

MINING AND METALLURGY INSTITUTE BOR and TEHNICAL FACULTY BOR, UNIVERSITY OF BELGRADE







# PROCÉEDINGS

Editors: Ana Kostov Milenko Ljubojev

3 – 5 October 2022. Hotel "Albo" Bor, Serbia

#### MINING AND METALLURGY INSTITUTE BOR

and



TEHNICAL FACULTY BOR, UNIVERSITY OF BELGRADE



53<sup>rd</sup> International October Conference on Mining and Metallurgy

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#### ELECTRONIC TRANSPORT PROPERTIES OF THE Bi<sub>0.5</sub>As<sub>1.5</sub>Te<sub>2.98</sub>Se<sub>0.02</sub> SINGLE CRYSTAL: PART II

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#### Abstract

This work present the effect of arsenic on the BiTeSe bulk single crystal synthesis by the Bridgman method.

Bulk single crystal growth was achieved by the spontaneous nucleation. The bismuth telluride thermoelectric material with the composition of  $Bi_{0.5}As_{1.5}Te_{2.98}Se_{0.02}$  was obtained. During the experiment, the values of conductivity ( $\sigma$ ), resistivity ( $\rho$ ), Hall coefficient ( $R_{H}$ ), magnetic resistance ( $\Delta R$ ) and vertical/horizontal resistance ratio ( $\alpha$ ) were studied using the Hall Effect system, based on the Van der Pauw method. The Hall Effect was measured at temperature of liquid nitrogen with silver contacts with the applied magnetic field strength of 0.37 T at current intensities of 1, 5 and 10 mA.

Keywords: electronic transport properties, Hall and Van der Pauw method, single crystal

#### **1 INTRODUCTION**

Electrotechnical and electronic materials are applied in electrical engineering and electronics. The development of technique requires finding the new materials with predetermined requirements regarding their properties [1]. That is why a lot of work is being done on the improvement of already existing and development the new technological procedures. The study of the semiconductors properties saw an upswing with the production of sufficiently pure semiconductor materials. Certain elements such as: silicon, germanium, selenium, tellurium, boron, etc. possess the semiconductor properties. Semiconductors are defined on the basis of value of their specific electrical resistance and the energy gap value, eg. values of specific electrical resistance for the semiconductors range from  $10^{-6} \Omega m$  to about  $10^{10} \Omega m$ . By doping, a usable semiconductor can be created. The ssemiconductors compounds were obtained by combining the trivalent and pentavalent elements. The II-VI compounds, made by combining the stoichiometric ratio of divalent and hexavalent elements, were also studied a lot. Compounds of the type  $V_2VI_3$ ,  $III_2VI_3$ ,  $II_2V_3$  were developed and studied, too.

Actually, the used thermoelectric (TE) material at room temperature is Bi<sub>2</sub>Te<sub>3</sub>.

In order to evaluate the performance of the  $Bi_2Te_3$  thermoelectric material, the energy conversion efficiency is evaluated by a figure of merit, ZT [ $K^1$ ].

The figure of merit (ZT) is defined as:

$$ZT = \frac{\sigma S^2}{\kappa} * T \tag{1}$$

where T is the temperature, S is the Seebeck coefficient,  $\sigma$  is the electronic conductivity and k is the total thermal conductivity which is the sum of electronic (k<sub>e</sub>) and lattice



thermal ( $k_L$ ) conductivities [2]. The performance of thermoelectric materials is quantified by a dimensionless thermoelectric figure of merit, ZT, which is defined by the Seebeck coefficient, electric conductivity, and thermal conductivity. A challenge to create the high ZT thermoelectric materials lies in achieving simultaneously high  $\sigma$ , high S, and low  $\kappa$ . There is a strong correlation of these three parameters.

The great efforts have been devoted to the research of thermoelectric materials.

Thermoelectric (TE) materials can play an important role in reducing the carbon emission by converting the waste heat into electricity [3]. Bismuth telluride and its alloys are the best-known TE materials for the ambient temperature applications [4-8].

This work is focused on the electronic transport properties of  $Bi_{0.5}As_{1.5}Te_{2.98}Se_{0.02}$ single crystal. The first measurements were conducted at temperature of liquid nitrogen with silver contacts and at currents of: 0.05; 0.1 and 0.5 mA. Also, this work gives a report of the values of conductivity ( $\sigma$ ), resistivity ( $\rho$ ), Hall coefficient ( $R_H$ ), magnetic resistance ( $\Delta R$ ) and vertical/horizontal resistance ratio ( $\alpha$ ) using a Hall Effect system on the basis of the Van der Pauw method at current intensities of 1, 5 and 10 mA.

Herein, As-doping effects is reported on the electronic transport properties of a single crystal bulk.

