THERMOELECTRIC PROPRIETIES OF LEAD TELLURIDE MICROWIRES

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Introduction

In recent years, in physics of narrow-gap semiconductors, much attention is paid to semiconductors of the group A^4B^6 and solid solutions on their basis. Practical significance of these substances is determined by the possibility to produce a series of electronic devices, such as thermoelectric, optoelectronic, strain-gauge, etc.

The present-day methods of planar technology for preparation of films make it possible to control the type of conductivity and concentration of carriers in the process of epitaxial growth. However, the structure of films obtained is not ideal due to defects at the material-support interface.

Therefore, it is of interest to obtain and to study quasi-one-dimensional crystalline structures—microwires in glass isolation.

Obtaining of PbTe microwires

As the initial material for obtaining of microwires, we used compounds of lead telluride (PbTe) with conductivity of *p*- or *n*-type. Initial compounds were synthesized in the form of polycrystals or single crystals grown by the Bridgman-Stockbarger method [1]. PbTe compounds were synthesized by consecutive melting of chemical elements in quartz glass ampoules [2]. Initial components Pb-0000 and tellurium purified by the method of zone crystallization were placed into quartz glass ampoules with a diameter of 2-2.5 mm and a length of 120 mm. Before the placing, the chemical elements had been chemically processed, washed, and dried. To avoid sticking of the compound to the ampoule walls, the inner surface of the ampoules was treated with pure alcohol or acetone. After grinding, compounds the introduced into the ampoule to obtain microwires by the Ulitovsky method [3]. The microwires obtained were spooled on a coil with a diameter of 30 cm at a rate of 10-15 m/min.

Results of the study of the microwires showed that there are many cracks along their length and that their number does not depend on sort of the glass and on technology of obtaining. The main causes of the cracks are the high rate of crystallization and the difference in the linear temperature coefficient of the glass and PbTe. The high rate of crystallization favors supercooling in the melt, which results in the appearance of spontaneous crystallization centers, which leads to the creation of new centers of crystallization. The melt in the glass capillary, being between two crystallization centers, is attracted between them in the process of crystallization, which results in the formation of microcracks in the microwires.

In addition, in the process of formation of microcracks, the atmospheric pressure inside the capillary decreases leading to elimination of intrinsic vapors of the PbTe melt, which also results in the formation of microcracks in the microwires.

Therefore, to obtain microwires of PbTe, we applied the method that involves the filling of preliminarily evacuated glass capillaries with a melt of PbTe by means of inert gas pressure as shown in Fig. 1 [1]. For this purpose, evacuated quartz ampoule *1* with weighted amount of PbTe *2*, above which sealed quartz capillaries *3* are placed, is introduced into a high-temperature furnace in such a way that capillaries with the material are situated in the operating zone of the furnace (its temperature is 10-15° higher than the melting temperature of the material). After melting of the material and the immersion of the capillaries in the melt, an excess pressure is generated by means of inert gas (P = (6. 8 - 49) x 10^4 Pa); as a result, the capillaries are filled with the melt. Afterwards, the oriented crystallization of the melt in the capillaries takes place.

