UDC 621.362.2

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DESIGNING OF SEGMENTED THERMOELECTRIC MODULES BASED ON Bi2Te3 / PbTe MATERIALS

*Abstract***.** The results of computer simulation of segmented thermoelectric generator modules are presented. Found that use of Bi_2Te_3-based materials as cold sections and *PbTe*-based materials as hot sections for two-section modules in the temperature range 303-773K, allows to multiply their efficiency by 1.28 as compared to single-section ones.

Introduction

In recent years there has been increasing attention to researches aimed at finding ways to improve the efficiency of thermoelectric energy conversion. A promising is the recuperation of low-grade waste heat of industry, automobile transport, etc., in particular the conversion into electrical energy by means of thermoelectricity [1-3]. Among the thermoelectric materials which are used to create generator modules on the level of hot temperatures to 773 K materials based on Bi_2Te_3 and *PbTe* are conventional [4, 5]. However, the widespread practical application based on them generators is constrained by insufficiently high efficiency.

The aim of this paper is design and evaluation of the possibility increasing the efficiency of thermoelectric modules based on Bi_2Te_3 and *PbTe* by the use of sectional thermoelements.

Methods of calculations

By analysis of the impact technology of reception thermoelectric materials on their thermoelectric properties found that materials which produced by spark plasma sintering (SPS) [6], in comparison with other methods (hot pressing, zone melting, extrusion), are characterized highly homogeneous, mechanical strength and high quality factor values. Given this characteristic was conducted selection of materials for researches:

- for legs of the *n*-type conduction is chosen: $Bi_2(Te_{1-x}Se_x)_3$ $(0 \leq x \leq 1)$ [7] and $Sb_xPb_{1-x}Te_{0.88}S_{0.12}$ $(0 \leq x \leq 0.008)$ [8];
- for legs of the *p*-type conduction is chosen: $(Bi_2Te_3)_x(Sb_2Te_3)_{1-x}$ $(0.16 \leq x \leq 0.24)$ [9] and *PbTe*+2*mol%Na*+*x mol%SrTe* $(0 \leq x \leq 4)$ [10].

Design and calculation of the generator modules characteristics were carried out by a computer program COMSOL Multiphysics [11], using the experimental temperature dependences of thermoelectric parameters: thermoelectric power, electrical conductivity and thermal conductivity of listed above materials. The temperature dependences of the thermoelectric parameters were approximated by polynomials whose coefficients were introduced to the program as input data.

A model of thermoelement, which describes the temperature and potential distributions along the height of its branches, is developed. The equation of the physical field's distribution in the thermoelement received from the laws of energy conservation $div\vec{W} = 0$ and electric charge $div\vec{j} = 0$ following way:

$$\vec{W} = \vec{q} + U\vec{j} \,, \tag{1}$$

$$\begin{cases} \vec{q} = \kappa\vec{\nabla}T + \alpha T\vec{j}, \\ \vec{j} = -\sigma\vec{\nabla}U - \sigma\alpha\vec{\nabla}T, \end{cases} , \tag{2}$$

where $\vec{W}$ – energy flux density; $\vec{j}$ – electric current density; U – electric potential; T – temperature; α, σ, κ – Seebeck coefficient, electrical conductivity and thermal conductivity.

Considering equations (2) into (1) was obtained:

$$\vec{W} = -(\kappa + \alpha^2\sigma T + \alpha U\sigma)\vec{\nabla}T - (\alpha\sigma T + U\sigma)\vec{\nabla}U \,. \tag{3}$$

Then the conservation laws take on the form:

$$\begin{cases} -\vec{\nabla}\left[(\kappa + \alpha^2\sigma T + \alpha U\sigma)\vec{\nabla}T\right] - \vec{\nabla}\left[(\alpha\sigma T + U\sigma)\vec{\nabla}U\right] = 0, \\ -\vec{\nabla}(\sigma\alpha\vec{\nabla}T) - \vec{\nabla}(\sigma\vec{\nabla}U) = 0. \end{cases} \tag{4}$$

Equations (4) – second order partial differential equations of the unknown functions U and T, which it was necessary to compare with one of the standard forms of the COMSOL Multiphysics program:

$$\nabla(-C\nabla M) = 0. \tag{5}$$

Taking into account that $M = \begin{bmatrix} U \\ T \end{bmatrix}$, $C = \begin{bmatrix} C_{11} & C_{12} \\ C_{21} & C_{22} \end{bmatrix}$ equation (5) will take the form:

$$\begin{cases} \nabla(-C_{11}\nabla U) + \nabla(-C_{12}\nabla T) = 0, \\ \nabla(-C_{21}\nabla U) + \nabla(-C_{22}\nabla T) = 0. \end{cases} \tag{6}$$

