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ABSTRACT

Of the dissertation for the degree of Doctor of Philosophy in Physics

INVESTIGATION OF ELECTRICAL INSTABILITY OF TlS AND TlSe CRYSTALS EXPOSED TO GAMMA IRRADIATION

Speciality: 2225.01 – Radiation materials science

Field of science: Physics

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GENERAL CHARACTERISTICS OF THE WORK

The subject's actuality. A thorough investigation into how semiconductor materials behave (properties) when exposed to radiation in the penetrating radiation fields of semiconductor electronic equipment is required (satellites, running reactors, etc.). Two linked difficulties should be resolved during these studies: the practical challenge of extending the service life of devices under these circumstances, and the physical problem of semiconductor defect generation. Investigating the mechanisms of defect creation, the impact of radiation on the structural structure, and the impact of defects on the physical processes in semiconductors are currently of utmost importance. By studying the influence of the introduction of defects and the type of radiation on them, it is possible to determine the basic laws that govern the change in the electrical properties of semiconductor crystals. As a result, solving the physical issue should also solve the problem under investigation, i.e., it should make the materials and gadgets made of them more radiation resistant.

The successful usage of devices utilizing lithium-ion-based current sources in electronics, energy, and industry has raised interest in ion-conducting materials in recent years. It is vital to research the characteristics of ion transport in these systems since the functioning of these devices depends on the electrochemical reactions that take place at the interface between the crystalline electrolyte and the electrode. Impedance spectroscopy is a useful technique in the domains of electrochemistry, physics, and materials science. One of the most informative methods for studying electrochemical and electrophysical processes in ion-conducting materials is impedance spectroscopy, which is used in various fields of electrochemistry, physics, and materials science. The technique of impedance spectroscopy finds extensive application in both fundamental and applied research. Superionic conductors are used in micro- and nanoelectronics, which stimulates interest in discovering novel compounds where the conductivity is largely influenced by the ionic component. Currently, the value of ionic conductivity in materials research is primarily determined via impedance spectroscopy.

Highly polarized cations in the crystalline structure, such as Pb^{2+}, Bi^{3+}, Tl^{+}, etc., and the existence of structural components, are important factors in the creation of ionic conductivity in solid bodies.

The purpose of the dissertation work: was to determine the jump conductivity, superion conductivity, and Poole-Frenkel effect properties in TlS and TlSe crystals using impedance spectroscopy and investigate the impact of γ-quanta on these properties.

The following problems were fixed in order to reach the target:

- Synthesis of TlS and TlSe and cultivation of single crystals;
- Investigation of electrical conductivity of exposed to irradiated with γ-quanta TlS and TlSe crystals at temperatures of 100-450 K and frequency intervals of 25-10^6 Hz;
- Investigation of the Poole-Frenkel effect of exposed to irradiated with γ-quanta TlS and TlSe crystals;
- Research of superionic conductivity of exposed to irradiated with γ-quanta TlS and TlSe crystals;
- Investigation of dielectric relaxation phenomena of exposed to irradiated with γ-quanta TlS and TlSe crystals;
- Construction (measurement) and analysis of complex impedance spectra of exposed to irradiated with γ-quanta TlS and TlSe crystals.

Research object and methods:

The studied TlS and TlSe single crystals were grown by Bridgman-Stockbarger method. After the synthesis process, the substance is filled inside the quartz ampoule and deaerated.

After determining the cultivation and brewing temperature, the temperature of the melting zone in the electric furnace is automatically heated by controlling the RIF-101 thermal regulator according to the selected temperature range. The temperature is recorded with the help of a Pt-Pt/Rh thermocouple connected to a 2-coordinate potentiometer, and after 2-3 hours of stabilization of the system (ampoule), the furnace starts moving at a speed of 0.2 cm/min to the end of the brewing zone. The temperature of the brewing zone was chosen to be ≈60% of the melting temperature of the studied substances. After the temperature in the zone and the necessary temperature gradient are found according to the selected temperature, the quartz ampoule filled with the substance to be grown is connected to the driving mechanism and inserted into the furnace. It is stabilized for 1.5-2 hours and the container is moved. In this case, the crystallization process begins.

Conductivity studies were conducted using the four-probe method. A digital impedance E7-25 measuring device was used to carry out these studies. Electrical conductivity measurements were performed in a wide temperature (100-450 K) and frequency (25-10^6 Hz) range. The measurements were performed using the quasi-stationary continuous heating mode in a nitrogen cryostat in the direction perpendicular to the crystallographic "c" axis.

In order to study the mechanism of current flow in semiconductor materials, the Volt-Ampere characteristics of the studied materials were investigated. B7-30 electrometer and a constant current source were used for the study. The experiments were conducted in the temperature range of 100-300 K and in the crystallographic 110 and 001 directions. Silver paste was used as current contacts.

The main scientific propositions defended include the following:

1. In the temperature range of 100-300 K, it was determined that the electrical conductivity of primary and exposed to irradiated with γ-quanta TlS and TlSe crystals has a jumping character.
2. The deviation from linearity in the Volt-Ampere characteristics of primary and exposed to irradiated with γ-quanta TlS and TlSe crystals obeys the Poole-Frenkel thermal-field theory.
3. In the temperature dependence of electrical conductivity ($\sigma(T)$) of primary and irradiated with γ-quanta TlS and TlSe crystals, the transition of the crystal to the superionic phase is observed at temperatures above room temperature.
4. The frequency dispersion of the dielectric permittivity and the relaxation properties of the dielectric loss angle of primary and

exposed to irradiated TlS and TlSe crystals with γ-quanta are observed. The charge transport mechanism has a jump character for localized states near the Fermi level at a frequency of 10^4-10^5 Hz.

5. As a result of the study of the complex impedance spectra of TlS and TlSe crystals, it was determined that Warburg's diffusion impedance is created at high temperatures and as a result of the influence of radiation.

The scientific innovations obtained as a result of the research are as follows: For the first time in the presented dissertation work:

1. The electrical conductivity of primary and exposed to irradiated with γ-quanta TlS and TlSe crystals in the temperature range of 100-300 K was determined to be subject to a hopping mechanism and was explained within the Mott approximation.
2. The deviation from linearity in the Volt-Ampere characteristics of primary and exposed to irradiated with γ-quanta TlS and TlSe crystals is explained within the framework of the Poole-Frenkel thermal-field theory.
3. It was determined that in the temperature dependence of the electrical conductivity of TlS and TlSe crystals irradiated with γ-quanta (σ(T)), a sharp increase in the value of electrical conductivity was observed in the temperature range of 300-450 K. The observed sharp increase is related to the diffusion of Tl^{+1} ions on vacancies in the thallium sublattice.
4. As a result of the analysis of complex impedance spectra of TlS and TlSe crystals, it was determined that Warburg's diffusion impedance was formed after γ-irradiation.
5. Frequency dispersion of dielectric permittivity and dielectric loss angle relaxation properties of primary and exposed to irradiated with γ-quanta TlS and TlSe crystals were determined. It was shown that the jumping increase in electrical conductivity at T=350 K and 10^5-10^6 Hz frequencies is due to the transition of the system to the superionic state.

