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MANUFACTURING OF HALF-ELEMENTS BY DEEP ALLOYING AND THE METHOD OF PRESSING POWDERS BY SELECTING THE OPTIMAL PRESSING MODE

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Abstract:

This article presents research on improving the thermoelectric properties of semiconductor thermoelectric materials using powder pressing and heat treatment methods. A technology for obtaining homogeneous samples of a given concentration of charge carriers is described. To expand the operating temperature range, p-type materials based on $Sb_2Te_3 – Bi_2Te_3$ were deeply alloyed with cadmium.

Keywords: thermoelectric power sources, thermoelectric material, cold pressing, hot pressing, annealing

Introduction

Increasingly, there is a growing circle of engineers and power engineers who are interested in creating autonomous, compact, maintenance-free, environmentally friendly and efficient power supplies. Thermoelectric power sources can be used to power meteorological stations, spacecraft and marine navigation buoys [1-5]. With an increase in the efficiency of thermoelectric materials and an increase in the efficiency of thermoelectric converters, the prospect of turning thermoelectric generators from auxiliary into the main sources of electricity for a number of land-based, marine and space units becomes more and more real.

The practical application of thermoelectric materials essentially depends on several technological criteria, in addition to the above physical characteristics. These include the mechanical properties of the material and its ability to withstand repeated heating and cooling. Therefore, the temperature of the hot junction of the thermoelement is chosen to be much lower than the melting point of the starting material. And thermoelectric materials used in the areas of medium and high temperatures must have a low vapor pressure and the ability to firmly retain alloying additives. Thermoelectric material exposed to radioactive radiation, in the case of using a thermoelectric converter in combination with a nuclear heat source (reactor, radioactive isotope), should not change its characteristics during the entire service life of the installation.

Method

Currently, a lot of work is underway to complicate the composition of alloys and solid solutions containing three or more components. To expand the range of operating temperatures, deep alloying of the most effective solid solutions and alloys based on them is carried out, and new types of materials with high thermoelectric characteristics are being sought. The thermoelectric properties of low-temperature thermoelements depend to a large extent on the sample fabrication technology (single crystals, directional polycrystals, and pressed samples) [6]. Cast samples, in principle, should have higher efficiency values, but due to the liquidation inhomogeneity, a large spread in parameters from point to point and from sample to sample is obtained [7]. Obtaining homogeneous samples of a given concentration of charge carriers is a difficult task. Significantly more homogeneous in the microscopic sense are obtained samples made by the method of powder metallurgy. The mechanical properties of such samples also exceed the properties of crystals, which are easily stratified along cleavage planes. However, even here it is required to choose the optimal pressing mode: pressure, temperature and holding time. Each of these factors affects the quality of samples. All parameters of the pressing mode varied quite widely: pressure from 2,5 t/cm^2 to 11 t/cm^2 [8], temperature from 300°C to 400°C. Then the resulting ingots were sintered at a temperature of 390°C for 17 hours and kept at room temperature for 5 minutes. Fractions from 0,125 to 0,5 mm were used for pressing.

Discussion of Results

To obtain n- and p-type ingots, materials of the following composition are prepared: for n-type 85 wt.% Bi_2Te_3 - 15 wt.% Bi_2Se_3 and for p-type 70 wt.% Sb_2Te_3 - 30 wt.% Bi_2Te_3. Doping of p-type thermoelectric material was carried out by adding cadmium. Cadmium, in a ternary hole alloy, has higher mobility values compared to Zn, Sn, Pb. These impurities (Zn, Sn, Pb), like Cd, are acceptor additives, but with an increase in the concentration of current carriers in a ternary hole alloy, these impurities lead to a drop in mobility [9–11]. Therefore, we used cadmium as an alloying additive in the ternary alloy. Cadmium was placed in the crucible after each layer of tellurium. As a result of the research, it was found that for a thermoelectric material having a composition of 70 wt.% Sb_2Te_3 - 30 wt.% Bi_2Te_3 doped with 0,25 wt.% Cd, the thermoelectric figure of merit varies from $1,9\cdot10^{-3}$ K^{-1} (293 K) to $0,3\cdot10^{-3}$ K^{-1} (573K). The maximum value of thermoelectric figure of merit $Z=2.1\cdot10^{-3}$ K^{-1} (323 K) when using the powder pressing method. In the original component, cadmium was introduced in an amount of 0,1 wt.% to 0,4 wt.%. Figure 1 shows the thermoelectric properties of the obtained alloyed materials.

