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DETERMINATION OF THE THERMOELECTRIC PARAMETERS OF MATERIALS FORMING PART OF GENERATOR THERMOELECTRIC MODULES

A method is proposed for determining the thermoelectric parameters of materials forming part of generator thermoelectric modules. A detailed physical model of this method is considered and the results of estimating possible values of errors are presented. The effectiveness of various methods for reducing errors, in particular, the use of gradient radiation shields and thermal switches, has been studied. Bibl. 8, Fig 6, Table 1.
Key words: measurement, electric conductivity, thermoEMF, thermal conductivity, figure of merit, thermoelectric module.

Introducion

Further progress in thermoelectricity largely depends on the quality of thermoelectric material, which is determined by the figure of merit *Z* of the material on which the efficiency of thermoelectric power converters depends, namely the efficiency of generators, the maximum temperature difference and the coefficient of performance of coolers, and the heating coefficient of heaters.

In this case, the most effective are experimental methods of material optimization, which reduce to creating a set of material samples of different composition and with different concentrations of impurities, measuring their electrical conductivity, thermoEMF, thermal conductivity and determining the figure of merit *Z*. In this procedure, the correct measurement of these material parameters plays a decisive role.

Preliminary studies [1-7] carried out at the Institute of Thermoelectricity of the National Academy of Sciences and the Ministry of Education and Science of Ukraine made it possible to develop methods and create equipment for precise determination of the properties of thermoelectric materials by the absolute method, the accuracy of which exceeds the accuracy of world analogues by a factor of 3-5.

The task of creating measuring equipment for studying the parameters of materials used in ready-made thermoelectric power converters remains important. This information is necessary both for optimizing the thermoelectric material for its specific applications, upgrading the design of the thermoelectric converters as such, improving the technology of creating interconnect junctions, and also expanding the possibilities for quality control of the finished product.

The most suitable for solving this problem is the absolute method for measuring the parameters of generator thermoelectric modules and equipment ALTEC-10002 on its basis.

The purpose of this work is to develop a method for determining the thermoelectric parameters of materials forming part of generator thermoelectric modules, estimate possible values of errors of this method and determine conditions for their minimization.

Description of the method for determining the σ, α, κ, *Z* of the legs material when measuring parameters of a generator thermoelectric module

The method proposed to determine the average values σ, α, κ, *Z* of the legs material of which the module is composed is as follows:

- determination of electrical conductivity σ by the measured value of the AC resistance of the module and the known module design;

-- determination of the Seebeck coefficient α by the measured values of module EMF and temperature difference between the heater and the heat sink;

- determination of thermal conductivity κ by the measured values of heat flow through the module (using a heat meter) and temperature difference between the heater and the heat sink.

The average values of electrical conductivity, thermoEMF, thermal conductivity and figure of merit of the material of thermoelectric module legs are determined by the formulae

$$\sigma = \frac{1}{R_M / 2N} \frac{h_1}{a_1 \cdot b_1}, \quad (1)$$

$$\alpha = \frac{E / 2N}{\left(T\text{г}_0 - Tx_0\right)}, \quad (2)$$

$$\kappa = \frac{Q / 2N}{\left(T\text{г}_0 - Tx_0\right)} \frac{h_1}{a_1 \cdot b_1}, \quad (3)$$

$$Z = \frac{\alpha^2 \sigma}{\kappa}, \quad (4)$$

where R_M is the AC module resistance; a_1 x b_1 is the cross-section of legs; h_1 is the height of legs; N is the number of pairs; E is the EMF of module; Th_0 is the temperature of the heat equalizing plate located on the hot side of module; Tc_0 is the temperature on heat meter located on the cold side of module; Q is heat flow through the module measured by heat meter.

However, the values of σ, α, κ, *Z* obtained by formulae (1) - (4) will be inaccurate, because these formulae do not take into account temperature differences between the heater (cooler) and the module, temperature differences on ceramic plates and interconnects, contact and interconnect electrical resistances, heat loss to the environment by convection, radiation and through thermocouple wires and module wires, etc. To estimate possible values of errors, it is necessary to consider a detailed physical model of measurements, which is shown in Fig. 1.

