprotected samples was reduced by more than 40%. This shows that future research needs to pay more attention to interface engineering and long-term protection to promote its practical application.

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