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ABSTRACT

of the dissertation for the degree of Doctor of Philosophy

“THERMOELECTRIC AND MAGNETOTHERMOELECTRIC PROPERTIES OF γ -IRRADIATED $Bi_{0,85}Sb_{0,15}$ SOLID SOLUTION”

Speciality: 2225.01 – Radiation materials science

Field of science: Physics

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GENERAL DESCRIPTION OF WORK

Relevance and development of the topic. Single crystals of Bi–Sb solid solutions are among the most suitable materials for low-temperature (below ~200 K) electronic coolers. However, due to their layered crystal structure, these materials exhibit low mechanical strength, which significantly limits their range of applications.Recent studies have demonstrated that extruded materials based on the Bi–Sb system possess high mechanical strength and well-developed deformation texture, while their thermoelectric properties approach those of single-crystal specimens. Such solid solutions are therefore considered indispensable for the development of low-temperature energy converters. The high performance characteristics of electronic devices fabricated on the basis of the Bi–Sb system make it possible to employ them in various fields of technology, including under radiation conditions. Nevertheless, in a radiation environment, the formation of radiation-induced defects in the semiconductor materials used in such devices affects their thermal and other physical properties, and consequently influences the operational parameters of the devices.

Ensuring the operability of various energy converters under the influence of natural and artificial ionizing radiation, as well as expanding the scope of their application in different fields of technology, constitutes an urgent scientific and practical problem.

The implementation of such studies makes it possible to obtain valuable information on the distribution of dopants, radiation-induced defects, and electroneutral particles in semiconductor materials with different structural perfection, as well as on the degree of texture formation depending on grain size. At the same time, the investigation of electrical and thermal properties in bulk nanostructured materials with various degrees of texture, both unannealed and optimally thermally annealed, provides broader insights into the correlation between structure and physical properties, the scattering mechanisms of electrons and phonons by defects, and related phenomena.

A review of the literature has revealed that the influence of gamma quanta on the electrophysical properties of Bi–Sb solid solution specimens has not yet been studied.

Therefore, the investigation of the effect of radiation-induced defects on the electrical properties of extruded Bi–Sb solid solutions is of particular interest.

The present dissertation represents a scientific study directed towards solving the aforementioned issues, which are of significant scientific and practical importance.

Object and subject of research. The object of the research is bulk nanostructured $Bi_{0,85}Sb_{0,15}$ solid solution extrusion specimens irradiated with gamma quanta at various doses, including those modified with acceptor (Pb), donor (Te), and ZrO_2 additives. The subject of the research is the influence of gamma radiation on charge and heat transport phenomena in the mentioned $Bi_{0.85}Sb_{0.15}$ solid solution extrusion specimens, investigated over a wide range of temperatures and magnetic fields.

Goals and objectives of the research. The aim of the dissertation is to determine the mechanisms of the influence of radiation-induced defects on heat and charge transport phenomena in extruded specimens of $\mathbf{Bi_{0,85}Sb_{0,15}}$ solid solutions alloyed with dopants of various nature, over a wide range of temperatures and magnetic field intensities, as well as to explore the prospects for creating electronic devices based on these materials for applications in different areas of optoelectronics.

To achieve this aim, the following objectives have been set:

- to develop the fabrication technology of $Bi_{0.85}Sb_{0.15}$ solid solution extrusion specimens, both undoped and alloyed with dopants of different nature;
- to establish the technology for irradiating the obtained specimens with various doses of gamma quanta;
- to investigate, in both irradiated and unirradiated specimens, the effect of donor, acceptor, and electroneutral particles, grain size, thermal annealing, and radiation defects on thermo- and magnetothermoelectric properties over a wide range of temperatures and magnetic field intensities;

- to study heat and charge transport phenomena in the investigated materials over a broad interval of temperatures and magnetic field intensities;
- based on the obtained results, to examine the potential application of extruded $Bi_{0,85}Sb_{0,15}$ solid solution specimens in the development of electronic devices.

Investigation methods:

- development of the fabrication technology for extruded bulk nanostructured $Bi_{0,85}Sb_{0,15}$ solid solution specimens, both pure and alloyed with dopants of different nature, as well as with electroneutral particles (ZrO_2);
- preparation and thermal treatment of specimens for studying the influence of annealing on their thermo- and magnetothermoelectric properties;
- establishment of the methodology for irradiating the obtained specimens with various doses of gamma quanta;
- preparation of specimens for the investigation of electrical and thermal properties over a wide temperature range and under variable magnetic fields;
- study of charge and heat transport phenomena in the obtained specimens within a broad range of temperatures and magnetic field intensities.

Main Scientific Provisions Submitted for Defense:

1. The mechanisms of the influence of γ-irradiation at various doses on the thermo- and magnetothermoelectric properties of $Bi_{0,85}Sb_{0,15}$ solid solution extrusion specimens have been identified over a wide range of temperatures and magnetic field intensities.
2. The dependence of the electrical and thermal parameters of bulk nanostructured $Bi_{0,85}Sb_{0,15}$ solid solution extrusion specimens on grain size and thermal treatment has been established, associated with defects related to grain size and boundaries, the degree of grain alignment (texture) during extrusion, and changes in recrystallization and orientation of crystallites during thermal processing.
3. In $Bi_{0,85}Sb_{0,15}$ 〈Te〉 specimens doped with tellurium atoms and irradiated with various doses of gamma quanta, a significant increase in specific electrical conductivity at low temperatures was observed,

along with a change in the character of the temperature dependence of electrical conductivity compared to unirradiated specimens of the same composition. For gamma-irradiated tellurium-doped specimens, the temperature dependences characteristic of the impurity conduction region were revealed. The temperature dependence of conductivity is determined by the temperature dependence of mobility (μ) and variations in carrier concentration (n).