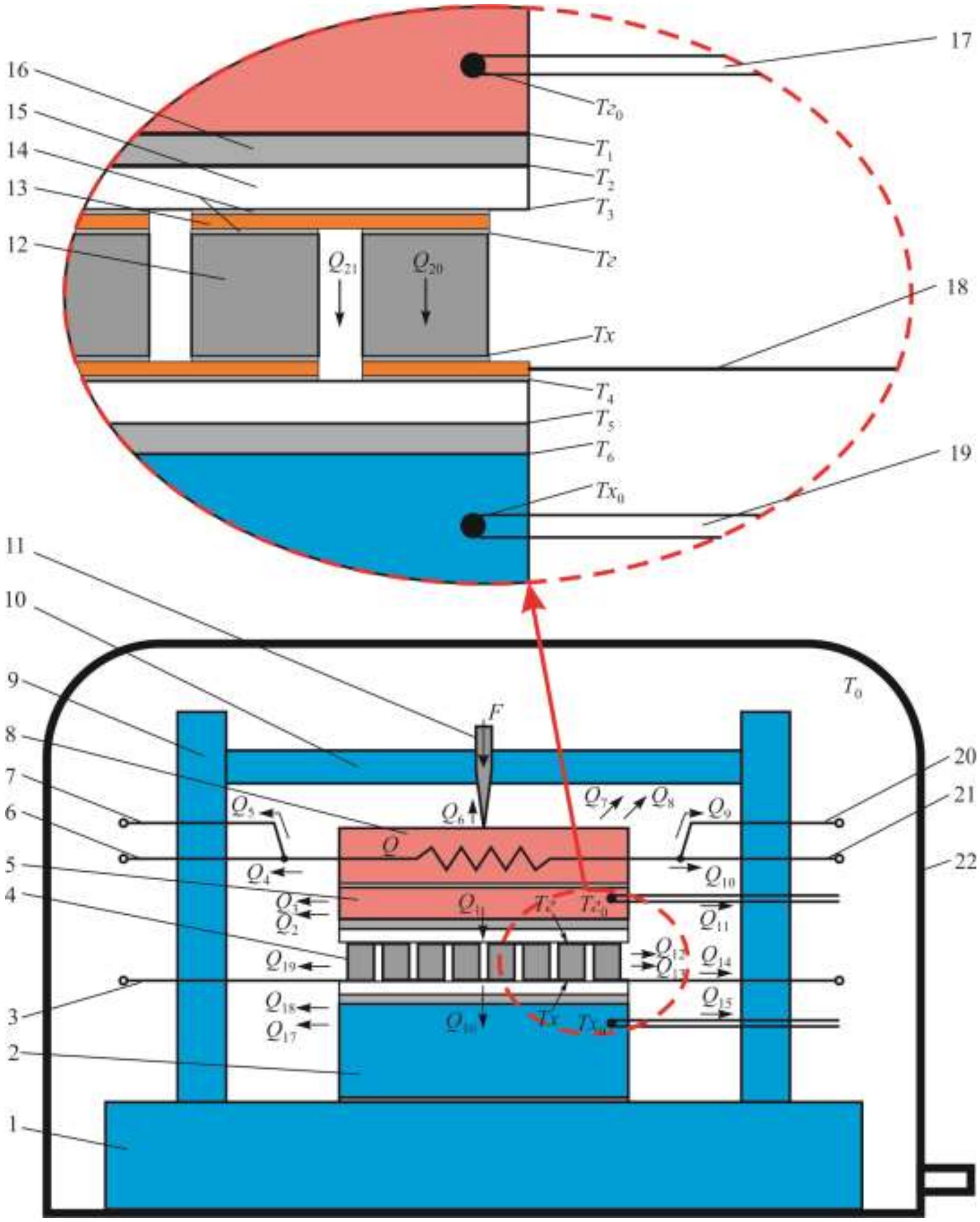

Fig. 1 –Schematic for determining the thermoelectric parameters of materials when measuring the parameters of generator thermoelectric modules:
1 – device for providing the required "cold" temperature of module;
2 – heat meter; 3, 18 – module wires; 4 – module; 5 – heat equalizing plate;
6, 21 – heater wires; 7, 20 – potential heater wires; 8 – module heater; 9 – clampi ng mechanism racks; 10 – clamping bar; 11- clamping screw; 12 – material legs;
13 – interconnect plates; 14 – electric contact resistances; 15 – ceramic plates;
16 – thermal contact resistances; 17, 19 – thermocouples; 22 – bell jar.

In Fig. 1: Q is heat released by heater 8; Q_1 is heat transferred from heater 8 to the "hot" side of module 4; Q_2 is heat transferred from heat equalizing plate 5 to the environment by convection; Q_3 is heat transferred from heat equalizing plate 5 to the environment by radiation; Q_4, Q_{10} is heat transferred from heater 8 to the environment through heater wires; Q_5, Q_9 is heat transferred from heater 8 to the environment through potential heater wires; Q_6 is heat transferred from heater 8 to the environment through the module clamp; Q_7 is heat transferred from heater 8 to the environment by convection; Q_8 is heat transferred from heater 8 to the environment by radiation; Q_{11} is heat transferred from heat equalizing plate 5 to the environment through the wires of thermocouple 17; Q_{12} is heat transferred from module 4 to the environment by convection; Q_{13} is heat transferred from module 4 to the environment by radiation; Q_{14}, Q_{19} is heat transferred from module 4 to the environment through module wires 3 and 18; Q_{15} is heat transferred from heat meter 2 to the environment through the wires of thermocouple 19; Q_{16} is heat transferred from the "cold" side of module 4 to heat meter 2; Q_{17} is heat transferred from heat meter 2 to the environment by convection; Q_{18} is heat transferred from heat meter 2 to the environment by radiation; Q_{20} is heat that passed through module legs 13; Q_{21} is heat transferred from the "hot" to "cold" ceramics in the gap between the legs.

