IRSTI 29.19.16 https://doi.org/10.26577/JPEOS20261282

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DIVERGENT MECHANISMS OF THE EFFECTS OF ELECTRON AND GAMMA IRRADIATION ON THE STRUCTURAL AND THERMOELECTRIC PROPERTIES OF BI_2TE_3–SB_2TE_3 THIN FILMS

The advancement of space exploration and radioisotope thermoelectric generator (RTG) technologies demands the development of flexible thermoelectric materials capable of stable operation under extreme radiation environments. However, the divergent dynamic effects of corpuscular and photon-type radiation on the structural properties of chalcogenide solid solutions deposited on polymer substrates remain insufficiently understood. In this study, 25% Bi_2Te_3–75% Sb_2Te_3 thin films with a thickness of 0.790 μm, grown on polyethylene terephthalate (PET) substrates via thermal vacuum evaporation, were subjected to electron beam irradiation (up to 1×10^{15} e/cm²) and gamma irradiation (up to 1×10^{7} R). The morphological, structural, and electrophysical parameters of the specimens were systematically analyzed using atomic force microscopy (AFM), X-ray diffraction (XRD), Raman spectroscopy, and a two-probe measurement technique. The results revealed a fundamental difference in the degradation mechanisms depending on the nature of irradiation. At the critical electron fluence of 1×10^{15} e/cm², severe radiation damage and surface atom sputtering led to the formation of an amorphous-metallized surface layer, resulting in a sharp decrease in the Seebeck coefficient. In contrast, gamma irradiation fully preserved the surface morphology while generating homogeneous bulk Frenkel pairs via the Compton effect. This induced successive compensation and decompensation (defect coagulation) cycles strictly governed by the Pisarenko–Mott relation, with electrical conductivity dropping to 0.12×10^{3} $\Omega^{-1}\cdot m^{-1}$ at 1×10^{6} R and recovering to 0.84×10^{3} $\Omega^{-1}\cdot m^{-1}$ at 1×10^{7} R. It is concluded that gamma irradiation serves as a bulk defect engineering tool, whereas high-dose electron bombardment irreversibly degrades thermoelectric efficiency through surface erosion.

Keywords: Bi_2Te_3–Sb_2Te_3 thin films, thermoelectric properties, electron beam irradiation, gamma irradiation, radiation hardness, defect engineering, surface amorphization.

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Электронды және гамма-сәулеленудің Bi_2Te_3–Sb_2Te_3 жұқа үлдірлерінің құрылымдық және термоэлектрлік қасиеттеріне әсер етуінің әртүрлі механизмдері

Ғарышты зерттеудің және радиоизотопты термоэлектрлік генераторлар (РИТЭГ) технологияларының дамуы экстремалды радиациялық ортада тұрақты жұмыс істеуге қабілетті икемді термоэлектрлік материалдарды жасауды талап етеді. Дегенмен, корпускулалық және фотондық сәулеленудің полимерлі төсеніштерге қондырылған халькогенидті қатты ерітінділердің құрылымдық қасиеттеріне әртүрлі динамикалық әсерлері әлі де жеткілікті зерттелмеген. Бұл жұмыста термиялық вакуумдық булану әдісімен полиэтилентерефталат (ПЭТ) төсеніштерінде өсірілген, қалыңдығы 0,790 мкм болатын 25% Bi_2Te_3–75% Sb_2Te_3 жұқа үлдірлері электронды шоқпен сәулеленуге (1×10^{15} э/см² флюенске дейін) және гамма-сәулеленуге (1×10^{7} Р дозаға дейін) ұшыратылды. Үлгілердің морфологиялық, құрылымдық және электрофизикалық параметрлері атомдық-күштік микроскопия (АКМ), рентгендік дифракция (РД), Раман спектроскопиясы және екі зондты өлшеу әдісі арқылы жүйелі түрде талданды. Нәтижелер сәулеленудің табиғатына байланысты деградация механизмдеріндегі іргелі айырмашылықты көрсетті. 1×10^{15} э/см² критикалық электрондық флюенсінде қатты радиациялық зақымданулар мен беткі атомдардың шашырауы аморфты-металданған беттік қабаттың түзілуіне әкелді, бұл Зеебек коэффициентінің күрт төмендеуін тудырды. Керісінше, гамма-сәулелену Комптон эффектісі есебінен біртекті көлемдік Френкель жұптарын тудыра отырып, беткі морфологияны толығымен сақтап қалды. Бұл Писаренко – Мотт қатысына қатаң бағынатын дәйекті компенсация және декомпенсация (ақаулардың коагуляциясы) циклдерін тудырды, мұнда электр өткізгіштік 1×10^{6} Р дозада дейін төмендеп, 1×10^7 Р дозада 0.84×10^3 Ом$^{-1}$·м$^{-1}$-ге дейін қалпына келді. Гамма-сәулелену көлемдік ақаулар инженериясының құралы ретінде қызмет етеді, ал жоғары дозалы электронды атқылау беттік эрозия салдарынан термоэлектрлік тиімділікті қайтымсыз төмендетеді деген қорытынды жасалды.