4. At low irradiation doses, simple defects such as interstitial atoms and vacancies are generated in $Bi_{0,85}Sb_{0,15}$ 〈Pb〉 specimens, leading to an increase in free carrier concentration (n), electrical conductivity (σ), and the electronic contribution to thermal conductivity (χ_e), as well as a decrease in the thermoelectric power (α) and Hall coefficient (R_H). With increasing irradiation dose, complex defects (clusters of point defects) form, resulting in a decrease in carrier concentration and corresponding changes in σ, α, χ, and R_H. In specimens doped with 0.05 at.% Pb and irradiated at different doses, the signs of α and R_H coefficients at low temperatures remain unchanged compared to unirradiated specimens.

5. In specimens with complex doping by Pb and Te atoms, the presence of Te dopants compensating the acceptor effect of Pb explains the variation in electron concentration. The values of σ and α were compared with those obtained for $Bi_{0,85}Sb_{0,15}$ without Pb doping. The influence of thermal treatment on the concentration of structural defects in the specimens was determined, while in complexly doped samples the electronic contribution (χ_e) plays a decisive role in thermal conductivity.

6. The addition of 0.5 wt.% ZrO_2 modifier to extruded $Bi_{0.85}Sb_{0.15}$ solid solution specimens with optimal electron concentration resulted in a material with thermoelectric efficiency at ~77 K equal to ~$6.4\cdot10^{-3}$ K^{-1}. The study revealed the potential application of this material for the development of more efficient low-temperature cooling devices.

Scientific Novelty of the Research

For the first time, the present dissertation establishes the following:

1. In $Bi_{0,85}Sb_{0,15}$ solid solution extrusion specimens irradiated with gamma quanta at various doses, radiation defects acting as donor

centers are generated at low doses. As a result, the concentration of free electrons (n) and the specific electrical conductivity (σ) increase, while the thermoelectric power coefficient (α) decreases. These defects scatter charge carriers and thereby reduce their mobility (μ). With increasing irradiation dose, the concentration of defects also increases, leading to the trapping of free carriers at radiation defect levels.

2. Investigation of the dependence of the electrical and thermal parameters of bulk nanostructured $Bi_{0,85}Sb_{0,15}$ solid solution extrusion specimens on grain size and thermal treatment has shown that grain size, defects associated with grain boundaries, the degree of texture formed during extrusion, and the recrystallization and reorientation of crystallites during thermal processing play a decisive role in carrier and phonon scattering at ~77 K.

3. In tellurium-doped $Bi_{0,85}Sb_{0,15}$ specimens irradiated with different doses of gamma quanta, a significant increase in specific electrical conductivity at low temperatures was observed, along with a change in the character of its dependence compared to unirradiated specimens. The temperature dependences typical of the impurity conduction region were revealed.

4. In $Bi_{0,85}Sb_{0,15}$ ⟨Pb⟩ solid solution specimens, irradiation with low doses of gamma quanta leads to the formation of simple defects such as interstitial atoms and vacancies. This results in an increase in free electron concentration (n), electrical conductivity (σ), and thermal conductivity (χ), while the thermoelectric power (α) and Hall coefficient (R_H) decrease. With higher irradiation doses, the formation of complex defects (clusters of point defects) occurs, leading to a decrease in carrier concentration and corresponding changes in σ, α, χ, and R_H.

5. In $Bi_{0,85}Sb_{0,15}$ solid solution specimens with complex doping by Pb and Te atoms, Te dopants compensate for the acceptor effects of Pb, leading to an increase in electron concentration. As a result, the values of σ and α approach those obtained for undoped $Bi_{0,85}Sb_{0,15}$

6. It was established that the addition of 0.5 wt.% ZrO_2 modifier to $Bi_{0,85}Sb_{0,15}$ results in a reduction of the phonon contribution to thermal conductivity at ~77 K, thereby decreasing the total thermal

conductivity. During isothermal thermal treatment, recrystallization occurs in both modified and unmodified specimens, leading to a decrease in the concentration of structural defects formed during plastic deformation and to an increase in charge carrier mobility (μ). The addition of 0.5 wt.% ZrO_2 modifier to $Bi_{0,85}Sb_{0,15}$ extrusion specimens made it possible to obtain a material with a thermoelectric efficiency of $\sim 6.4 \cdot 10^{-3}$ K^{-1} at ~77 K, which enhances the performance parameters of low-temperature coolers based on this material.

The theoretical and practical significance of the research: Thermoelectric energy converters are often operated under radiation conditions. The formation of radiation-induced defects affects the physical properties of semiconductor materials and alters the performance parameters of devices based on them. The study of such materials makes it possible to evaluate the operability of energy converters under the influence of natural and artificial ionizing radiation, as well as to assess the prospects for their application in various fields of industry, in special-purpose systems, and in space technologies.

Approbation and application:

The scientific results obtained in the dissertation were presented at several international and local scientific events in accordance with the different directions of the research and were brought to scientific discussions. The results of the studies were reported at various conferences and were evaluated by specialists.

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Soft Magnetic Alloys in Information Technologies and Military Industry. – Baku: October 9–10, – 2019, – pp. 174–178.

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✓ Tagiev, M.M., Abdullayeva, I.A. Influence of Gamma Radiation on Electrical Properties of Extruded Samples of Bi0.85Sb0.15<Te> Solid Solutions // Electronic Processes in Organic and Inorganic Materials (ICEPOM-12). – Kamianets-Podilskyi, Ukraine: June 1–5, – 2020, – pp. 94–95.

✓ Tagiyev, M.M., Barkhalov, B.Sh., Abdinova, G.D., Abdullayeva, I.A. Magnetothermoelectric Properties of Extruded Samples of Bi0.85Sb0.15 + 0.01 Pb +Tex($0 \leq x \leq 0.1$) Solid Solution // ICTPE-2020. The 16th International Conference on Technical and Physical Problems of Engineering. – Istanbul, Turkey: October 12–13, – 2020, – pp. 89–94.