Effective means of reducing the values of the above heat fluxes are a gradient thermal shield and thermal switches on all the wires and the clamp (Fig. 2).

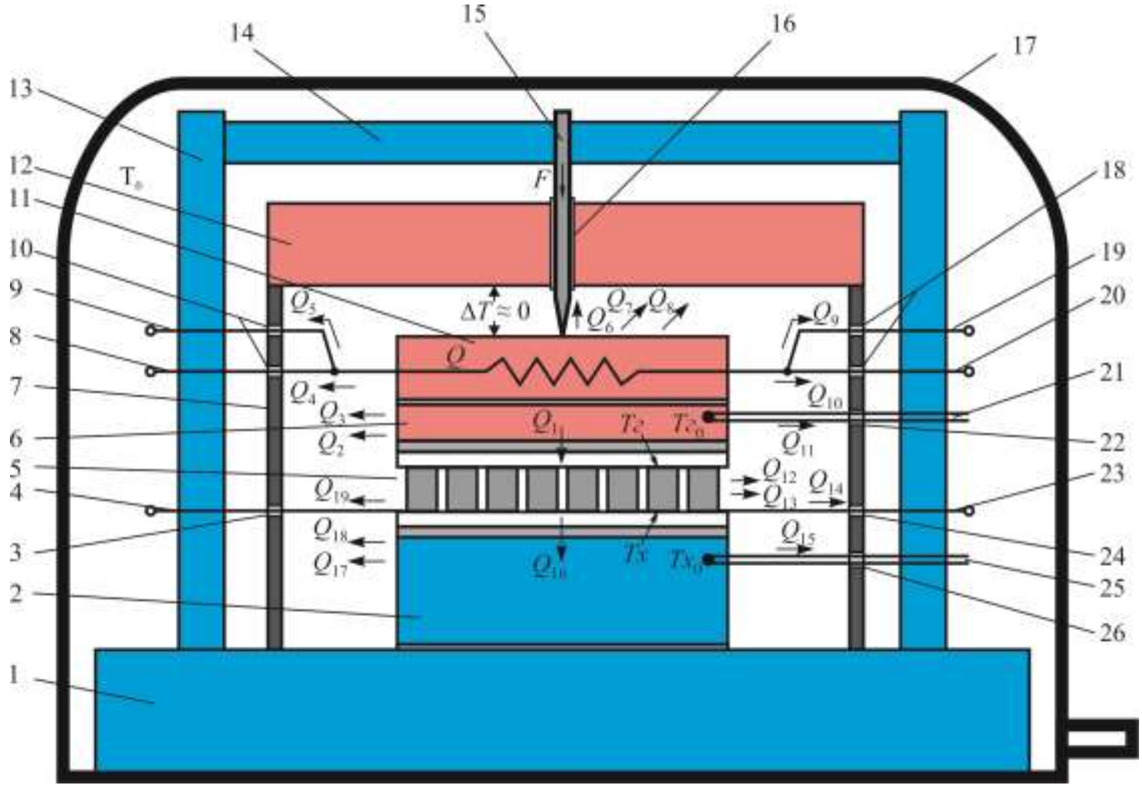

Fig. 2 – Schematic for determining the thermoelectric parameters of materials when measuring the parameters of generator thermoelectric modules with the use of thermal switches and radiation shield: 1 – device for assuring the necessary "cold" temperature of module; 2 – heat meter; 3, 10, 16, 18, 22, 24, 26 – thermal switches; 4, 23 – module wires; 5 – module; 6 – heat equalizing plate; 7 – gradient radiation shield; 8, 20 –heat wires; 9, 19 –potential heater wires; 11 – module heater; 12 –shield heater; 13 – clamping mechanism racks; 14 – clamping bar; 15 – clamping screw; 17 – bell jar; 21, 25 – thermocouples.

