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DESIGN OF THERMOELECTRIC STAGED MODULES WITH SEGMENTED LEGS BASED ON Bi_2Te_3-*PbTe-TAGS*

The results of computer design of generator staged modules based on Bi_2Te_3*-PbTe-TAGS with segmented legs for improved efficiency of thermoelectric power conversion are presented. A three-dimensional model was used to find the optimal parameters of leg segment materials for each stage and the inter-stage temperature whereby the efficiency of staged module achieves maximum value. Comparative energy characteristics of such modules in the operating temperature range 30 – 500* °C *are given. Design was performed with regard to the temperature dependences of material parameters, the thermal and electrical losses on segment contacts and stage connections.*

Key words: computer design, three-dimensional model, staged generator modules, segmented legs, efficiency, thermoelement.

Introduction

At present, traditional methods for improvement of thermoelectric power conversion efficiency are mainly limited to creation of materials with high values of figure of merit *ZT* [1, 2]. However, despite numerous investigations, no real improvement of the figure of merit has been achieved in recent 40 years [3, 4]. For crystalline thermoelectric materials currently used in industry (materials based on *Bi-Te*, *Pb-Te*, *Ag-Sb-Ge-Si*), the values of *ZT* 1.0-1.6 were achieved as far back as the mid of the previous century [5]. Whereas nanostructured materials, though possessing higher *ZT* values as compared to crystalline ones, are economically unsound because of the high cost of their manufacturing techniques [6]. In this connection, of current concern become basically new approaches to thermoelectric conversion efficiency improvement that are not related to increase in material figure of merit, but are aimed at developing rational recovery schemes of supplied heat [7-9].

Practical application of such schemes has resulted in creation of staged thermoelectric modules based on Bi_2Te_3-*PbTe-TAGS*, where each stage is oriented at its own level of operating temperatures, which allowed improving the efficiency of thermoelectric conversion by a factor of ~1.5 [10-12]. In turn, for further efficiency improvement of staged structures none of the variants of thermal schemes is used yet.

On the other hand, the efficiency of thermoelectric converters can be increased by making thermoelement legs in the form of segments, rather than of homogeneous materials. In so doing, for each segment one can choose different material in the corresponding temperature range, so as to assure maximum figure of merit of leg as a whole [13, 14].

So, the purpose of our work is to estimate the possibility of efficiency improvement of staged thermoelectric modules of materials based on Bi_2Te_3-*PbTe-TAGS* through use of segmented legs in thermoelements of each stage.

Physical model of a staged module with segmented legs and its description

A physical model of a staged module with segmented legs is given in Fig. 1, *a*. Each stage is composed of thermoelements in the form of segments of different thermoelectric materials, connected electrically in series and thermally in parallel. For computer design use was made of a model of elementary structural unit of such module, namely a block of two thermoelements composed of *N* segments. Each thermoelement is optimized for the level of working temperatures of the “hot” and “cold” stage, respectively (Fig. 1, *b*).

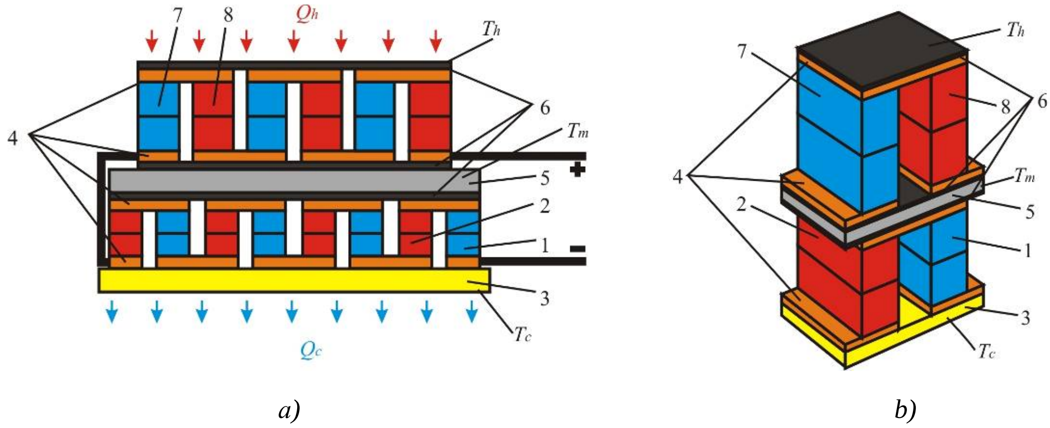


Fig. 1. Physical model of a two-staged module (a) and its elementary structural unit (b):
1, 2 – segmented n-andi p-legs of the cold stage; 3 – ceramic plate;
4 – connecting plates; 5 –heat-conductive plate;
6 – electric insulation; 7, 8 – segmented n-and p-legs of the hot stage.

The model takes into account contact resistance between the legs, leg segments and connecting plates. The temperatures on the heat releasing and heat absorbing thermoelement surfaces T_h and T_c are fixed, the lateral surface is adiabatically isolated. Parameters of thermoelectric materials are a function of temperature and concentration of doping impurities. The space between the legs is filled with the air of thermal conductivity κ_{air}. To determine maximum efficiency, it is necessary to find optimal distribution of current density in the stages and optimal inter-stage temperature.

The formulated problem is solved by a numerical method of successive approximations with the use of “Comsol Multiphysics” program [15]. The choice of geometrical dimensions of thermoelements helps to achieve the electrical compatibility of stages and matching between stages in thermal flux.