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✓ Taghiyev, M.M., Abdinova, G.D., Abdullayeva, I.A. Influence of heat treatment on the galvanomagnetic properties of bulk nanostructured Bi0.85Sb0.15 solid solution samples // Physica.SPb. Proceedings. – Saint Petersburg: October 18–22, – 2021, – pp. 122–123.

✓ Abdullayeva, I.A., Abdinova, G.J., Tagiyev, M.M. Influence of Gamma Radiation on Electrical and Thermal Properties of Extruded Samples of Bi0.85Sb0.15 Solid Solution Modified with ZrO2 // ICTPE-2021. The 17th International Conference on

Technical and Physical Problems of Engineering. – Istanbul, Turkey: October 18–19, – 2021, – pp. 72–78.

- ✓ Tagiyev, M.M., Abdinova, G.J., Abdullayeva, I.A. Influence of γ-radiation on the electrical properties of extruded samples of Bi0.85Sb0.15 <Pb> solid solution // Turkish Physical Society. The 37th International Physics Congress. – Bodrum, Turkey: September 1–5, – 2021, – p. 227.
- ✓ Tagiyev, M.M., Abdinova, G.D., Abdullayeva, I.A., Piriyeva, T.I., Aliyeva, Kh.F. Influence of heat treatment on the thermoelectric properties of extruded Bi0.85Sb0.15 samples // Actual Problems of Modern Natural and Economic Sciences. – Ganja, Azerbaijan: May 6–7, – 2021, Vol. 2, – pp. 364–367.
- ✓ Tagiyev, M.M., Abdinova, G.D., Abdullayeva, I.A. Influence of γ-radiation on the magnetoelectric properties of extruded Bi0.85Sb0.15 solid solution samples // XXVI International Scientific and Technical Conference on Photoelectronics and Night Vision Devices. – Moscow, Russia: May 25–27, – 2022, – pp. 432–433.
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- ✓ Tagiyev, M.M., Aliyeva, Kh.F., Abdullayeva, I.A., Abdinova, G.D., Piriyeva, T.I. Anisotropy of electrical properties of extruded

$Bi_{0.85}Sb_{0.15}$ samples irradiated with γ-radiation // Actual Problems of Modern Natural and Economic Sciences. International Conference Dedicated to the 100th Anniversary of National Leader Heydar Aliyev. – Ganja, Azerbaijan: May 5–6, – 2023, Vol. V, – pp. 106–109.

- ✓ Abdullayeva, I.A., Samedov, O.A., Tagiyev, M.M. Influence of gamma irradiation on the anisotropy of electrical properties of $Bi_{0.85}Sb_{0.15}$-based solid solution samples // Radiation Technologies and Their Application. Scientific-Technical Conference Dedicated to the 100th Anniversary of National Leader Heydar Aliyev. – Baku, Azerbaijan: May 5, – 2023, – pp. 65–67.
- ✓ Tagiyev, M.M., Aliyeva, Kh.F., Abdinova, G.D., Abdullayeva, I.A., Piriyeva, T.I. Electrical properties of extruded $Bi_{0.85}Sb_{0.15}$ solid solution samples modified with γ-quanta // Actual Problems of Physics of Metals and Alloys. Republican Scientific-Practical Conference. – Azerbaijan University of Architecture and Construction: February 6–7, – 2025, – pp. 114–119.

Publications: The main content and scientific results of the dissertation have been published in 13 articles in international journals and in 8 abstracts in the proceedings of international scientific conferences**,** as well as in 4 articles and 10 abstracts published within the country**.** In total, the principal results of the dissertation are reflected in 35 scientific works, including 17 articles (12 of which were published in journals indexed in the Web of Science and Scopus databases) and 18 conference abstracts.

The name of the organization where the dissertation work was performed: The present dissertation was conducted at the *"Radiation Physics of Disordered Solids" Laboratory of the Institute of Radiation Problems* of the Ministry of Science and Education of the Republic of Azerbaijan, and at the *"Solid-State Electronics" Laboratory of the Institute of Physics* of the Ministry of Science and Education of the Republic of Azerbaijan.

Personal attendance of the author: The dissertation was independently carried out by the applicant. The relevance of the research was substantiated on the basis of an extensive review of the literature. The applicant determined and applied the appropriate research methods for solving the stated scientific problems, conducted the planned experiments at all stages of the investigation, and systematized the obtained results. Furthermore, the applicant performed a comprehensive analysis of the experimental findings, presented the research outcomes at scientific conferences, prepared scientific articles based on the results, and completed the compilation of the dissertation.

The total volume of the dissertation includes a separate character count for each of its structural units. The dissertation consists of an introduction, four chapters, a conclusion, a list of references, and abbreviations, covering a total of 175 pages. The total volume of the main text (excluding figures, tables, and the list of references) amounts to 196669 characters. The character count of the respective sections is as follows: Introduction – 30179; Chapter I – 33865; Chapter II – 23579; Chapter III – 35847; Chapter IV – 69048; Conclusion – 4151. The research work contains 30 figures and 9 tables, and the list of references includes 138 sources.

THE CONTENT OF THE DISSERTATION

In the Introduction, the relevance of the dissertation topic is substantiated. The objectives of the research, the main tasks to be solved, the scientific novelty, the practical significance, the main provisions submitted for defense, as well as the objects and methods of research are presented. In addition, the introduction provides a concise summary of the main content of the dissertation chapters.

Chapter I presents a review of studies devoted to the physical properties of solid solutions based on the Bi–Sb system. This review analyzes the scientific works of various authors and summarizes information on the properties, electronic spectrum, thermoelectric and magnetic characteristics of Bi–Sb-based materials. Particular attention is given to the regularities of charge and heat transport phenomena at low temperatures in Bi–Sb systems and to their promising thermoelectric performance.

