ELEKTRO-GENERATINQ DOPED ALLOY – INFLUENCE OF RESONANT CONDITIONS OF TIN AND COMPLEX IMPURITY CdCl$_2$ ON OPTIMUM PHYSICAL PROPERTIES OF TELLURIDE OF BISMUTH AND ITS FIRM SOLUTIONS

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ABSTRACT

In a firm solution (Bi$_2$Te$_3$-96mol % - Bi$_2$Se$_3$ 4 mol%) <Sn, CdCl$_2$> on received by a method of the vertical directed crystallization in crystals are investigated electric conductivity (δ) along a layer (0001), factor Zeebek (α) and thermoelectric efficiency. The received crystals had high uniformity only in a primary part of length of ingot (L=30mm); the section of an ingot made Ø(8-20) mm. The data on existence of impurity conditions of tin located on a background of a valence zone, has been confirmed by the analysis and comparison of the results received by us and known literary data. Besides that due to a complex impurity Sn+CdCl$_2$ it was possible to keep not only flat, but also exceed thermo-electromotive factor in samples (Bi-Te-Se) <Sn, <CdCl$_2$> in comparison with not alloyed tin.

Keyboard: factor Zeebek, mono-crystals, кinetic, temperature, electronic, structure.

I. INTRODUCTION

Last 15-20 years the non-traditional direction on improvement of thermoelectric parameters due to use of the unusual phenomena with resonant conditions of tin in a valence zone of telluride of bismuth has appeared. These conditions of tin appreciably modify physical properties of compound Bi$_2$Te$_3$ <Sn> [1-5]. Presence of resonant conditions exceeds electric uniformity of mono-crystals even brought up by Czochralski method, with additional charging by a liquid phase from floating crucible [5]. Necessary researches on influence Sn on thermoelectric and kinetic properties of firm solutions on the basis of telluride of bismuth are carried out by authors of the work [6]. In this article [6] the results of research of physical properties of mono-crystals of firm solutions (Bi$_2$Te$_{3-x}$ Se$_x$) <Sn> (x=0,06 ; 0,12). Kinetic factors are investigated: δ, δ, Hall R$_{321}$ and Nerns-Ettingsgauzen in a range of temperatures 77-400К, and also distribution of thermo-electromotive factor with the help micro hot probe on a surface of crystals changes character of their temperature dependences [6]. What characteristic changes are they?

First of all these features are reflected in temperature dependences R$_{321}$, α, Q$_{123}$, δ. So for example, temperature dependences R$_{321}$ change from "usual" for not alloyed firm solutions up to strongly falling with increase in temperature at doping of tin atoms. Temperature dependences of factor Zeebek and in samples with Sn become more flat in comparison with not alloyed samples. At temperatures T <200°К and T> 380°К samples Bi$_2$Te$_3$ <Sn> have the big thermo-electromotive factors .

In researched firm solutions authors [6] connect features of experimental data on the kinetic phenomena with displays impurity conditions of tin located on a background of a valence zone, is similar to p-Bi$_2$Te$_3$ <Sn>. These conditions are partially filled with electrons. At the increase of temperature the degree of filling impurity conditions changes, that results in change of temperature dependences α and R$_{321}$ in comparison with not alloyed firm solutions.

Features of electro-physical properties of firm solutions Bi$_2$Te$_3$-Se$_x$ alloyed by tin are also shown in structure [7]. By the method of appearing through electronic microscopy it is revealed, that a prevailing kind of defects in the given mono-crystals received by Czochralsk method, are those located in a plane of basis (0001) of disposition. The estimated density of dispositions makes 108-109 cm . A plane of sliding of dispositions is the plane of basis. From the analysis of pictures presence of defects of packing and very small dislocation loops is revealed. Observable defects of crystal structure do not render essential influence on electro-physical properties of firm solutions Bi$_{2-y}$Sn$_y$Te$_{3-x}$Se$_x$ [7]. The structure of samples was described by chemical formula Bi$_{2-y}$Sn$_y$Te$_{3-x}$Se$_x$ [7], where x=0,05 both 0,12; y=0 and 0,01.

Conditions for formation of the fixed units of dislocation lines that promote duplication of dispositions on the mechanism of Franc – Reed do not arise. Experimentally are not fixed the dislocation configurations, capable to work as sources of Franc - Reed. Dislocation loops are obviously caused by the formation of dot defects - internodal atoms and vacancies. The authors [7] do not exclude the possibility of formation education of internodal loops and their

simultaneous presence with vacancy loops. It is necessary to note, that the establishment of quantitative ratio between these loops demands more detailed studying.

In separate sites of crystals the block structure is established. On a surface of contact of the areas having structural discrepancy, dispositions and dislocation grids are revealed at simultaneously observable parallel moire pattern. The moire pattern is caused by occurrence of elastic deformation owing to formation of hexagonal grids of dispositions. Under influence of deformation, the compression (stretching) of a basic plane is possible. Presence of insignificant area of a crystal with moire pattern, is probably connected to variations of structure or a congestion of the dot defects causing structural discrepancy [7].

