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Effect of Gamma Irradiation on Structural and optical properties of Thin Films of a-$Cd_5Se_{95-x}Zn_x$

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Abstract: We report the gamma irradiation effect on the structural and optical properties of a-$Cd_5Se_{95x}Zn_x$ (x=0, 2, 4) thin films. Thin films were prepared by thermal evaporation technique at a base pressure of 10^{-5} torr. The investigated thin films of ~ 200 nm thickness are then irradiated by 25-100 kGy doses of ^{60}co gamma rays. The elemental composition of as-deposited thin films of $Cd_5Se_{95-x}Zn_x$ (x=0, 2, 4) were verified by RBS spectrometry shows good agreement with the actual elemental composition. XRD, PL and FT-IR study confirms the crystallinity of the investigated thin films increases after gamma irradiation. The optical parameters were estimated from optical absorption spectra data measured from UV-vis-spectrophotometer. It was found that the value of optical band gap of investigated thin films decreases and the corresponding absorption coefficient increases up to 75 kGy dose of gamma irradiation. This post irradiation change in the values of optical band gap and absorption coefficient were interpreted in terms of increase in crystallinity of the material after gamma irradiation.

Keywords: Thin films; Chalcogenides; Gamma irradiation; Structural properties; Optical properties.

1. Introduction

Amorphous natured chalcogenide semiconductor materials have revolutionized the world of optoelectronics owing to their low phonon energy, photo-induced anisotropy, high linear refractive index, chemical stability and possibility of variation in physical properties with composition. Now-a-days chalcogenide glasses are considered as an attractive material for high energy irradiation detector in dosimetric system for wide industrial applications due to a close relationship of irradiation induced effects with absorbed doses [2]. These kinds of irradiation detectors allows a low barrier of information bleaching temperature (< 350 ℃) as compared to widely used closed oxide glasses (< 500 ℃). In particular the ternary alloy of CdSeZn is an attractive candidate for all these important technological applications like laser screen materials in projection color TV's [3, 4], Laser diodes, Electroluminescent, Photovoltaic's, Photolumniscent diodes, LED's and nuclear radiation detectors [5-9].

The properties of materials are further modified by using different irradiation techniques like laser irradiation, swift heavy ion irradiation, gamma-ray irradiation etc in order to enhance the performance of various devices. The main job of these irradiation techniques is to dissipate much more energy into to the localized region of the material. This dissipation of the energy is responsible for micro-structural change occurring in the material that in turn changes the other properties of the material. The present study deals with the effect of gamma-rays irradiation on structural and optical properties of thin films of a-$Cd_5Se_{95-x}Zn_x$(x=0, 2, 4). Gamma irradiation of solid materials produces microstructural changes which intern changes the optical and other properties of the material [10-13]. When gamma radiation interacts with the material, two processes occurs at the same time i.e., defect creation and defect annihilation. Thus the changes in the properties of the material will depends up on these two processes or on the other hand one can say depends on the irradiation dose of gamma radiation. This is because at higher doses of gamma radiation the number of defects created becomes more than the number of defects annihilated, the reverse is true at lower doses of gamma radiation. Thus gamma irradiation of materials may cause re-crystallization or amorphization depends on the nature of the material and irradiation dose of gamma radiation. From above discussion it becomes clear that the probability of recrystallization is more at lower doses of gamma radiation whereas the amorphization of the material is more at higher doses of gamma radiation. The relative

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effects were investigated experimentally in this study.

2. Experimental details

Thermal quenching technique has been for the preparation of bulk samples of amorphous $Cd_5Se_{95-x}Zn_x$ (x = 0, 2, 4). The mixture of $Cd_5Se_{95-x}Zn_x$ (x = 0, 2, 4) from highly pure 99.9999% Cd, Se and Zn was weighed according to atomic percentages and sealed in quartz ampoules kept at pressure of 10^{-5} torr. The sealed ampoules were kept inside the rotating furnace for 15h at 900°C temperature for obtaining the homogeneous melt of the material. After 15h, the ampoules were quenched in ice cooled water for obtaining amorphous state of the material. Thin films of investigated material have been prepared by thermal evaporation technique on quartz substrates at room temperature under a pressure of ~10^{-5} torr through molybdenum boat. The quartz substrates was first cleaned in ultrasonic bath and then by acetone. For achieving metastable equilibrium, the films were kept inside the deposition chamber for 24h. The thickness was measured to be ~ 200nm by using ellipsometry technique. These thin films of $Cd_5Se_{95-x}Zn_x$ (x = 0, 2, 4) were irradiated by 25-100kGy dose of ^{60}Co gamma radiation (available at Interuniversity Accelerator Center New Delhi, India).

