Measurement of γ-ray transmission factors of semiconductor crystals at various annealing temperature and time

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In the present study, the change according to the annealing temperature and time of γ-ray transmission factors or transmissivity of InSe, InSe:Sn semiconductor crystals that prepared by not evaporated onto the stage (glass) and InSe, InSe:Mn, InSe:Fe, InSe:Ag, InSe: Cd, InSe:Sn and InSe:Gd semiconductor crystals that prepared by evaporated onto the stage (glass), have been examined. Gamma-rays of $^{241}$Am passed through crystals have been detected by a high-resolution Si(Li) detector and by using energy dispersive X-ray fluorescence spectrometer (EDXRF). Undoped-InSe and Mn, Fe, Ag, Cd, Sn, Gd doped InSe semiconductor crystals have been grown by using the Bridgman/Stockbarger. Evaporated onto the stage crystals have been prepared by using thin-film coatings system with thermal evaporation method. The structural and lattice parameters of the InSe and InSe:Sn semiconductors have been analyzed by using X-ray diffractometer (XRD). Transmission factors have been given graphically against the annealing temperature and time for time range 0 (unannealed) - 60 min with a step of 10 min. Also, transmission factors have been measured for annealing temperature range 50-(combustion temperature of the crystal) with a step of 50°C for not evaporated onto the stage semiconductor crystals. Transmission factors have been measured for annealing temperature range 60°C-(cracking temperature of the stage) with a step of 60°C for evaporated onto the stage crystals. Results are presented and discussed in the present paper.

Keywords: Transmission factors, Crystal growth, InSe, Semiconductor crystals, Bridgman/Stockbarger technique, EDXRF, XRD

1 Introduction

X and γ-ray transmission measurements are used in a wide variety of industrial and medical applications. These applications include measurements of the composition of multi-phase flow in piping from oil wells, thickness gauges for production of flat-rolled steel and aluminium, level measurements in vessels, bone density measurements, inspection of the explosive charge in artillery shells, monitoring of the ash in coal on conveyor belts, monitoring the amount of tobacco in cigarettes, and soil density gauges. The single crystals of A$^{III}$B$^{VI}$ type compounds, in particular of indium selenide (InSe) crystal in a layered structure, where each layer contains two In and two Se close-packed sub layers in the stacking sequence Se-In-In-Se. The energy band structure of InSe:Mn, InSe:Fe, InSe:Ag, InSe: Cd, InSe:Sn and InSe:Gd binary semiconductor alloy is similar to that of the binary analog InSe semiconductor compound.

InSe single crystals belonging to the family of III-VI compound semiconductors and used for many technological applications have been grown and their structural and morphological analyses have been carried out at room temperature. These are very important in solar energy transports and other technology applications. The interest in this crystal has been increasing in recent years. The interaction with matter of electromagnetic radiation for experimental work has been an important issue. In recent years, the interactions with crystals of electromagnetic radiation have been observed by many researchers.

Sreekumar et al. have reported the effect of indium concentration and annealing temperature (100-400°C) on the structural and optoelectronic properties of indium selenide thin films grown using a stack elemental layer technique. Gürbulak have grown undoped-InSe and Gd doped InSe (InSe:Gd) single crystals by using the Stockbarger method. Absorption measurements have been carried out in InSe and InSe:Gd samples in the temperature range 10-320 K with a step of 10 K. Erzenegölü et al. have measured the mass attenuation coefficients of InSe and InSe having different holmium concentrations in the energy region 15.746-40.930 keV using a Si(Li) detector. InSe and InSe:holmium (0.0025), InSe:holmium (0.0050), InSe:holmium (0.025) and InSe: holmium (0.05) crystals were grown by the
Bridgman-Stockbarger method. The measured values are compared with the theoretical ones obtained using WinXCom. İçelli et al., the mass attenuation coefficients of undoped n-type InSe, and Gd, Ho, Er doped n-InSe single crystals using a Si(Li) detector in the energy region 15.746-40.930 keV. Viswanathan et al., have analyzed the properties of deposited InSe thin films, deposited onto well cleaned glass substrates under a vacuum of 10^{-5} Torr using X-ray diffraction, Rutherford back scattering, energy dispersive analysis of X-rays, optical transmittance and current-voltage (120-390 K) measurements.

The variation according to the annealing temperature and time of γ-ray transmission factors prepared by evaporated onto the stage and not evaporated onto the stage semiconductor crystals, has been studied. The transmission factor versus annealing time and temperature for InSe and InSe:Mn, InSe:Fe, InSe:Ag, InSe:Cd, InSe:Sn and InSe:Gd at 59.5 keV has been measured.

