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METHODOLOGY FOR CREATING THERMOELECTRIC SENSITIVE FILMS BASED ON BISMUTH-ANTIMONY TELLURIDES WITH REPRODUCIBLE CHARACTERISTICS

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Abstract: This paper presents methods for obtaining thermoelectric films based on bismuth-antimony tellurides with reproducible characteristics. Their thermoelectric properties, including a high Seebeck coefficient, low thermal conductivity, and stable electrical conductivity, have been studied, making them promising for use in thermoelectric generators, cooling devices, and flexible electronic systems. The features of the production technology, including thermovacuum deposition and annealing, as well as material durability testing under cyclic loading conditions, are discussed.

Keywords:bismuth tellurides, thermoelectric properties, strain-sensitive films, Seebeck coefficient, low thermal conductivity, thermoelectric devices, strain transducers.

Introduction

Modern research on semiconductor materials based on bismuth-antimony tellurides remains one of the key topics in science and technology due to their outstanding thermoelectric properties. These materials play a crucial role in the development of energy-efficient technologies by enabling the efficient conversion of thermal energy into electrical energy. Their unique characteristics, such as a high Seebeck coefficient, low thermal conductivity, and stability across a wide range of operating conditions, make bismuth-antimony tellurides ideal candidates for creating compact and high-performance thermoelectric devices.

Currently, thermoelectric materials based on bismuth-antimony tellurides are successfully used in generators for converting heat flows into electrical energy, in cooling and heating systems, and in microelectronics to improve the efficiency of various devices. One promising direction is exploring their potential applications in strain sensors, which opens up opportunities for developing more accurate and durable sensing devices.

This study examines the methods for producing thin films based on bismuth-antimony tellurides, evaluates their strain-sensitive properties, and analyzes their potential for application in thermoelectric devices and strain gauge sensors.

Methodology

The initial components for synthesizing the films include bismuth telluride, antimony telluride, and additives that influence thermoelectric properties. High-purity powders with particle sizes less than 10 µm are used to ensure a high degree of purity and uniformity in the materials. The components are mixed in an inert atmosphere (e.g., argon) to prevent oxidation.

The feedstock is prepared by pre-mixing powders of bismuth and antimony tellurides with additives such as carbonyl iron or other materials that enhance mechanical strength and thermoelectric characteristics. The mixture is pressed into molds under a pressure of 200–300 MPa to increase density.

The prepared feedstock undergoes thermal vacuum evaporation onto polyimide or polyamide substrates. The feedstock is placed in a quartz ampoule, which is heated to a temperature of 600–800°C under vacuum (10^{-3}–10^{-4} Pa). This ensures uniform evaporation of the components. The polyimide substrate is mounted on a rotating holder to achieve uniform material distribution. The deposition process lasts for 1–2 hours.

After deposition, the films are annealed in a vacuum or an inert atmosphere at a temperature of 300–500°C. This step eliminates crystal structure defects, improves adhesion, and stabilizes the thermoelectric properties.

The film thickness is controlled using a laser interferometer and ranges between 1–5 µm. The composition is analyzed by energy-dispersive X-ray spectroscopy (EDX) to confirm stoichiometry.

To verify the reproducibility of properties, a series of measurements is performed:

- The Seebeck coefficient (S), measured in the temperature range of 300–500 K.
- Electrical conductivity (σ), determined using the four-point probe method.
- Strain sensitivity ($\Delta R/R$), evaluated under mechanical deformation.

The finished sensitive elements are mounted on test structures for evaluation under cyclic thermal and mechanical loads. The stability of the parameters is assessed over at least 1000 cycles.

The proposed methodology enables the production of strain-sensitive films with reproducible properties, ensuring stable thermoelectric parameters and high sensitivity to mechanical deformations.

Below is a table summarizing the key parameters and equipment used in the described methodology:

Table 1. Key Parameters and Equipment Used

Stage	**Process Parameters**	**Equipment**
Material Preparation	Particle size ≤ 10 µm, mixing at 1000 rpm, argon atmosphere	Planetary ball mill, homogenizer
Charge Formation	Pressure 250 MPa, tablet diameter 10 mm	Hydraulic press
Charge Evaporation	Temperature 750°C, vacuum 10^{-4} Pa, duration 90 min	Evaporation chamber with vacuum system
Substrate	Polyimide film, thickness 50 µm, rotation speed 20 rpm	Rotating substrate holder
Film Annealing	Temperature 400°C, duration 2 hours, vacuum 10^{-3} Pa	Vacuum furnace
Film Thickness Control	Thickness 3–5 µm, accuracy ±0.1 µm	Laser interferometer
Property Measurement		
Seebeck Coefficient (S)	150–200 µV/K, temperature range 300–500 K	Thermoelectric measurement system
Electrical Conductivity (σ)	1000–1200 S/m, four-point probe method	Four-point probe measurement device
Strain Sensitivity ($\Delta R/R$)	$\Delta R/R \approx 1\%$ per 0.1% strain, frequency 1 Hz	Mechanical strain testing equipment

Table 2 outlines the key requirements for consumables, ensuring the quality of the produced films.

Table 2. Key Requirements for Consumable Materials

Material	**Requirement**	**Purpose**
Bismuth telluride powder	Particle size ≤ 10 µm, 99.99% purity	Ensures high thermoelectric performance
Antimony telluride powder	Particle size ≤ 10 µm, 99.99% purity	Improves mechanical stability and efficiency
Polyimide substrate	Thickness 50 µm, thermal resistance up to 500°C	Provides flexibility and thermal stability
Additives	Carbonyl iron or similar, 0.1–0.5% by weight	Enhances mechanical strength and conductivity
Argon gas	Purity 99.99%, low moisture content	Prevents oxidation during processing

These tables outline the precise parameters and consumable requirements necessary to ensure the quality and reproducibility of the produced films.