At an average dose rate of 1 kGy/h. The elemental composition and thickness measurement of as-deposited thin films of $Cd_5Se_{95-x}Zn_x$ (x = 0, 2, 4) were confirmed by Rutherford backscattering spectroscopy (RBS). A beam of 2 MeV from He^+ source was used. Regaku X-ray diffractometer has been used for structural study of $Cd_5Se_{95-x}Zn_x$ (x = 0, 2, 4). Further the structural characterization of as-deposited and gamma irradiated thin films of $Cd_5Se_{95-x}Zn_x$ (x = 0, 2, 4) were done by FT-IR (Nicolet, Protégé 460 MS-632). PL measurements of as-deposited and 75kGy Gamma irradiated films have been done at room temperature in the wavelength range of 420–700 nm using a Perkin Elmer LS055 spectrometer (Perkin Elmer LS055 with excitation wavelength of 350 nm). The optical characterization before and after irradiation of thin films of $Cd_5Se_{95-x}Zn_x$ (x = 0, 2, 4) were done by using VU-vis-spectrophotometer (Model:comspec M550) in the wavelength range 190-1100nm.

3. Results and discussions

3.1 RBS Study

RBS spectrometry is a non-destructive and sensitive technique for quantitative analysis and for thickness measurements. Fig.1 shows the RBS spectra of as-deposited thin films of $Cd_5Se_{95-x}Zn_x$(x=0, 2, 4). The elemental composition and thickness measurement analysis have been done with the help of RUMP software. As-deposited thin films of $Cd_5Se_{95-x}Zn_x$(x=0, 2, 4) shows good stoichiometry as analyzed by RBS simulation Se (90%), Cd (10%) for $Cd_5Se_{95-x}Zn_x$(x=0), Se (90%), Cd (6%),Zn (4%) for $Cd_5Se_{95-x}Zn_x$(x=2) and Se (88%), Cd (5%), Zn (7%) for $Cd_5Se_{95-x}Zn_x$(x=4). The thickness of the as-deposited films of $Cd_5Se_{95-x}Zn_x$(x=0, 2, 4) evaluated by RBS are 197 nm, 195 nm and 210 nm respectively, which also shows good agreement with the thickness measured by ellipsometry technique (200nm).

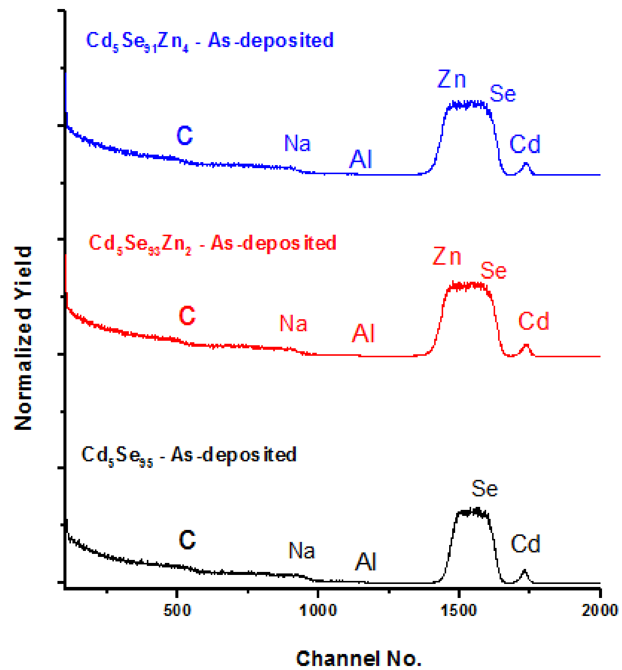


Fig.1 Rutherford's back scattering spectra of as-deposited thin films of $Cd_5Se_{95-x}Zn_x$(x=0, 2, 4) respectively.