#### **2 EXPERIMENTAL**

Bulk single crystal ingot of  $Bi_{0.5}As_{1.5}Te_{2.98}Se_{0.02}$  was synthesized using the Bridgman method by spontaneous nucleation. The maximum synthesis temperature was of about 600°C. High purity elements (5 N) were used as the source material. Bismuth (Sigma – Aldrich, 99.999%), arsenic (Koch-Light Laboratories Ltd Colnbrook Bucks England, 99.999%), Tellurium (Sigma – Aldrich, 99.999%) and selenium (Alfa Aesar, 99.999%) were taken in a proportion 2:3.

The temperature gradient was 2°C/mm in the zone of heating. In the zone of cooling, the temperature gradient was 5°C/mm. The ingot was grown at the speed of 2.2 mm/h.

The Hall effect measurements were conducted using the Hall Effect Measurement System Ecopia HMS-3000 at the Faculty of Technical Sciences, University of Novi Sad in Serbia.

Software for the Hall Effect measurement system (Ecopia, HMS-3000) automatically calculated the conductivity, resistivity, A-C cross Hall coefficient, B-D cross Hall coefficient, magnetic resistance and vertical/horizontal resistance ratio. Calculations were done on the basis of voltage, obtained by the Van der Pauw laws and input data was entered into the software (sample thickness D, current intensity I, the magnetic induction of permanent magnet B). The measured samples were cleaned in acetone before they are used for measurements.

#### **3 RESULTS AND DISCUSSION**

Samples used for the Hall effect measurements were prepared to be in the form of thin disc cut perpendicular to the long axis of a single crystal ingot. All samples were carefully inspected for cavities and scratches and polished if necessary. All measurements were carried out at temperature of liquid nitrogen with silver contacts. The source of magnetic field applied perpendicular to the Hall element was a permanent magnet of 0.37 T. The Hall effect measurements were done to obtain the electronic transport properties.



**Table 1** Results of the Hall and Van der Pauw method for sample 5/6 (⊥) at temperature of liquid nitrogen with silver contacts

Current intensity <i>I</i> [mA]	Conductivity σ [1/Ωcm]	Resistivity ρ [Ωcm]	A-C Cross Hall coefficient R <sub>H1</sub> [cm <sup>3</sup> /C]	B-D Cross Hall coefficient R <sub>H2</sub> [cm <sup>3</sup> /C]	Magnetic resistance $\Delta R [\Omega]$	Vertical/ Horizontal resistance ratio α
1	$1.154 \text{x} 10^3$	8.669x10 <sup>-4</sup>	$-2.429 \times 10^{0}$	7.758x10 <sup>-1</sup>	1.439x10 <sup>-3</sup>	-2.321x10 <sup>-1</sup>
5	$1.294 \times 10^{3}$	7.730x10 <sup>-4</sup>	2.841x10 <sup>-1</sup>	-8.974x10 <sup>-1</sup>	3.498x10 <sup>-4</sup>	$-1.242 \times 10^{0}$
10	$2.956 \times 10^3$	3.383x10 <sup>-4</sup>	-1.787x10 <sup>-1</sup>	3.820x10 <sup>-1</sup>	5.109x10 <sup>-4</sup>	6.103x10 <sup>-1</sup>

The electronic transport properties of  $Bi_{0.5}As_{1.5}Te_{2.98}Se_{0.02}$  single crystal were measured. The results of electronic transport properties i.e. conductivity, resistivity, A-C cross Hall coefficient, B-D cross Hall coefficient, magnetic resistance and vertical/horizontal resistance ratio for the studied sample 5/6 ( $\perp$ ) is given in Table 1.

Sample, tested by the Hall and Van der Pauw method, was cut from a part of the ingot normally to the crystallization direction ( $\perp$ ). In the following, this sample will be referred to as 5/6 ( $\perp$ ). The sample 5/6 ( $\perp$ ) of circular cross-section is 1.55 mm thick.

Measurements were made at currents of: 1; 5 and 10 mA. The values of the A-C cross Hall coefficient at current intesity of 1 and 10 mA are negative, indicating that the samples are n-type and that the majority of charge carriers are electrons. For the current intesity of 5 mA, the value of the A-C cross Hall coefficient is positive, indicating that the sample is of a p-type and that the majority of charge carriers are hols. The values of the B-D cross Hall coefficient at current intesity of 1 and 10 mA are positive for the same sample, indicating that sample is of a p-type and that the majority of charge carriers are hols. For the current intesity of 5 mA the value of the game sample, indicating that sample is of a p-type and that the majority of charge carriers are hols. For the current intesity of 5 mA the value of the B-D cross Hall coefficient is negative, indicating that samples are of a n-type and that the majority of charge carriers are electrons.

#### **4 CONCLUSION**

The main objective of this study was to obtain  $(BiAs)_2$  (TeSe)<sub>3</sub> bulk single crystal by the spontaneous nucleation. The crystal was characterized with the Hall and Van der Pauw method and grown using the Bridgman method.

After added a small amount of As-doping on Bi sites, the electronic transport properties could not be enhanced.

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