The chapter also discusses approaches to improving the mechanical properties of thermoelectric materials and enhancing the reliability of energy converters based on them.[1]

Based on the analysis conducted, the main purpose of the dissertation was formulated: the improvement of the structural, thermoelectric, and magnetothermoelectric properties of Bi–Sb solid solutions, the development of new high-efficiency materials, and the investigation of the effects of gamma irradiation on these materials.

Chapter II provides a detailed description of the experimental methodology used in the research. Special attention is given to the preparation technology of $Bi_{0,85}Sb_{0,15}$ solid solutions, their irradiation methods, and sample processing. The samples were synthesized based on the Bi–Sb system and subsequently subjected to mechanical processing by extrusion. It is noted that the specimens were irradiated with γ-quanta at various doses and, in some cases, additionally doped with different chemical elements. Such modification leads to the formation of new defects in the materials, alters the interaction mechanisms of electronic and phononic subsystems, and consequently changes their electrical and thermal conductivities. The use of different irradiation doses allowed a comparative analysis of the changes in material properties. Within the experimental methodology, electrical and thermal parameters of the samples—electrical conductivity, Seebeck coefficient, thermal conductivity, and magnetothermoelectric effects—were measured over a wide temperature range, including both low and high temperatures. In addition, the properties of the samples were investigated under magnetic fields of varying intensities. This approach enabled a deeper understanding of the kinetics of charge carriers in Bi–Sb-based materials.

Chapter III presents the results of studying the influence of grain size and thermal treatment on the thermo- and magnetothermoelectric properties of undoped $Bi_{0,85}Sb_{0,15}$ solid solution extrusion samples and those modified by various doses of

[1] Grabov, V.M., Uryupin, Sh.N. Influence of dendritic inhomogeneity on the thermoelectric properties of $Bi_{0,88}Sb_{0,12}$ crystals // Semiconductors, - 2022, Vol. 56, No. 2, -pp. 145–148.

gamma irradiation, as well as bulk nanostructured $Bi_{0,85}Sb_{0,15}$ solid solution samples with different grain sizes, over a wide range of temperatures and magnetic field intensities.

Initially, the electrical and thermal properties of pure and unirradiated $Bi_{0,85}Sb_{0,15}$ solid solutions were investigated across wide ranges of temperature and magnetic field.

The comparative analysis presented in this chapter revealed the efficiency of various modification methods for controlling the properties of Bi–Sb-based thermoelectric materials.

Extruded $Bi_{0,85}Sb_{0,15}$ samples, both unirradiated and irradiated with gamma quanta at different doses, and subjected to thermal treatment at ~503 K for 2 hours after extrusion, were studied for their specific electrical conductivity (σ), Seebeck coefficient (α), and Hall coefficient (R_H) in the temperature range of ~77–300 K and under magnetic fields up to ~74×10^4 A/m. The obtained results are presented in Table 1.
It was found that at ~77 K, the specific electrical conductivity decreases approximately threefold with increasing magnetic field intensity, while the Seebeck coefficient increases by about 35%.

Under the influence of gamma irradiation, changes in the defect subsystem of extruded $Bi_{0,85}Sb_{0,15}$ solid solutions lead to modifications in the spectrum of localized levels and in electron scattering processes, which, in turn, result in corresponding changes in the electrical parameters.

The influence of thermal treatment and nanoscale effects on the electrical and thermal properties of bulk nanostructured $Bi_{0.85}Sb_{0.15}$ solid solution samples was also investigated in the ~77–300 K temperature range (Table 2).

Based on the values of R_H and σ, the Hall mobility of charge carriers was calculated using the relation $\mu=R_H\cdot\sigma$. It has been established that with an increase in nanoparticle size and after thermal treatment, the exponent m in the dependence $\mu\sim T^{-m}$ increases from 1.42–1.57 for untreated samples to 1.81–2.6 for thermally treated samples. After thermal treatment, the increase in at R_H ~77 K, except for the sample with grain size of ~$2\cdot10^5$ nm, is associated with changes in the parameter A in the expression $R_H=A/en$ (where e is the electron charge), which characterizes the scattering mechanism, as well as with variations in defect concentration.[2]

Table 1.

Values of electrical conductivity (σ), thermoelectric power (Seebeck coefficient, α), Hall coefficient (R_H), charge carrier concentration (n), and mobility (μ) of $Bi_{0,85}Sb_{0,15}$ samples irradiated with gamma quanta.

Radiation doses (Mrad)	At 77 K					At 300 K				
	σ ($\Omega^{-1}cm^{-1}$)	α (μV/K)	$R_H \cdot 10^{-8}$ (cm^3/ C)	n· 10^{18} (cm^{-3})	μ (cm^2/ V·s)	σ ($\Omega^{-1}cm^{-1}$)	α (μV/K)	$R_H \cdot 10^{-8}$ (cm^3/ C)	n· 10^{18} (cm^{-3})	μ (cm^2/ V·s)
0	5250	-182	-26,5	0,6	139125	7520	95	1,43	10,5	10528
1	8481	-137	-1,26	11,9	10686	6524	89	1,26	11,9	8220
10	4840	-161	-60	2,5	290400	6890	101	17,4	0,9	119886
50	4552	-188	-85	0,2	386920	6448	90	33	0,5	212784

[2] Krabs, G. Fundamentals of Crystal Chemistry of Inorganic Compounds. Moscow: Mir, -1971. -304 p.

Table 2.
Values of charge carrier concentration (n), mobility (μ), and the phonon contribution to thermal conductivity (χ_L) in nanostructured extruded samples of $Bi_{0,85}Sb_{0,15}$ solid solutions with different grain sizes.