3.2 XRD Studies

Regaku X-ray diffractometer was employing for structural analysis of a-$Cd_5Se_{95-x}Zn_x$ (x = 0, 2, 4) before and after gamma irradiation. Cu target was used as a source of X-rays with λ= 1.54056A^0 (Cu$K\alpha_1$). The scanning angle was in the range of 20^0 to 60^0 with scan speed 2^0/min. The absence of any peak in the XRD pattern of Cd_5Se_{95} (Fig not shown) before and after gamma irradiation confirms the amorphous nature of the material. However peak values of as-deposited thin of $Cd_5Se_{93}Zn_2$ and $Cd_5Se_{91}Zn_4$match's well hexagonal phase when compares with the standard diffraction data file card no. 89-2715. Reported study also shows similar type of results[16].

The peak corresponding to plane (100) is very clear and abundant indicates preferential crystallite growth in this particular direction. It was found that the peak intensity increases after 75 kGy dose of gamma irradiation of $Cd_5Se_{95-x}Zn_x$ (x = 2, 4) thin films as shown in Fig.2. The crystallite size of $Cd_5Se_{91}Zn_4$ was calculated by using Scherer's formula [17] i.e., D= $\frac{0.94\lambda}{\beta Cos\theta}$. Where 'D' is the crystallite size, 'λ' is the wavelength of X-ray used, 'β' is the full width at half maximum (FWHM) and 'θ' is the Bragg's angle of reflection. The crystallite size of $Cd_5Se_{91}Zn_4$ increases from 35.30 nm to 44.90 nm after 75KGy gamma irradiation dose. Also the dislocation density 'δ' (length of dislocation lines per unit volume) of $Cd_5Se_{91}Zn_4$ has been calculated by using the relation [18], $\delta = \frac{1}{D^2}$ where 'D' is the crystallite size. The dislocation density decreases from $0.802\times 10^{15}(m^{-2})$ to 0.496×10^{15} (m^{-2}) after 75 kGy gamma irradiation indicates the disorder of the films decreases.