Mathematical description of the model

For the description of heat and electric current fluxes in such thermoelement we used the laws of conservation of energy

$$\mathrm{div}\vec{W} = 0\,, \tag{1}$$

and electric charge

$$\mathrm{div}\,\vec{j} = 0\,, \tag{2}$$

where

$$\vec{W} = \vec{q} + U\vec{j}\,, \tag{3}$$

$$\vec{q} = \kappa\nabla T + \alpha T\vec{j}\,, \tag{4}$$

$$\vec{j} = -\sigma\nabla U - \sigma\alpha\nabla T\,, \tag{5}$$

where $\vec{W}$ is energy flux density, $\vec{j}$ is electric current density, U is electric potential, T is temperature, α, σ, κ are the Seebeck coefficient, electric conductivity and thermal conductivity of materials.

Taking into account (3) - (5), we obtain:

$$\vec{W} = -(\kappa + \alpha^2 \sigma T + \alpha U \sigma)\nabla T - (\alpha \sigma T + U\sigma)\nabla U . \tag{6}$$

Then the laws of conservation (1), (2) acquire the form:

$$-\nabla\left[(\kappa + \alpha^2 \sigma T + \alpha U \sigma)\nabla T\right] - \nabla\left[(\alpha \sigma T + U\sigma)\nabla U\right] = 0 , \tag{7}$$

$$-\nabla(\sigma\alpha\nabla T) - \nabla(\sigma \nabla U) = 0 . \tag{8}$$

Exactly these nonlinear second-order differential equations in partial derivatives (7) and (8) determine the distributions of temperature T and potential U in materials of the legs, the contact, connecting and isolation layers of thermoelement.

The boundary conditions for solving of equations (7) and (8) were selected as follows. The temperatures of heat absorbing and heat releasing thermoelement surfaces T_h = 200 °C and T_c = 30 °C were recorded. The zero potential value was assigned on the connecting plate of *n*-type leg. On the other connecting plate of *p*-type leg, the value U was assigned, that is half of thermopower generated by thermoelement. In turn, the value of generated thermopower was determined by a system of equations (7) and (8) in the absence of current through thermoelement.

At the boundaries of the legs and contact layer, the contact layer and connecting plates, the insulation and connecting plates, conditions of equality of temperatures and heat fluxes were taken into account.

The general equation of "Comsol Multiphysics" program is of the form:

$$\nabla\left(-C\nabla M + \alpha M + \gamma\right) + \delta M + \beta \nabla M = f , \tag{9}$$

where

$$C = \begin{bmatrix} C_{11} & C_{12} \\ C_{21} & C_{22} \end{bmatrix},\ \alpha = \begin{bmatrix} \alpha_{11} & \alpha_{12} \\ \alpha_{21} & \alpha_{22} \end{bmatrix},\ \gamma = \begin{bmatrix} \gamma_{11} & \gamma_{12} \\ \gamma_{21} & \gamma_{22} \end{bmatrix},\ \delta = \begin{bmatrix} \delta_{11} & \delta_{12} \\ \delta_{21} & \delta_{22} \end{bmatrix},$$
$$\beta = \begin{bmatrix} \beta_{11} & \beta_{12} \\ \beta_{21} & \beta_{22} \end{bmatrix},\ f = \begin{bmatrix} f_1 \\ f_2 \end{bmatrix},\ M = \begin{bmatrix} T \\ U \end{bmatrix}. \tag{10}$$

From the analysis of equations (7) – (10) it follows that equation (9) can be simplified as follows:

$$\nabla\left(-C\nabla M\right) = 0 . \tag{11}$$

A differential equation for matrix components M is given by:

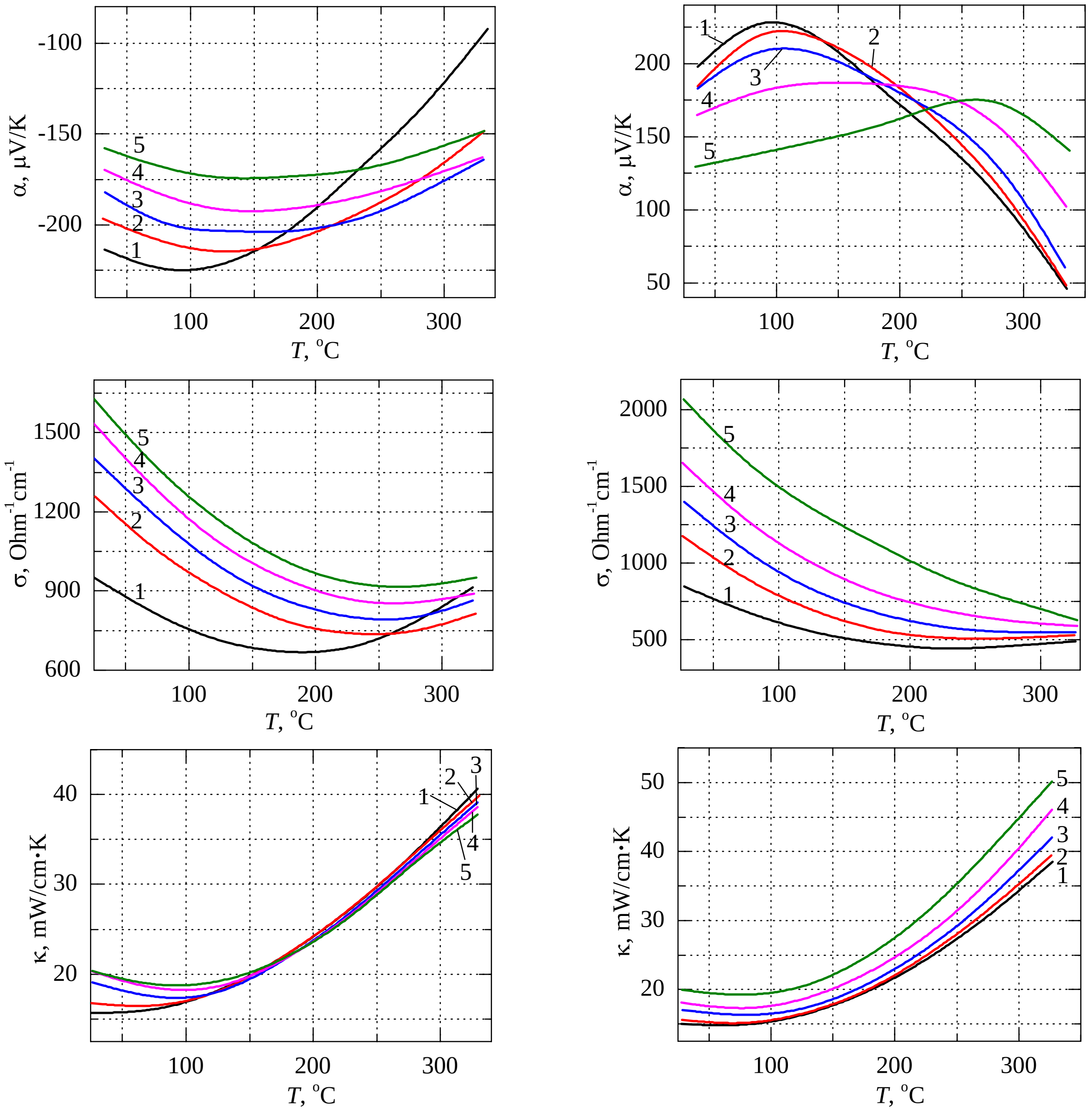
$$\left.\begin{array}{l} \nabla\left(-C_{11}\nabla T\right) + \nabla\left(-C_{12}\nabla U\right) = 0 \\ \nabla\left(-C_{21}\nabla T\right) + \nabla\left(-C_{22}\nabla U\right) = 0 \end{array}\right\}. \tag{12}$$

Comparison of the laws of conservation in the form of (7), (8) to equations (12) yields coefficients for computer model:

$$C = \begin{pmatrix} \kappa + \alpha^2\sigma T + \alpha U \sigma & \alpha\sigma T + U\sigma \\ \sigma\alpha & \sigma \end{pmatrix}. \tag{13}$$

Investigation results

The above method was used to calculate the energy characteristics of two-staged modules with two- and three-segmented legs. As the input data, the experimentally measured in [16-18] temperature dependences of thermoelectric parameters α, σ, κ of materials based on *n*- and *p-Bi_2Te_3* for the cold stage and *n-PbTe* and *p-TAGS* for the hot stage, respectively, were used.



a) *b)*

Fig. 2. Temperature dependences of thermoelectric parameters of the cold stage materials with different electric conductivity values σ_0^n, σ_0^p at T=300 K [16]:
a) n- $(Bi_2Te_3)_{0.90}(Sb_2Te_3)_{0.05}(Sb_2Se_3)_{0.05}$, doped with iodine
(1 – σ_0^n =970 Ohm^{-1}cm^{-1}; 2 – σ_0^n =1250 Ohm^{-1}cm^{-1};
3 – σ_0^n =1400 Ohm^{-1}cm^{-1}; 4 – σ_0^n =1550 Ohm^{-1}cm^{-1}; 5 – σ_0^n =1650 Ohm^{-1}cm^{-1});
b) p- $(Bi_2Te_3)_{0.25}(Sb_2Te_3)_{0.72}(Sb_2Se_3)_{0.03}$, doped with lead
(1 – σ_0^p =880 Ohm^{-1}cm^{-1}; 2 – σ_0^p =1100 Ohm^{-1}cm^{-1}; 3 – σ_0^p =1380 Ohm^{-1}cm^{-1};
4 – σ_0^p =1660 Ohm^{-1}cm^{-1}; 5 – σ_0^p =2000 Ohm^{-1}cm^{-1}).

The temperature dependences given in Figs. 2, 3 were approximated by two-dimensional polynomials (Fig. 4, 5) in the form of $\alpha^{n,p} = \alpha^{n,p}(x_0^{n,p}, T)$, $\sigma^{n,p} = \sigma^{n,p}(x_0^{n,p}, T)$, $\kappa^{n,p} = \kappa^{n,p}(x_0^{n,p}, T)$ whose coefficients were introduced into computer program.

