

Electrical Resistivity Peculiarities of the Nanograined Bi_2Te_3 Material

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(Received 30 September 2015; published online 24 December 2015)

The hot quasiisostatic pressure method was applied to sinter the nanograined Bi_2Te_3 material. The samples with various mean grain size of 64, 61, 56 and 51 nm were prepared by changing the pressure of sintering. It was found that the specific electrical resistivity of the material under study increases when the mean grain size decreases. The Hall effect was measured to extract the concentration and mobility values of the charge carries. It was found that the electron concentration decreases as the mean grain size decreases while the electron mobility has extreme dependence on the grain size.

Keywords: Nanograined materials, Electrical resistivity, The Hall effect, Size effects.

PACS numbers: 72.10.Fk, 72.15. – v

1. INTRODUCTION

It is very well known that the various nanograined materials show specific electrical properties [1]. It is for instance known that the scaling down corresponding to grain size decreasing in the nanograined materials will decrease their specific electrical conductivity due to strong electron scattering at the grain boundaries [2].

At present, the nanograined materials based on Bi_2Te_3 are considered as ones of perspective thermoelectric materials [3-5]. Low enough lattice thermal conductivity, but high enough electrical conductivity must be simultaneously combined for such materials. It is believed that thermoelectric efficiency in the nanograined samples can be increased via reducing the lattice thermal conductivity, because numerical grain boundaries act as effective centers for phonon scattering. It is clear that the grain boundaries will then involve decreasing the specific electrical conductivity due to the size effect. In order to study the grain structure effect on electrical properties the samples of the nanograined Bi_2Te_3 material must be sintered and applied for research.

The goal of this paper is to study and analyze the grain structure effect on the specific electrical resistivity of the nanograined material based on Bi_2Te_3 with various mean grain size.

2. EXPERIMENTAL PROCEDURE

In order to prepare the nanograined material, the nanosized Bi_2Te_3 powder was first synthesized. The microwave-solvothermal synthesis in closed reactor ERTEC (Model 02-02) was applied. The microwave-assisted heating technique is characterized by short time of synthesis, small particle size of the products, narrow particle size distribution and high purity [6].

As starting components the analytical grade Bi_2O_3 , TeO_2 powders and ethylene glycol were applied. In order to induce microwave assisted reactions a 300 W microwave oven with a 2450 kHz working frequency were chosen.

The ethylene glycol was used as both the reducing agent in the reaction and the solvent. The optimal synthesis conditions were as temperature of 523 K, pressure of 15 atm., duration of synthesis of 50 min and the ratio of Bi_2O_3 and TeO_2 of 1 : 1.

The synthesized nanopowders were then hot quasiisostatically pressed (HQIP) by a toroidal press. The powder was placed in a graphite matrix with hexagonal BN powder as a media spreading the quasiisostatic pressure to the material under pressing. The samples with various grain structures were prepared by using various HQIP-pressure values of 2, 4, 6 and 8 GPa. The HQIP-temperature was equal to 773 K for all the HQIP-pressure values.

A scanning electron microscope Quanta 200 3D was applied to analyze the grain structure of the material under study.

The specific electrical resistivity was measured by four-probed method at 1 mA dc current.

3. RESULTS AND DISCUSSION

It was observed that all the samples sintered under various HQIP-pressure values have the nanograined structures. The SEM-images of the nanograined structures taken for all the HQIP-pressure values were used to plot the histograms of grain size distribution. These histograms were found to be fitted a lognormal distribution. Analysis of these histograms was used to estimate a mean grain size, D_m . The D_m values were 56, 64, 61 and 51 nm for the HQIP-pressure values of 2, 4, 6 and 8 GPa. Detailed analysis of the distribution histograms of the material under study is presented in Ref. [1].

It is important noted that the samples with various mean grain size have quite different values of electrical resistivity. The $\rho(D_m)$ dependence taken at room temperature is shown in Fig. 1.

One can see that the ρ values increase when the D_m values decrease. So, grain size effect on electrical resistivity specific for the nanograined materials [1] takes place for the material under study.

It is very well known that electrical resistivity of materials is determined by the concentration, n , and mobility, μ , of the charge carries. In order to extract both concentration and mobility contributions to electrical resistivity in Fig. 1 the Hall effect was measured.

First of all, a sign of the Hall constant allowed us to conclude that major charge carries are electrons for all the samples under study. Next, the Hall constant values and electrical resistivity values were used to extract the

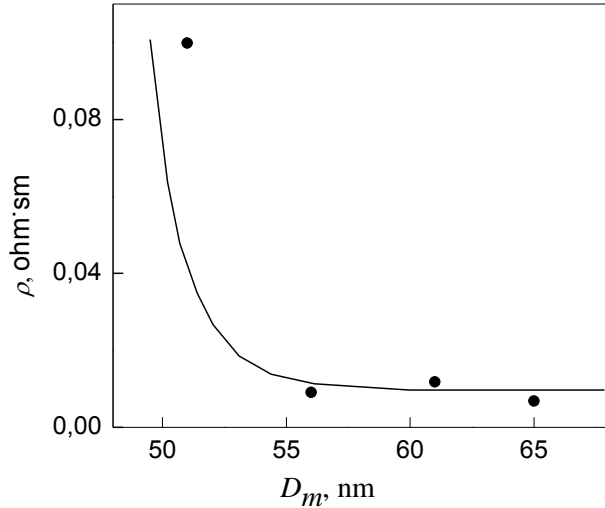


Fig. 1 – The $\rho(D_m)$ dependence for the samples sintered under various HQIP-pressures