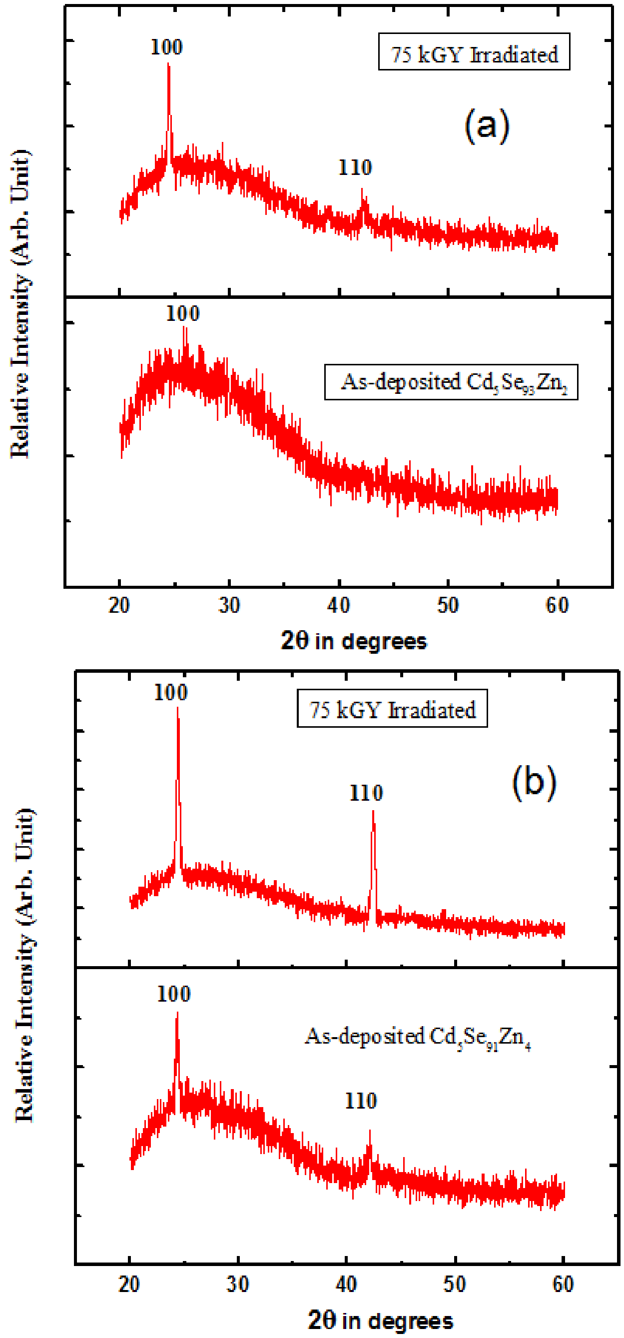


Fig.2 (a, b) Shows XRD spectra of as-deposited and 75 kGy gamma irradiated thin films of $Cd_5Se_{95-x}Zn_x$ (x=2, 4) respectively.

3.3 PL Study

The micro-structural change occurs due to gamma irradiation are identified by the peaks and bands in the PL spectroscopy. The defect population, band edge transition, crystalline quality etc. can be determined by the PL The PL spectra of as-deposited and gamma irradiated thin films of $Cd_5Se_{95-x}Zn_x$ (x = 0, 2, 4) has been studied in the wavelength range 350 nm–750 nm as shown in Fig.3.

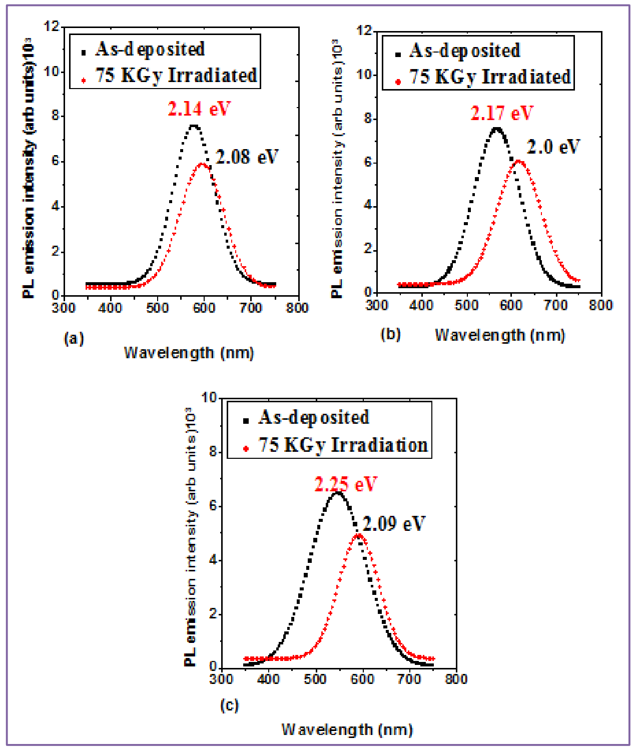


Fig.3 (a, b, c) PL spectra of as-deposited and 75 kGy gamma irradiated thin films of $Cd_5Se_{95-x}Zn_x$ (x = 0, 2, 4) with excitation wavelength 420 nm.

The yellow emission centered around 2.14 eV, 2.17 eV and 2.25 eV are being identified as the luminescence due to as-deposited thin films of $Cd_5Se_{95-x}Zn_x$ (x = 0, 2, 4) respectively [19]. The area under the peak will be proportional to the defect density present in the sample. Fig.3 shows the reduction of the peak intensity (reduction of the area under peak) after 75 kGy gamma irradiation in all cases of $Cd_5Se_{95-x}Zn_x$ (x = 0, 2, 4) indicates the concentration of defects are reduced due defect annealing. Here also this type of change is found more in case of Zn doped samples. Also the shift of the peak observed in PL spectra verifies the reduction of optical band gap from 2.14 eV–2.08 eV, 2.17 eV-2.00 eV and 2.25 eV–2.09 eV after 75 kGy gamma irradiation of $Cd_5Se_{95-x}Zn_x$ (x = 0, 2, 4) respectively. Thus PL spectra of investigated thin films show good agreement with results obtained from UV-vis-spectrophotometry Study (discussed below).