Түйін сөздер: Bi_2Te_3–Sb_2Te_3 жұқа үлдірлері, термоэлектрлік қасиеттер, электронды шоқпен сәулелену, гамма-сәулелену, радиациялық төзімділік, ақаулар инженериясы, беттің аморфтануы.

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Разнонаправленные механизмы влияния электронного и гамма-облучения на структурные и термоэлектрические свойства тонких пленок Bi_2Te_3–Sb_2Te_3

Развитие космических исследований и технологий радиоизотопных термоэлектрических генераторов (РИТЭГ) требует создания гибких термоэлектрических материалов, способных к стабильной работе в условиях экстремального радиационного воздействия. Однако разнонаправленные динамические эффекты корпускулярного и фотонного излучений на структурные свойства халькогенидных твердых растворов, нанесенных на полимерные подложки, остаются недостаточно изученными. В данной работе тонкие пленки состава 25% Bi_2Te_3–75% Sb_2Te_3 толщиной 0,790 мкм, выращенные на подложках из полиэтилентерефталата (ПЭТ) методом термического вакуумного испарения, подвергались облучению электронным пучком (флюенс до1×10^{15} экв/см^2) и гамма-излучению (доза до 1×10^7 Р). Морфологические, структурные и электрофизические параметры образцов были систематически проанализированы с использованием атомно-силовой микроскопии (АСМ), рентгеновской дифракции (РД), спектроскопии комбинационного рассеяния (КР) и двухзондового метода измерений. Результаты выявили фундаментальное различие в механизмах деградации в зависимости от природы излучения. При критическом флюенсе электронов 1×10^{15} экв/см^2 сильные радиационные повреждения и распыление поверхностных атомов привели к формированию аморфизованно-металлизированного поверхностного слоя, что вызвало резкое падение коэффициента Зеебека. Напротив, гамма-облучение полностью сохранило морфологию поверхности, генерируя при этом однородные объемные пары Френкеля за счет эффекта Комптона. Это индуцировало последовательные циклы компенсации и декомпенсации (коагуляции дефектов), строго подчиняющиеся соотношению Писаренко — Мотта, при этом электропроводность падала до 0.12×10^3 Ом$^{-1}$·м$^{-1}$ при дозе 1×10^6 Р и восстанавливалась до 0.84×10^3 Ом$^{-1}$·м$^{-1}$ при 1×10^7 Р. Сделан вывод, что гамма-облучение служит инструментом инженерии объемных дефектов, тогда как высокодозная электронная бомбардировка необратимо снижает термоэлектрическую эффективность вследствие эрозии поверхности.

Ключевые слова: тонкие пленки Bi_2Te_3–Sb_2Te_3, термоэлектрические свойства, облучение электронным пучком, гамма-облучение, радиационная стойкость, инженерия дефектов, аморфизация поверхности.