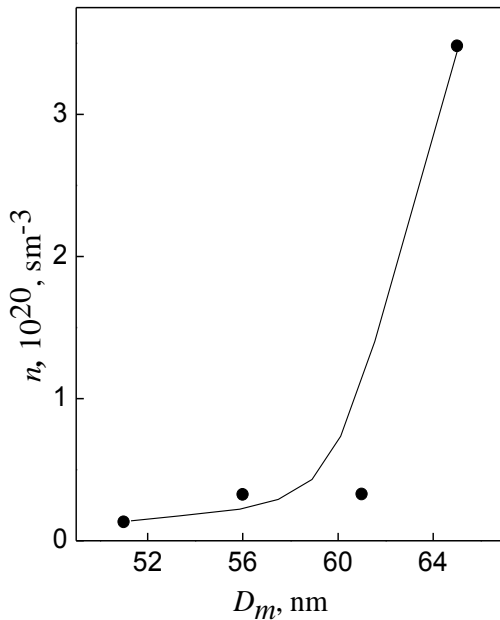


Fig. 2 – The $n(D_m)$ dependence for the samples sintered under various HQIP-pressures

$n(D_m)$ and $\mu(D_m)$ dependences shown in Fig. 2 and 3, respectively.

The charge carries concentration decreases as D_m decreases too, while the $\mu(D_m)$ dependence is extreme one with maximum μ value at $D_m = 56$ nm. According to Ref. [7], electrons as major charge carries may appear in Bi_2Te_3 due to Te vacancy forming during the material preparation. A number of the Te vacancies is believed to decrease as the HQIP-pressure increases. In this case, decrease of the Te vacancies will lead to decrease of the charge carries concentration.

The extreme $\mu(D_m)$ dependence can be determined by two mechanisms at least. In general case the total mobility of the charge carries is known to consist of a few contributions.

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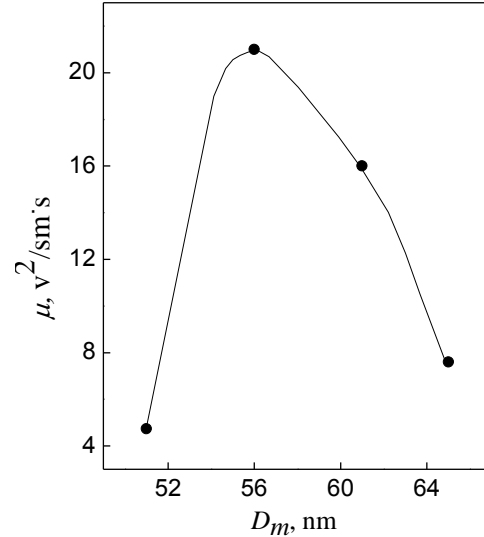


Fig. 3 – The $\mu(D_m)$ dependence for the samples sintered under various HQIP-pressures

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In accordance with the Matthiessen rule, one can write an expression for the inverse total mobility of the charge carries as

$$\mu^{-1} = \sum_i \mu_i^{-1}, \quad (1)$$

where μ_i^{-1} is the inverse mobility for the i -th contribution.

For instance, taking into account the specific contributions to mobility for our research we can rewrite the Matthiessen rule as

$$\mu^{-1} = \mu_{ph}^{-1} + \mu_{imp}^{-1} + \mu_{def}^{-1} + \mu_b^{-1}, \quad (2)$$

where μ_{ph}^{-1} , μ_{imp}^{-1} , μ_{def}^{-1} and μ_b^{-1} are corresponding to the electron-phonon interaction, the electron scattering by the charged impurities, the electron scattering by the neutral defects and the electron scattering by the grain boundaries, respectively.

In order to explain the $\mu(D_m)$ dependence in Figure 3, let us believe that μ_{ph} and μ_{def} are the same contributions for all the samples with various men grain size. So, the electron scattering by the charged impurities and the grain boundaries should be taken into account.

The electron scattering by the charged impurities should be increased as D_m decreases, because n increases as D_m decreases (Fig. 2). As was mentioned above, electrons in Bi_2Te_3 appear due to the Te vacancy forming. On the other hand, the electron scattering by the grain scattering will become more effective process as the mean grain size decreases. For instance, the $\mu(D_m)$ dependence can be expressed as [8]

$$\mu_b = \frac{l_e/D_m}{1+l_e/D_m} \mu_a, \quad (3)$$

where l_e is the free electron path and μ_a is the electron mobility of the bulk material having no grain boundaries.

In contrast to the $\mu_{imp}^{-1}(D_m)$ dependence, the $\mu_b^{-1}(D_m)$ dependence decreases as D_m decreases. So, combination of the $\mu_{imp}^{-1}(D_m)$ and $\mu_b^{-1}(D_m)$ dependences will lead to extreme D_m -dependence of the total electron mobility.

4. CONCLUSIONS

The peculiarities of the grain structure and electrical properties of the samples of the nanograined material based on Bi_2Te_3 with various values of the mean grain size have been studied. It was found that the electrical resistivity decreased as the mean grain size increased.

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Such kind of electrical behavior is due to changes of the electron concentration and electron mobility during the sintering of material under study by hot quasiisostatic pressure with various value of pressure.

ACKNOWLEDGEMENTS

All the studies were carried out by the scientific equipment of the joint research center "Diagnostics of structure and properties of nanomaterials" of Belgorod National State University which financially supported with Ministry of Science and Education of RF under project No 14.594.21.0010, unique code RFMEFI59414X0010. This work was also financially supported by the Ministry of Education and Science of the Russian Federation under projects No 2014/420-1 and 3.308.2014/K.