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AUTOMATED MEASURING SYSTEM "ALTEC-10003" FOR THE DETERMINATION OF THERMOELECTRIC PROPERTIES OF MATERIAL INGOTS

The results of development of automated system "ALTEC-10003" intended for automation of the process of measuring the properties of thermoelectric material shaped as rods and data processing are presented. Control unit comprises a multi-channel analog-digital converter, measuring probes travel system, thermal control system and power supplies for measuring unit components. Measurement process control, processing and display of the results are done with the aid of computer to which measuring unit is connected via standard USB channel. The results are displayed as plots and tables. Examples of using the elaborated measuring system control to determine the distributions of material thermoelectric properties in the rods, as well as the analysis of precision and reproducibility of the results are presented.

Key words: electric conductivity, thermoEMF, thermal conductivity, error, thermoelectric material, automation.

Introduction

General characterization of the problem.

Bismuth compounds (Bi_2Te_3, Bi_2Se_3, Bi_2Sb_3) and their solid solutions remain the best materials used in thermoelectric modules for refrigeration. The widely accepted methods for obtaining such thermoelectric materials under industrial conditions are zone recrystallization and extrusion [1]. In the former case, due to segregation of impurities in the process of growth and other factors, the resulting material is inhomogeneous, especially on the rod ends. The extruded material is characterized by considerable distortions of homogeneity at the beginning of a rod, when process conditions are not stable yet.

Therefore, of paramount importance in the manufacture of modules is quality control of thermoelectric material. In this connection, one of the most important tasks when creating quality control equipment is its speed and independence of human factors. For this purpose, full automation of measurement processes is helpful.

Analysis of the literature. The choice of material with the required properties under laboratory conditions is generally done by measuring the distributions of electric conductivity and thermoEMF along the ingot.

The measurement of electric conductivity is based on a two-probe measurement method whereby current is passed through the end surfaces of the ingot, and the electric potential on its surface is measured by two mobile probes with a known distance between them [2, 3]. The electric conductivity is calculated by the values of current and potential difference between the probes with regard to geometrical dimensions (cross-section area of the ingot and the distance between the probes). This method is generally accepted for the investigation of semiconductor material rods (international standard SEMI MF397-02 «Test Method for Resistivity of Silicon Bars Using a Two-Point Probe»).

The measurement of the Seebeck coefficient is based on the hot probe method [4]. One of the two probes is heated relative to the other, and thermoEMF is created between them at contact to ingot. The Seebeck coefficient is calculated as the ratio between the generated thermoEMF and temperature difference between the probes.

To increase the efficiency of express measurements, the electric conductivity and thermoEMF are measured in a single cycle, with one lowering of probes, one of them being heated (Fig. 1). To eliminate thermoEMF in the calculation of electric conductivity, measurements are made with two current directions or on a sign-variable rectangular current meander.

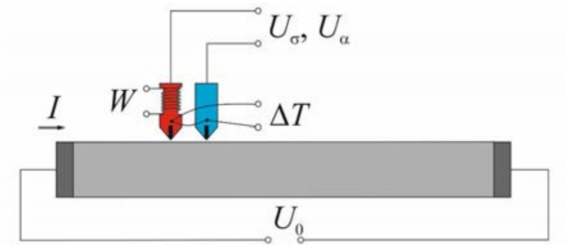


Fig. 1. Schematic for measuring electric conductivity and thermoEMF of ingots.

At the Institute of Thermoelectricity of the NAS and MES of Ukraine, a series of investigations were performed aimed at creating high-precision methods and equipment for measuring the properties of thermoelectric material ingots. These investigations resulted in new physical methods for reducing measurement errors and equipment on their basis for precise measurement of electric conductivity and thermoEMF of materials shaped as ingots [5 – 9]. The following errors of the developed equipment were achieved: electric conductivity – up to 1 %, thermoEMF – to 1.5 %. Also, a procedure for measuring thermal conductivity of ingots under dynamic conditions was created.

For an ingot 300 mm long at a step of 10 mm with the four ingot rotation angles it is necessary to perform over one hundred measurements. When positioning the probes and measuring in manual mode, full measurement cycle may require up to four hours. Moreover, in this case human errors in operator's work are possible when positioning the probes, taking device readings, calculating, plotting of graphs, etc.

Therefore, of particular importance in the development of such equipment is automation of measurement process, which allows not only avoiding human errors, but also increasing considerably measurement speed. Moreover, automation contributes to identity of measurement conditions, hence to measurement precision increase.

