# Synthesis and investigation of BiTeSe single crystal doped with As obtained using bridgman method

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***Abstract:*** *Researches in this paper included synthesis and characterization of bismuth telluride single crystal doped with arsenic, obtained using Bridgman method. Compounds based on bismuth telluride are very important materials for thermoelectric refrigerators and devices for electricity production. For the monocrystal characterization, SEM - EDS, Hall and Van der Pauw method were used. The results presented in paper show the synthesis of monocrystal ingot, BiTeSe doped with arsenic. An analysis of energy dispersive spectrometry (EDS) was used to determine the chemical composition of the samples studied, as well as checking and confirming the homogeneity of the samples. Measurements of X-ray diffraction (XRD) showed that the resulting crystalline ingot represent a single crystal and confirm the compound of $Bi_2Te_3$ type. Mobility, concentration, resistivity/conductivity, of majority of charge bearers and Hall coefficient of single crystal, were determined using a Hall Effect measurement system based on the Van der Pauw method. For the sample of BiTeSe doped with arsenic Hall effect was measured at room temperature with an applied magnetic field strength of 0.37 T at different current intensities. Further characterization of the BiTeSe sample doped with arsenic was not performed, because the expected improvement in the mobility of this sample in comparison with the theoretical value of the n type $Bi_2Te_3$, was not obtained.*

***Keywords:*** *BISMUTH AND TELLURIUM SINGLE CRYSTAL, SEMICONDUCTOR, BRIDGEMANN METHOD, DOPING, SEM - EDS, HALL AND VAN DER PAUW METHOD*

## *1. Introduction*

Thermoelectric transport properties of doped $Bi_2Te_3$ have been examined for a long time [1 - 7].

Special attention has been paid to increase the thermoelectric figure of merit (ZT) for enabling the widespread use of this method for directly converting heat into electricity [8 - 12].

A parameter that evaluates the quality of thermoelectric materials, thermoelectric figure of merit, *Z*, is determined by the dimensionless value, *ZT*, [13 -15] which is defined as:

$$ZT = \frac{S^2 \cdot \sigma \cdot T}{k} = \frac{S^2 \cdot T}{k \cdot \rho} = \frac{S^2 \cdot T}{(k_e + k_l) \cdot \rho}$$

where: $S$ - Seebeck coefficient, $\sigma$ - electrical conductivity, $k$ - thermal conductivity, $T$ - absolute temperature, $\rho$ - electrical resistance.

Thermal conductivity has two components: electronic conductivity, $k_e$, and lattice conductivity, $k_l$. The ratio $\frac{S^2}{\rho}$ is defined as the power factor and determines the electrical properties. Combinations of material properties required for thermoelectric materials to have quality and usable properties are also a challenge for scientists.

## *2. Experimental*

Single crystal ingot of $Bi_2Te_3$ doped with arsenic was synthesized using the Bridgman method at the maximum temperature of about 600ºC. High purity elements (5 N) were used as the source material. Tellurium (Sigma – Aldrich, 99.999%), bismuth (Sigma – Aldrich, 99.999%), selenium (Alfa Aesar, 99.999%) and arsenic (Koch-Light Laboratories Ltd Colnbrook Bucks England, 99.999%) were taken in a certain proportion.

The temperature gradient was 2ºC/mm in the zone of heating and 5ºC/mm in the zone of cooling. The ingot was grown at the speed of 2.2 mm/h.

The content of Bi, Te, Se and As in sample was obtained using Energy Dispersive X-ray Spectroscopy (EDS) analysis on a JEOL JSM 6610LV device equipped, which uses W wire or $LaB_6$ as the electron source and has Se, BSE, CL i EDS detectors. Measurements were made in the middle of the sample.