Practical significance of the work:
The results obtained in the dissertation can be used as a suitable material for the preparation of electronic converters, feeding microbatteries, ionistors (capacitors with extremely large capacity), memory cores.

Approbation of the work:

The results of the dissertation were presented at the following conferences: "Materials of the IX republican scientific conference “Fizikanın aktual problemləri” (Baku 2016); “International Conference Modern Trends in Physics” (Baku,2017); Международная Конференция, посвященная 60-летию Института физики ДНЦРАН и 110-летию Х.И. Амирханова, «Фазовые переходы, критические и нелинеиные явления в конденсированных средах» (МахачКала 2017);“International Conference on Theoretical and nanoscience and Nanotechnology: Fundamentals and Applications Toronto, Ontario” (Canada, Toronto 2017); 11-я Международная Конференция «Ядерная и радиационная физика» Международная Конференция «Ядро-2017» (Алматы 2017); “7th International Conference MTP-(Baku, 2021); All-Republic conference on "Problems of Physics and Astronomy" dedicated to the "Year of Shusha" of masters and young researchers. BSU (Baku 2021).

The name of the organization where the dissertation work was carried out: The presented dissertation work was carried out in the "Radiation Physics of Ferroelectrics" laboratory of the Institute of Radiation Problems of the Ministry of Science and Education of the Republic of Azerbaijan.

Published works: 18 scientific works, including 11 articles, 7 conference materials have been published in republican and foreign scientific journals related to the subject of the dissertation work.

Structure and scope of the dissertation. The dissertation consists of the introduction, four chapters, conclusions and a list of cited reference. The submitted dissertation work consists of 61 figures, 5 tables and 182000 characters.

THE MAIN CONTENTS OF THE WORK

In the introduction, the relevance of the subject of the dissertation is justified, the purpose of the dissertation, its scientific innovation is substantiated, and its practical importance is indicated, and information is also given on the main provisions, approbation rate, and publications. The main content of the work by chapters is briefly described in the introduction.

In the first chapter of the dissertation work, literature materials related to the electrophysical properties of thallium-based chain structure chalcogenides included in the class of A^3B^6 type compounds were analyzed.

Methods of growing TlS and TlSe single crystals are given **in Chapter II** of the dissertation work. Also, in this chapter, the scheme and working principle of research devices for studying electrical and dielectric properties, the research method of γ-rays effect and the impedance spectrum measurement method are explained.

In Chapter III of the dissertation work, the results of the temperature dependence of the electrical conductivity and dielectric permeability of the TlS crystal irradiated with γ-quanta in the temperature range of 100-450 K and frequency range of 25-10^6 Hz, Volt-Ampere characteristics and ion conductivity by impedance spectroscopy method are given.

The properties of the conductivity of the TlS crystal irradiated with doses of primary and 0,15 MGy; 0,25 MGy; 0,35 MGy and 0,75 MGy were studied in the temperature range of 100-300 K, and the temperature dependences of the electrical conductivity in Arrhenius coordinates are shown in Figure 1.

Based on the results of the dependence of electrical conductivity on temperature, the existence of jumpy conductivity before and after irradiation was observed and temperature intervals were determined.

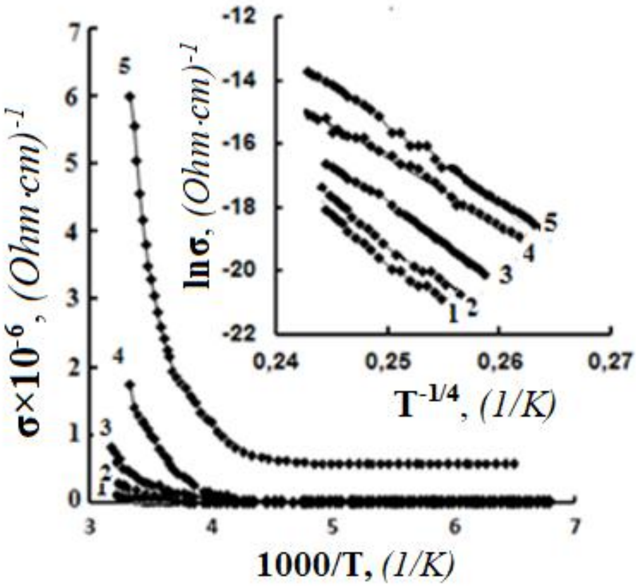

Figure 1. Temperature dependence of the electrical conductivity of primary and irradiated with different doses TlS crystal with different doses. The dependence of lnσ on $T^{-1/4}$ in Mott coordinates is given in the appendix above the figure. Curve 1 and curves 2, 3, 4, 5 correspond to radiation at a dose of 0.15 MGy, 0.25 MGy, 0.35 MGy, 0.75 MGy, respectively.

The temperature dependence of the electrical conductivity of the studied TlS crystal before and after gamma irradiation showed an exponential dependence in the temperature range of 190÷280 K. In the mentioned temperature range, thermally activated charge carriers have a dominant conductivity in the allowed zone. As can be seen from the insets above the figure, the experimental points in the dependence of lnσ on $T^{-1/4}$in the Mott coordinates in the temperature range 190<T<280 K are collected along a straight line. The observation of the linear dependence allows us to say that the charge carrier process in the temperature range of 190÷280 K in the primary and irradiated TlS crystal with doses of 0.15 MGy, 0.25 MGy, 0.35 Mgy, 0.75 MGy is carried out through jump conduction on local levels existing in the forbidden zone.

As it is known from the literatures, a number of irregularities appear in the crystal structure of semiconductors under the influence of radiation rays. Radiation defects caused by fast electrons and γ-rays in semiconductors are of special interest. Data obtained from the literature show that point defects such as vacancies or internodal atoms are formed when the studied materials are irradiated with gamma rays with an energy of 1 MeV.

These defects in turn create energy levels localized in the forbidden zone. According to Moot's theory, conductivity occurs by jumping over localized levels.

Based on Mott's theory, the calculated values of the parameters of the breakthrough permeability are given as a dose dependence (Fig. 2). As can be seen from the curves, the values of the jump conduction parameters in the TlS crystal vary significantly depending on the radiation dose, but the mechanism of conduction remains jump. A slight increase in the values of the density of localized states (N_F), concentration of traps (N_t) near the Fermi level, and a decrease in the energy difference (ΔE) and the length of the jump (R) are observed after irradiation. This leads to an increase in the price of conductivity.

It has been shown that the generation of radiation defects in the TlS compound after irradiation with gamma quanta leads to the emergence of new localized states [3].