3.4 FT-IR

FT-IR spectroscopy is a powerful and non-destructive technique which gives us various information about quality or consistency of a sample and amount of components in a mixture. The present FT-IR spectrum matches with library spectrum of Silicon dioxide by using eFT-IR software.

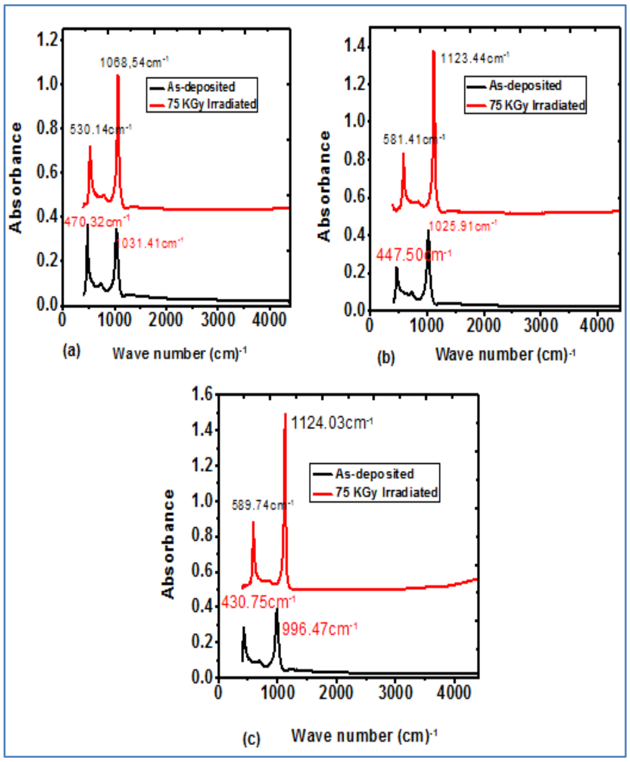


Fig.4 (a, b, c) FT-IR spectra of as-deposited and 75 kGy gamma irradiated thin films of $Cd_5Se_{95-x}Zn_x$(x=0, 2, 4) respectively.

Fig.4 shows FT-IR spectrum of as-deposited and 75 kGy gamma irradiated thin films of $Cd_5Se_{95-x}Zn_x$ (x = 0, 2, 4) respectively in the wave number range 400 cm^{-1}-4400 cm^{-1}(mid-infrared range).The peaks at 470.32 cm^{-1} and 1031.41 cm^{-1} of as-deposited $Cd_5Se_{95-x}Zn_x$ (x = 0) are the Se-Cd-Se bending vibrations and strong asymmetric vibrations when compares with the Sio_2 spectrum [20]. As-deposited $Cd_5Se_{95-x}Zn_x$ (x = 2) show the peaks at 447.50 $cm^{-1}$and 1025.91 cm^{-1} while that of as- deposited $Cd_5Se_{95-x}Zn_x$ (x = 4) show at 430.75 $cm^{-1}$and 996.47 cm^{-1} are the corresponding peaks of Cd-Se-Zn bending and asymmetric vibrations respectively [20]. Also it has been observed that the peak intensity increases and the corresponding peak shifts towards higher wavenumber side i.e., 530.14 cm^{-1} and 1068.54 cm^{-1} for $Cd_5Se_{95-x}Zn_x$ (x = 0), 581.41 cm^{-1} and 1123.44 cm^{-1} for $Cd_5Se_{95-x}Zn_x$ (x = 2), 589.74 cm^{-1} and 1124.03 cm^{-1} for $Cd_5Se_{95-x}Zn_x$ (x = 4) after 75 kGy dose of gamma irradiation in other words we can say that the bond strength increases after 75 kGy dose of gamma irradiation. According to Beer's law the peak intensity or the area under peak is proportional to the atomic concentration in absorption mode of FT-IR spectrum [22]. Thus the atomic concentration increases after irradiation for all cases of $Cd_5Se_{95-x}Zn_x$ (x = 0, 2, 4). That is why the XRD shows increase in particle size film after irradiation because the atoms come close to each other in other words we will say annealing effect is produced due to gamma ray irradiation and favors the increase in crystallinity of the investigated thin films. This type change is found much more in case of higher Zn doped CdSe thin films. This may be due to addition of Zn, the bonding arrangement changes.