1. Introduction

The global energy crisis and the growing demand for environmentally clean technologies have stimulated unprecedented scientific interest in thermoelectric (TE) materials and devices that directly convert thermal energy into electrical energy. For thermoelectric devices operating in the near-room-temperature range (300–500 K), solid solutions based on Group V–VI chalcogenides – particularly bismuth and antimony tellurides (Bi_2Te_3, Sb_2Te_3) – represent the most promising and well-established materials. The distinctive layered rhombohedral crystal structure ($R\bar{3}m$ space group) of these materials enables independent control of charge carrier and phonon scattering mechanisms, thereby ensuring a high thermoelectric figure of merit (ZT) (Silva et al., 2025; Musri, et al., 2024). In recent years, the rapid advancement of wearable electronics, autonomous sensor networks, and lightweight radioisotope thermoelectric generators (RTGs) designed for deep-space exploration has created a strong impetus to transition from conventional bulk crystals to thin-film structures grown on flexible polymer substrates such as polyethylene terephthalate (PET) or polyimide (Ma et al., 2024; Ali et al., 2026).

Thermoelectric modules deployed in space environments or operated in proximity to nuclear reactors are continuously exposed to extreme external factors, particularly high-energy ionizing radiation. The interaction of ionizing radiation with the semiconductor lattice generates point defects – including vacancies, interstitial atoms, and antisite defects (collectively termed Frenkel-type disorder) – within the material bulk. These defects fundamentally alter the density of states and charge carrier scattering centers, exerting a profound influence on the macroscopic electrophysical properties of the material (Zhang et al., 2022, Yang et al.,2024).

The scientific literature has demonstrated, through numerous bulk crystal studies, that low-dose ionizing irradiation can be employed as a "defect engineering" tool to artificially enhance thermoelectric power (Seebeck coefficient) by deliberately shifting the Fermi level (Zheng et al., 2020; Xu et al., 2021). However, the thermodynamic and kinetic response of thin films to irradiation differs fundamentally from that of bulk crystals. The significantly elevated surface-to-volume ratio in thin films gives rise to pronounced surface energy effects, grain boundary diffuse scattering, and charging of interface states during irradiation (Park et al., 2026).

Nevertheless, the distinction between the dynamic effects of particle beams (high-kinetic-energy electrons) and photon radiation (gamma quanta) on ternary chalcogenide thin films remains insufficiently elucidated in the international scientific literature. Most existing studies address either electron bombardment or gamma irradiation in isolation, and a unified physical model providing a comparative analysis of the surface-versus-bulk damage partitioning between these two types of irradiation is currently lacking (Qasrawi et al., 2015; Aliyev et al., 2025). In particular, the open question of whether the sharp decrease in thermoelectric efficiency under extreme radiation doses arises from radiation-induced surface sputtering and amorphization or from purely bulk compensation effects has yet to be resolved. Addressing this problem is of paramount importance for predicting the long-term reliability and radiation hardness of flexible thermoelectric generators operating in radiotoxic environments.

To address the aforementioned scientific gap, the present work is dedicated to the systematic investigation of the effects of progressively increasing electron and gamma irradiation on the structural, morphological, and electrophysical parameters of 25% Bi_2Te_3–75% Sb_2Te_3 stoichiometric thin films grown on PET substrates. Through the synchronized application of atomic force microscopy (AFM), Raman spectroscopy, and comprehensive electrophysical measurements, the complete evolutionary chain of radiation-induced processes in the material – from radiation hardening to irreversible radiation damage – was traced. The study provides scientific evidence for the distinct degradation mechanisms that arise depending on the nature of irradiation: electron beams act as an agent causing surface metallization and erosion, while gamma rays serve as a tool inducing bulk defect coagulation while preserving surface integrity. These findings contribute to a deeper understanding of the radiation stability of flexible thermoelectric materials and open new prospects for their application in extreme environments