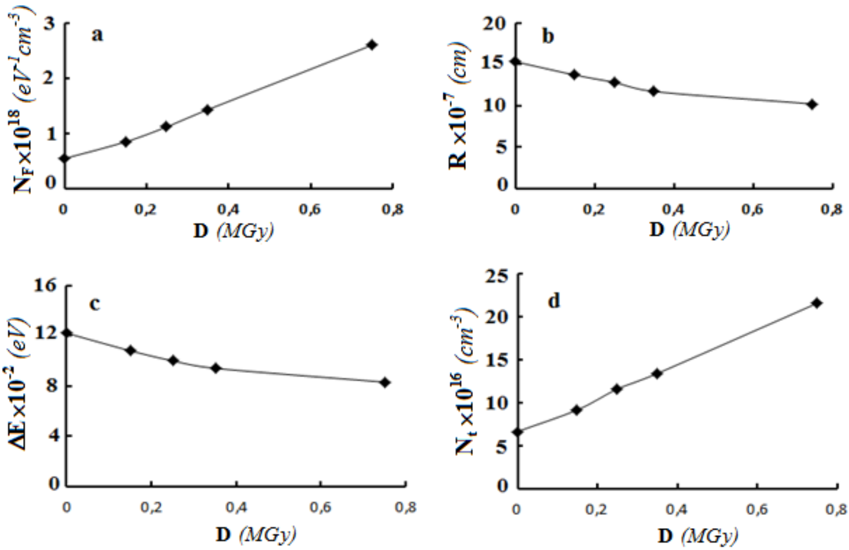

Figure 2. Dose dependence of jump conductance parameters in TlS crystal exposed to gamma irradiation. **a –** density of localized states (N_F), **b –** jump length (R), **c –** energy difference of localized states (ΔE), **d –** concentration of deep traps (N_t) on radiation dose.

In this chapter, the Volt-Ampere characteristic of non-irradiated and irradiated TlS crystal in the temperature range of 200÷300K and the value of the external electric field in the range of 0÷3000 V/cm was studied.

Primary and non-linear ($J \sim U^n$) parts are observed in the Volt-Amperes characteristic of non-irradiated and 0.25 MGy irradiated TlS crystal at different temperatures and electric field values.

The dependence of the electrical conductivity on the field intensity for a TlS crystal irradiated at doses of primary and 0.25 MGy in a strong electric field is in good agreement with the Frenkel formula.

$log\sigma - E, log\sigma - \sqrt{E}$ dependences were established for TlS crystals irradiated at doses of primary and 0,25 MGy. Based on established dependencies, it was determined that the $log\,\sigma - \sqrt{E}$

dependence obeys a linear regularity. It's on its turn, agrees well with the theoretical expression of the β-Frenkel coefficient. It was also determined that the temperature dependence of the Frenkel coefficient β, determined from the dependence of lnσ on $E^{1/2}$, obeys the expression $\beta = \frac{\sqrt{e^3}}{kT\sqrt{\pi\varepsilon\varepsilon_0}}$, and the extrapolation of the line obtained from the dependence of $\beta \sim 10^3/T$ passes through the coordinate origin.

The length of the free path (λ), the Frenkel coefficient (β) and the distance from the traps to the maximum of the potential barrier (x_m) in the framework of the Poole-Frenkel effect at the studied temperatures in the TlS crystal that was not irradiated and irradiated at doses of primary and 0.25 MGy, as well as the concentration of ionized centers (N_t) values were calculated and it was shown that the concentration of ionization centers formed due to radiation defects after gamma irradiation increases. It was determined that the deviation from linearity in the Volt-Ampere characteristic is explained by the Pool-Frenkel effect.

Also in this chapter, the properties of ion conductivity of non-irradiated and irradiated TlS crystal in the temperature range of 300-450K were studied.

Temperature dependence of electrical conductivity of primary and irradiated TlS samples at doses of 0.25 MGy, 0.75 MGy is given in figure 3. As can be seen from the curves, a jump-like increase in the value of conductivity is observed at a certain critical value of temperature at temperatures above room temperature. This is also associated with the predominance of ions in conductivity at temperatures above room temperature. One of the conditions confirming this fact is that the $10^3/T$ dependence of ln(σ·T) obeys a linear regularity. Primary and 0.25 MGy; The dependence of ln(σ·T) on $10^3/T$ was established based on the results obtained from the experiment on the temperature dependence of the electrical conductivity of the TlS crystal irradiated at doses of 0.75 MGy (Fig. 3 appendix). It was determined that this dependence follows a linear regularity in the studied crystals.

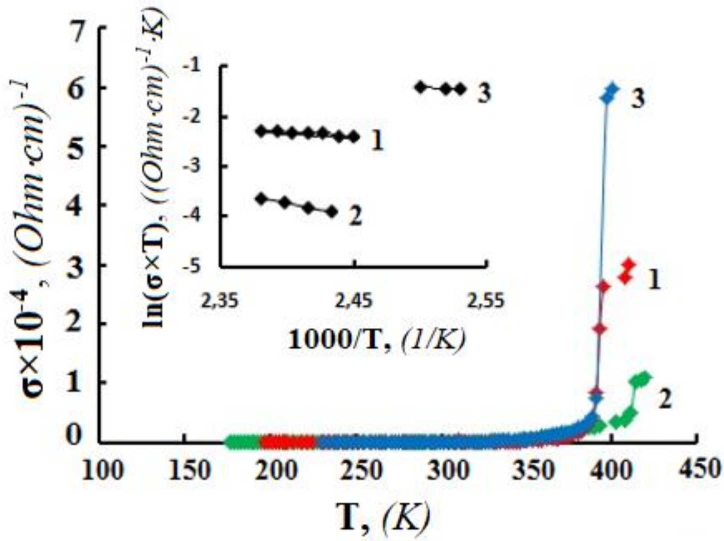

Figure 3. Temperature dependence of the electrical conductivity of the TlS crystal primary and irradiated with γ-quanta; 1-0 MGy, 2-0.25 MGy, 3-0.75 MGy. 1000/T dependence of lnσ is given in the insets above the figure.

The observed increase in the value of electrical conductivity in the TlS crystal at temperatures above room temperature can be explained by a sharp increase in the number of Tl^{+1} ions in the structure.

It is also known that the temperature dependence of the dielectric permeability in ionic conductive materials is followed by an exponential increase. In the studied sample, at a certain value of the temperature, a sudden increase in the dielectric permeability occurs. The sharp increase observed in temperature dependence of dielectric permittivity in TlS crystal occurs at temperatures T_{Kr}= 388 K before irradiation, T_{Kr} = 405 K after 0.25 MGy dose irradiation, and T_{Kr} = 388 K after 0.75 MGy dose irradiation.

In the studied TlS crystal, the value of the dielectric constant at higher temperatures in dependence of ε(T) increases in comparison with the values at lower temperatures. Such behavior observed in temperature dependence of dielectric permittivity in TlS crystal before and after irradiation is related to movement of ions along defects.

In the third chapter of the dissertation work, the results of dielectric relaxation of TlS crystal irradiated at different doses and the temperature dependence of the dielectric loss angle are also given.

Complex studies of the dielectric permeability of the TlS crystal were carried out on a digital E7-25 immittance measuring device at a frequency of $25\text{-}10^6$ Hz and a temperature interval of 100-450 K. The amplitude of the field size did not exceed 5 V/cm.

It was determined that the value of the dielectric constant at low temperatures in the unirradiated and irradiated TlS compound with gamma quanta is small (in TlS crystal, ε = 10 for 0 MGy; ε = 100 for 0.25 MGy; ε = 200 for 0.75 MGy) and remains unchanged in the studied frequency range. With the subsequent increase in temperature, an increase in the value of dielectric permeability is observed at all frequencies of the measurement range. Also, maxima are observed in the temperature dependence of the dielectric permittivity, and with the increase of the frequency of the field, these maxima shift towards the higher temperature region.