II. EXPERIMENTAL RESUTS AND THEIR DISCUSSION.TECHNOLOGY

Monocrystals were raised by a method vertically directed crystallization from components (B1, Te, Se) and impurity (Sn, CdCl$_2$). In spite of the fact that the used impurities were of high cleanliness, for the receptions of reliable results bismuth and tellurium have been subjected to additional clearing. Bismuth at temperature 500°C was poured through very thin aperture from one part of an
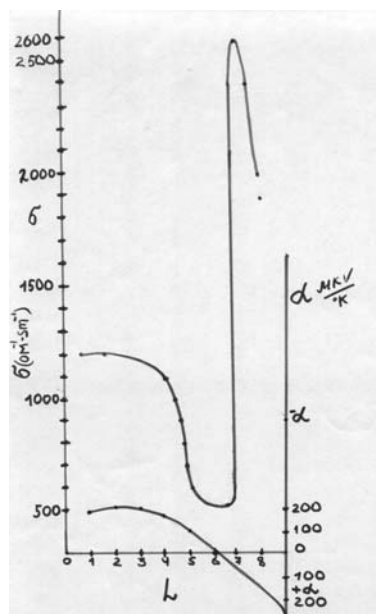

Fiq. Dependence α and σ from length of a sample (sm)

ampoule in another twice. Tellurium was sublimated at temperature 480°C also two times. Only after that synthesis was carried out at temperature 700°C together with impurity. Preliminary initial components were degassing in quartz ampoules at residual pressure of 10^{-3} mm.Hg. The received ingots after synthesis were located in graphitization ampoules in diameter (8-5) mm for carrying out of the area-directed crystallization. Crystallization was carried out at speed of moving of a zone 1cm/hour.

Further the received crystals were cut on samples in length 20-25mm. On such crystals thermoelectric properties were measured in a direction parallel to planes of crystals cohesion.

However in the beginning we shall characterize the interface structures received on a basic plane (0001) (Bi$_2$Te$_3$-4 mol % Bi$_2$Se$_3$).

III. CONCLUSION

On the given surface, investigated by us with raster electronic microscope, very fine disperse particles (tin and tellurium) were found out. They arise spontaneously in the closed system and are equilibrium in separate sites along length of a crystal. We do not bring these pictures. They will be a subject of the further researches. " Self-organizing " of such structures we consider as spontaneous occurrence of the macro-scopical order in initial homogeneous system. Table of electroconductivity dependence (δ) and thermo-electromotive (α) along the length of a sample shows homogeneous distribution only on an initial half of length. Further there is an abrupt change both α, and δ the type of conductivity of a crystal changes. There is a redistribution of a superfluous impurity of tin along a sample. In an initial stage of crystallization a dominating impurity, most likely, is chlorine; the impurity of tin is pushed aside in second half. Change testifies to it such as conductivity and high hole concentration. The initial stage of crystallization (at structure of impurity CdCl$_2$ – (0,09 % weight) and tin (0,1% weight) is shown by uniform distribution of these impurity. Reception of a firm solution testifies to it (Bi$_2$Te$_3$ • 96mol % - Bi$_2$Se$_3$• 4mol %) <CdCl$_2$ and Sn> with high thermoelectric efficiency (Z=α^{2}δ/K, here K-full heat conductivity of a sample).

Impurity doping process (CdCl$_2$ and Sn) allows to receive high thermoelectric parameters (Z> 3,0 • 10^{-3} K^{-1}) not only at T=300 ° K, but willows an interval 250-350 K. As well as at impurity doping process by tin of system Bi$_{2-y}$Sn$_y$Te$_{3-x}$Se$_x$ [6] observes the following features of physical properties of crystals:

- Temperature dependences α in samples with tin and CdCl$_2$ +Sn become more flat in comparison with not alloyed samples;

- At impurity doping process complex impurity CdCl$_2$ + Sn the increase turns out

Factor Zeebek (α) at T=250-350°K in comparison only with Sn.

Influence of tin affects and at impurity doping process of a double impurity (CdCl$_2$

+Sn). Thus, and in systems (Bi$_2$Te$_3$-Bi$_2$Se$_3$) <CdCl$_2$ • Sn> are shown impurity resonant conditions of the tin, located on a background valence zones.

In comparison with the data [6] in which high uniformity is found out

Electric properties of mono-crystals Bi$_{2-y}$Sn$_y$Te$_{3-x}$Se$_x$ in investigated us systems along length strong change is revealed physical properties. However received in an initial part of an ingot samples also were homogeneous and had optimum concentration elektrons. For an example we shall result parameters along three centimetric

Parts:

α = (195-200) (MKV / ° K)

$\delta = (1200\text{-}1150)$ (OM$^{-1}\cdot$ cm^{-1}) At cultivation of samples in length more than 20 cm uniformity grew only at increase of quantity of donor additives CdCl$_2$ Probably reception of higher characteristics of crystals with the complex additiveCdCl$_2$.

The technique of the directed vertical crystallization is more favorable commercially. It allows to raise simultaneously crystals (0=15-25) mm with high speed (more V =2 cm/hour) and a plenty of zones.

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