2. Materials and methods

Thin films based on the 25% Bi_2Te_3–75% Sb_2Te_3 stoichiometric solid solution were deposited by thermal vacuum evaporation using a UVN-71 P2 universal vacuum coating system. To minimize the adverse effects of external impurities and residual gases and to ensure high film purity, the vacuum chamber was evacuated to a base pressure of 1.33×10^{-3} Pa prior to deposition. Mechanically and thermally stable polyethylene terephthalate (PET; trade name: Lavsan) polymer sheets with a thickness of 50–125 μm served as the substrate material. To establish thermodynamic equilibrium and optimal crystallization conditions, the substrate temperature was maintained at 100 ± 2 °C throughout the deposition process.

Evaporation was carried out using a resistive tantalum (Ta) crucible. To ensure stable evaporation kinetics, the crucible was heated to 610 °C. The source-to-substrate distance was optimized based on kinetic theory to guarantee a uniform distribution of the evaporated material across the substrate surface, and was fixed at 8 cm. The initial charge mass for each deposition run was 70 mg, and the total deposition time was 8 min. These parameters consistently yielded thin films with an average thickness of 0.790 μm. Silver (Ag) contacts were subsequently deposited by thermal evaporation onto both edges of the as-grown specimens for electrical measurements.

The synthesized specimens were divided into two groups for comparative analysis. The first group was subjected to electron beam irradiation at three fluence levels (1×10^{13}, 5×10^{14}, and 1×10^{15} e/cm²) using an electron accelerator. The second group was exposed to gamma irradiation at three dose levels (1×10^{6}, 5×10^{6}, and 1×10^{7} R) using a ^{60}Co isotope source.

Vibrational modes and phonon characteristics were investigated by Raman spectroscopy. Surface morphology and roughness parameters were analyzed in both 2D and 3D topographic imaging modes using an atomic force microscope (AFM). Electrical properties – including electrical conductivity (σ), electrical resistivity (ρ), and the temperature-gradient-dependent Seebeck coefficient (α) – were measured at room temperature using a standard two-probe method.

3. Results and discussion

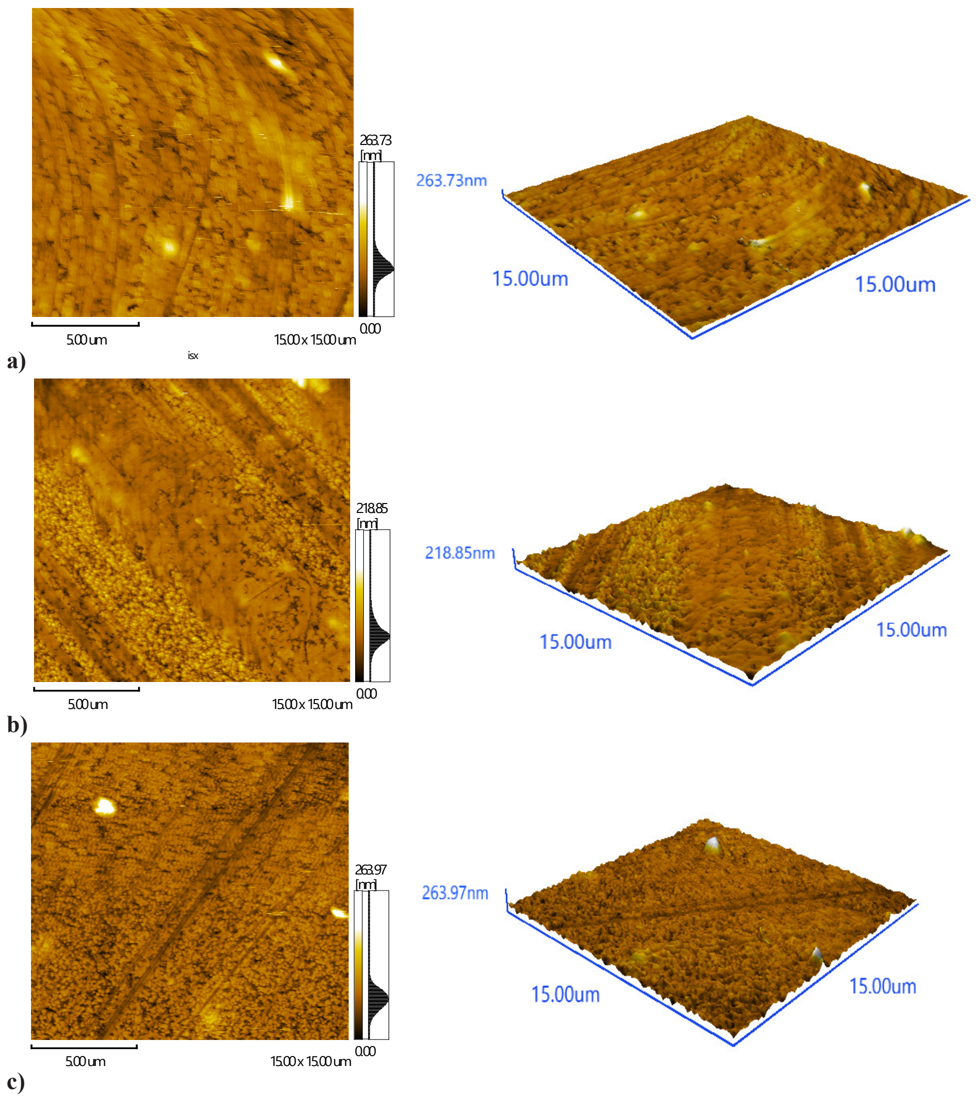
3.1. Morphological and thermoelectric parameters of the as-deposited (unirradiated) specimens