By approaching the temperature at which the observed ionic conductivity occurs in the studied crystals, additional relaxors consisting of monovalent Tl^{+1} ions are formed,, which in turn leads to an increase in the dielectric coefficient. Also, it should be taken into account that as the value of ionic conductivity increases at higher temperatures, the concentration of ions between the blocking contact and the ion crystal will increase, which will lead to the generation of a diffuse current of ions in the opposite direction. At this time, as the temperature at which ionic conductivity is observed approaches, the ions accumulated in the contact cause an increase in the dielectric coefficient of the system.

Also, the tangent of the dielectric loss angle of the TlS crystal irradiated at doses of primary and 0.25 MGy, 0.75 MGy was studied in the frequency range of $25 \div 10^6$ Hz and the temperature range of $100 \div 450$ K, and the obtained results are depicted in figure 4.

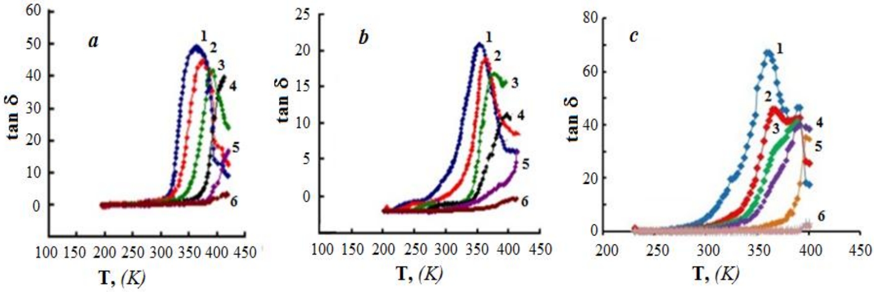

Figure 4. Temperature dependence of the dielectric loss angle tangent for TlS crystal irradiated at doses of primary and 0.25 MGy, 0.75 MGy. Measurements were made at frequencies of 1–25 Hz, 2–100 Hz, 3–1 kHz, 4–10 kHz; 5–100 kHz, 6–1 MHz. a-0 MGy, b-0.25 MGy, c- 0.75 MGy.

As can be seen from the grapHz, as the temperature increases in the dependence of tgδ(T), the value of the dielectric loss angle increases and takes a maximum value at a certain temperature. With the subsequent increase in temperature, a relaxation-like decrease is observed. As its frequency increases, the value of the peak corresponding to the maximum decreases and shifts towards the upper temperature region. This situation observed in the tgδ(ν,T) and ε (ν,T) dependences of the studied TlS crystal is characteristic of Debye type relaxation processes. The results obtained from the dependences of tgδ(ν,T) and ε (ν,T) are related to the presence of weakly connected electric charges in the crystal lattice.

The analyzes show that the temperatures at which the observed peaks in the ε(T) dependence occur lag behind the temperatures corresponding to the maxima of the tgδ(T) dependence. Since these anomalies are observed in the TlS crystal in the frequency range of

25÷1 MHz and at temperatures of 330÷450 K, it is not difficult to study them experimentally.

On the other hand, as it is known from the literature, the obtained results allow us to find the frequencies of possible jumps in the potential hole. At this time, it is also possible to determine the frequences of possible jumps by establishing dependence of lgf_{max} on 1/T from the maxima observed in the temperature dependence of tgδ. The frequency of the particle bouncing off the barrier was determined by constructing the extrapolation of the straight line in the $1/T \to 0$ approximation.

For the studied TlS crystal, the frequency of particle jumping from the barrier is $\nu = 4 \cdot 10^{12}$ Hz for 0 MGy; $\nu = 2 \cdot 10^{12}$ Hz for 0.25 MGy and $\nu = 10^{12}$ Hz for 0.75 MGy. These obtained values correspond to the region of the terahertz spectrum and fall into the phonon mode region of the crystal oscillation spectrum.

It is known that $(Tl^{3+}S^{2-}{}_{2})^{-}$ chains consist of Tl – S chain stretched in the direction of the tetragonal "c" axis. A favorable factor in the structure is the presence of large spaces for Tl^{+} ions to participate in conductivity. These voids in the crystal structure can significantly increase ionic conductivity. Based on crystallochemical considerations, it can be assumed that Tl^{+1} cations are the most mobile ions in the structure of the TlS compound.

In the third chapter of the dissertation work, the complex impedance and relaxation processes of the TlS crystal irradiated with gamma quanta in the frequency range of $25 \div 10^{6}$ Hz were investigated.

Real (Z′) and imaginary (Z″) parts of impedance of TlS crystal irradiated at doses of primary and 0.25 MGy and 0.75 MGy were measured. Based on the obtained results, images of the complex impedance hodograph were constructed and are given in figure 5.

As can be seen from the curves, in non-irradiated, 0.25 MGy and 0.75 MGy doses of irradiated TlS crystal, the arc of the complex plane hodograph at the point of intersection of Z' and Z" depicts a curve inclined to the true axis near the maximum semicircle (Fig. 5).

These hodograph curves correspond to the parallel equivalent substitution scheme and the energy transport is characterized by a single relaxation time. The imaginary part of the complex impedance, shows a maximum at certain frequencies $f_{(max)}$ and corresponds to the condition $C_{eff}R_{eff}\omega_{max} = 1$. Here, C_{eff} and R_{eff} are the effective parameters of the equivalent circuit, and $\omega_{max} = 2\pi f_{max}$ is the circular frequency. The maximum (tip) of the impedance hodograph corresponds to the resonance frequency (ω_{max}).

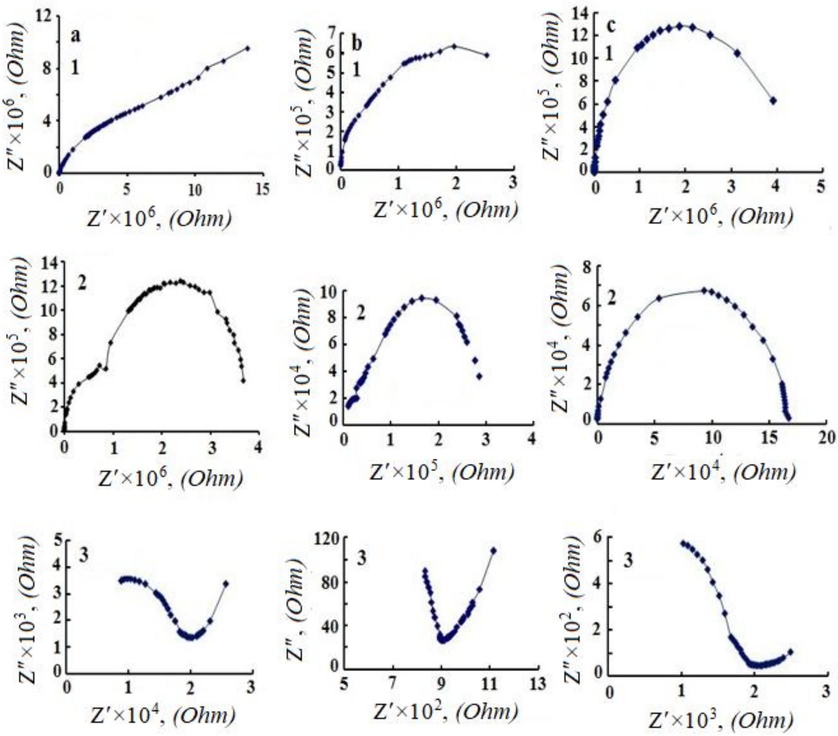

Figure 5. Impedance hodograph Z"(Z') in the complex impedance plane for a TlS crystal. Measurements 1-300 K, 2-350 K, and 3-400 K, a-0; b-0.25 MGy; and c-0.75 MGy dose was carried out.