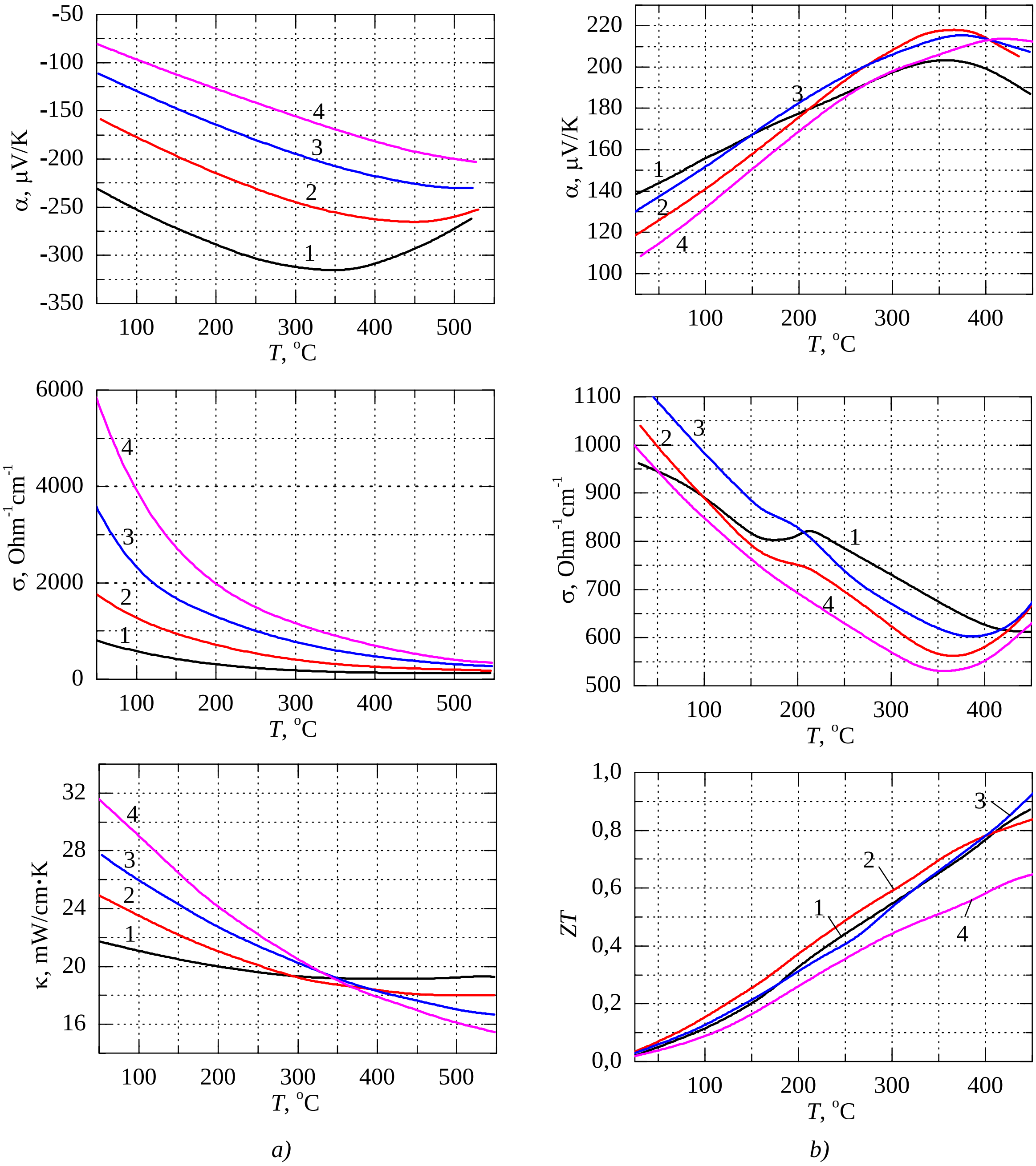


Fig. 3. Temperature dependences of thermoelectric parameters of the hot stage materials n-PbTe and p-TAGS:
a) PbTe <x mol.% PbI_2> (1 – x=0.01; 2 – x=0.03; 3 – x=0.055; 4 – x=0.1) [17];
b) $(GeTe)_{80}(Ag_ySb_{2-y}Te_{3-y})_{20}$ (1 – y= 0.8; 2 – y= 1.0; 3 – y= 1.2; 4 – y= 1.4) [18].