AFM topographic images (Fig. 1a) revealed a dense, void-free, well-defined granular surface structure formed via the Volmer–Weber island growth mechanism. The root-mean-square (RMS) surface roughness (Rq) was 17.76 nm. Raman spectra (Fig. 3) confirmed the distinct presence of the Eg^2 (~110–120 cm^{-1}) and A_1g^2 (~150–160 cm^{-1}) vibrational modes, indicating the formation of quintuple-layer structural units bonded through covalent and van der Waals interactions.

Electrophysical analysis of the as-deposited specimens yielded an electrical conductivity of $\sigma \approx 6.67\times10^3\ \Omega^{-1}\cdot m^{-1}$. The positive value of the Seebeck coefficient (+160 to +168 μV/K) indicates that charge transport is dominated by holes (p-type conductivity) generated by antisite defects (Sb_pTe, Bi_pTe).

Figure 1

a) 2D and 3D AFM topographic images of the unirradiated thin film; b) 2D and 3D topographic images after electron beam irradiation at 1×10^{15} e/cm²; c) 2D and 3D topographic images after gamma irradiation at 1×10^7 R.

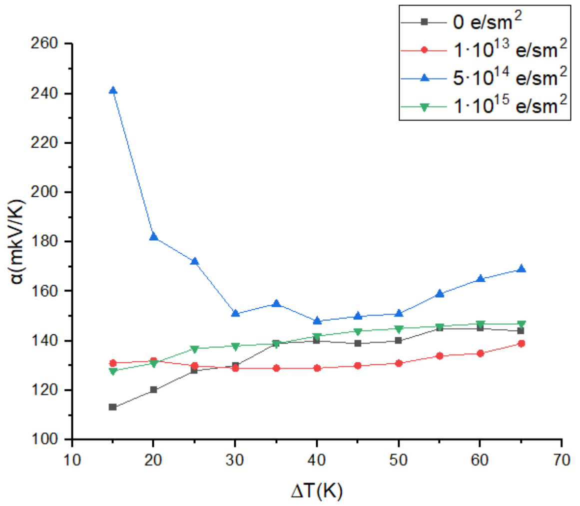


3.2. Destructive mechanisms and surface amorphization under electron beam irradiation

The interaction of the electron beam with the thin films induced strictly dose-dependent anomalies through localized kinetic energy release ("thermal spikes") and elastic collisions.