By comparing (4) and (6) equations we obtain coefficient values for computer program which depend on thermoelectric characteristics of the materials:

$$\begin{cases} C_{11} = \alpha\sigma T + U\sigma, \\ C_{12} = \kappa + \alpha^2\sigma T + \alpha\sigma U, \\ C_{21} = \sigma, \\ C_{22} = \sigma\alpha. \end{cases} \tag{7}$$

Sectional generator modules calculation

Results of the calculations of modules with homogeneous materials with number of thermoelements – 56 couples, height branches of 5.6 mm and cross sectional area of branches 4.3 × 1.8 mm^2 (module design is shown on Figure 1 a):

- module based on Bi_2Te_3 materials for the operating temperature range 303-500 K, where for *n*-leg was used $Bi_2(Te_{0.8}Se_{0.2})_3$ and $(Bi_2Te_3)_{0.24}(Sb_2Te_3)_{0.76}$ for the *p*-leg was calculated. Chosen concentration of alloying additives corresponds to the highest quality factor of the material. The maximum efficiency of the module is 7.5% and electric power – 4 watts.
- module based on *PbTe* materials for the operating temperature range 303-773 K, where for *n*-leg was used $Sb_{0.004}Pb_{0.996}Te_{0.88}S_{0.12}$, and *PbTe*+2*mol%Na*+4*mol%SrTe* for the *p*-leg was calculated. Chosen concentration of alloying additives corresponds to the highest quality factor of the material. The maximum efficiency of the module is 10.5 % and electric power – 13.6 watts. It should be noted that at temperature drops 303-500 K the module efficiency is only 3.8 %. This means that *PbTe* based materials efficiently use at the cold side temperature above 500 K.

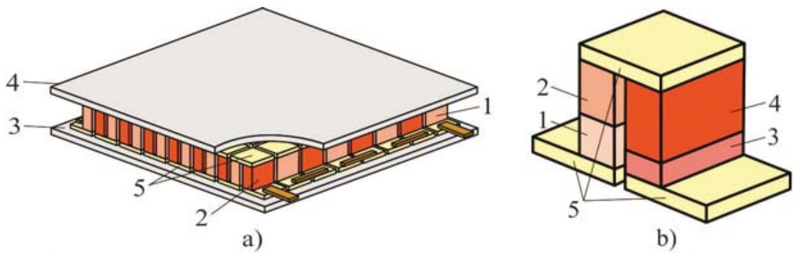

Fig. 1. Scheme of: a) design of the single-section module (1 – branch of *n*-type, 2 – branch of *p*-type conductivity; 3, 4 – cold and hot ceramics, respectively; 5 – commutation of branches thermoelements), b) two-section thermoelement (1, 2 – cold and hot sections of *n*-type leg conductivity; 3, 4 – the hot and cold sections of *p*-branch, respectively; 5 – commutation of legs).

Thus, it was appropriate to carry out calculations of two-section module (scheme its thermoelement is filed in a Figure 1 b), using as a cold sections Bi_2Te_3 based materials and *PbTe* based materials for hot sections. However, at developing sectional thermoelements there is a problem of electrical conformity sections. Since the sections are connected sequentially then the ratio of electromotive forces to the internal resistance should be constant, or will be parasitic losses of electric power at internal resistances of the sections with high resistance and low electromotive force. Consequently, the current passing in the sections of thermoelements branches must be the same. For maximum efficiency mode current can be written as

$$I = \frac{\alpha_i \cdot \Delta T_i}{r_i \cdot (1+\sqrt{1+Z_i \cdot \overline{T_i}})} = \frac{Z_i \cdot Q_i^{\lambda}}{\alpha_i \cdot (1+\sqrt{1+Z_i \cdot \overline{T_i}})} = const, \tag{8}$$

where r_i – internal resistance, Z_i – quality factor of the i-th section. In the approximation that the heat conductivity Q_i^{λ} in adjacent sections differ little, equation (8) is performed only when the ratio of quality factor Z_i to Seebeck coefficient α_i for material sections is equally: $Z_i/\alpha_i = const$. It should be noted that for maximum efficiency mode sectional module exact matching condition sections of materials is the equality for each section of the so-called conformity factor of sections materials s_i

$$s_i = \frac{Z_i}{(1+\sqrt{1+Z_i \cdot \overline{T_i}}) \cdot \alpha_i}. \tag{9}$$

It is evident that from the approximation of $Q_i^{\lambda} \approx const$ in (8) get (9). At the case of significant variation s values, sections works under less than ideal modes and efficiency of the thermoelement is reduced.

Calculations conformity factors Bi_2Te_3 based materials at temperature range 303-500 К, *PbTe* based materials at temperature range 303-773 К for *n*-leg and 303-823 К for *p*-type conductivity were conducted using the relation

$$s(T) = \frac{Z(T)}{(1+\sqrt{1+Z(T)\cdot T}) \cdot \alpha(T)}. \tag{10}$$

The results of calculations of the temperature dependence of conformity factors investigated materials with different degrees of doping are given in Fig. 2.