Relaxation times f(max) of the frequencies correspond to the maximum of Z". Frequencies corresponding to the onset of frequency dispersion were determined for the non-irradiated and irradiated TlS crystal at doses of 0 MGy, 0.25 MGy and 0.75 MGy. An increase in the frequency $f_{(max)}$ corresponding to the Z" maximum is observed for the studied crystals at higher temperatures and after irradiation with gamma quanta. In the investigated compound TlS, the curves obtained before irradiation have a semicircular shape centered on the true axis. In this case, charge transfer is characterized by a relaxation period. The obtained hodograph curve corresponds to a homogeneous sample with low resistance and non-blocking contacts. This corresponds to a parallel RC-chain in the equivalent substitution circuit (Fig. 6 a). The hodograph curves obtained from the study of the complex impedance spectrum after irradiation at doses of 0.25 MGy and 0.75 MGy in TlS crystal have the shape of a semicircle at higher frequencies. In the area of low frequencies, it is reflected by "rays". The "rays" observed in the Naqvist curves are most likely due to the presence of both capacitance polarization near the electrode and polarization resistance as a result of charge accumulation in the region of the electric double layer (Fig. 6 b). The resulting output models the linear diffusion impedance caused by gamma radiation, known as the Warburg diffuse impedance. At high temperatures and after γ-irradiation, the transition of the crystal into a superionic state is due to the diffusion of Tl^{+1} ions on vacancies. Also, the frequency dispersion of the real (ε´) and imaginary (ε") part of the dielectric permittivity of TlS crystal irradiated at doses of unirradiated and 0.25 MGy and 0.75 MGy, tangent of the dielectric loss angle (tgδ), ac-conductivity (σ_{as}) 25 Hz - 10^6 MHz frequency interval was studied and the obtained results are given i**n the third chapter** of the dissertation. Dielectric measurements were made at temperatures of 300 K, 350 K, 400 K.

As a result of the study of the frequency dependence of the real and imaginary parts of the dielectric permittivity, it was determined that ε′ decreases weakly with increasing frequency (25 -10^6 Hz), at high frequencies ($f>10^3$ Hz) ε′ is weakly dependent on *f*, and at frequencies of 10^6 Hz ~10. It is shown that it receives a value of ~10.0. Variation of ε′ and ε″ as a function of frequency indicates the presence of relaxation dispersion of dielectric constant in TlS crystal.

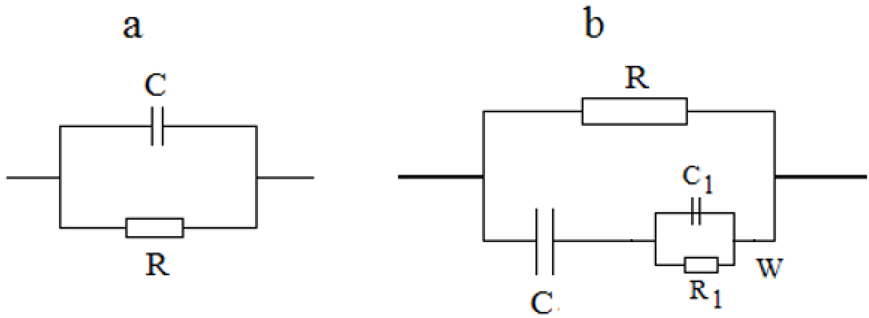

Figure 6. Equivalent circuit of complex impedance hodograph of unirradiated and irradiated TlS crystal with gamma quanta. (a–before irradiation, b–after γ-irradiation). W is Warburg's diffuse impedance, R is the resistance of the sample, and R_1 is the resistance in the saturable region.

At the same frequencies, a stronger dispersion is observed in the frequency dependence of the imaginary part of the dielectric constant. It should be noted that at a temperature of 300 K, the frequency dependence of the real and imaginary part of the dielectric permeability changes weakly, while at temperatures above room temperature, a sharp increase in the absolute values of ε′ və ε″ is observed. It is known that activation processes in dielectrics with relaxation polarization lead to a maximum in the frequency dependence of the imaginary part of the complex conductivity (ε"). In the TlS crystal, it was observed that at T = 400 K and a dose of 0.75 MGy, the complex conductivity undergoes a sharp increase, which is associated with an increase in the concentration of ion charge carriers.

Also, the frequency dependences of the tangent of the dielectric loss angle of the unirradiated and irradiated TlS crystal with gamma quanta were studied and the obtained results were interpreted in the third chapter of the dissertation. The maximum is observed in the dependence of tgδ(f) in the interval of frequency $f{\sim}10^3$ Hz, and it decreases to the minimum in the interval of frequency $10^5 \div 10^6$ Hz. The value of frequency dispersion ($f_p = 10^3$ Hz) and relaxation time ($\tau = 10^{-3}$ sec) was calculated in the studied crystals. As a result of the study of frequency dependences of the tangent of the dielectric loss angle, it is shown that TlS crystals have relaxation losses and current conductivity. This chapter also presents the results of the study of the frequency dependence of the ac-conductivity of the non-irradiated and irradiated TlS crystal with gamma quanta. After irradiation, a sharp increase in the frequency dependence of the conductivity of the investigated crystals is observed. The increase in the conductivity value is due to the fact that the crystals are already in the superionic phase at high temperatures and after gamma irradiation. This, in turn, is related to the irregularity of the system with the participation of ions in conductivity at high temperature.

In the third chapter of the dissertation, the results of the construction of complex impedance spectra of TlS crystal irradiated at different doses and their research are given. It is known that the most complete information about the processes occurring at the boundary of a material with superionic conductivity and a metal can be obtained using impedance spectroscopy methods.

As a result of the investigation of the complex impedance spectra, it was determined that with the increase in the frequency of the measurement area, the presence of dispersion is observed, as a result of decreasing values of the real Z'(ν) and imaginary Z"(ν) parts of the impedance. A sharp decrease in Z'(ν)and Z"(ν) is observed in the range of frequencies below $25\text{-}10^3$ Hz, but with the subsequent increase in frequency, a weakening of the frequency dependence is observed.