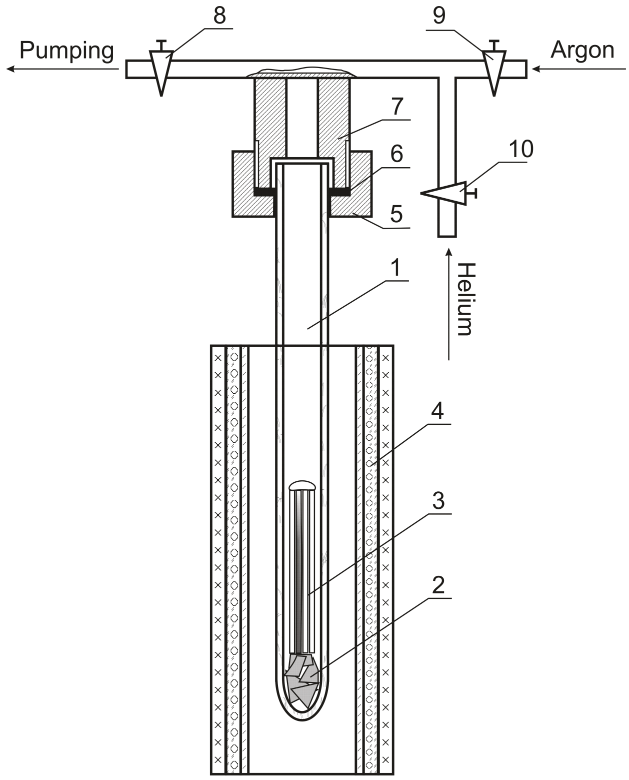

Fig. 1. Diagram of the installation for obtaining of PbTe microwires: *1* is the quartz ampoule; *2* is the PbTe material; *3* is the quartz capillaries; *4* is the furnace; *5, 6, 7* are the coupling device; *8, 9, 10* are taps for evacuation and introduction of inter gas.

It should be mentioned that high rates of growth of the microwires (higher than 15 cm/h) lead to the appearance of microcracks along the length of microwires as well as in the case of the Ulitovsky method. Low rates (lower than 1 cm/h) result in the formation of step-like pits due to evaporation of material from the microwire surface.

Qualitative PbTe microwires were obtained in the following mode: the growth rate was 1-10 cm/h; the temperature gradient was 10-15°/cm; the maximum melting temperature was maintained to be 10-15° higher than the temperature of crystallization; the excess of inert gas pressure was (6.8 - 49) x 10^4 Pa [4].

Electrothermal properties of PbTe microwires

Crystalline microwires of lead telluride (PbTe) with the *n*-type electric conductivity were obtained from the initial material being a chemical compound with an excess of lead with respect to the stoichiometric composition; the ones of the *p*-type, with an excess of tellurium [1].

Temperature dependence of resistivity of PbTe microwires of the *n*- and *p*-types (Figs. 2 and 3, respectively) in the temperature range of 80-350 K showed the following:

- The resistivity exhibits the metallic behavior; that is, it increases with growing temperature; the intrinsic conductivity for the crystals studied in this temperature range does not occur.
- The resistivity depends on microwire diameter; as the diameters decreases, the resistivity ρ decreases as well.

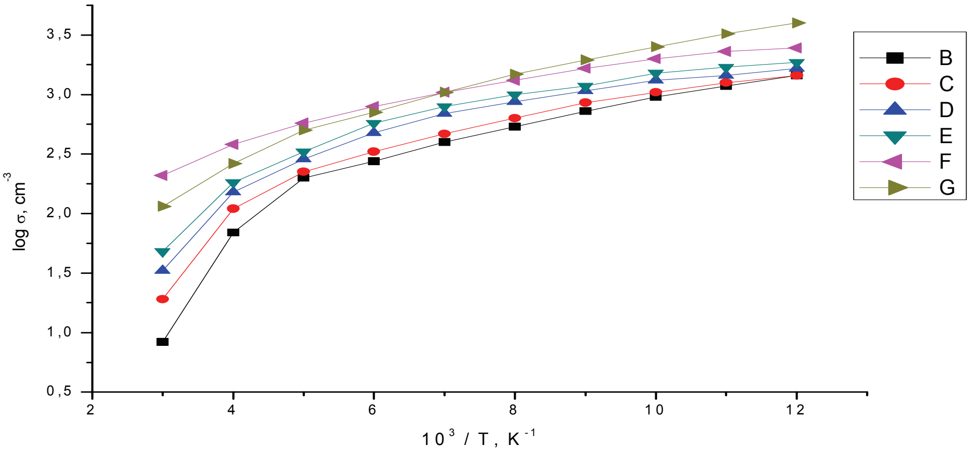

Fig. 2. Dependence of resistivity on temperature for PbTe microwires with the *n*-type conductivity: B - d = 125.0 μ; C - d = 92.0 μ; D - d = 51.0 μ; E - d = 33.0 μ; G - d = 15.0 μ; F - n = 1.2 10^{18} cm^{-3} at the temperature T = 77 K.