In Fig. 2: Q is heat released by heater 11; Q_1 is heat transferred from heater 11 to the "hot" side of module 5; Q_2 is heat transferred from heat equalizing plate 6 to shield 7 by convection; Q_3 is heat transferred from heat equalizing plate 6 to shield 7 by radiation; Q_4, Q_{10} is heat transferred from heater 11 to shield 7 through heater wires; Q_5, Q_9 is heat transferred from heater 11 to shield 7 through potential heater wires; Q_6 is heat transferred from heater 11 to shield 7 through module clamp; Q_7 is heat transferred from heater 11 to shield 7 by convection; Q_8 is heat transferred from heater 11 to shield 7 by radiation; Q_{11} is heat transferred from heat equalizing plate 6 to shield 7 through the wires of thermocouple 21; Q_{12} is heat transferred from module 5 to shield 7 by convection; Q_{13} is heat transferred from module 5 to shield 7 by radiation; Q_{14}, Q_{19} is heat transferred from module 5 to shield 7 through module wires 4 and 23; Q_{15} is heat transferred from heat meter 2 to shield 7 through the wires of thermocouple 25; Q_{16} is heat transferred from the "cold" side of module 5 to heat meter 2; Q_{17} is heat transferred from heat meter 2 to shield 7 by convection; Q_{18} is heat transferred from heat meter 2 to shield 7 by radiation.

Estimation of possible error values of the proposed method

Errors in determining electrical conductivity

When determining the average value of the electrical conductivity of the legs of a thermoelectric module, determined by formula (1), the total AC resistance of the module R_M is used, which, in addition to the resistance of the legs R_1,, also includes the interconnect resistance R_2, the contact resistance R_3 and the resistance of the wires R_4

$$R_M = R_1 + R_2 + R_3 + R_4 . \tag{5}$$

To estimate possible errors, the parameters of the thermoelectric generator module Altec-1061 were used as an example:

– number of pairs – N = 56;
– height of legs h_1 = 3 mm;
– cross-section of legs a_1 x b_1 = 1.8 mm x 4.2 mm;
– ceramics thickness h_2 = 0.65 mm;
– ceramics area a_2 x b_2 = 40 mm x 40 mm;
– interconnect thickness h_3 = 0.25 mm;
– electrical conductivity of legs material σ = 2000 $\text{Ohm}^{-1}\cdot\text{cm}^{-1}$.

For the above values of module geometry and material properties: R_1 = 0.222 Ohm; $R_2 \approx$ 0.004 Ohm; R_3 = 0.03 Ohm (with the value of specific electrical contact resistance 10^{-5} Ohm·cm); R_4 = 0.005 Ohm (for two wires of diameter 1 mm and length10 cm); $R_M \approx$ 0.261 Ohm.

Therefore, the error in determining the electrical conductivity due to disregard for contact resistance will be ~ 13.5%; interconnect resistance ~ 1.8%; wires resistance ~ 2.3%. The errors associated with the accuracy of information about the geometric dimensions of the legs will be determined by the manufacturing technology of the legs and methods for their geometry control. These errors can be reduced by introducing appropriate corrections calculated for a given module design or determined experimentally.

Errors in determining thermoEMF

The errors in determining the Sebeck coefficient of the material of thermoelectric module legs will arise due to the fact that formula (2) should include not the temperature difference ($Th_0 - Tc_0$) on the heat exchangers in contact with the module, but the temperature difference (Th - Tc) directly on the legs

$$\alpha = \frac{E/2N}{(T\textit{г} - Tx)}. \quad (6)$$

Temperature difference on the legs ($Th - Tc$) can be found as

$$(T\textit{г} - Tx) = (T\textit{г}_0 - Tx_0) - (T\textit{г}_0 - T_1) - (T_1 - T_2) - (T_2 - T_3) - (T_3 - T\textit{г}) - (Tx - T_4) -$$

$$- (T_4 - T_5) - (T_5 - T_6) - (T_6 - Tx_0). \quad (7)$$

where ($Th_0 - T_1$) is the temperature difference on the part of the heat equalizing plate between the thermocouple and the surface of this plate in contact with the module; ($T_1 - T_2$) and ($T_5 - T_6$) – temperature differences on the thermal contact resistances of the "hot" and "cold" sides of the module; ($T_2 - T_3$) and ($T_4 - T_5$) - temperature differences on the ceramic plates of the "hot" and "cold" sides of the module; ($T_3 - Th$) and ($T_c - T_4$) - temperature differences on the interconnects of the "hot" and "cold" sides of the module; ($T_6 - Tc_0$) – temperature difference on the part of the heat meter between the thermocouple and the surface in contact with the module.