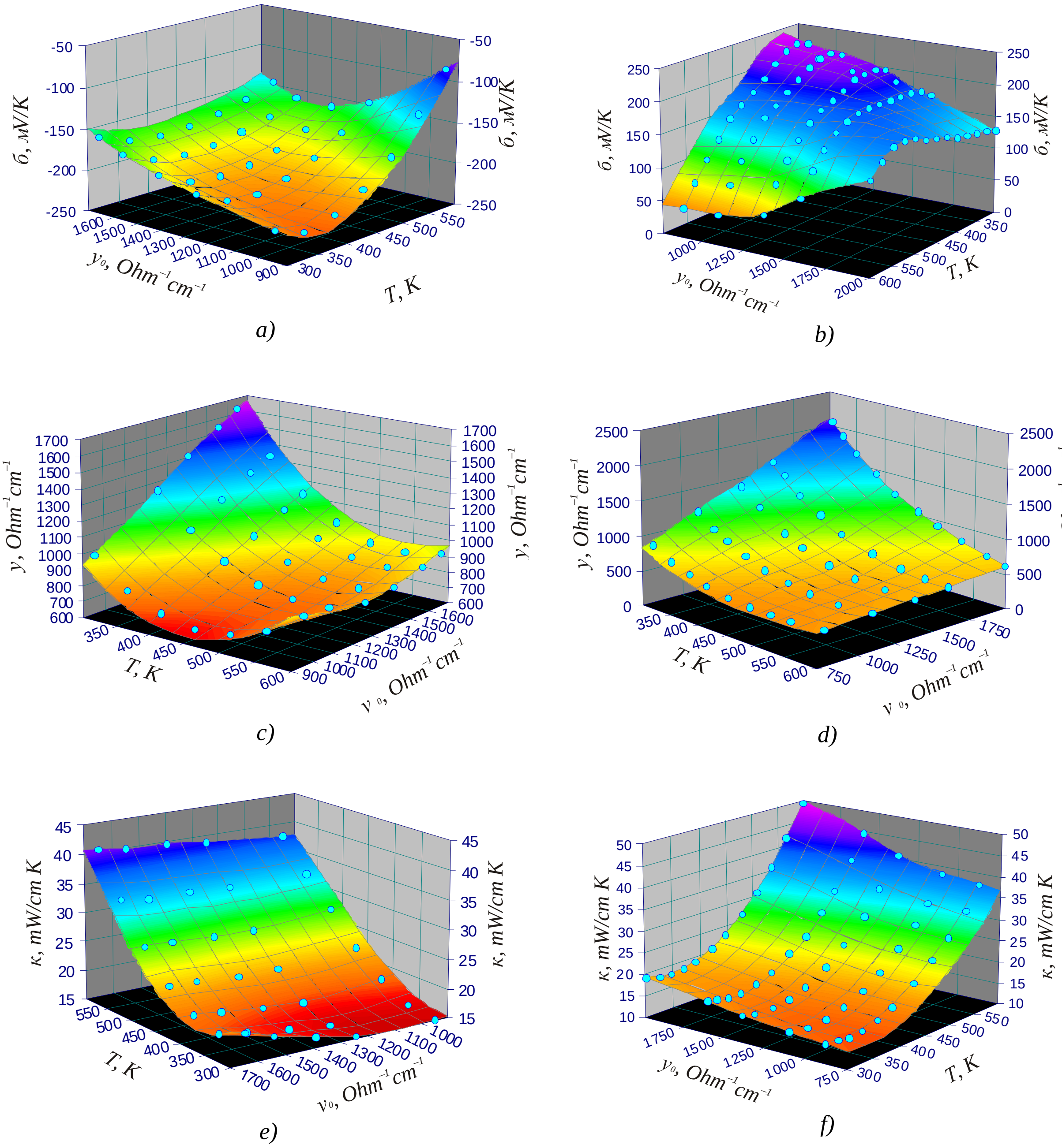


Fig. 4. Temperature-concentration dependences of thermoelectric parameters of the cold stage materials a), c), e) n-$(Bi_2Te_3)_{0.90}(Sb_2Te_3)_{0.05}(Sb_2Se_3)_{0.05}$, doped with iodine. b), d), f) p-$(Bi_2Te_3)_{0.25}(Sb_2Te_3)_{0.72}(Sb_2Se_3)_{0.03}$, doped with lead.