Particle sizes , (nm)	At 77 K					
	Non-annealed samples			Annealed samples		
	χ_l (Vt/m·K)	μ (cm²/ V·s)	n· 10¹⁸ (cm⁻³)	χ_l Vt/m·K	μ (cm²/ V·s)	n· 10¹⁸ (cm⁻³)
$2 \cdot 10^5$	2,98	31833	0,7	3,54	54799	0,5
950	3,28	6080	5,2	3,33	9458	5,2
650	3,09	3026	15,6	3,39	14966	3,1
400	1,93	5485	12,5	2,14	6329	15,6
32	2,02	4340	15,6	2,68	4996	16,9
15	2,78	1800	31,3	2,25	2213	34,7

It has been established that at low temperatures (below ~200 K), a decrease in grain size in extruded samples leads to a reduction in the phonon contribution to thermal conductivity (χ_l). The decrease in the phonon component of thermal conductivity exhibits a non-monotonic character; that is, in both untreated and thermally treated samples, phonon thermal conductivity decreases with grain size reduction and subsequently increases. With decreasing crystallite size, the concentration of grain boundaries increases, which results in an increase in carrier concentration and enhanced scattering at the boundaries, thereby reducing the electron (charge carrier) mobility in the samples.

The dependence of thermal conductivity on grain size and thermal treatment in nanostructured extruded $Bi_{0,85}Sb_{0,15}$ solid solution samples can be satisfactorily explained by structural modifications that occur during extrusion and thermal processing, which are consistent with the observed variations in electrical parameters. It has been established that in these samples, heat is primarily transported by conduction electrons and phonons.

In Chapter IV of the dissertation, comprehensive research findings are presented for extruded samples of $Bi_{0,85}Sb_{0,15}$ solid solutions modified by various methods. This chapter mainly examines the influence of γ-quanta at different doses on the electrical and thermal properties of donor-doped (tellurium), acceptor-doped (lead), complex Pb–Te co-doped, and neutral-particle-modified (ZrO_2) samples, as well as pure and Te-doped samples, over a wide range of temperatures and magnetic fields.

As shown in Table 3, with the exception of the sample irradiated at a dose of 1 Mrad, an increase in radiation dose leads to a decrease in charge carrier concentration in undoped samples, while mobility increases across the entire temperature range studied. At low irradiation doses (1 Mrad), both Te-doped and undoped samples exhibit a slight increase in carrier concentration (n) and a reduction in mobility (μ).

With increasing irradiation dose, n decreases significantly, while μ increases. These variations in n and μ correlate well with the observed changes in σ and α.

Based on the obtained results, it is considered that at low irradiation doses (1 Mrad) in $Bi_{0,85}Sb_{0,15}$<Te> solid solution samples, radiation-induced defects acting as donor centers are formed. As a result, the concentration of free electrons (n) and, accordingly, the specific electrical conductivity (σ) increase, while the Seebeck coefficient (α) decreases. These defects scatter charge carriers, thereby reducing their mobility (μ). With increasing irradiation dose, the concentration of defects also increases, leading to carrier trapping at radiation defect levels. Consequently, the defect-controlled carrier concentration (n) and the conductivity (σ) of the sample decrease, the Fermi level shifts deeper into the forbidden band, while the Seebeck coefficient and mobility increase. The temperature dependences of the electrical parameters of the extruded $Bi_{0,85}Sb_{0,15}$<Te> solid solution samples irradiated with gamma quanta can be explained by the temperature dependences of carrier mobility (μ) and concentration (n).

Table 3.

Values of the electrical parameters of extruded $Bi_{0,85}Sb_{0,15}$<Te> > solid solution samples irradiated with different doses of gamma radiation.

Radiation doses (Mrad)	Compositions	77 K					300 K				
		σ ($\Omega^{-1}cm^{-1}$)	α (μV/K)	$R_H \cdot 10^{-8}$ (cm^3/ C)	μ (cm^2/ V·s)	n· 10^{18} (cm^{-3})	σ ($\Omega^{-1}cm^{-1}$)	α (μV/K)	$R_H \cdot 10^{-8}$ (cm^3/ C)	μ (cm^2/ V·s)	n· 10^{18} (cm^{-3})
0	$Bi_{0,85}Sb_{0,15}$	5250	-182	-26,5	139125	0,24	7520	-95	-1,43	10754	4,4
	$Bi_{0,85}Sb_{0,15}$ <Te>	7574	-161	-23,97	181549	0,3	9079	-95	-1,15	10441	5,4
1	$Bi_{0,85}Sb_{0,15}$	8481	-121	-1,26	10686	4,96	6524	-89	-1,26	8220	5
	$Bi_{0,85}Sb_{0,15}$ <Te>	15477	-50	-1,08	16715	5,79	7035	-80	-1,08	7598	5,8
10	$Bi_{0,85}Sb_{0,15}$	4240	-161	-60	254400	0,1	6890	-101	-17,4	119886	0,36
	$Bi_{0,85}Sb_{0,15}$ <Te>	12084	-56	-11,6	140174	0,54	6101	-66	-21,1	128731	0,3
50	$Bi_{0,85}Sb_{0,15}$	4552	-188	-85	386920	0,07	6448	-90	-34,3	221166	0,18
	$Bi_{0,85}Sb_{0,15}$ <Te>	15371	-47	-16,2	249010	0,39	7233	-79	-30,1	154381	0,21

The experimental results on the influence of lead doping and γ-radiation on the electrical and thermal properties of extruded $Bi_{0.85}Sb_{0.15}$ solid solution samples are presented in Figure 1. Irradiation significantly affects the electrical and thermal properties of $Bi_{0.85}Sb_{0.15}$ samples doped with various concentrations of lead. As can be seen, γ-irradiation exerts different effects on the specific electrical conductivity of samples with different Pb concentrations. The most pronounced changes in σ occur at irradiation doses up to ~10 Mrad. For instance, at ~77 K, under a 1 Mrad γ-irradiation dose, the σ of the undoped sample increases from 5230 to 8431 $\Omega^{-1}\cdot cm^{-1}$. With further increase of the dose up to 10 Mrad, σ decreases to 4240 $\Omega^{-1}\cdot cm^{-1}$, and with subsequent irradiation up to 50 Mrad, it increases slightly again, reaching 4552 $\Omega^{-1}\cdot cm^{-1}$.