Figure 2

Seebeck coefficient as a function of temperature gradient for the unirradiated and electron-beam-irradiated 25% Bi_2Te_3–75% Sb_2Te_3 thin films at various dose levels



At the low fluence of 1×10^{13} e/cm², tellurium vacancies (V_pTe) with donor character were rapidly generated within the film bulk. These vacancies compensated the existing holes (compensation effect), sharply reducing electrical conductivity. However, in accordance with the Pisarenko–Mott relation, the decrease in carrier concentration increased the entropy per charge carrier, leading to an anomalous enhancement of the Seebeck coefficient to +270 μV/K – a manifestation of "defect engineering." At the intermediate fluence of 5×10^{14} e/cm², kinetic processes promoted defect clustering and the "radiation hardening" phenomenon, partially stabilizing the lattice properties.

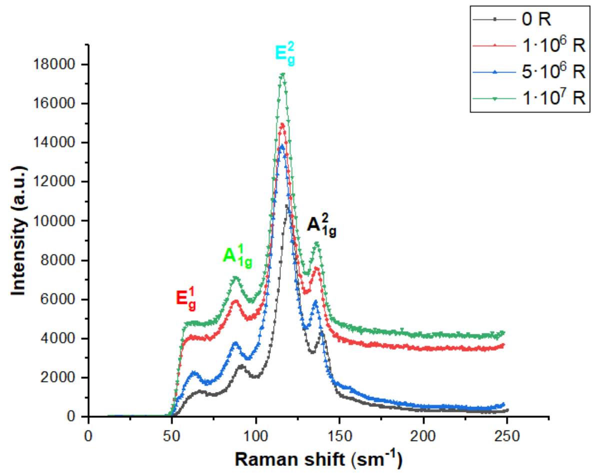
At the critical fluence of 1×10^{15} e/cm², severe radiation degradation was observed. As shown in Fig. 1b, electron bombardment sputtered surface atoms into the vacuum. AFM images visually confirmed the transformation of the surface into a porous, sharp needle-like residual relief. The vibrational modes in the Raman spectrum were completely suppressed, transitioning to a broad amorphous background. As a result of the massive loss of Te atoms from the surface, a Bi/Sb-rich, partially amorphous metallized layer was formed. This highly conductive metallized surface layer effectively short-circuited ("shunted") the underlying semiconducting bulk. Consequently, the thermoelectric properties deteriorated sharply, with the Seebeck coefficient decreasing to +150 μV/K.

3.3. Isotropic evolution and defect coagulation under gamma irradiation

The interaction of gamma radiation with the film material differs fundamentally from that of electrons, generating purely bulk processes without surface erosion (Fig. 1c). Gamma photons penetrated deep into the film bulk via Compton scattering, generating secondary electrons. AFM results confirmed that even at the critical dose of 1×10^7 R, the original granular surface morphology and roughness (Rq = 19.3 nm) were fully preserved.

Figure 3

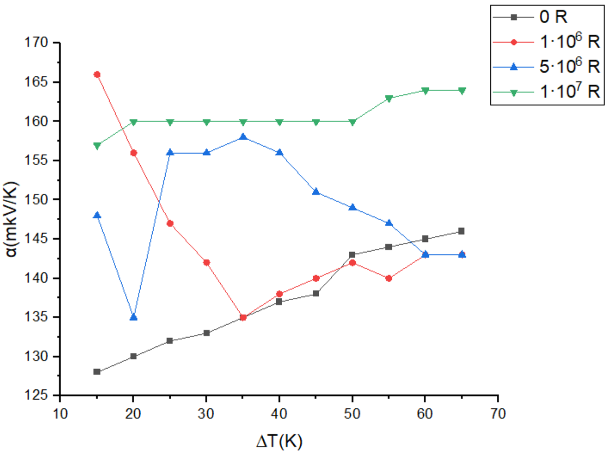
Raman spectra of the unirradiated and electron-beam-irradiated 25% Bi_2Te_3–75% Sb_2Te_3 thin films at various dose levels



In the Raman spectra, the intensities of the A_1g and Eg vibrational modes decreased but did not vanish, indicating the preservation of long-range structural order in the thin film.