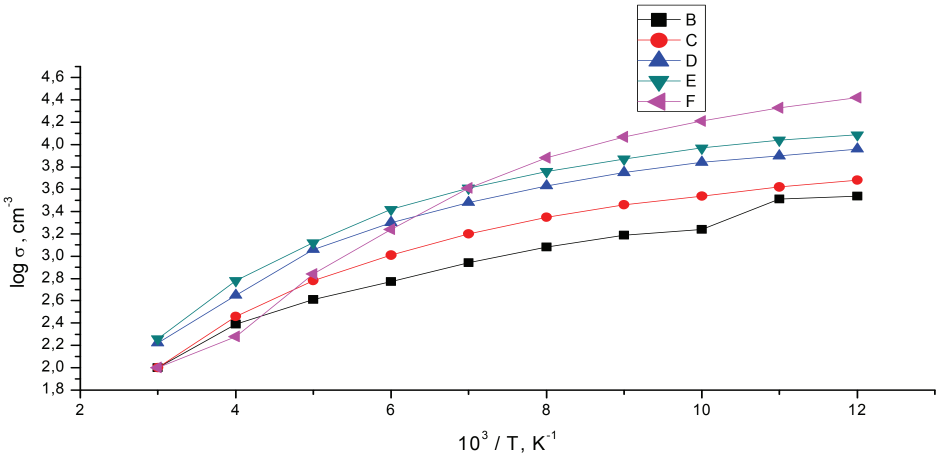

Fig. 3. Dependence of resistivity on temperature for PbTe microwires with the *p*-type conductivity: B - d = 103.0 μ; C - d = 45.0 μ; D - d = 29.3 μ; E - d = 17.8 μ; F - p = 1.47 10^{18} cm^{-3} at T = 77 K.

The decrease in the resistivity due to the decrease in the microwire diameter can be explained as follows. On the basis of the above, for current carriers of the p- or n-type, the dependences between resistivity (ρ), concentration (n), and carrier mobility (μ) is given by the following expression

$$\rho = (e\, n\, \mu)^{-1}. \quad (1)$$

It follows from this expression that resistivity can decrease due to the increase in concentration (n) or in mobility (μ).

The studies of the temperature dependence of thermopower for the microwires with different diameters showed (Figs. 4 and 5, respectively) that, as the microwire diameter decreases, the thermopower coefficient (α) hardly changes at all.

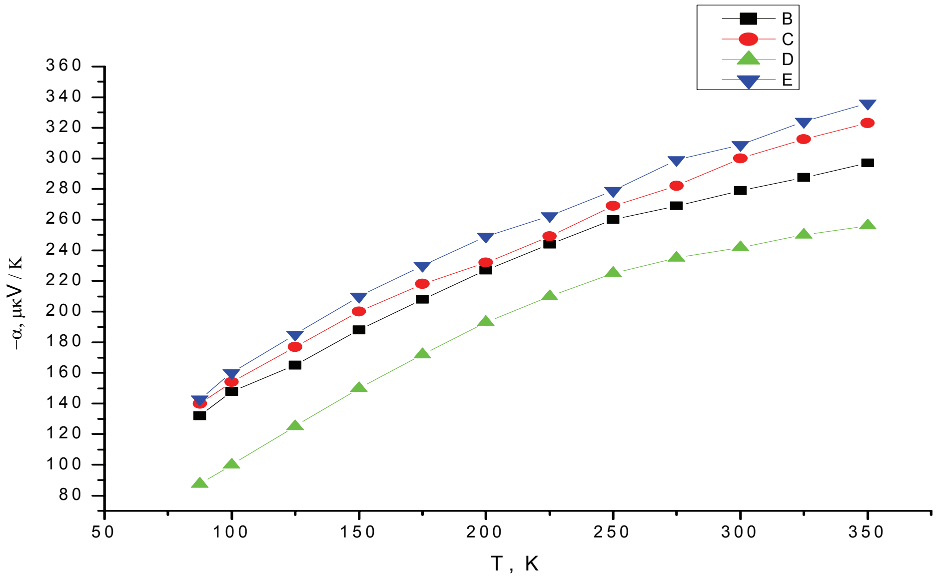

Fig. 4. Temperature dependence of the thermopower coefficient of PbTe microwires with the *n*-type conductivity: B - d = 125.0 µ; C - d = 51.0 µ; E - d = 15.0 µ; D - n = 1.2 10^{18} cm^{-3} at a temperature of 77 K.