Complex studies of the frequency dependences of the TlS crystal irradiated at doses of unirradiated and 0.25 MGy; 0.75 MGy at 300, 350 and 400 K were carried out: real and imaginary parts of the complex dielectric constant; ac-conductivity; real and imaginary parts of the complex impedance Z*(f). It was determined that the dielectric constant is the frequency dispersion and the dielectric losses have a relaxation character. It has been shown that the charge transport mechanism has a breakthrough character for cases localized near the Fermi level. It was also established that Tl^{1+} ions diffusing in the case of ionic conductivity in the crystal are responsible for the existence of Warburg's diffuse impedance expressed as rays in the hodograpHz of the studied TlS crystals.

In Chapter IV of the dissertation work, the results obtained from the study of the temperature dependence of the electrical conductivity, dielectric permeability of the TlSe crystal irradiated with different doses in the temperature range of 100-450 K and frequency of 25-10^6 Hz, as well as the ionic conductivity by the impedance spectroscopy method were presented.

The dependence of the electrical conductivity of the TlSe crystal irradiated with a dose of unirradiated and 0.25 MGy; 0.75 MGy on temperature ($\sigma(T)$) was studied in the temperature range of 100-300 K. The temperature dependences of the electrical conductivity of TlSe crystal irradiated at a dose of unirradiated and 0.25 MGy; 0.75 MGy in Arrhenius coordinates have been investigated. It was shown that the temperature dependence of the electrical conductivity is exponential in the temperature range of ~190-240 K for the original TlSe crystal, ~185-250 K for the TlSe crystal irradiated with a dose of 0.25 MGy, and ~180-260 K for the TlSe crystal irradiated with a dose of 0.75 MGy. The dependence of $ln\sigma$ on $T^{-1/4}$ in the regions of exponential dependence was established and it was shown that the dependence follows a linear law. According to this condition, almost unirradiated and 0.25 MGy; In the TlSe crystal irradiated with doses of 0.75 MGy, it was determined that the conductivity had a jumping character in the mentioned temperature range. Permeability parameters in the Mott approximation framework: The density of localized states (N_F), the concentration of deep traps (N_t), the energy difference of localized states (ΔE) and the average length of carrier jumps (R) near the Fermi level were calculated and listed in table 1. Dose dependencies are also given.

Table 1. Calculated parameters of conductivity of TlSe crystals in the Mott approximation. (T = 240 K)

Dose (MGy)	$\mathbf{N_F}$ ($eV^{-1}\cdot cm^{-3}$)	**R** (cm)	**ΔE** (eV)	$\mathbf{N_t}$ (cm^{-3})
0	1.52×10^{18}	1.15×10^{-6}	0.101	1.53×10^{17}
0.25	1.75×10^{18}	1.11×10^{-6}	0.099	1.73×10^{17}
0.75	2.27×10^{18}	1.05×10^{-6}	0.095	2.15×10^{17}

It is shown that the density of localized states (N_F), the concentration of deep traps (N_t) increases depending on the radiation dose, while the energy difference of localized states (ΔE) and the average length of charge jumps (R) decrease depending on the radiation dose. In our opinion, this change is related to the generation of radiation defects.

Also, the electrical conductivity of the TlSe compound exposed to radiation in a strong electric field (10^3 V/sm) and in the temperature range of 100-300 K was studied within the framework of the Poole-Frenkel effect, and the obtained results are given in the fourth chapter of the dissertation.

VAC of primary and 0.25 MGy irradiated TlSe crystal at different temperatures was studied. Linear part and deviations from linearity were observed in VAC at different values of temperature and electric field ($J \sim U^n$). It was determined that with the increase in temperature and after irradiation with a dose of 0.25 MGy, the ohmic region decreases, and at this time, the transition voltage values decrease in the quadratic region. This is due to the increase in the concentration of charge carriers as a result of the formation of radiation defects.

As is known from the literature, it is possible to determine a number of parameters by implementing Frenkel's law. These important parameters are the β-Frenkel coefficient, λ- the length of the free path, x_m- the distance from the traps to the maximum of the potential barrier, N_t - the concentration of traps. These parameters were calculated in non-irradiated and 0.25 MGy irradiated TlSe samples and are listed in table 2.

Thus, the deviation from linearity in the Volt-Ampere characteristics of non-irradiated and γ-quantum-irradiated TlSe crystals was explained within the framework of the Poole-Frenkel thermal-field theory.

The ion conductivity of the TlSe crystal in the temperature range of 300-450 K and the results of the effect of γ-rays on this conductivity were investigated.

The electrical conductivity of TlSe crystal irradiated with doses of primary and 0.25; 0.75 MGy was studied in the temperature range of 300-450 K. Based on the obtained results, it was determined that at a certain value of temperature, the value of conductivity increases by several orders.

One of the important conditions of superionic conductors is that the 1/T dependence of ln(σ·T) obeys a linear regularity in the region where a sharp increase is observed, i.e., where ionic conductivity occurs. The (1/T) dependence of ln(σ·T) in the studied TlSe crystal obeys a linear regularity. The reason for this is related to the predominance of ions in conductivity at temperatures above the critical temperature. As we have mentioned, this change can be explained by the sharp increase in the number of Tl ions with high mobility, during which the phase transition to the superion state occurs.

The ionic conductivity is defined by the expression $\sigma \cdot T = \sigma_0 \cdot \exp(-\Delta E_a / k T)$, where ΔE_a is the activation energy of the conductivity. The values of the activation energy of the studied TlSe crystal were calculated before and after gamma irradiation, and 0 MGy-E_a =0.032 eV; 0,25 MGy-E_a =0.04 eV; 0,75 MGy-E_a =0.012 eV before irradiation; values were obtained.

Table 2. Values of parameters calculated within the framework of the Poole-Frenkel effect of TlSe crystal before and after irradiation.

T *(K)*	**Dose** *(MGy)*	**β** $(cm/V)^{1/2}$	**λ** *(cm)*	$\mathbf{x_m}$ *(cm)*	$\mathbf{N_t}$ (cm^{-3})
200	0	0.017	6.58×10^{-6}	1.21×10^{-6}	1.14×10^{17}
	0.25	0.015	1.35×10^{-6}	5.32×10^{-7}	5.42×10^{17}
230	0	0.012	7.72×10^{-6}	1.63×10^{-6}	3.52×10^{16}
	0.25	0.014	3.22×10^{-6}	8.22×10^{-7}	3.52×10^{17}
300	0	0.011	31.9×10^{-6}	2.32×10^{-6}	7.42×10^{16}
	0.25	0.009	14.2×10^{-6}	1.08×10^{-6}	1.31×10^{17}

Also in the fourth chapter of the dissertation work, the ratio of electronic and ion components of the total conductivity in TlSe compound, which was not irradiated at 300, 350 K and irradiated with γ-quantum at a dose of 0.25 MGy, was determined by the Wagner polarization method. It is possible to estimate the share of ions in mixed electron-ionic compounds by studying the time dependence of electric conductivity in constant current.

Figure 7 shows the time dependence of the conductivity of the TlSe compound irradiated at a dose of primary and 0.25 MGy at temperatures of 300, 350 K. As electrodes, silver contacts were used, which block the contribution of ions to the conductivity, and a constant potential difference was applied.