The purpose of the work is creation of measurement and probe travel control system for automation of processes of determination of thermoelectric properties of materials, processing and display of their results.

Description of measuring equipment design

The basic specifications in the development of automated measuring equipment were as follows: ingot length – 50 – 400 mm, their diameter – 6 – 30 mm; minimum discreteness of coordinate measurement along the ingot axis – 0.1 mm; minimum discreteness of ingot rotation angle measurement – 1 degree. The installation must be operated by computer which sets measuring tasks, the necessary calculations and their averaging, builds the plots, fills in the tables, saves and prints the results.

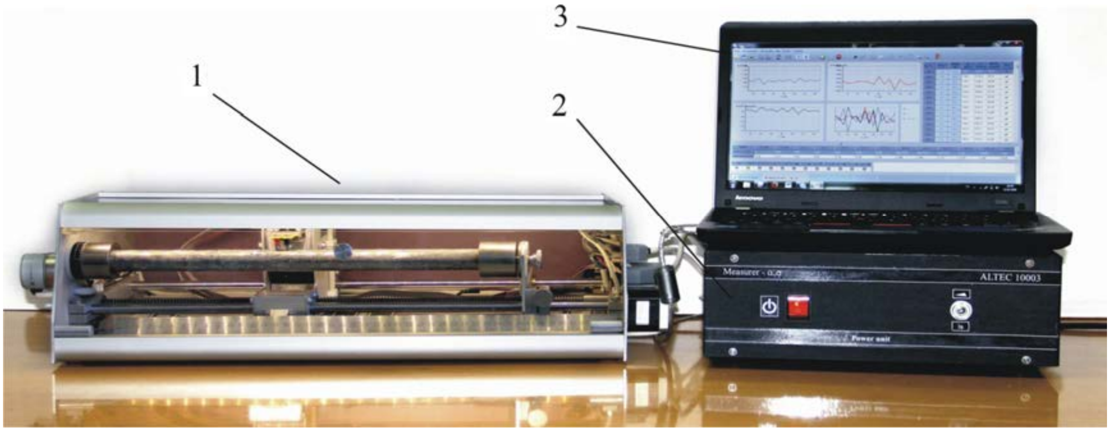


Fig. 2. Installation for measuring thermoelectric parameters of semiconductor material rods “ALTEC-10003”.
1 – measuring unit (rod holder); 2 – control unit;
3 – computer with software support.

The design requires constant distance between the probes, hot probe temperatures, probes hold-down force to sample, equal time of measuring thermoEMF and time of current supply when measuring electric conductivity. To assure equal temperature conditions of measurement, the installation unit involves thermal stabilization system adapted to heating 5 – 10 degrees above room temperature.

The whole measurement process is controlled by high-level computer programs working together with low-level microprograms of control unit AD converter.

The appearance of “ALTEC-10003” installation is given in Fig. 2. It is composed of three units, namely measuring – rod holder, control unit and computer. Its block-diagram is shown in Fig. 3.

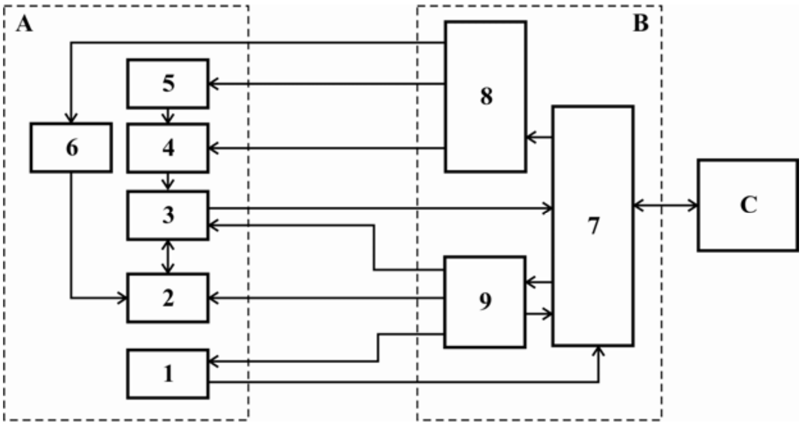


Fig. 3. Block-diagram of installation “ALTEC-10003” for measuring thermoelectric properties of material rods. A – measuring unit, B – control unit, C – computer;
1 – housing thermostat, 2 – thermoelectric material rod, 3 – measuring probes,
4 – probe travel mechanism, 5 – carriage travel mechanism, 6 – rod rotation mechanism,
7 – microcontroller with embedded AD converter; 8 – power supplies and drivers of step motors,
9 – measuring unit.