Doping $Bi_{0.85}Sb_{0.15}$ with 0.001 at.% Pb reduces σ to 4938 $\Omega^{-1}\cdot cm^{-1}$. In this sample, under a 1 Mrad dose of γ-irradiation, σ increases significantly (by about 2.5 times). With increasing irradiation up to 10 Mrad, σ increases by ~2.2 times, and at higher doses (50 Mrad), its growth slows down (by ~50%). For samples doped with 0.005, 0.01, and 0.05 at.% Pb, the dependence of σ on irradiation dose shows almost the same trend: at low doses (up to 10 Mrad), σ increases slightly, while at higher doses, it remains nearly unchanged.

These changes in σ correlate satisfactorily with the variations of the Seebeck and Hall coefficients under the influence of γ-irradiation.

At temperatures below ~130 K (down to ~77 K), the Seebeck coefficient (α) and Hall coefficient (R_H) of the unirradiated sample doped with 0.05 at.% Pb are positive, indicating that the sample exhibits p-type conductivity. In contrast, across the entire investigated temperature range (~77–300 K), the $Bi_{0.85}Sb_{0.15}$ solid solution samples doped with 0.05 at.% Pb and irradiated at all doses exhibit n-type conductivity, i.e, the signs of α and R_H remain unchanged.

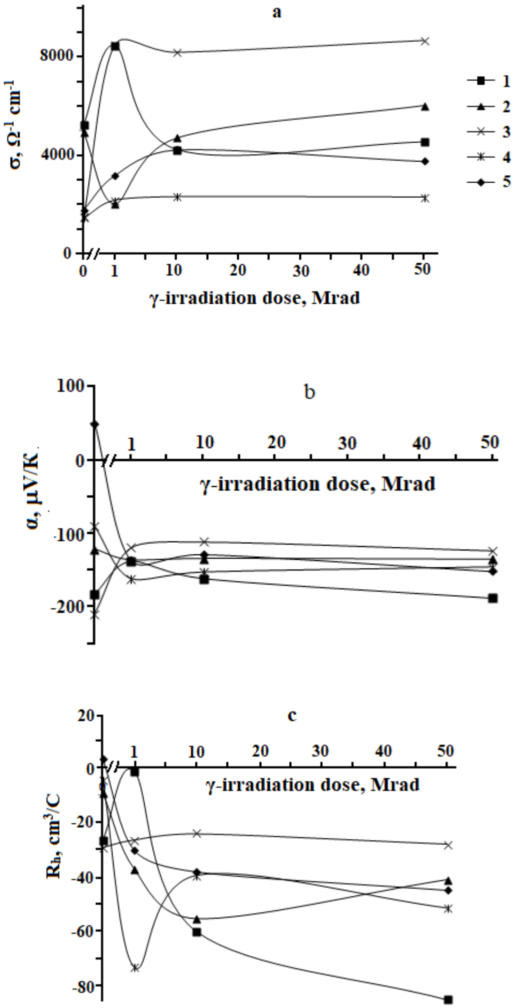

Figure 1. Dependences of specific electrical conductivity σ (a), Seebeck coefficient α (b), and Hall coefficient R_H (c) on irradiation dose for extruded $Bi_{0,85}Sb_{0,15}$solid solution samples doped with lead. 1 – undoped; 2, 3, 4, 5 – samples doped with 0.001, 0.005, 0.01, and 0.05 at.% Pb, respectively.

To investigate the influence of complex doping on the thermo- and magnetothermoelectric properties of $Bi_{0.85}Sb_{0.15}$ solid solution crystals, extruded samples with grain sizes of ~630 μm doped with acceptor (Pb) and donor (Te) impurities were prepared, and their electrical and thermal properties were studied. The obtained results are presented in Table 4.

When electroactive impurities Pb and Te are introduced into extruded $Bi_{0,85}Sb_{0,15}$ solid solution samples, the Lorenz number (L) may also change. However, calculations show that this variation (~10%) is relatively small compared to the change in σ (~3.7 times). This provides grounds to conclude that, in the case of complex doping of $Bi_{0,85}Sb_{0,15}$ solid solutions, the electronic component of thermal conductivity (χ) plays the decisive role in the variation of the total thermal conductivity.[3]

It has been established that Te dopants, which compensate the acceptor effect of Pb, lead to an increase in the electron concentration in the samples. As a result, the values of σ and α approach those obtained for undoped $Bi_{0,85}Sb_{0,15}$ solid solutions. The carrier mobility (and consequently the electrical parameters, electronic contribution to thermal conductivity, as well as their dependences on temperature and magnetic field intensity) and the lattice contribution to thermal conductivity are significantly influenced by structural defects "healed" through thermal treatment. With increasing temperature, the scattering of phonons by structural defects and impurity centers becomes weaker.

To clarify the influence of modification (in small amounts) and thermal treatment on the thermoelectric and magnetothermoelectric properties of solid solutions of the Bi–Sb system, extruded samples of $Bi_{0,85}Sb_{0,15}$ modified with 0.5 wt.% ZrO_2 were prepared. Their specific electrical conductivity (σ), Seebeck coefficient (α), Hall coefficient (R_H), and thermal conductivity (χ) were investigated in the temperature range of ~77–300 K and in magnetic fields up to ~7.4×10^4 A/m. The obtained results are presented in Table 5.

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[3] Gitsu, D.V., Golban, I.M., Lantser, V.G., Muntianu, F.M. *Transport Phenomena in Bismuth and Its Alloys*. Chișinău: Shtiintsa, -1983. –266 p.