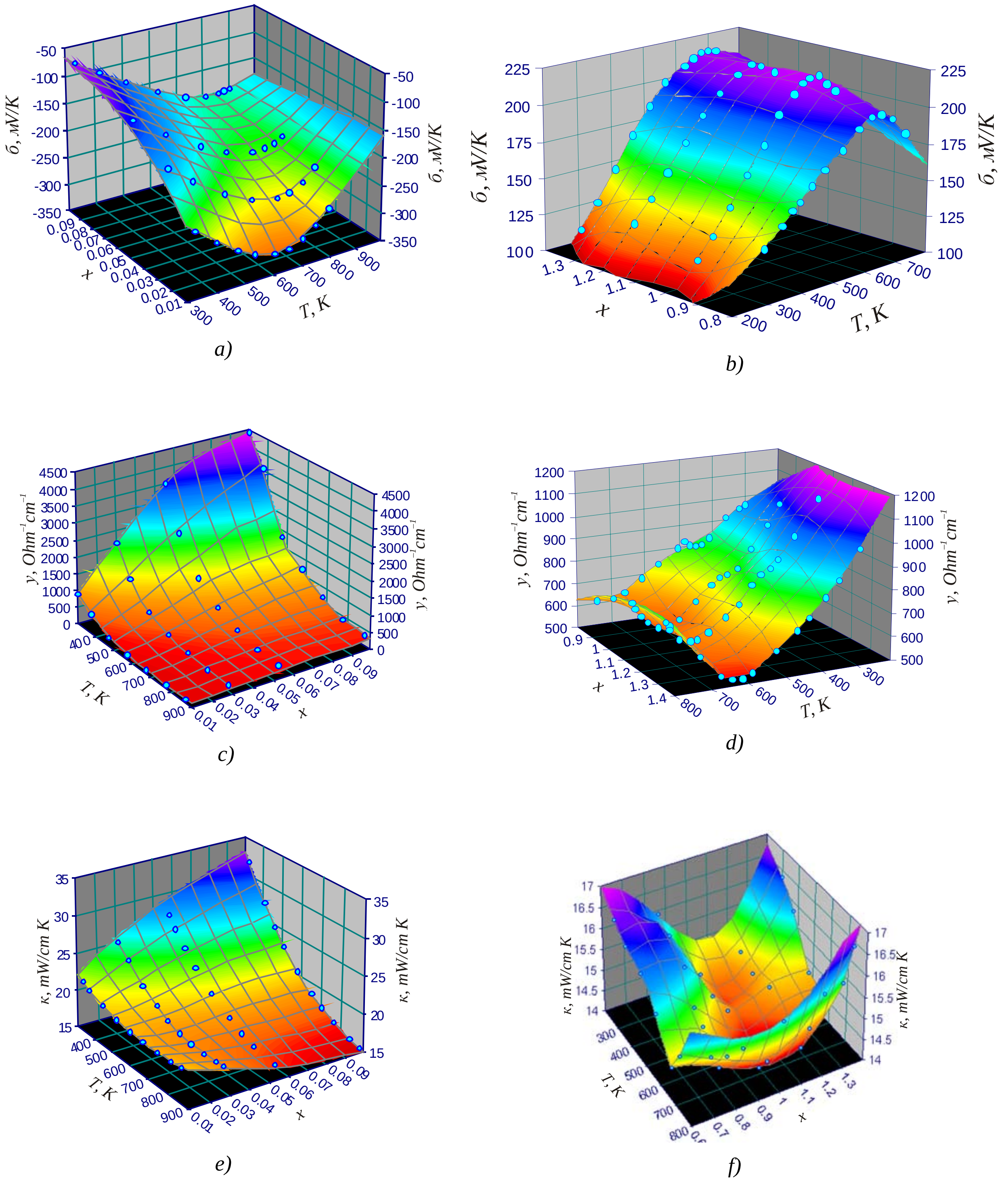
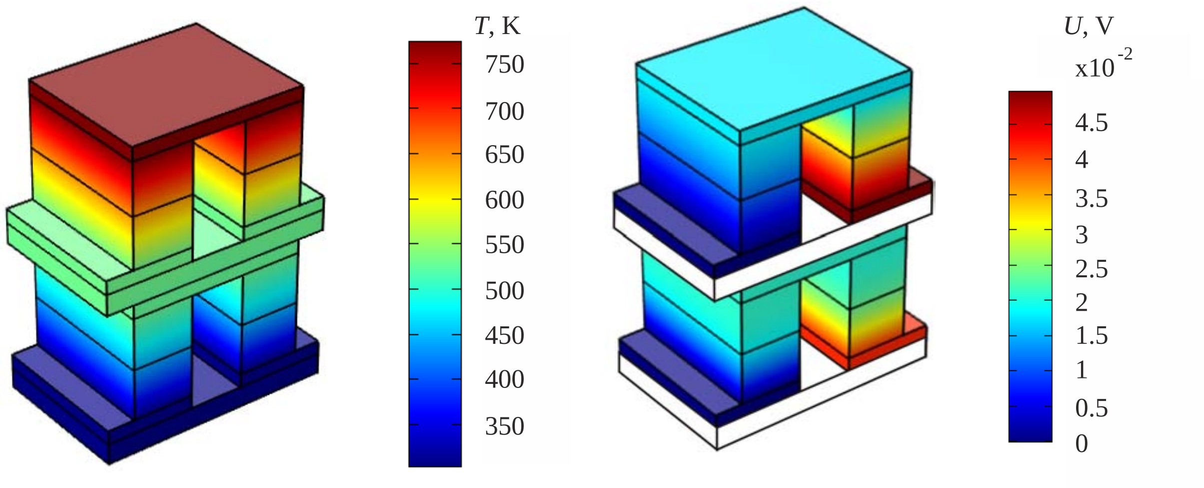


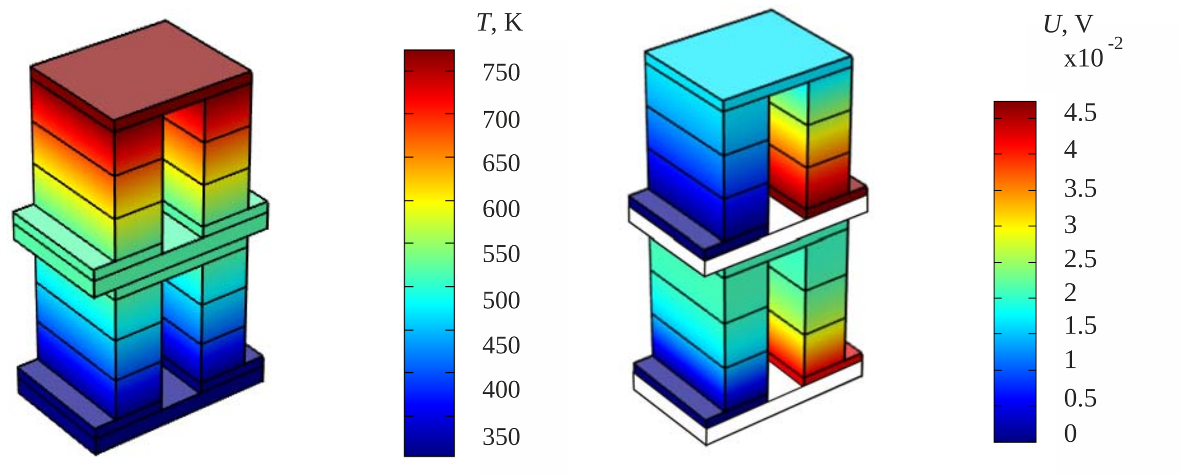
Fig. 5. Temperature-concentration dependences of thermoelectric parameters of the hot stage materials: a), c), e) n-PbTe < x mol.% PbI_2 > ;b), d), f) p-$(Ag_{0.5}Sb_{0.5}Te1_{1-x}(Pb_{0.16}Ge_{0.84}Te)_x$.