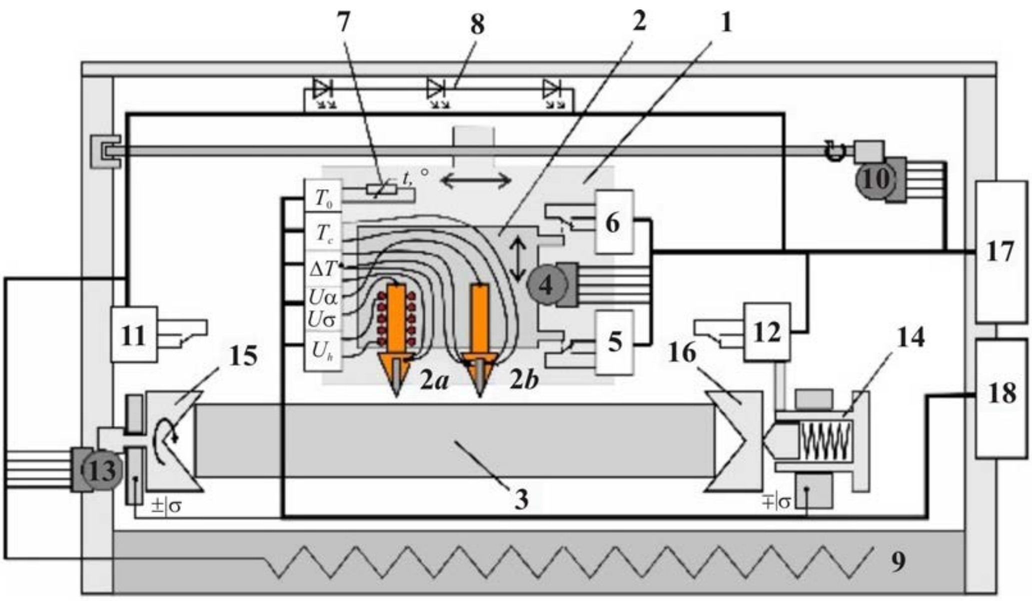


Fig. 4. Functional diagram of measuring unit of installation "ALTEC-10003".
1 – mobile carriage, 2 – measuring unit, 2a – "hot" probe, 2в – "cold" probe,
3 – thermoelectric material rod, 4 – step motor of probe lifting and lowering mechanism,
5, 6 – limit switches of the up and down positions of probes, 7 – ambient temperature sensor,
8 – LED backlight of measuring unit, 9 –heater of measuring unit case,
10 – step motor of mechanism for longitudinal travel of carriage, 11, 12 – limit switches
of mechanism for longitudinal travel of carriage, 13 – step motor of rod rotation mechanism,
14 – rod pressure and fixation mechanism, 15, 16 – cone-shaped current leads,
17, 18 – sockets for connection of measuring unit to measurement control unit.

The main unit in the installation is measuring unit which performs the whole process of primary measurement and consists of rod holder and measuring probes. Its functional diagram is shown in Fig. 4.

Measuring unit is arranged in aluminum housing with a front transparent hinged cover which besides being a structural component performs the function of thermostat. Its bottom accommodates heating elements making it possible to maintain air temperature inside the housing at a level of 300 K. When measuring, thermoelectric material rod is installed into a holder that consists of two coaxial current leads one of which (the right one) is mobile along the rod axis and has hold-down and fixation members.

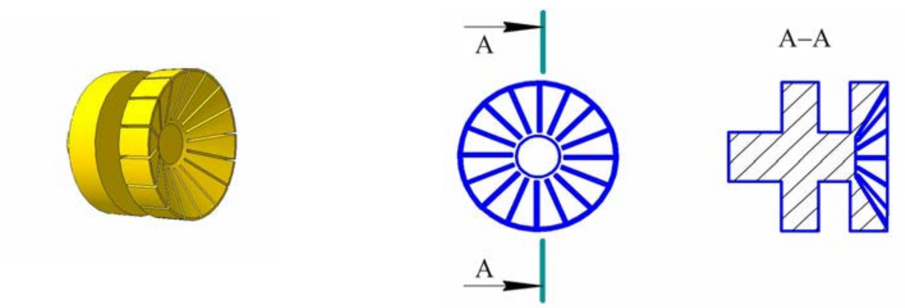


Fig. 5. Appearance and design of current leads.