3.5 Optical study

The optical absorption spectrum is one of the most important tools for developing the energy band diagram. Fig. 5(a, b, c) shows an absorption spectra of pristine and thin films of $Cd_5Se_{95-x}Zn_x$ (x = 0, 2, 4) in the wavelength range 190-1100 nm.

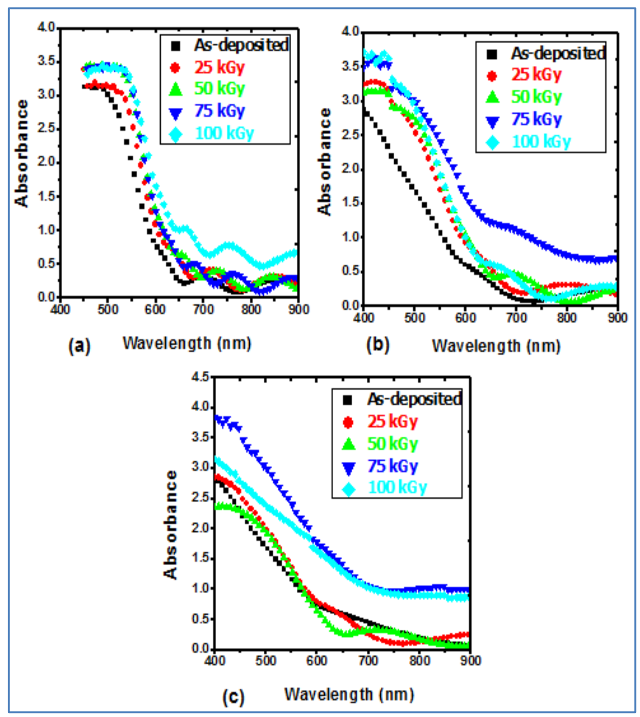
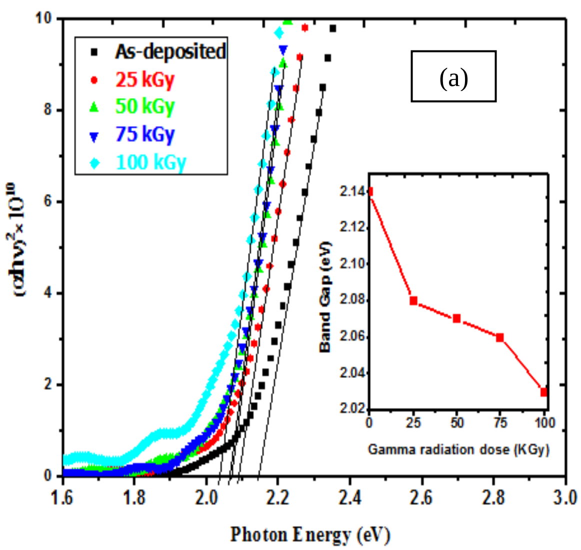


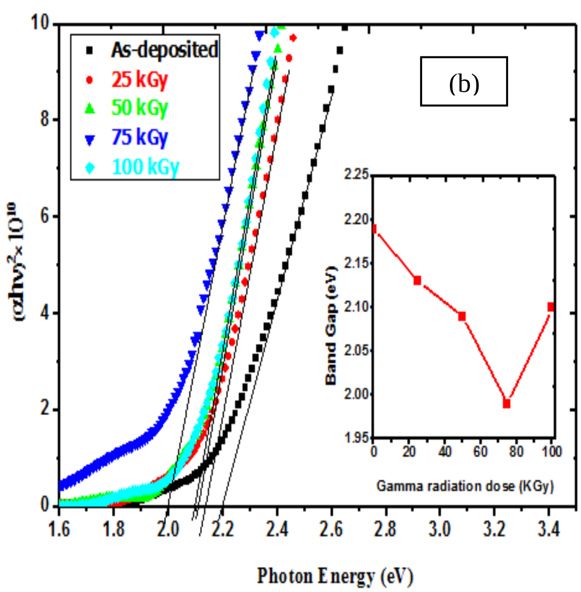
Fig.5 (a, b, c) the variation of absorbance with wavelength of as-deposited and gamma irradiated thin films of $Cd_5Se_{95-x}Zn_x$ (x=0, 2, 4) respectively.