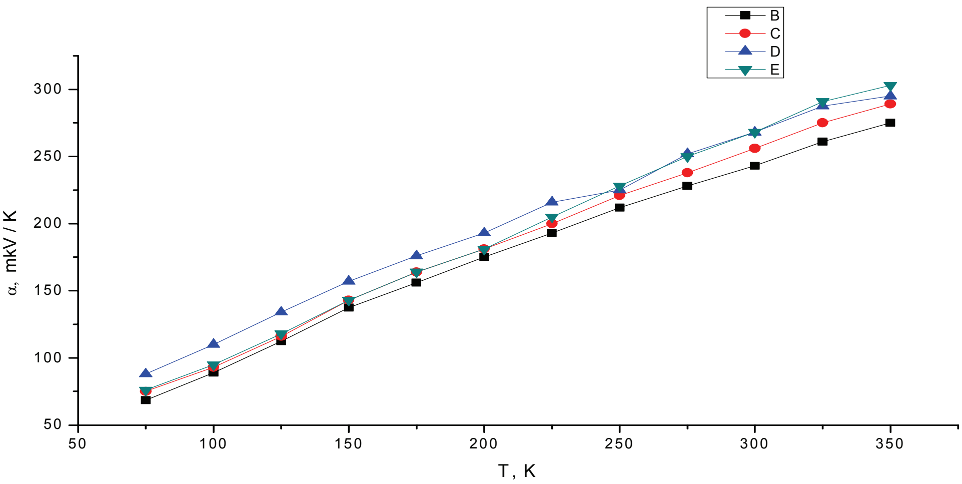

Fig. 5. Temperature dependence of the thermopower coefficient of PbTe microwires with the *p*-type conductivity: B - d = 103.0 µ; C - d = 45.0 µ; D - d = 17.8 µ; E - p = 1. 47 10^{18} cm^{-3} at temperature T = 77.

It follows from the relation of the coefficient α, which is

$$\alpha_{p(n)} = \pm k/e\,[A + \ln(2(2\pi m_{p(n)}kT)^{3/2} / p\,(n)\,h^3)], \qquad (2)$$

that the concentration of carriers of the p(n) type in the samples does not depend on diameter of microwires within the studied range. Thus, it follows from the obtained results that the decrease in the resistivity ρ is due to the increase in the mobility of carriers. Therefore, the increase in the mobility with decreasing diameters counts in favor of the enhancement of the structure perfection and the improvement of the homogeneity of microwires with decreasing diameter.

The comparison of the results of measurements of resistivity and thermopower coefficient of microwires and bulk crystals allows making the conclusion that the values for ρ and α range within the limits for the bulk crystals grown by the Bridgman method. This shows that the microwires obtained by this method retain the properties characteristic of bulk crystals.

Heat efficiency of PbTe microwires

According to the thermoelectricity energy theory, the efficiency of the thermal element branch material is determined by

$$Z = \alpha^2 \sigma / \kappa, \quad (3)$$

where Z is the heat efficiency; α is the thermopower; σ is the electric conductivity; κ is the heat conductivity of the material. One can see from the formula that the efficiency depends directly on the product $\alpha^2\sigma$ and is inversely proportional to the heat conductivity κ. Therefore, we will fix on the behavior of the parameter $\alpha^2\sigma$—the so-called thermoelectric power. Figures 6 and 7 show the temperature dependences of thermoelectric power for microwires of *n*- (Fig. 6) and *p*-type (Fig. 7).