For self-centering of the rods current leads are made in the form of cut cones (Fig. 5). They have radial grooves which divide hold-down surface into 16 sector parts assuring contact to the real shape of rod ends. This provides for at least 16 points of electric contact for each rod end with current leads. Mounted on the side walls of the housing are means for longitudinal travel of measuring probes and rotation of the rod itself about its axis.

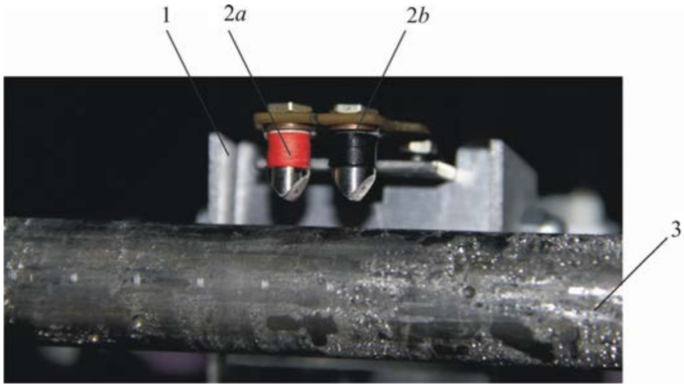


Fig. 6. Measuring unit.
1 – mobile carriage, 2a – "hot" probe, 2b – "cold" probe, 3 – ingot.

Measuring unit is mounted on a mobile carriage (Fig. 6). It accommodates measuring head with two probes. The head is designed to perform electric and temperature measurements and has two rigidly fixed knife-edged probes: one with heating – "hot probe" (2*a*), the other without heating – "cold probe" (2*b*). To reduce the errors in temperature measurement of rod surface contact zone, the probes are made of high thermal conductivity material (copper). For greater durability the probes have embedded knife-edged tungsten plates.

Also, the carriage has probe lifting and lowering mechanism consisting of step motor with a reducer and limit switches of the up and down probe positions.

Automation of measurement process is done by control unit which controls travel of probes in measuring unit, provides for stabilized voltages and currents to electric circuit elements of measuring unit. The voltages and thermoEMF are measured with the aid of an 8-channel 24-digit A/D converter that transfers measured voltages via USB interface to computer. Control unit comprises measuring unit, microcontroller unit and power unit. Power unit components for control of step motors are composed of three identical drivers of stem motors assuring travel of measuring probes in the horizontal and vertical directions and rod rotation.

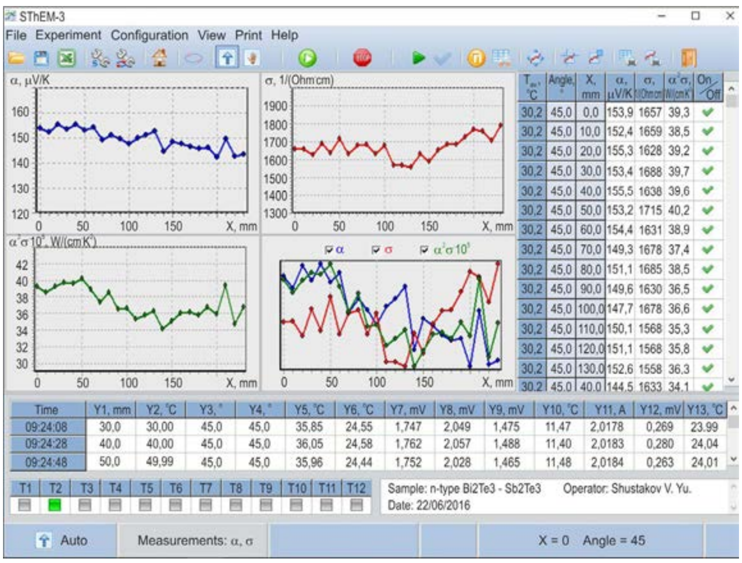


Fig. 7. Main window of measurement control program "SThEM-3".

The installation is controlled by computer with "SThEM-3" (Semiconductor ThermoElectric Material) software, developed jointly with "Tereks" Scientific Industrial Enterprise (Kyiv, Ukraine). The program allows online measurement, processing of measured result, displaying data in the form of plots and tables, saving them in computer, exporting to MS Excel, and printing-out.

Measurement control program "SThEM-3" has a standard structure adopted in Windows operational system. The main window of the program is shown in Fig. 7. It comprises means for control of measurement process (call buttons of experiment control box, on/off indicators of current through the sample, power supply to hot probe heater, etc), the area of plotting measured results, the tables with measured values and calculated values of rod properties. Also displayed is information on the rod entered by operator himself.