To estimate the values of these differences, the value of the heat flux Q_1 passing through the module was initially estimated. Without taking into account heat losses from the module and when a temperature difference of 10 K is created on the legs, the heat flux through one leg will be 0.0504 W, and through the entire module – 5.645 W (with the thermal conductivity of the material leg κ = 2 W/(m·K). Then the temperature differences on each of the elements can be estimated as:

$-(T\textit{г}_0 - T_1) = \dfrac{Q_1}{\kappa_{Cu} \cdot \dfrac{a_2 \cdot b_2}{h_{Cu}}} = 0.018K$ (with the distance between the thermocouple and the surface in contact with module h_{Cu} = 2 mm);

$-(T_1 - T_2) = \dfrac{Q_1}{K_{конт.}} = 0.282K$ (with the value of contact thermal resistance $K_{cont.}$ = 20 W/K);

$-(T_2 - T_3) = \dfrac{Q_1}{\kappa_{Al_2O_3} \cdot \dfrac{a_2 \cdot b_2}{h_2}} = 0.153K$;

$-(T_3 - T\textit{г}) = \dfrac{Q_1}{\kappa_{Cu} \cdot \dfrac{2N \cdot a_1 \cdot b_1}{h_3}} = 0.004K$ (on the assumption that heat flux Q_1 is uniformly distributed between 2N interconnect areas with the cross-section equal to leg cross-section and the height equal to interconnect thickness);

$-(Tx - T_4) = (T_3 - T\textit{г}) = 0.004K$;

$-(T_4 - T_5) = (T_2 - T_3) = 0.153K$;

$-(T_5 - T_6) = (T_1 - T_2) = 0.282K$;

$-(T_6 - Tx) = (T\textit{г}_0 - T_1) = 0.018K$.

Thus, the temperature difference measured by thermocouples will be ($Th_0 - Tc_0$) = 10.914 K, which is 9.1% more than the difference on the legs. In this case, the greatest contribution to the error is made by contact thermal resistance (5.6%) and thermal resistance of ceramic plates (3%). These errors can be significantly reduced by introducing corrections determined experimentally.

Errors in determining thermal conductivity

The error in determining the thermal conductivity of the material according to formula (3) will consist of errors in determining the temperature difference on the legs (without introducing corrections ~ 9.1%, according to the calculations given in clause 2.2), errors in measuring the geometric dimensions of the legs and errors in determining the heat flux passed through the legs.

Heat fluxes from the surface of the heater and the heat equalizing plate ($Q_2 - Q_{11}$) can be ignored, since the heat meter measures the heat released from the "cold" side of the module.

Transfer of heat in the gap between the legs by radiation (Q_{21})

$$Q_{21} \approx \varepsilon_1 \sigma_B S\left(T_3^4 - T_4^4\right), \tag{8}$$

where ε_1 is the emissivity of the inner ceramic surface; $\sigma_B = 5.67 \cdot 10^{-8}$ W/(m$^2 \cdot$K^4) is Stephan-Boltzmann constant; $S = (a_1 \cdot b_1 - 2N \cdot a_2 \cdot b_2)$ is the total area of gap between the legs.

Loss of heat Q_{12} from the lateral surface of the module by convection

$$Q_{12} \approx H_{conv}(h_1 + 2h_2 + 2h_3)(2a_2 + 2b_2)\left(\frac{Tг + Tx}{2} - \bar{T}_{екр.}\right), \tag{9}$$

where H_{conv} is coefficient of convective heat exchange, $\bar{T}_{екр.}$ is the average temperature of shield surface which opposite to the surface of module.

Loss of heat Q_{13} з from the lateral surface of the module by radiation

$$Q_{13} \approx \varepsilon_2 \sigma_B (h_1 + 2h_2 + 2h_3)(2a_2 + 2b_2)\left(\left(\frac{Tг + Tx}{2}\right)^4 - \bar{T}^4_{екр.}\right), \tag{10}$$

where ε_2 is emissivity of the lateral surface of the module.

Heat loss from the module through the wires (Q_{14} and Q_{19})

$$Q_{14} = Q_{19} = \kappa_{Cu}\frac{S_4}{L_4}\left(T_4 - T_{ключа}\right), \tag{11}$$

where S_4 is cross-section of the wire; L_4 is the length of the wire; T_{switch} is the temperature of thermal switch.

Loss of heat Q_{15} from the heat meter through thermocouple wires

$$Q_{15} = \kappa_5\frac{S_5}{L_5}\left(Tx_0 - T_0\right) + \kappa_6\frac{S_6}{L_6}\left(Tx_0 - T_{ключа}\right), \tag{12}$$

where S_5 anda S_6 are cross-sections of thermocouple wires; L_5 and L_6 are lengths of thermocouple wires; κ_5 and κ_6 are thermal conductivities of thermocouple wires.

Loss of heat Q_{17} from the heat meter by convection

$$Q_{17} = H_{conv}(h_4)(2a_2 + 2b_2)\left(\frac{Tx_0 + T_0}{2} - T_{екр}\right), \tag{13}$$

where H_{conv} is convective heat transfer coefficient, h_4 is heat meter height.