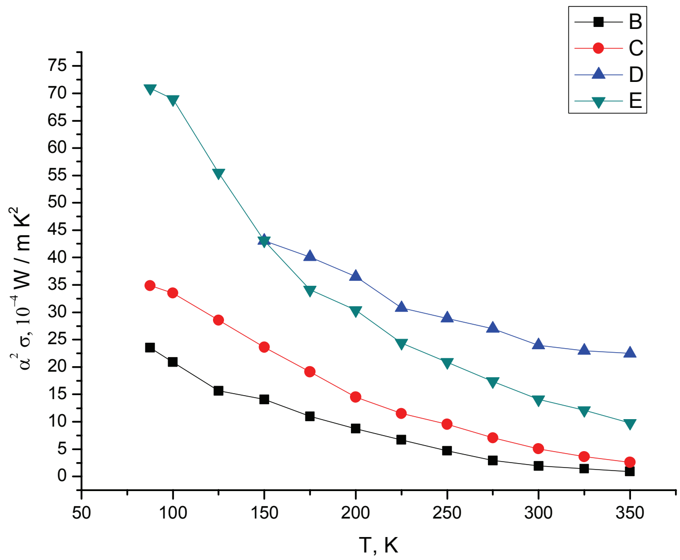

Fig. 6. Temperature dependence of the thermopower of *n*-type PbTe microwires: B - d = 125.0 μ; C - d = 51.0 μ; D - n = 1.2 10^{18} cm^{-3} at a temperature of 77 K; E - d = 15.0 μ.

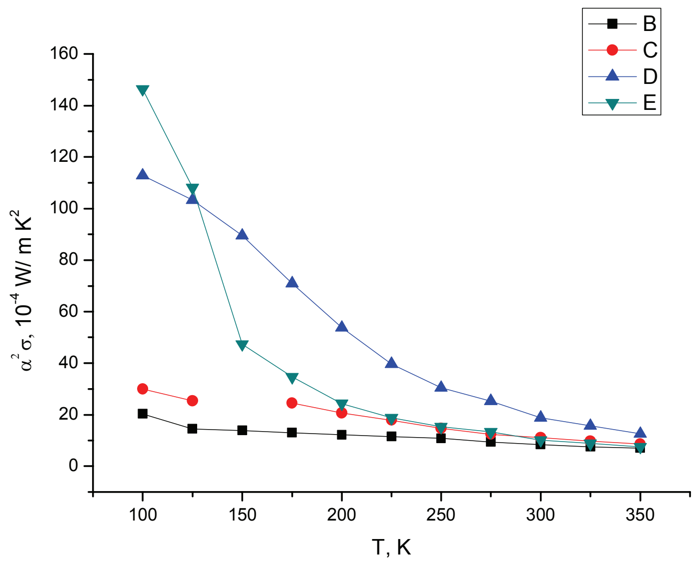

Fig. 7. Temperature dependence of the thermopower of *p*-type PbTe microwires B - d = 103.0 μ; C - d = 45.0 μ; E - p = 1.47 10^{18} cm^{-3} at a temperature of 77 K; D - d = 17.8 μ.

The calculation showed that if we compare the heat conductivity of microwires to the conductivity of bulk samples, then the efficiency of the material of *n*-type is Z = 0.7 10^{-3} K^{-1} at a temperature of 300 K.

Conclusions

1. A technology for obtaining of quartz-glass covered microwires of thermoelectric material PbTe with conductivity of the *p*- and *n*-type, with a core diameter of 10-150 μ, has been developed.
2. A series of measurements of electrical and thermoelectrical properties of microwires with different diameters of the crystalline core in the temperature range 77-350 K has been carried out.
3. Analysis of the results of measurements of resistivity and thermopower in comparison with properties of bulk materials obtained by the Bridgman method has shown that the numerical values of α and ρ for microwires are within the same limits. This means that obtaining of PbTe microwires by the method of capillary filling preserves the same properties as in bulk materials obtained by melting (deviation from stoichiometry, similar limits of solubility of components, high concentration of current carriers).
4. The heat efficiency of the PbTe material is Z = 0.7 10^{-3} K^{-1} at a temperature of 300 K.

References

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