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

## *3. Results and Discussion*

The sample tested by the EDS method was cut from the ingot normal to the crystallization direction (⊥). In the following, this pattern will be referred to as 5/3 (⊥). The same sample was also used for the Hall and Van der Pauw measurements.

| | | | | | |
|---|---|---|---|---|---|
| 1 | 2 | 3 | 4 | 5 | 6 |

***Figure 1.*** *Schematic representation of the location from which the 5/3 (⊥) sample was cut from the ingot*

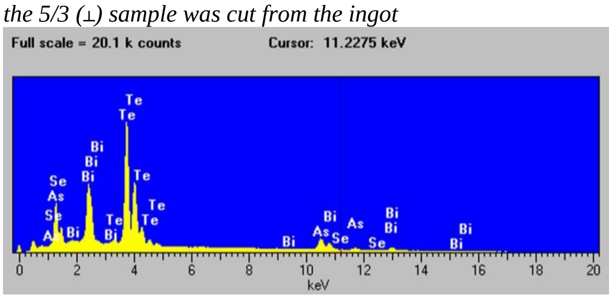


***Figure 2****. EDS picture of sample 5/3 (⊥)*

Concentrations of elements in studied point are function of the peak areas at EDS diagram (figure 2.).

The experimental results of EDS chemical analysis of sample 5/3 (⊥) are shown in Table 1. The table shows that Se was not detected.

***Table 1.*** *Results of EDS analysis of sample 5/3 (⊥)*

| Element | Measurement values (atomic%) | | | | | | | Average value |
|---|---|---|---|---|---|---|---|---|
| Bi | 16.39 | 20.06 | 17.82 | 21.46 | 23.06 | 20.02 | 17.45 | **19.46571** |
| Te | 68.74 | 62.71 | 63.7 | 62.88 | 62.36 | 62.94 | 62.59 | **63.70286** |
| As | 18.89 | 19.63 | 21.28 | 17.82 | 16.36 | 19.56 | 22.72 | **19.46571** |
| Se | **None** | | | | | | | |

The samples used for measurements were prepared to be in the form of thin disc (Figure 3.) 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 room temperature (T = 300 K). The source of magnetic field applied perpendicular to the Hall element was a permanent magnet of 0.37 T. Hall effect measurements were done to obtain transport properties.

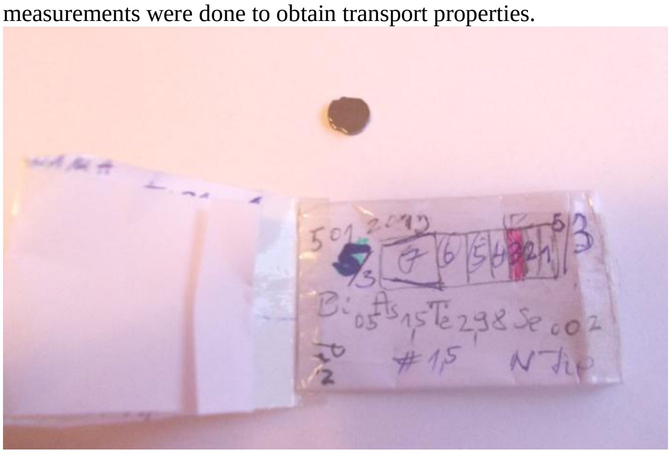

***Figure 3.*** *Cross-sectional view of a circular sample 5/3 (⊥), cut from the ingot*

The samples tested by the Hall and Van der Pauw methods were cut from different parts of the ingot normally to the crystallization direction (⊥). In the following, these samples will be referred to as 5/3 (⊥) and 5/6 (⊥), respectively.

The sample 5/3 (⊥) of circular cross-section is 1.5 mm thick

The concentration of the carriers and the mobility of the charge carriers for the studied samples 5/3 (⊥) and 5/6 (⊥) are given in Table 2. and Table 3.

Measurements were made at currents of: 0.1; 0.5; 1; and 5 mA. The concentration of charge carriers of both samples ranges from $10^{18}$ to $10^{19}$ $cm^{-3}$. The values of the Hall coefficient are negative, indicating that the samples are n-type and that the majority of charge carriers are electrons. The fact that the samples are n type is also confirmed by the hot spot method. The mobility of the majority charge carriers $\mu$ is less than the value of the mobility of bismuth telluride n type which is 1200 $cm^2V^{-1}s^{-1}$ [16]. A large dependence of the mobility of majority charge carriers is observed on the change in current intensity, so that the mobility decreases with increasing current intensity. The samples are n type and the transport parameters refer to electrons as majority carriers.