Table 4.

Values of specific electrical conductivity (σ), Seebeck coefficient (α), Hall coefficient (R_H), thermal conductivity (χ), carrier mobility (μ), and carrier concentration (n) at ~77 K for extruded $Bi_{0,85}Sb_{0,15}$ **+ 0.01 at.% Pb solid solution samples as a function of tellurium dopant concentration.**

Note.

№	~77 K											
	Non-annealed samples						Annealed samples					
	σ ($\Omega^{-1}cm^{-1}$)	α (μV/K)	$R_H \cdot 10^{-8}$ (cm^3/C)	$\chi \cdot 10^2$ (W/m·K)	μ (cm^2/ V·s)	$n \cdot 10^{18}$ (cm^{-3})	σ ($\Omega^{-1}cm^{-1}$)	α (μV/K)	$R_H \cdot 10^{-8}$ (cm^3/C)	$\chi \cdot 10^2$ (W/m·K)	μ (cm^2/ V·s)	$n \cdot 10^{18}$ (cm^{-3})
1	2393	-173	-14,83	2,9	35488	0,4	5387	-178	-25,3	3,02	136291	0,25
2	1273	-40	-0,69	3,08	878	9,1	1462	-90	-4,33	3,04	6331	1,4
3	1591	-148	-7,4	3,05	11773	0,8	2020	-171	-11,1	3,03	22422	0,56
4	2750	-127	-6,71	3,05	18453	0,9	1604	-154	-10,6	2,39	17002	0,58
5	11823	-44	-0,69	4,42	8158	9,1	13752	-30	-0,72	5	9901	0,7
6	10823	-45	-0,45	4,92	4870	13,9	17026	-57	-0,69	5,3	11748	9,1
7	17506	-8	-0,23	5,6	4026	27,2	15600	-3	-0,1	5,7	1560	62,5

1 – undoped sample; 2 – $Bi_{0,85}Sb_{0,15}$+ 0.01 at.% Pb; 3 – $Bi_{0,85}Sb_{0,15}$+ 0.01 at.% Pb + 0.0001 at.% Te; 4 – $Bi_{0,85}Sb_{0,15}$+ 0.01 at.% Pb + 0.0005 at.% Te; 5 – $Bi_{0,85}Sb_{0,15}$+ 0.01 at.% Pb + 0.005 at.% Te; 6 – $Bi_{0,85}Sb_{0,15}$+ 0.01 at.% Pb + 0.01 at.% Te; 7 – $Bi_{0,85}Sb_{0,15}$+ 0.01 at.% Pb + 0.1 at.% Te.

It was established that during isothermal thermal treatment, recrystallization occurs in the unmodified samples, leading to a decrease in the concentration of structural defects formed during plastic deformation. Consequently, the mobility (μ) of charge carriers increases. When 0.5 at.% ZrO_2 modifier is added to $Bi_{0,85}Sb_{0,15}$, the phonon component of thermal conductivity decreases at ~77 K, resulting in a reduction of the total thermal conductivity.[4]

It was further determined that recrystallization occurs in both unmodified and ZrO_2 - modified samples during thermal treatment, reducing the concentration of structural defects formed during plastic deformation and thus increasing the charge carrier mobility (μ). The introduction of 0.5 at.% ZrO_2 modifier into $Bi_{0,85}Sb_{0,15}$ reduces the phonon contribution to the thermal conductivity at ~77 K, which leads to an overall reduction in thermal conductivity. At ~77 K, heat transfer in ZrO_2-doped $Bi_{0,85}Sb_{0,15}$ solid solution samples is mainly mediated by lattice vibrations (phonons).

The addition of 0.5 at.% ZrO_2 modifier to extruded $Bi_{0,85}Sb_{0,15}$ enables the production of a material with a thermoelectric efficiency of $\sim 6{,}4 \cdot 10^{-3}$ K^{-1} at ~77 K, making it suitable for the development of more effective low-temperature cooling devices.

This also creates opportunities for the development of more efficient energy converters based on this material. The obtained value significantly exceeds the thermoelectric efficiency Z ($\sim 5{,}8 \cdot 10^{-3}$ K^{-1}) of unmodified $Bi_{0,85}Sb_{0,15}$ solid solution samples at ~77 K.
The temperature dependence of the Hall coefficient (R_H) in modified $Bi_{0,85}Sb_{0,15}$<Te> extruded samples shows that, in samples irradiated with various doses of gamma quanta containing the modifier, the value of R_H is higher compared to unirradiated samples. This is attributed to changes in charge carrier mobility. For unirradiated samples—particularly those that are undoped and unmodified—the most pronounced variations in the Hall coefficient and carrier concentration occur mainly within the 80–200 K temperature range.

[4] Dubrovina, A.N., Kruglov, V.V., Rybina, L.N., Tout, A.S. Influence of extrusion conditions on the thermoelectric power coefficient of $Bi_2Te_{2.7}Se_{0.3}$-based materials // Elektronnaya Tekhnika. Materials, 1989, No. 1(238), pp. 33–36.

In the 200–300 K range, however, the variations in R_H become relatively weaker as temperature increases.

Table 5.

Values of the electrical and thermal parameters of extruded $Bi_{0,85}Sb_{0,15}$ solid solution samples modified with ZrO_2.