2 Experimental Details

The schematically arrangement of the experimental set-up used in the present work is shown in Fig. 1. It consists of a 3.7×10^9 Bq (100mCi) Am-241 point source, which essentially emits monoenergetic (59.5 keV) γ-rays. The intensities of γ-rays were measured using a high-resolution Si(Li) detector (FWHM of 160 eV at 5.96 keV) and the data were collected into 4096 channels of a multichannel analyzer. Undoped InSe and Mn, Fe, Ag, Cd, Sn, Gd doped InSe semiconductor crystals were grown by using Bridgman/Stockbarger method. InSe, InSe:Mn, InSe:Fe, InSe:Ag, InSe:Cd, InSe:Sn and InSe:Gd semiconductor crystals have been evaporated onto the stage to obtain homogeneous samples. Evaporated onto the glass crystals have been prepared using thin-film coatings system with thermal evaporation method. Measurements have been made on non-porous section of evaporated onto the stage crystals. The temperature of annealing furnace was between 0-1200°C. The net counts without annealing and with annealing were obtained at the same and experimental conditions.

A parallel beam of monoenergetic X or γ-ray passing through matter is attenuated due to absorption and scattering. Attenuation due to absorption follows the Beer-Lambert’s law:

\[ I = I_0 \exp(-\mu x) \]  

... (1)

where \( I_0 \) and \( I \) are the unattenuated and attenuated photon intensities, respectively and \( \mu (\text{cm}^{-1}) \) is the linear attenuation coefficient of the material. Also, where \( I/I_0 \) is the transmission factor or transmissivity \( (T) \). The net counts without absorber \( (I_0) \) and with absorber \( (I) \) were obtained at the same and experimental conditions. Representative spectrum of 59.5 keV γ-rays passed through InSe:Cd is shown Fig. 2.

For the constituent of polycrystalline A_{III}B_{VI}, the first important step in obtaining high quality crystals is the purity of the basic elements which are being involved in the structure. These elements were weighed in a stoichiometric ratio accurate to 0.1 mg. The total mass of the elements was about 40 g. The basic criteria for this choice were, firstly a sufficient need to justify the cost of one run, secondly minimal loss of the material in case of breakage.

This stoichiometric ratio necessary to produce 40 g InSe was calculated using the following relationships:

\[ M_{\text{In}} = (M_{\text{Se}}/A_{\text{Se}})A_{\text{In}} \]  

... (2)

with total mass:

\[ M_{\text{In}} = M_{\text{Se}}=40 \text{ g} \]  

... (3)
where \( M \) and \( A \) are total and atomic masses, respectively.

InSe and doped InSe (0.01 wt \% Mn, Fe, Ag, Cd, Sn and Gd) semiconductor crystals were grown by using the Bridgman/Stockbarger method. The crystals used in this study were grown in our crystal growth laboratory. The ingots had no cracks or voids on the surface. No polishing or cleaning treatments were carried out on the cleaved faces of these samples because of the natural mirror-like cleavage faces. They were cleaved into perpendicular planes of naturally cleaved planes. The prepared InSe and doped InSe crystal ingots were 10 mm in diameter and about 60 mm in length. The melting point \( 660 \pm 5 \)°C of the InSe compound were determined from the phase diagram.

3 Results and Discussion

The structural and lattice parameters of the undoped InSe and InSe:Sn semiconductors were analyzed using X-ray diffractometer (XRD) using Cu-K\( \alpha \) radiation with a wavelength of \( \lambda=1.54050 \)Å (Rigaku Miniflex). The values of 2\( \theta \) were altered between 10° and 90° with the step of 0.1 deg/s. Figure 3 shows the XRD pattern of the undoped InSe and InSe:Sn semiconductors. It is found that the InSe and InSe:Sn crystals had hexagonal structure, quite close to 2\( \theta \) peak values and dominant diffraction peak around 2\( \theta=66.777 \)° for the both was the (0012) plane. This result is found to be in good agreement with that obtained in Refs (9,10).