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***Table 2.*** *The results of the Hall and Van der Pauw method for the sample 5/3 (⊥)*

| Current intesity $I$ [mA] | Specific conductivity $\sigma$ [1/Ωcm] | Specific resistivity $\rho$ [Ωcm] | Bulk carrier concentration $n_b$ [/cm$^3$] | Sheet carrier concentration $n_S$ [/cm$^2$] | Mobility $\mu$ [cm$^2$/Vs] | Average Hall coefficient $R_H$ [cm$^3$/C] |
|---|---|---|---|---|---|---|
| 0.1 | $2.447\cdot10^{1}$ | $4.086\cdot10^{-2}$ | $-3.033\cdot10^{18}$ | $-4.550\cdot10^{17}$ | $5.036\cdot10^{1}$ | $-2.058\cdot10^{0}$ |
| 0.5 | $2.900\cdot10^{1}$ | $3.448\cdot10^{-2}$ | $-2.575\cdot10^{18}$ | $-3.862\cdot10^{17}$ | $7.030\cdot10^{1}$ | $-2.424\cdot10^{0}$ |
| 1 | $2.881\cdot10^{1}$ | $3.471\cdot10^{-2}$ | $-3.044\cdot10^{19}$ | $-4.566\cdot10^{18}$ | $5.908\cdot10^{0}$ | $-2.051\cdot10^{-1}$ |
| 5 | $2.914\cdot10^{1}$ | $3.432\cdot10^{-2}$ | $-2.442\cdot10^{19}$ | $-3.662\cdot10^{18}$ | $7.450\cdot10^{0}$ | $-2.557\cdot10^{-1}$ |

The sample 5/6 (⊥) of circular cross-section is 1.55 mm thick.

| | | | | | |
|---|---|---|---|---|---|
| 1 | 2 | 3 | 4 | 5 | 6 |

***Figure 4.*** *Schematic representation of the location from which the 5/6 (⊥) sample was cut from the ingot*

***Table 3.*** *The results of the Hall and Van der Pauw method for the sample 5/6 (⊥)*

| Current intesity $I$ [mA] | Specific conductivity $\sigma$ [1/Ωcm] | Specific resistivity $\rho$ [Ωcm] | Bulk carrier concentration $n_b$ [/cm$^3$] | Sheet carrier concentration $n_S$ [/cm$^2$] | Mobility $\mu$ [cm$^2$/Vs] | Average Hall coefficient $R_H$ [cm$^3$/C] |
|---|---|---|---|---|---|---|
| 0.1 | $1.484\cdot10^{2}$ | $6.738\cdot10^{-3}$ | $-1.499\cdot10^{18}$ | $-2.324\cdot10^{17}$ | $6.180\cdot10^{2}$ | $-4.164\cdot10^{0}$ |
| 0.5 | $2.401\cdot10^{2}$ | $4.165\cdot10^{-3}$ | $-5.412\cdot10^{18}$ | $-8.388\cdot10^{17}$ | $2.770\cdot10^{2}$ | $-1.153\cdot10^{0}$ |
| 1 | $2.974\cdot10^{2}$ | $3.363\cdot10^{-3}$ | $-2.321\cdot10^{19}$ | $-3.597\cdot10^{18}$ | $7.999\cdot10^{1}$ | $-2.690\cdot10^{-1}$ |
| 5 | $3.620\cdot10^{2}$ | $2.762\cdot10^{-3}$ | $-5.342\cdot10^{19}$ | $-8.280\cdot10^{18}$ | $4.230\cdot10^{1}$ | $-1.168\cdot10^{-1}$ |

## 4. Conclusion

This paper was the result of testing the properties of an arsenic-doped bismuth telluride semiconductor monocrystalline compound. SEM-EDX, Hall's and Van der Pauw's methods were used for material characterization.

An arsenic-doped bismuth telluride monocrystal was synthesized. The electrical properties of this crystal were measured and the mobility and concentration of the majority charge carriers were observed. On the basis of the Hall coefficient, it was determined that in the monocrystal the majority carriers are electrons. The measured electrons mobility was significantly less than the electron mobility in pure bismuth telluride.

The results of these studies show that the arsenic-doped bismuth and tellurium monocrystal was successfully synthesized by the Bridgman method, and significantly complement existing knowledge of bismuth telluride single crystals.

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