As a result of simulation, the distributions of temperature and electric potential in a two-stage module with two-segmented and three-segmented legs were obtained (Figs. 6, 7).



a) *b)*

Fig. 6. Distribution of temperatures (a) and potential (b)
in a staged module with two-segmented legs.



a) *б)*

Fig. 7. Distribution of temperatures (a) and potential (b)
in a staged module with three-segmented legs.

Optimal parameters of segment materials for a staged module are given in Table 1. Optimization was carried out by determination of such impurity concentrations in each segment materials whereby the efficiency of a staged module in the temperature range of 30–250 °C and 250–500 °C reaches its maximum value. The values of contact resistances in the calculations were taken to be $5{\cdot}10^{-6}$ Ohm·cm on thermoelement junctions and $1{\cdot}10^{-5}$ Ohm·cm on the boundaries between leg segments.

Table 1

Optimal segment materials in a staged module

Module	Stage	Leg	Segment	Material	Concentration
Staged with two-segmented legs	cold	*n*-type	First	$(Bi_2Te_3)_{0.90}(Sb_2Te_3)_{0.05}(Sb_2Se_3)_{0.05}$, doped with iodine	σ_0 =1250 $Ohm^{-1}cm^{-1}$
			Second		σ_0 = 1400 $Ohm^{-1}cm^{-1}$
		p-type	First	$(Bi_2Te_3)_{0.25}(Sb_2Te_3)_{0.72}(Sb_2Se_3)_{0.03}$, doped with lead	σ_0 = 1100 $Ohm^{-1}cм^{-1}$
			Second		σ_0 = 2000 $Ohm^{-1}cm^{-1}$
	hot	*n*-type	First	*PbTe*+*x* mol.% PbI_2	*x* = 0.055
			Second		*x* = 0.1
		p-type	First	$(Ag_{0.5}Sb_{0.5}Te)_{1-x}(Pb_{0.16}Ge_{0.84}Te)_x$	*x* = 0.58
			Second		*x* = 0.6
Staged with three-segmented legs	cold	*n*-type	First	$(Bi_2Te_3)_{0.90}(Sb_2Te_3)_{0.05}(Sb_2Se_3)_{0.05}$, doped with iodine	σ_0 = 1380 $Ohm^{-1}cm^{-1}$
			Second		σ_0 = 1660 $Ohm^{-1}cm^{-1}$
			Third		σ_0 = 2000 $Ohm^{-1}cm^{-1}$
		p-type	First	$(Bi_2Te_3)_{0.25}(Sb_2Te_3)_{0.72}(Sb_2Se_3)_{0.03}$, doped with lead	σ_0 = 1250 $Ohm^{-1}cm^{-1}$
			Second		σ_0 = 1400 $Ohm^{-1}cm^{-1}$
			Third		σ_0 = 1550 $Ohm^{-1}cm^{-1}$
	hot	*n*-type	First	*PbTe*+*x* mol.% PbI_2	*x* = 0.6
			Second		*x* = 0.6
			Third		*x* = 0.6
		p-type	First	$(Ag_{0.5}Sb_{0.5}Te)_{1-x}(Pb_{0.16}Ge_{0.84}Te)_x$	*x* = 0.055
			Second		*x* = 0.055
			Third		*x* = 0.1

Fig. 8 *a*, *b* shows the efficiency of a staged module with two- and three-segmented legs as a function of inter-stage temperature.

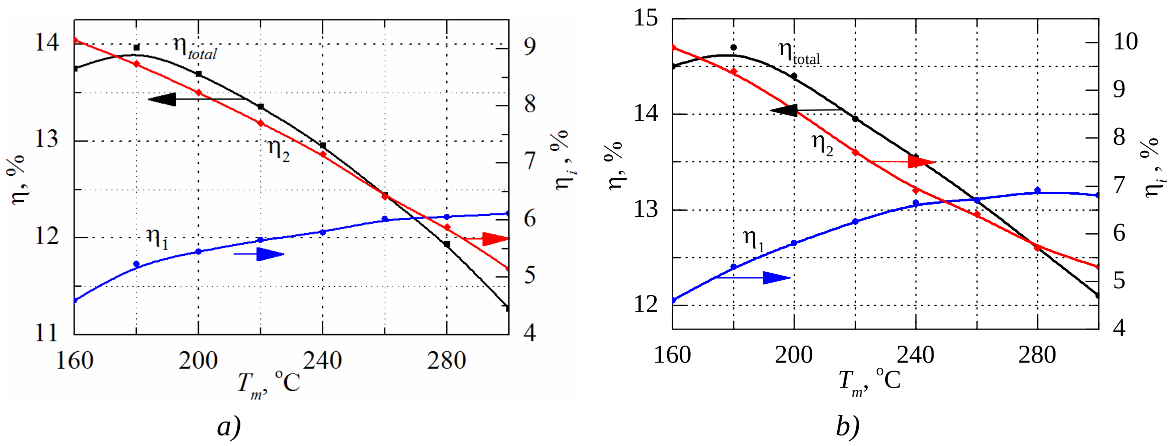


Fig. 8. Efficiency of the cold η_1 and hot η_2 stages,
as well as total efficiency η_{total} of a two-stage module with:
a) two-segmented legs; b) three-segmented legs
versus inter-stage temperature T_m (T_h=500°C, T_c= 30°C).