Compositions	77 K									300 K								
	σ ($\Omega^{-1}cm^{-1}$)	α ($\mu V/K$)	χ (W/m·K)	$\chi_e 10^{-2}$ (W/m·K)	χ_l (W/m·K)	$R_H \cdot 10^{-8}$ (cm^3/C)	μ ($cm^2/V \cdot s$)	$n \cdot 10^{18}$ (cm^{-3})	$Z \cdot 10^{-3}$ (K^{-1})	σ ($\Omega^{-1}cm^{-1}$)	α ($\mu V/K$)	χ (W/m·K)	$\chi_e 10^{-2}$ (W/m·K)	χ_l (W/m·K)	$R_H \cdot 10^{-8}$ (cm^3/C)	μ ($cm^2/V \cdot s$)	$n \cdot 10^{18}$ (cm^{-3})	$Z \cdot 10^{-3}$ (K^{-1})
Non-annealed samples																		
$Bi_{0,85}Sb_{0,15}$	2414	172	2,64	0,33	2,64	24,4	58902	0,025	2,7	6456	94	5,26	0,34	5,26	1,06	6843	5,9	1,08
$Bi_{0,85}Sb_{0,15}$ +0,5%ZrO_2	2985	182	2,54	0,4	2,54	32,1	95819	0,019	3,8	7858	103	4,95	0,41	4,95	1,21	9508	5,1	1,68
Annealed samples																		
$Bi_{0,85}Sb_{0,15}$	5250	182	3,02	0,7	3,01	26,5	139125	0,023	5,8	8456	95	5,96	0,45	5,92	1,43	12092	4,3	1,28
$Bi_{0,85}Sb_{0,15}$ +0,5%ZrO_2	4825	194	2,83	0,6	2,82	43,85	211576	0,014	6,42	9205	97	5,31	0,49	5,26	1,67	15372	3,7	1,6

MAIN SCIENTIFIC RESULTS

1. At low irradiation doses (1 Mrad), radiation defects that act as donor centers are generated in $Bi_{0,85}Sb_{0,15}$ solid solution samples. As a result, the free electron concentration (n) and electrical conductivity (σ) increase, while the thermoelectric coefficient (α) decreases. These defects scatter charge carriers, thereby reducing their mobility (μ). With increasing irradiation dose, the concentration of defects also increases, leading to the trapping of free carriers at radiation defect levels. Consequently, the carrier concentration (n) and σ of the samples decrease, the Fermi level shifts deeper into the forbidden band, while the Seebeck coefficient (α) and mobility increase. It has been shown that the increase in total thermal conductivity with irradiation dose is associated with the rise in the lattice contribution.
2. The dependence of the electrical and thermal parameters of bulk nanostructured extruded $Bi_{0,85}Sb_{0,15}$ solid solution samples on grain size and thermal treatment has been established. Grain size, boundary-related defects, texture (degree of grain alignment) during extrusion, and the recrystallization and reorientation of crystallites during thermal treatment predominantly govern carrier and phonon scattering at ~77 K. Thermal treatment decreases n and increases μ, χ, χ_e , and magnetoresistance. The increase in grain size raises the energy required for grain rotation, reducing deformation-induced texture in the samples and weakening the dependence of the lattice thermal conductivity (χ_l) on grain size. Structural changes occurring during extrusion and thermal treatment directly influence variations in electrical and thermal parameters.
3. In samples doped with tellurium atoms and irradiated with different doses of gamma quanta at low temperatures, the electrical conductivity significantly increases compared to unirradiated samples. The character of the temperature dependence changes, and dopant conductivity regions become evident. The temperature dependence of conductivity is attributed to variations in carrier mobility and concentration.

4. At low doses of gamma irradiation, simple defects such as interstitial atoms and vacancies form in $Bi_{0,85}Sb_{0,15}$<Pb> solid solution samples, leading to increases in free carrier concentration (n), electrical conductivity (σ), and thermal conductivity (χ), while the Seebeck coefficient (α) and Hall coefficient (R_H) decrease. With further increases in irradiation dose, complex defects (clusters of point defects) form, resulting in a decrease in carrier concentration and corresponding changes in σ, α, χ, and R_H . It has been established that in $Bi_{0,85}Sb_{0,15}$ solid solutions doped with 0.05 at.% Pb, the Seebeck and Hall coefficients of unirradiated samples change sign at ~130 K, but in irradiated samples at different doses of gamma quanta, no sign reversal of α and R_H is observed at low temperatures.

5. It has been determined that tellurium dopants, which compensate for the acceptor effect of lead, increase electron concentration in the samples. As a result, the values of σ and α approach those of undoped $Bi_{0,85}Sb_{0,15}$. Thermal treatment reduces structural defect concentrations, thereby significantly affecting carrier mobility and the lattice contribution to thermal conductivity. During complex doping of $Bi_{0,85}Sb_{0,15}$, the electronic component of thermal conductivity (χ_e) plays a decisive role in the variation of total thermal conductivity. With increasing temperature, phonon scattering at structural defects and dopant centers diminishes.

6. It has been established that during isothermal thermal treatment, recrystallization occurs both in unmodified samples and in those containing modifiers, reducing structural defect concentration created by plastic deformation and increasing carrier mobility (μ). When 0.5 at.% ZrO_2 modifier is added to $Bi_{0,85}Sb_{0,15}$, the phonon component of thermal conductivity decreases at ~77 K, leading to a reduction in overall thermal conductivity. In unmodified $Bi_{0,85}Sb_{0,15}$ samples with ZrO_2, heat transport at ~77 K is dominated by lattice vibrations. The addition of 0.5 at.% ZrO_2 modifier to extruded $Bi_{0.85}Sb_{0.15}$ solid solution samples with optimal carrier concentration makes it possible to obtain a material with thermoelectric efficiency of $\sim 6,4\cdot10^{-3}$ K^{-1} at ~77 K, thus enabling the creation of more efficient low-temperature electronic coolers.

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Author's personal contribution to the co-authored works: In publications [1–35], the problem formulation and its solution, as well as the execution of the experimental research, were carried out by the applicant (author); the analysis of the results obtained and the preparation of the scientific articles were performed jointly with the co-authors.

The defense will be held ___30___ ___October___ 2025, at 14:30 the meeting of the FD1.21 Dissertation Council operating under the Institute of Radiation Problems of the Ministry of Science and Education of the Republic of Azerbaijan.

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