Loss of heat Q_{19} from the heat meter by radiation

$$Q_{18} = \varepsilon_4 \sigma_B (h_4)(2a_2 + 2b_2)\left(\left(\frac{Tx_0 + T_0}{2}\right)^4 - T_{екр}^4\right), \tag{14}$$

where ε_4 is the emissivity of the heat meter lateral surface.

The following parameters were used to calculate possible values of these heat losses: emissivity - 0.7; diameter of wires - 1 mm; length of wires (before contact with the thermal switch) – 10 cm; diameter of thermocouple wires – 0.2 mm; length of thermocouple wires (before contact with the thermal switch) - 10 cm; number of pairs - 56; legs height - 3 mm; legs cross section - 1.8 mm x 4.2 mm; ceramics thickness - 0.65 mm; ceramics area - 40 mm x 40 mm; interconnects thickness - 0.25 mm.

Losses through the wires and the clamp

Table 1 shows the results of estimation of possible errors in determining the heat flux through the legs of material, caused by heat losses through the wires of the module, heater and thermocouples, as well as through the clamp. They were obtained for different temperature differences ΔT_{switch} between the contact point of the wire with the module (or module heater) and with the thermal switch, i.e. depending on the efficiency of the thermal switch.

Table 1.

Results of estimation of possible errors in determining the heat flux through the legs of material, caused by heat losses through the wires of the module, heater and thermocouples, as well as through the clamp.

№	**Name of losses**	**δQ, % – the ratio of heat losses in the element to heat flux through the legs**			
		ΔT_{switch} = 0.1 K	ΔT_{switch} = 0.2 K	ΔT_{switch} = 0.5 K	ΔT_{switch} = 1 K
1	Losses in the module wires	0.011	0.022	0.056	0.111
2	Losses in the thermocouple wires	< 0.001	< 0.001	< 0.001	< 0.001
3	Losses in the heater wires	< 0.001	< 0.001	< 0.001	< 0.001
4	Losses in the clamp	0.0174	0.0348	0.0869	0.1738

Thus, the largest losses are on the clamp, but they do not affect the readings of the heat meter, so they can be ignored.

The heat losses through the wires when using a thermal switch with an accuracy of at least 1 K are within 0.11%, and the losses through the thermocouple wires, the heater current and potential wires together do not exceed 0.01%.

Radiation in the gap between the legs

According to the results of calculations, the radiation in the gap between the legs is a decisive factor that affects the accuracy of determining the thermal conductivity of the legs.

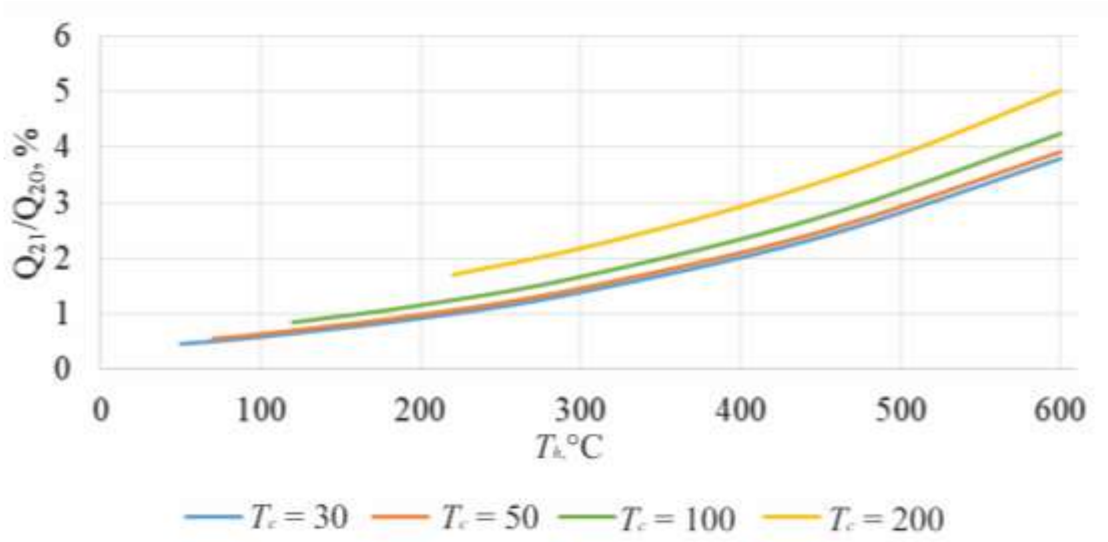

Fig. 3. The ratio between the heat transferred through the module legs and the heat transferred by radiation in the gap between them (for different Tc and Th)

For the temperature intervals of the cold side of the module *Tc* from 30 to 200 °C and hot *Th* - from 30 to 600 °C, heat losses in the gap between the legs grow together with an increase in *Th* and *Tc* and amount to 5% of the total heat flux through the legs with a corresponding difference (3). However, these losses can be determined for a known design and taken into account in the form of corrections.