As can be seen from the figure, the time dependence of electrical conductivity in a constant field is nonlinear. As a result, a total current of ions and electrons flows during measurements at the initial instant of time; charge transfer in the stationary state is carried out only by electrons. Thus, the result of the mutual compensation of the volume charge region near the silver contact interface, which has a blocking nature, is the decrease of the electric current with time in the constant field. As can be seen from Fig. 7, in a TlSe crystal irradiated with a dose of 0.25 MGy, ~ 80% ion current passes through the sample in a stationary state at a temperature of 350K.

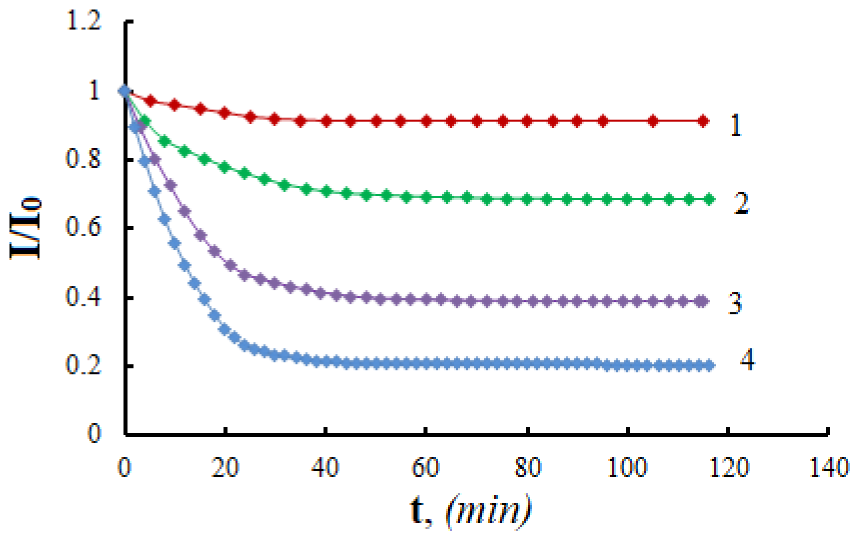

Figure 7. Time dependence of electrical conductivity I/I_0 of TlSe compound. Measurements were carried out using silver electrodes at temperatures of 300 K and 350 K and doses of 0, 0.25 MGy. Curves 1 and 2-300 K were 0 MGy, curves 3 and 4-350 K were 0.25 MGy.

This chapter also presents the results of studies of temperature and frequency dependence of ε and tgδ unirradiated and gamma irradiated TlSe crystal. It was noted that with the increase in the frequency of the field, the maxima observed in the dependences of tgδ(T) and ε(T) shift towards higher temperature and a decrease in their value is observed. This is related to the relaxation nature of the studied crystal and allows us to say that there are weakly connected electric charges in the crystal lattice.

The results obtained from the calculation allow us to find the activation energies of the jump and their frequencies for the possible jumps in the potential hole.

On the other hand, these parameters can be easily found by establishing the dependence of lgf_{max} on 1/T in non-irradiated and irradiated TlSe crystal with gamma quanta. Figure 8 shows the dependence of lgf_{max} on 1/T, and by extrapolating the straight line from the approximation 1/T→0 in the TlSe crystal, the frequency of the particle jumping from the barrier is determined.

Primary and 0.25 MGy; 0.75 MGy irradiated TlSe crystals, this frequency is $\nu = 4\cdot 10^{12}$ Hz for the unirradiated sample, $\nu = 3\cdot 10^{12}$ Hz for the 0.25 MGy irradiated sample, and $\nu = 2\cdot 10^{12}$ Hz for the 0.75 MGy irradiated sample. As we mentioned, these values of the frequency correspond to the outer part of the infrared spectrum and fall into the phonon mode region of the crystal's oscillation spectrum.

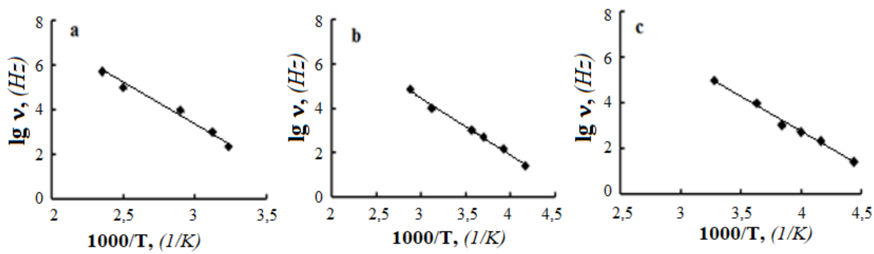

Figure 8. Dependence of the frequency of relaxation maxima of the tangent of the dielectric loss angle of the TlSe crystal irradiated at a dose of primary and 0.25 MGy; 0.75 MGy on the inverse value of the temperature. a-0 MGy, b-0.25 MGy, c-0.75 MGy.

The study of the phenomenon of charge transport in irregular systems in an alternating electric field is of practical and fundamental importance. At the same time, the main goal is to determine the load transport mechanism based on known theories. **In the fourth chapter,** the nature of the electrical conductivity of the TlSe crystal was studied in a wide range of temperature (300 K; 350 K; 400 K) and frequency (25-10^6 Hz) and the charge transport mechanism was determined.

Primary and 0.25 MGy; Frequency dependence of ac-conductivity of irradiated TlSe crystal at 300 K, 350 K, 400 K temperature was studied at 0.75 MGy doses. In the frequency interval of 10^5-10^6 Hz, the value of ac-conductivity of the studied crystals increases sharply by ~ 10 times. The effect of gamma radiation on the frequency dependence of electrical conductivity at doses of 0.25 MGy and 0.75 MGy is due to the fact that the studied crystal is in the superionic phase at temperatures above room temperature and at a frequency of 10^6, at this temperature and at this frequency, the conductivity is mainly ionized and the system is disordered.

The frequency dependence of the real and imaginary part of the dielectric permittivity of the unirradiated and irradiated TlSe crystal at different doses was studied and it was shown that the observation of the decrease in the values of the complex dielectric components with the increase in the frequency of the electric field measurement field reveals the dispersion properties. With increasing frequency, ε' weakly decreases at high frequencies (f >10^3 Hz). At high frequencies, for TlSe crystal, ε' decreases 10 times at 0 MGy dose, ε' decreases 2 times at 0.25 MGy dose, and ε' decreases 4 times at 0.75 MGy dose. The nature of the frequency change ε' and ε'' in the investigated TlSe crystal indicates the presence of relaxation dispersion of the dielectric constant.

Figure 9 shows the impedance hodograph curves of TlSe crystal irradiated at doses of primary and 0.25 MGy and 0.75 MGy at different temperatures. It has been shown that the hodograph curves can be divided into two parts, indicating the presence of a relaxation mechanism. The high-frequency part of the dependence of $Z''(Z')$ is related to the relaxation process. The relaxation mechanism in the low-frequency part of the impedance godagraph is related to the diffusion mechanism, which is related to the presence of the carrier concentration gradient in the surface region. Thus, the mechanism of energy loss in the studied TlSe sample consists of losses related to conduction and relaxation polarization.