Transmission factors versus annealing temperature and annealing time have been plotted in Fig. 4-19 for InSe, InSe:Mn, InSe:Fe, InSe:Ag, InSe:Cd, InSe:Sn and InSe:Gd semiconductor crystals.
Fig. 6 — Transmission factor versus annealing time of InSe, InSe:Fe and InSe:Ag that prepared by evaporated onto the stage

Fig. 7 — Transmission factor versus annealing time of InSe that prepared by not evaporated onto the stage

Fig. 8 — Transmission factor versus annealing time of InSe:Sn that prepared by not evaporated onto the stage

Fig. 9 — Transmission factor versus annealing time of InSe and InSe:Sn that prepared by not evaporated onto the stage

Fig. 10 — Transmission factor versus annealing temperature of InSe and InSe:Sn that prepared by not evaporated onto the stage

Fig. 11 — Transmission factor versus annealing temperature of InSe, InSe:Mn, InSe:Fe and InSe:Gd that prepared by evaporated onto the stage
Fig. 12 — Transmission factor versus annealing time of InSe (not evaporated onto the stage) and InSe:Sn (evaporated onto the stage)

Fig. 13 — Transmission factor versus annealing time of InSe and InSe:Sn that prepared by not evaporated onto the stage

Fig. 14 — Transmission factor versus annealing time of InSe:Sn (not evaporated onto the stage) and InSe:Sn (evaporated onto the stage)

Fig. 15 — Transmission factor versus annealing time of InSe and InSe:Sn that prepared by not evaporated onto the stage

Fig. 16 — Transmission factor versus annealing temperature of InSe and InSe:Sn that prepared by not evaporated onto the stage

Fig. 17 — Transmission factor versus annealing time of InSe that prepared by not evaporated onto the stage
In Figs 4 and 6, transmissivity has similar changes at the same temperature for different crystals. In addition to, transmissivity of InSe:Ag has different changes after 10 min as seen in Fig. 6. In Fig. 7, transmissivity before decreases for InSe at 32°C. It is seen that transmissivity decreases through minutes from 0 (unannealed) to 60, the other side it increases through minutes from 60 to 125. It is clear that there is turning point at 60 min. As seen in Figs 5, 8, 9, 12-15 and 17, transmission factor curves decrease as the annealing time is increased. In Figs 10, 11, 16 and 18 transmission factor curves decrease as the annealing temperature is increased, which suggests that semiconductor crystals become absorbent as they have a long annealing time and high annealing temperature. This situation may be related to the fact that at high temperature, the semiconductor material begins to lose its extrinsic characteristics and begins to behave more like an intrinsic semiconductor. Also, these results may be related to the scattering effect. That is, Coulomb interaction exists between the electrons or holes and produces scattering or collisions. However, at higher temperatures, the intrinsic carrier concentration increases and begins to dominate the electron concentration as well as the conductivity. These situations clearly suggest that the atoms in a crystal have a certain thermal energy, which is a function of temperature. This thermal energy causes the atoms to vibrate in a random manner about an equilibrium lattice point. This random thermal motion causes the distance between atoms to randomly fluctuate, slightly disrupting the perfect geometric arrangement of atoms. Foreign atoms, or impurity atoms, may be present in a crystal lattice. Impurity atoms may be located at normal lattice sites\textsuperscript{11}. The present results confirm the presentation of doped and temperature effects. All semiconductor crystals burned at a definite temperature and time. In Fig. 10, while max annealing temperature is 300°C for InSe, it is 350°C for InSe:Sn. As seen in Fig. 11, 16, 18 and 19, maximum annealing temperature is 180°C that cracking temperature of the stage (glass) for evaporated onto the stage crystals. In Figs 8, 12-15 and 17, there are similarities at different annealing temperatures to not evaporated onto the stage crystals and evaporated onto the stage. InSe:Mn, InSe:Fe, InSe: Cd, InSe:Sn and InSe:Gd at high annealing temperatures and long annealing time become more absorbent but InSe and InSe:Ag become more permeable. As seen in Figs 18 and 19, crystals doped with consecutive atomic number elements have shown similar changes and as symmetrical. Crystals doped with elements that have big atomic mass have shown more absorptive property than they have small. That is, the elements with small atomic mass are faster to diffuse.

In the present study, effort has been made to reduce the error sources in transmission measurements. In an ideal transmission experiment, all photons must be sent on absorber sample with a parallel beam. But in real experimental studies, there is the same errors. The errors in the evaluation of the area under the $\gamma$-ray peaks are ($\leq 0.9\%$). The errors in the determination of the thickness of sample are ($\leq 1\%$), in the counting statistic are ($\leq 1.5\%$), in the annealing temperatures are ($\leq 0.83\%$) and annealing times are ($\leq 1.7\%$).
To obtain and compare more sensitive values, more experimental study is needed connected with different crystals, doped materials, grown method except for Bridgman/Stockbarger method, thickness samples and energies. Also, the experiment can be repeated with cold of crystal and used for different ground for evaporated except for stage (glass).

References