As is evident from the above data, maximum efficiency of a staged module with two-segmented legs is 14% at inter-stage temperature 180°C. With this inter-stage temperature, the hot side temperature 500°C, and the cold side temperature 30°C, the distribution of efficiency between the stages is as follows: the cold stage – 5.2%, the hot stage – 8.8% (Fig. 8, *a*). In the case of three-segmented legs (Fig. 8, *b*) maximum efficiency of a staged module increases to 14.7 %, which is mainly related to increase in hot stage η (η_2 = 9.4%). The use of the third segment in the cold stage has no significant impact on the increase in total efficiency η_{total} of the module (η_1 = 5.3%).

Fig. 9 presents comparative dependences of the efficiency of a two-staged module based on *Bi_2Te_3-PbTe-TAGS* materials with the homogeneous, two- and three-segmented legs on the inter-stage temperature.

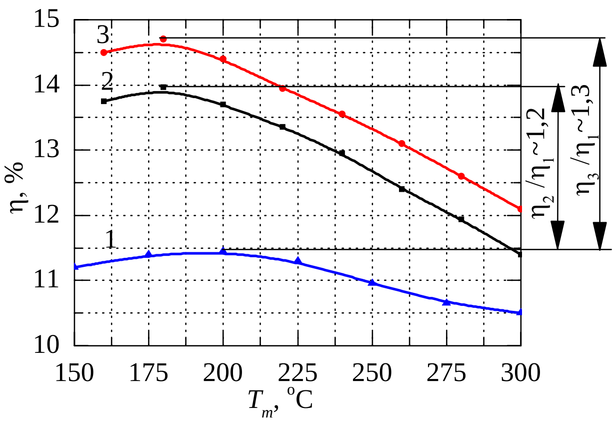


Fig. 9. The efficiency of a two-staged module with 1 – homogeneous legs; 2 – two-segmented legs; 3 –three-segmented legs versus inter-stage temperature T_m.

From the analysis of Fig. 9 it follows that maximum efficiency increase of a two-staged module of *Bi_2Te_3-PbTe-TAGS* in the operating temperature range 30-500 °C is observed when passing from one- to two-segmented leg and makes ~ 20%. Due to introduction of the third segment the efficiency of thermoelectric conversion can be increased by another 5%.

In order to investigate the influence of the number of segments on total module efficiency, similar calculations were performed for a two-stage module with four-segmented legs (Fig. 10).

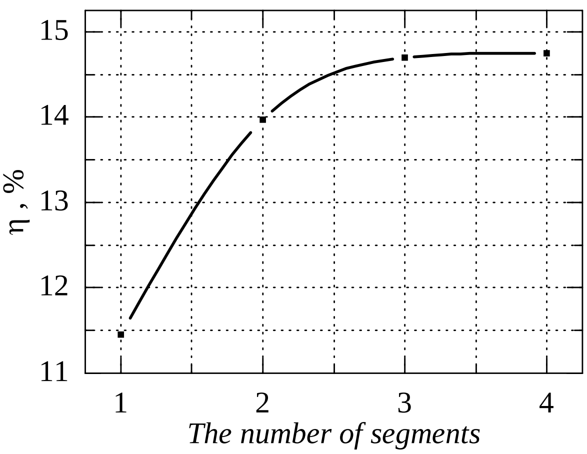


Fig. 10. Dependence of maximum efficiency of a two-stage module of materials based on Bi_2Te_3-PbTe-TAGS on the number of segments in thermoelement legs.

Calculations show that the use of four segments in the legs of each stage is unreasonable, since the efficiency of such module will make ~14.75%. The contribution of the fourth segment to maximum efficiency increase does not exceed 0.5% as compared to a module with three-segmented legs. Such efficiency behaviour is caused primarily by additional contact resistance at points of segments connection, which in turn leads to the Joule heat increase.

Based on the results of computer design, the electrical parameters of elementary structural unit of a two-staged module were calculated for the case of two- and three-segmented legs (Table 2).

Table 2

Electrical parameters of the elementary structural unit of a two-staged module with two- and three-segmented legs. Cross-section of legs is 1.8×4.3 mm

Number of segments	U, V	I, A	P, W	Height of stage legs, mm
2 segments	0.09	2.76	0.25	h_1=3 mm h_2=7 mm
3 segments	0.09	3.1	0.28	h_1=3 mm h_2=7,5 mm

Using the data given in Table 2, one can design a thermoelectric generator module with the required electrical characteristics. For instance, for creation of a two-staged module with two-segmented legs of electric power 5 W, on condition of a series connection of thermoelements, it is necessary to have 20 “blocks”. In so doing, the output electric voltage of such module will be 1.8 V.

Conclusions

1. The method for efficiency improvement of thermoelectric two-staged generator modules of materials based on *Bi_2Te_3-PbTe-TAGS* through use of segmented legs in thermoelements of each stage is presented .

2. Computer methods were used in the design and to determine the efficiency of such modules with two- and three-segmented legs in the operating temperature range 30–500°C. It is shown that maximum efficiency of staged modules with two- and three-segmented legs is achieved at inter-stage temperature 180 °C and makes 14% and 14.7%, respectively.

3. The use in module stages of thermoelements with two- and three-segmented legs allows improving the efficiency of thermoelectric power conversion by 20-25% as compared to a conventional staged module based on *Bi_2Te_3-PbTe-TAGS*. Further increase in the number of leg segments in stages is ineffective because of increased heat release at points of contact resistances.

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