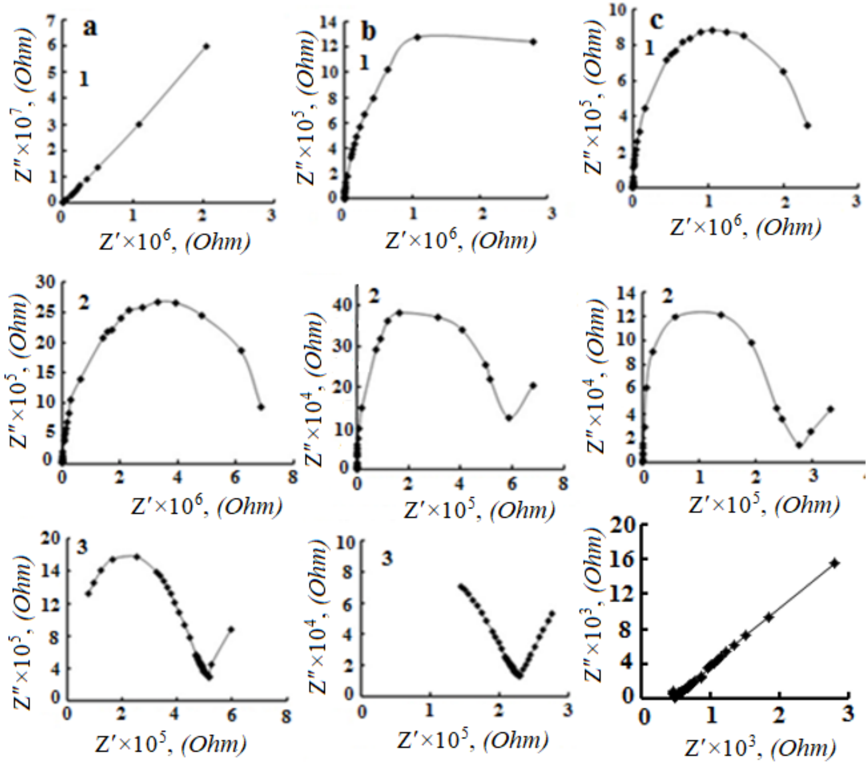

Figure 9. Complex impedance spectra (Z" - Z') in TlSe crystal irradiated at doses of 0; 0.25 and 0.75 MGy.

Based on the obtained hodograph curves, the equivalent circuit of the electric circuit is described using the impedance components. Such a hodograph is suitable for homogeneous samples with low resistance and no insulating contact. The equivalent circuit described corresponds to the low frequency range in the figure 6 a. The low-frequency region of the (Z"−Z') complex diagram corresponds to the beam. In the primary samples at 400 K, after gamma quantum irradiation at 350 K temperature, the hodograph curves of impedance observed the rays in the low-frequency region and these rays are associated with diffuse ion transport near the solid electrolyte and surface.

The charge transport process is defined by a single relaxation period prior to irradiation; nevertheless, additional "rays" were discovered in the low-frequency area of the crystals' hodograph curves following gamma-quantum irradiation. As we previously discussed, Warburg's diffuse impedance is connected to these rays in the hodograph curves.

Based on our earlier research, it was demonstrated that at temperatures higher than room temperature, the Tl^{+1} ion in TlSe crystals started to diffuse, dominating the ionic component of conductivity. Based on the theory that the applied sinusoidal signal does not reach the boundary of the charge carrier's diffuse layer in the frequency range, rays in the impedance diagram are indicative of the Warburg diffuse impedance. The crystal's shift to the superion state at temperatures higher than room temperature is connected to the measurement of the Warburg diffuse impedance.

MAIN RESULTS

1. The electrical conductivity of TlS and TlSe crystals, irradiated with different doses, in the temperature range of 100-300 K was studied and it was shown that the conductivity occurs with a hopping on localized energy levels. The values of the hopping conduction parameters for the localized states around the Fermi level were calculated and it was shown that the values of these parameters change significantly depending on the radiation dose, however the conduction mechanism remains hopping. For the TlS crystal, it was defined that in the dose range of 0 MGy – 0.75 MGy, the values of density of localized states near the Fermi level (N_F – $5.43\cdot10^{17}$–$2.6\cdot10^{18}$ $eV^{-1}\cdot cm^{-3}$) and the concentration of traps (N_t – $6.62\cdot10^{16}$–$2.17\cdot10^{17} cm^{-3}$) increase depending on the radiation dose, while the values of average hopping length (R – $1.53\cdot10^{-6}$ – $1.02\cdot10^{-6}$ cm) and energy difference of localized states (ΔE – 0.122 – 0.083 eV) decrease. However, for the TlSe crystal, in the dose range of 0 MGy – 0.75 MGy, the values of the density of localized states near the Fermi level (N_F – $1.52\cdot10^{18}$ – $2.27\cdot10^{18}$ $eV^{-1}\cdot cm^{-3}$), the concentration of traps (N_t – $1.53\cdot10^{17}$– $2.15\cdot10^{17} cm^{-3}$) increase depending on the radiation dose, while the values of average hopping length R – $1.15\cdot10^{-6}$ – $1.05\cdot10^{-6}$cm) and energy difference of localized states (ΔE – 0,101 – 0,095 eV) decreases.

2. It was determined that the deviation of the Volt-Ampere characteristics in the TlS and TlSe crystals that were non-irradiated and irradiated with γ-quanta, from linearity is explained in the Frenkel heat-field effect. The values of the parameters within the framework of the Poole-Frenkel effect were calculated and it was shown that the values of these parameters vary significantly depending on the radiation dose. After irradiation with gamma quanta, the value of the concentration of traps (N_t) increases, while the values of the mean-*free-path length* (λ), Frenkel coefficient (β) and the distance from the traps to the maximum of the potential barrier (x_m) decrease.

3. It was determined that the characteristics observed in the value of electrical conductivity (σ) of TlS and TlSe crystals, irradiated with γ-quanta, at temperatures above 388 K are related to the transition of the crystal to the superion state. The observed superionic conductivity occurs as a result of the diffusion of monovalent thallium ions through node vacancies in the thallium sublattice.

4. In TlSe crystal, that was non-irratiated and irradiated with a dose of 0.25 MGy, ion accumulation in constant electric field at different temperatures was calculated. It was shown that the share of ion accumulation in the conductivity before irradiation is about 10%, and after irradiation at a dose of 0.25 MGy, about 80% of the ion current passes through the sample in a stationary state at a temperature of 350 K. The decrease of the current in the constant electric field over time is explained by the mutual compensation of the volume charge regions near the blocking electrode-crystal boundary (interface).

5. In TlS and TlSe crystals, transition to the superionic phase stimulated by radiation and Warburg impedance in the electric double layer were observed. Thus, the "rays" observed in the godograph curves obtained after γ-irradiation were associated with Warburg's diffuse impedance. This is associated with the inability of ion carriers (Tl^{+1}) to pass through the blocking contact.

THE MAIN RESULTS OF THE DISSERTATION ARE PUBLISHED IN THE FOLLOWING ARTICLES AND THESES.

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