Radiation from the heater surface

Fig.4 shows the dependences of the heat losses Q_8 on the heater temperature *Th* at a fixed *Tc* = 30°C and different values of the temperature difference ΔT between the module heater and the shield heater.

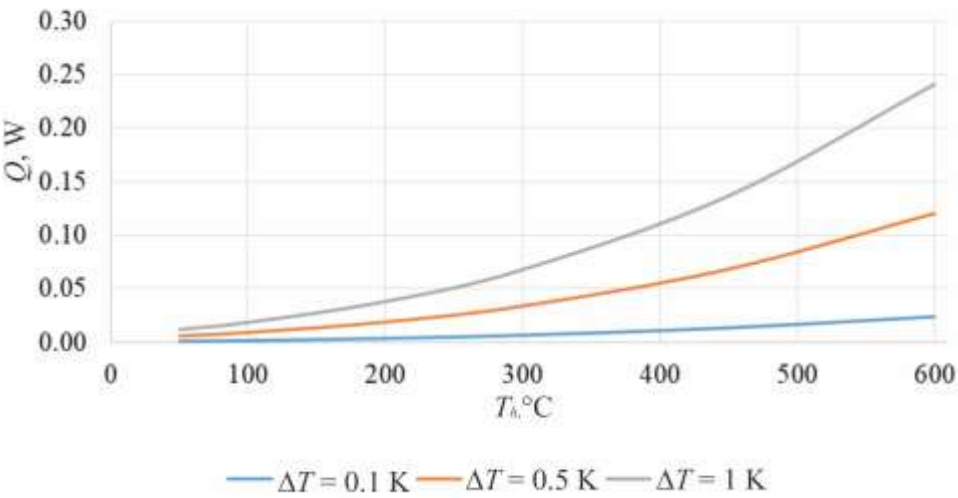

Fig. 4. Dependences of the value of heat losses Q_8 by radiation from the heater surface on the heater temperature Th (at a fixed Tc = 30°C and different values of temperature difference ΔT between the module heater and the shield heater).

The ratio of these losses to the heat flux through the legs at different temperature differences on the module is shown in Fig. 5. The difference between the heater and the shield is 1 K.

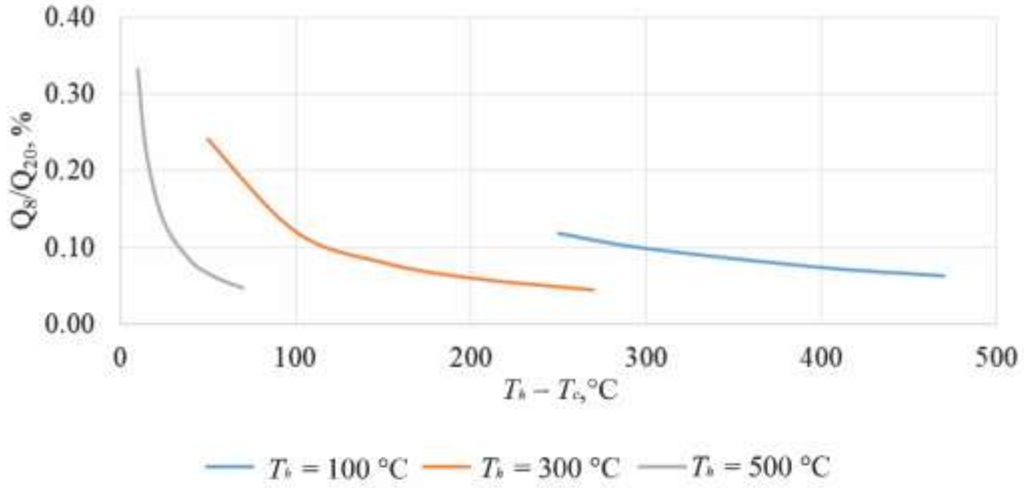

Fig. 5. The ratio between the heat transferred through module legs and the heat transferred by radiation from the heater surface for different values of temperature difference on the module (the temperature difference between the heaters of the module and the shield is 1 K).

It should be noted that radiation from the heater surface can be ignored, since it does not affect the readings of the heat meter located on the cold side of the module.

Radiation from the module surface

The ratio of radiation losses from the lateral surface of the module to the heat flux through the legs at different temperature differences on the module is shown in Fig. 6. The estimate was made for the case when the maximum discrepancy between the temperatures on the module surface and on the shield does not exceed 1K.