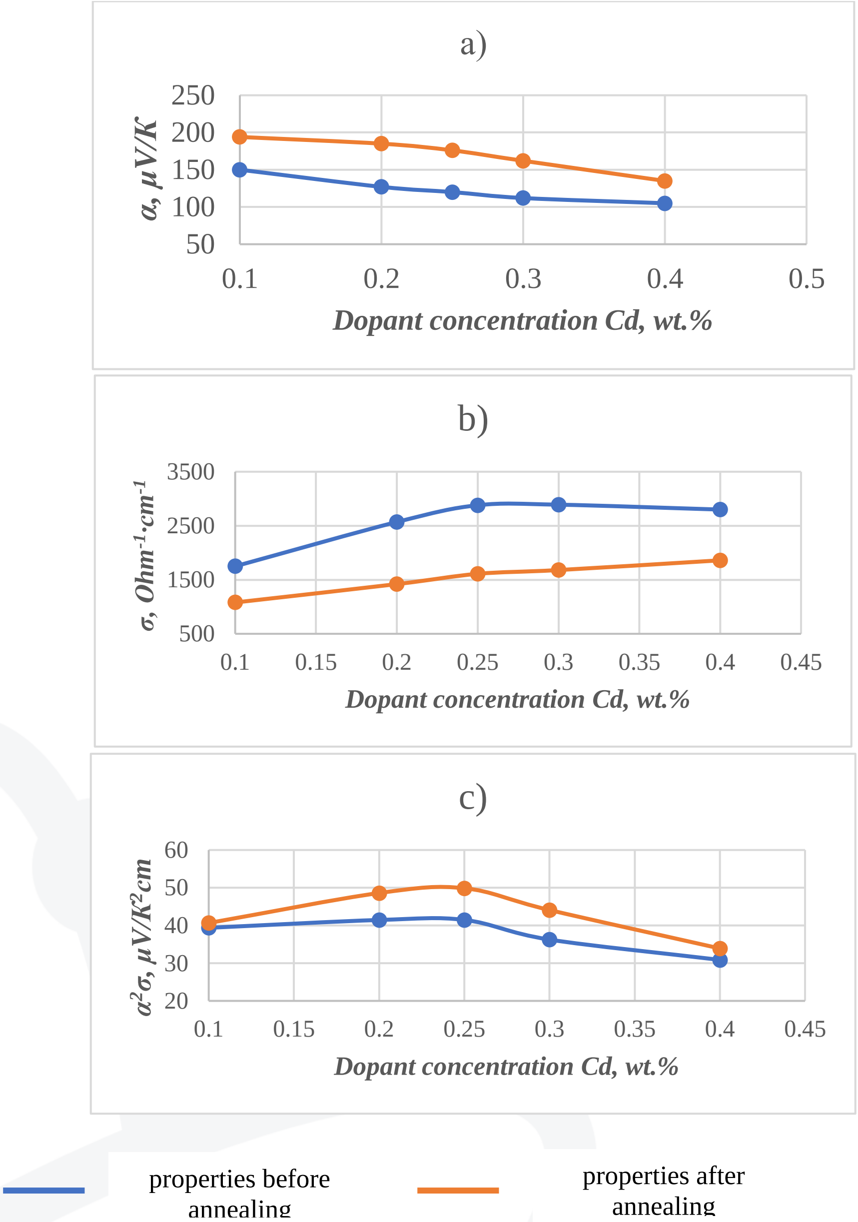

Fig.1. Graph of the dependence of the values of a) thermoelectric coefficient α, b) electrical conductivity σ, c) power factor $\alpha^2\sigma$ on the concentration of wt.% Cd.

It can be seen from the figure that the concentration of the dopant is linearly related to the change in the thermoelectric properties of the obtained materials. As is known, with an increase in the concentration of the dopant - cadmium - the electrical conductivity and thermopower coefficient change linearly. Of interest was the temperature dependence of the change in the thermoelectric properties of half-elements on the concentration of the introduced dopant - cadmium.

For the study, half-cells with different concentrations of cadmium were fabricated. The results of measurements of the thermoEMF coefficient and electrical conductivity of half-cells at room temperature before and after annealing are shown in Table 1.

Table 1. Thermoelectric properties of half-elements at room temperature depending on the concentration of the introduced dopant

Compound	Dopant concentration Cd, % wt.	Properties before annealing			Properties after annealing		
		σ Ohm^{-1}·cm^{-1}	α µV/K	$\alpha^2\sigma$ µV/K^{2}cm	σ Ohm^{-1}·cm^{-1}	α µV/K	$\alpha^2\sigma$ µV /K^{2}cm
30% mol. Bi_2Te_3 70% mol. Sb_2Te_3	0,1	1750	150	39	1080	194	40
	0,2	2570	127	41	1420	185	48
	0.25	2880	120	41	1610	176	49
	0,3	2890	112	36	1680	162	44
	0,4	2800	105	30	1860	135	33

Conclusion

It follows from the table that with an increase in the concentration of the introduced dopant, the electrical conductivity of half-elements before annealing increases, and the thermopower coefficient decreases. After annealing of half-elements, their electrical conductivity decreases, and the thermoEMF coefficient increases in comparison with unannealed half-elements. However, the same pattern of change in properties with a change in the concentration of the dopant is preserved in both annealed and unannealed half-elements, i.e., with an increase in the concentration of the dopant, the electrical conductivity increases, and the thermopower coefficient decreases.

In the study of electrical conductivity and thermoEMF coefficient with temperature change (Fig. 1), it was found that in the initial period with an increase in temperature, the electrical conductivity drops, and the thermoEMF coefficient increases. With a further increase in temperature, the thermopower coefficient reaches a maximum, and then decreases, while the electrical conductivity continues to decrease.

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