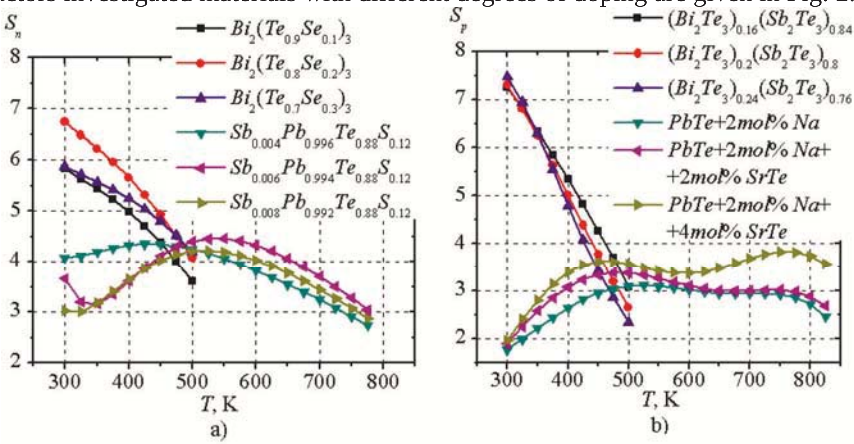

Fig. 2. Temperature dependence of conformity factors of Bi_2Te_3 and *PbTe* based materials with different concentrations of alloying additives for branches: a) *n*-type conductivity, b) *p*-type conductivity.

From the dependencies in Figure 2 is not difficult to determine the intersectional temperature for every branch of thermoelement. The concentration of dopant is chosen similar as in the calculation of homogeneous modules basis on Bi_2Te_3 and *PbTe* materials. Temperatures between sections of *n*- and *p*-type branches are: $T^n_{intersect}$ = 488 K, $T^p_{intersect}$ = 443 K.

Two-section module parameters are similar to modules from homogeneous materials. The heights of the sections under which the approaches the setpoint intersectional temperatures are determined: l^n_{cold} = 2.2 mm; l^n_{hot} = 3.4 mm; l^p_{cold} = 1.55 mm; l^p_{hot} = 4.05 mm. The determination of optimal geometrical parameters two-section Bi_2Te_3 / *PbTe* based thermoelement with excluding intersectional losses of electric power made it possible to calculate the main characteristics of the module, operating in the temperature range 303-773 K: maximum efficiency is 13.5% and electrical power – 17 watts. The temperature dependences of the efficiency and capacity from the hot side temperature are shown in Figure 3.

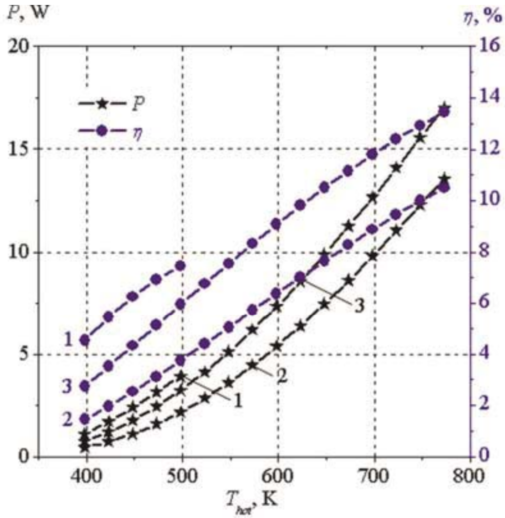

Fig. 3. Temperature dependences of the efficiency and electric power from the hot side temperature when the cold side is 303 K (1 – single-section module based on Bi_2Te_3 materials; 2 – single-section module based on *PbTe* materials; 3 – two-section module basis on Bi_2Te_3 / *PbTe* materials).

Figure 3 shows that maximum efficiency is achieved at a hot side temperature of 773 K for two-section module. However, if the temperature of the hot side does not exceed 500 K, then the best are the characteristics of single-section module based on Bi_2Te_3. Efficiency of two-section module with decreasing hot side temperature decreases faster than the efficiency of *PbTe* based module. This means that for two-section module with decreasing hot side temperature increases losses which are related to the increasing mismatch between the materials sections. That is, for each of the temperature range in which the module has to work, you need to calculate definite ratios between the heights of the sections in which conformity factors of investigated materials are the same.

Conclusions

The main characteristics of the thermoelectric generator modules from homogeneous materials based on Bi_2Te_3 and *PbTe*, working at the maximum possible temperature range for these materials are identified by the method of computer designing in the computer environment COMSOL Multiphysics. Parameters of the two-section module based on Bi_2Te_3 / *PbTe* materials with optimal relations of section heights are calculated.

Using of Bi_2Te_3-based materials as cold sections and *PbTe*-based materials as hot sections for two-section modules allows to raise efficiency of converting thermal energy in the temperature range 303-773K by 1.28 as compared to thermoelectric modules of homogeneous materials.

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Поступила 18.9.2013р.