The software allows working in "manual" and "automatic" modes.

In "manual" mode the user can install the probes at any point of the rod under study, perform the measurements and calculations with reference to the rod coordinates. In "manual" mode one can also determine the thermal conductivity of the rod. For this purpose, a special device shown in Fig. 8 must be placed on one of the rod ends.

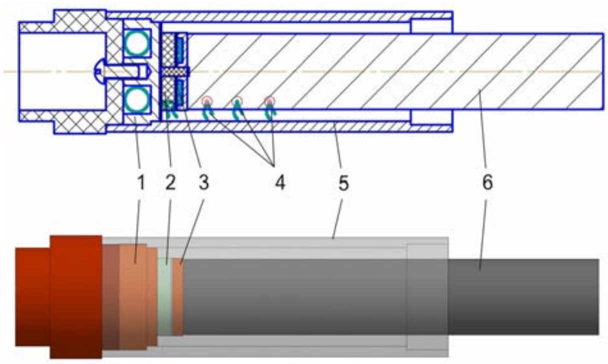


Fig. 8. Design of device for measuring thermal conductivity.

The device is composed of sample reference heater 3, thermoelectric material rod 6 with installed thermocouple probes 4. To reduce thermal losses and assure reproduction of equal thermal conditions when measuring, the device is provided with screen heater 1, differential "null-thermocouple" 2 and screen tube 5.

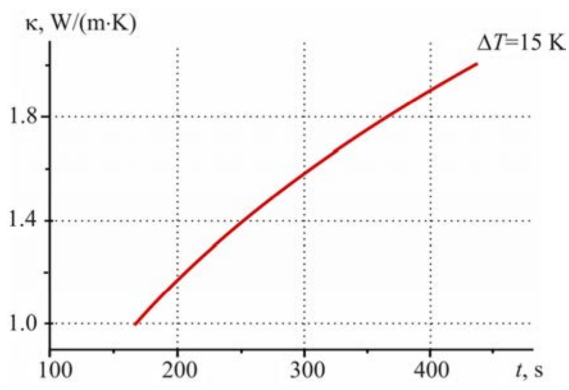


Fig. 9. Calibration dependence for determination of thermal conductivity.

Calibration dependence obtained by computer simulation for this device design is given in Fig. 9.

In "automatic" mode the user shapes the task as a table of coordinates according to which the installation automatically performs a set of measurements in this cyclogram, calculates electrophysical parameters of material with reference to the rod coordinates.

The calculated result is tabulated, and a plot of the value versus measurement coordinate is built simultaneously. In the table, one can average the results for all rotation angles, save and print out the tables and the plots.

It is worth mentioning that the hardware and software parts of the installation are developed with the possibility of measurements both on the rods and flat samples of any size, for instance, discs. All one has to do is to change sample holder and set the appropriate coefficients in the program.

Experimental studies on automated equipment "ALTEC-10003" to measure the properties of thermoelectric material ingots.

Jointly with State enterprise "Bukovinastandartmetrologiya", the program and procedure of metrological certification of installation "ALTEC-10003" was developed and approved. It was established that a relative error in the measurement of electric conductivity is not more than 0.5 %, in the measurement of thermoEMF – not more than 1 %, which corresponds to expected values obtained by means of computer simulation.

Also, the speed and productivity of equipment was determined. It was established that one measurement takes 20 sec. Accordingly, measurement of a rod 30 cm long with its four rotation angles and step 10 mm will require 40 min.

Table

Comparing thermal conductivity values measured on the installation "ALTEC-10003" to thermal conductivity values measured by the absolute method on the installation "ALTEC-10001"

Rod №	Thermal conductivity value measured on the installation "ALTEC-10003", κ, W/(m*K)	Thermal conductivity value measured on the installation "ALTEC-10001", κ_0, W/(m*K)	Deviation, %
n-type $Bi_2Te_3 – Sb_2Te_3$			
1	1.8	1.93	7.0
2	1.4	1.54	9.1
3	1.6	1.74	8.0
p-type $Bi_2Te_3 – Sb_2Te_3$			
4	1.6	1.77	9.6
5	1.7	1.85	8.0
6	1.8	1.93	6.7