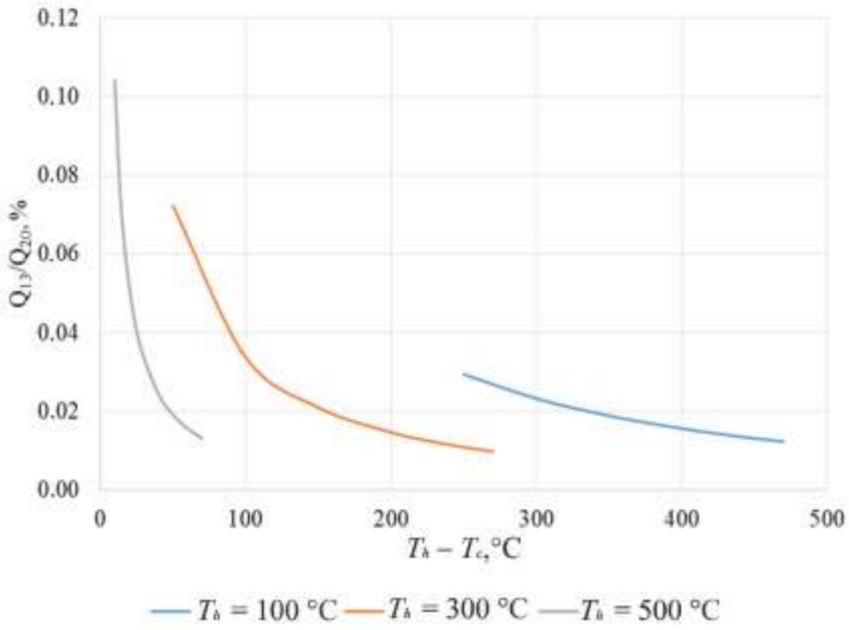

Fig. 6. The ratio between the heat transferred through module legs and the heat transferred by radiation from the module surface for different values of temperature difference on the module

Thus, the total amount of heat losses when using thermal switches and a radiation shield will be 5-7%.

The results obtained are the basis for upgrading the “ALTEC-10002” equipment for measuring the parameters of thermoelectric generator modules.

Conclusions

1. A method is proposed for determining the thermoelectric parameters of the legs material of a generator thermoelectric module when measuring its parameters by the absolute method. A detailed physical model of this method is considered and the results of estimating possible values of errors are presented by the example of a thermoelectric module of the type Altec-1061.
2. It is shown that when determining the electrical conductivity, the decisive factor leading to errors of ~ 13.5%, is disregard for contact electrical resistance. The impact of interconnect resistance will be ~ 1.8%, the resistance of the wires ~ 2.3%. To reduce the value of these errors, appropriate corrections should be made, calculated for a given module design or determined experimentally.
3. When determining the thermoEMF, the largest contribution to the error is made by the errors in determining the temperature difference on the legs, caused by the contact thermal resistance (5.6%) and the thermal resistance of ceramic plates (3%). These errors can be significantly reduced by introducing the appropriate corrections.
4. When determining thermal conductivity, in addition to errors in determining temperature difference on the legs, an additional factor is the presence of heat losses, the total value of which, when using thermal switches and a gradient radiation shield, will be up to 7%. The largest component in this case (up to 5%) will be heat loss by radiation in the gap between the legs. However, these losses for a known module design can be determined and taken into account in the form of corrections.

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Submitted 18.02.2021

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ВИЗНАЧЕННЯ ТЕРМОЕЛЕКТРИЧНИХ ПАРАМЕТРІВ МАТЕРІАЛІВ У СКЛАДІ ГЕНЕРАТОРНИХ ТЕРМОЕЛЕКТРИЧНИХ МОДУЛІВ

Запропоновано методику визначення термоелектричних параметрів матеріалів у складі генераторних термоелектричних модулів. Розглянуто детальну фізичну модель цієї методики та наведено результати оцінки можливих величин похибок. Досліджено ефективність застосування різних методів зниження похибок, зокрема застосування градієнтних радіаційних екранів та теплових ключів. Бібл. 8, рис. 6, табл. 1.
Ключові слова: вимірювання, електропровідність, термоЕРС, теплопровідність, добротність, термоелектричний модуль.

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ОПРЕДЕЛЕНИЕ ТЕРМОЭЛЕКТРИЧЕСКИХ ПАРАМЕТРОВ МАТЕРИАЛОВ В СОСТАВЕ ЕНЕРАТОРНЫХ ТЕРМОЭЛЕКТРИЧЕСКИХ МОДУЛЕЙ

Предложена методика определения термоэлектрических параметров материалов в составе генераторных термоэлектрических модулей. Рассмотрена подробная физическая модель этой методики и приведены результаты оценки возможных величин погрешностей. Исследована эффективность применения различных методов снижения погрешностей, в частности,

применения градиентных радиационных экранов и тепловых ключей. Библ. 8, рис. 6, табл. 1.
Ключевые слова: измерение, электропроводность, термоЭДС, теплопроводность, добротность, термоэлектрический модуль.

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Submitted 18.02.2021