Figure 4

Seebeck coefficient as a function of temperature gradient for the unirradiated and gamma-irradiated 25% Bi_2Te_3–75% Sb_2Te_3 thin films at various dose levels



At the reference dose of 1×10⁶ R, V_pTe donors generated in the bulk induced a compensation effect, reducing σ by a factor of 55 (to 0.12×10^3 $\Omega^{-1}\cdot m^{-1}$). Concurrently, the Seebeck coefficient exhibited an anomalous increase to ~185 μV/K. As the dose was increased to 5×10^6 R and 1×10^7 R, defect coagulation (decompensation) processes were activated within the bulk, and electrical conductivity recovered along a U-shaped trajectory to 0.84×10^3 $\Omega^{-1}\cdot m^{-1}$.

These non-monotonic variations in the Seebeck coefficient strictly obey the fundamental Pisarenko–Mott equation applicable to partially degenerate semiconductors (Eq. 1):

$$\alpha = \frac{8\pi^2 k_B^2}{3eh^2} m^* T \left(\frac{\pi}{3p}\right)^{2/3} \quad (1)$$

where kB is the Boltzmann constant, e is the elementary charge, h is Planck's constant, m* is the effective mass of charge carriers, T is the absolute temperature, and p is the hole concentration.

As is clearly evident from Eq. (1), $\alpha \sim p^{-2/3}$, meaning thermoelectric power increases as carrier concentration decreases. When the hole concentration (p) is reduced by compensation at 1×10^6 R, the entropy per carrier increases, driving the Seebeck coefficient to its maximum. At the extremely high dose of 1×10^7 R, the increase in concentration due to decompensation causes the Seebeck coefficient to decrease to +126 μV/K, in full accordance with Eq. (1). This finding mathematically confirms that gamma irradiation acts as a genuine bulk defect engineering tool. Since the surface and contacts remained intact throughout, the Ag–thin film interface guaranteed ideal ohmic current transport at all gamma dose levels.

4. Conclusions

The optimal deposition technology for 25% Bi_2Te_3–75% Sb_2Te_3 stoichiometric thin films with a thickness of 0.790 μm on PET polymer substrates via thermal vacuum evaporation was successfully developed. AFM results confirmed that the specimens possess a highly ordered crystalline lattice belonging to the $R\bar{3}m$ space group with excellent surface morphology, and that the Ag contacts form ohmic electrical contacts.

– The effect of electron beam irradiation was found to exhibit two sharply distinct regimes depending on the fluence. At 1×10^{13} e/cm², the Seebeck coefficient was enhanced to +270 μV/K via the compensation effect (defect engineering), whereas at the critical fluence of 1×10^{15} e/cm², the material completely lost its thermoelectric character due to massive surface sputtering, partial amorphization, and metallization.

– It was demonstrated that gamma irradiation generates homogeneous Frenkel pairs in the thin film bulk via the Compton effect without causing any surface damage. With increasing dose, defect coagulation and decompensation processes are activated, resulting in U-shaped recovery of electrical conductivity, with the Seebeck coefficient responding solely to the balance of bulk parameters.

– The totality of experimental evidence demonstrates that thin films based on bismuth and antimony chalcogenides exhibit extremely high radiation stability against high-dose gamma irradiation, rendering them optimal materials for operation in extreme radiotoxic zones and as elements of space-based RTGs. Particle beams, acting as an erosive and destructive agent, necessitate active surface protection of the device during long-term operation.

Author Contributions

Conceptualization, editing and methodology, K.E. Onarqulov; Validation, formal analysis, investigation, writing – original draft preparation, B.U. Omonov.

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Submitted for review 22.04.26
Accepted 29.05.26