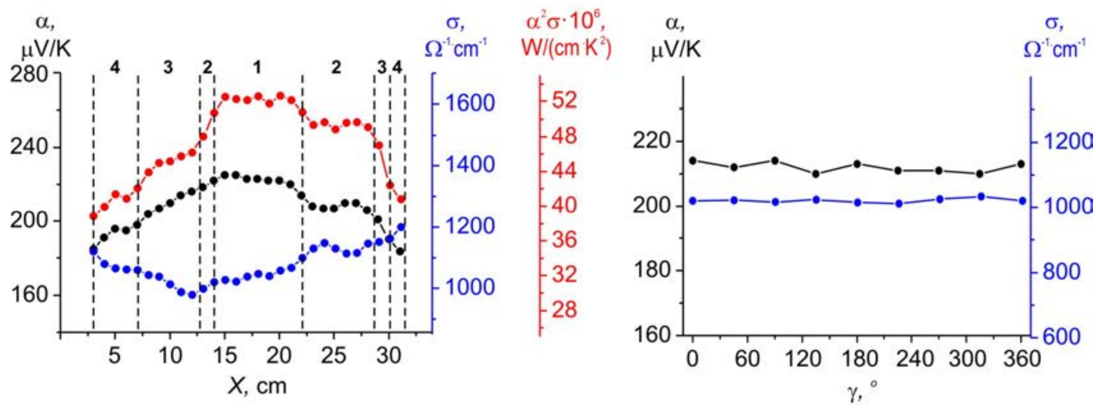
Also, investigations of error in measuring thermal conductivity were performed. For this purpose, thermal conductivity values obtained when measuring on the rods were compared to thermal conductivity values of samples cut of the same rods and measured on the installation "ALTEC-10001" by the absolute method. The results of comparison are listed in Table.

As can be seen from the table, the error in measuring thermal conductivity on the installation "ALTEC-10003" is different from the measurements by the absolute method by 7 – 10 %.

System automated measurements of rods are useful for the optimization of composition and production conditions of thermoelectric materials.

The use of such equipment is particularly attractive under production conditions. A single installation "ALTEC-10003" can realize quality control of nearly 1500 kg of thermoelectric material per year. The introduction of such control allows not only eliminating rejected rods from modules fabrication technology. Determination of $\alpha^2\sigma$ along the rods makes it possible to find reliably and with minimum losses the places of low-quality material on their ends that must be eliminated. The topography of $\alpha^2\sigma$ allows also determination of places on the rods where material quality is high, average or reduced. With regard to equal electric conductivity values such rejecting allows choosing thermoelectric material for the modules of enhanced, average and reduced quality. In so doing, the modules of enhanced quality must have better parameters than those made without using automated quality control.

The use of automated system "ALTEC-10003" under production conditions has proved its efficiency. Investigations were performed in "ALTEC-M" company. Typical dependences of rod properties are given in Fig. 10. In the figure, one can see ingot part 1 with maximum value $\alpha^2\sigma$, part 2 with the values $\alpha^2\sigma$ corresponding to averaged quality values of non-rejected modules, part 3 with the values $\alpha^2\sigma$ somewhat lower than the averaged and part 4 with the unsatisfactory values $\alpha^2\sigma$. The latter are eliminated from the technological process of modules manufacture. This quality distribution of material provides for manufacture of modules with the values ΔT_{max} 75 – 73 K from ingot part 1, 72 – 70 K from part 2 and 69 – 68 K from part 3.



a) *b)*

Fig. 10. Distributions of the Seebeck coefficient and electric conductivity in the n-type thermoelectric material based on $Bi_2Te_3 – Sb_2Te_3$, solid solution prepared by vertical zone melting method (a – along the length X of the rod, b – along the angle of rotation γ of the rod).

Thus, the use of automated measurements in the industrial manufacture of modules allows getting modules of enhanced quality and rejecting low-quality material.

Conclusions

1. State metrological certification of automated measuring complex "ALTEC-10003" has established that a relative error in the measurement of electric conductivity is not more than 0.5 % and not more than 1 % in the measurement of thermoEMF, which corresponds to the values obtained by means of computer simulation.
2. By comparing the results of measuring thermal conductivity of thermoelectric material ingots on the installation "ALTEC-10003" to thermal conductivity values of samples cut of these ingots and measured on the installation "ALTEC-10001" by the absolute method it was

established that the error of thermal conductivity determination by the developed device lies within 7 – 10 %.

3. The efficiency of using measuring complex "ALTEC-10003" under production conditions has been confirmed. It has been established that introduction of automated control of material quality allows not only withdrawing low-quality material from the technological process of modules manufacture , but also getting modules of enhanced quality with ΔT_{max} about 2 – 4 K higher.

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