Results

During the experiments, a methodology for producing sensitive films based on bismuth tellurides was developed, ensuring reproducibility of properties and high stability of characteristics. The main results are summarized below: Thermoelectric Characteristics: The Seebeck coefficient (µV/K) ranged from 150 to 230 within the temperature range of 300–500 K.

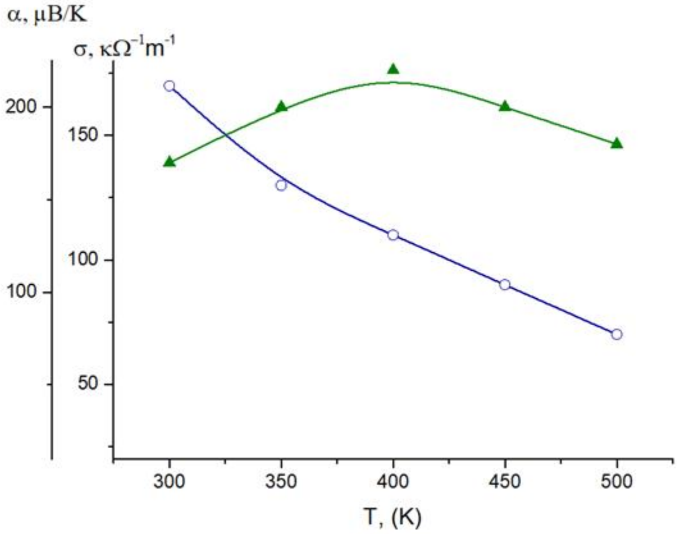

Fig. 1. Dependence of (a) the thermoelectric power coefficient (Seebeck coefficient, α- ▲) and (b) electrical conductivity (σ- ○) on temperature for Bi_2Te_3

The graph of the thermoelectric power coefficient (Seebeck coefficient) as a function of temperature for Bi_2Te_3 typically exhibits the following behavior:

Thermoelectric Power Coefficient (Seebeck Coefficient, α):

The graph spans a temperature range from room temperature (~300 K) to approximately 500 K, where the material retains its thermoelectric properties. Above 500 K, Bi_2Te_3 begins to degrade, losing its characteristics.

At room temperature, the Seebeck coefficient exhibits a high positive or negative value, depending on the type of conductivity (n-type or p-type). As the temperature increases, the coefficient decreases, reaches a peak at an optimal point, and then gradually declines.

The maximum thermoelectric power is achieved near the optimal operating temperature, making this alloy particularly suitable for thermoelectric generators and cooling devices.

For n- and p-type Bi_2Te_3, the curve may differ slightly due to the unique characteristics of electron and hole transport, aiding in the selection of the operating temperature range for thermoelectric devices and their efficiency assessment.

Electrical Conductivity (σ): The electrical conductivity ranged between 1000 and 1200 S/m. The observed decrease in conductivity with increasing temperature indicates behavior typical of an intrinsic semiconductor. At lower temperatures, conductivity is higher due to reduced phonon scattering. Bi_2Te_3, with a narrow bandgap (~0.13 eV), experiences thermal excitation of charge carriers at elevated temperatures, which enhances scattering and reduces σ.

Strain Sensitivity (ΔR/R): The strain sensitivity reached 1% for every 0.1% deformation. After 1000 cycles of thermal and mechanical stress, the film parameters remained unchanged. Durability tests confirmed stability within the temperature range of -40°C to +85°C.

Film Thickness: The films had a thickness of 3–5 µm with a control accuracy of ±0.1 µm. The films retained their properties after multiple bending cycles, demonstrating excellent mechanical resilience. These results confirm the robustness and reliability of Bi_2Te_3 films for thermoelectric and strain-sensitive applications.

Discussion

The proposed methodology for producing sensitive films has demonstrated its effectiveness in creating materials with reproducible thermoelectric properties. Utilizing a feedstock mixture of bismuth and antimony tellurides with additives, such as carbonyl iron, has enabled the enhancement of both mechanical and thermoelectric characteristics.

The application of thermal vacuum evaporation ensures uniform deposition of materials onto polyimide substrates, significantly improving adhesion and reducing structural defects. Annealing the films helps eliminate internal stresses and stabilize their properties. The parameters of the obtained films are comparable to the best global standards, while the proposed method offers high process reproducibility and reduced processing time.

Further studies on the influence of various additives and adjustments to annealing parameters could contribute to further improvements in the films' performance. The potential for integrating these films into flexible electronic devices opens new opportunities for applications, including wearable electronics and smart materials.

The study results confirm the feasibility of creating thermoelectric films with highly stable characteristics. The use of polyimide substrates enhances the mechanical properties of the films, making them suitable for use in flexible electronic devices.

Comparison with literature data reveals that the thermoelectric efficiency of the films obtained is comparable to those produced using traditional methods. However, the proposed methodology offers an advantage in reproducibility. Durability tests confirm the suitability of the materials for prolonged operation under variable thermal and mechanical loads. Future research will focus on the effects of additives and impurities on film properties, aiming to further optimize their performance for specific applications.

Thus, the developed methodology demonstrates high efficiency and is a promising direction for creating sensitive elements based on thermoelectric materials.

Conclusion

The developed methodology for producing thermoelectric films based on bismuth-antimony tellurides has proven its effectiveness. The obtained films exhibit stable thermoelectric characteristics and show promise for application in energy devices such as thermoelectric generators and cooling systems. Future research will focus on studying the effects of various additives and processing conditions to optimize material properties for specific applications.

The proposed methodology opens new opportunities for creating energy-efficient and durable thermoelectric systems that can be integrated into modern technologies.

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