The absorption coefficient has been calculated on the basis of experimentally recorded absorption spectra by employing the following equation [23, 24].

$$\alpha = \frac{A}{d} \quad (1)$$

Where 'A' is the absorbance and 'd' is the thickness of the film. The spectral dependence of absorption coefficient shows the value of absorption coefficient increases linearly with increase in photon energy for the a-$Cd_5Se_{95-x}Zn_x$ (x = 0, 2, 4) thin films. Due to gamma irradiation, the bond breaking and bond rearrangement taking place and constituently there is a change in the local structures of a-$Cd_5Se_{95-x}Zn_x$ (x = 0, 2, 4) thin films. This leads to the various subtle effects such as shift in the absorption edge and more substantial atomic and molecular reconfigurations which are associated with these changes in absorption coefficient. The value of absorption co-efficient increases up to 75 kGy gamma irradiation dose and decreases after 100 kGy gamma irradiation in all cases of $Cd_5Se_{95-x}Zn_x$ (x = 0, 2, 4) as shown in Table.1. The calculated value are of absorption coefficient are of the order of ~ 10^4 cm^{-1} comparable with the reported study.





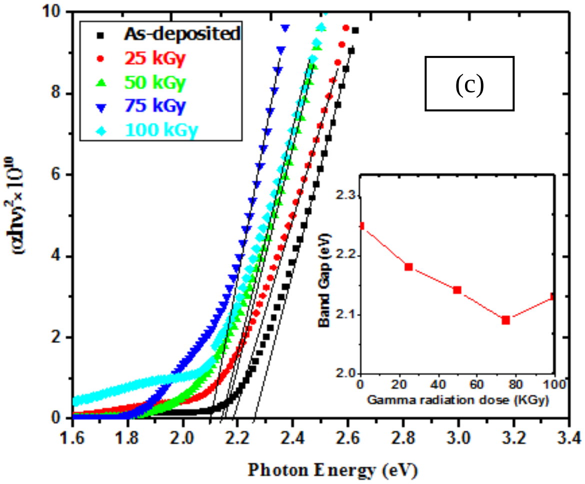


Fig.6 (a, b, c) Shows the variation of $(\alpha h\nu)^2$ with photon energy of as-deposited and gamma irradiated thin films of $Cd_5Se_{95-x}Zn_x$ (x=0, 2, 4) respectively.

Inserts show variation of optical band gap with Irradiation dose, inset shows variation of band gap with irradiation dose.

The absorption coefficient in the high absorption region ($\alpha \geq 10^4 cm^{-1}$) of amorphous semiconductors follows an exponential law according to Tauc [25]

$$\alpha h\nu = A\,(h\nu - E_g)^m \qquad (2)$$

Where 'A' is a constant and 'E_g' is the optical band gap of the semiconductor material. The value of 'm' decides the type of transition, where m = 1/2, 2, 3, 3/2 for direct allowed, indirect allowed, indirect forbidden, and direct forbidden transitions respectively. The present system obeys the rule of direct transition (i.e., m = 1/2) similar as per the reported study [19, 20]. The value of optical band gap was determined by plotting the graph between $(\alpha h\nu)^2$ and photon energy with the linear portion of the curve extrapolated to the x-axis. It has been found that the value of optical band gap increases due to addition of Zn as shown in Table.1. This may be due to the mixing of higher band gap material ZnSe with smaller band gap material CdSe [26]. Also this decrease of optical band gap may be due to the change in average bond energy because optical band gap is a bond sensitive property. Also it was found that the value of optical band gap decreases from 2.14 eV–2.03 eV for $Cd_5Se_{95-x}Zn_x$ (x = 0 i.e., Undoped Zn) after gamma irradiation as shown in Fig.6. For $Cd_5Se_{95-x}Zn_x$ (x = 2, 4) the optical band gap decreases from 2.19 eV–1.99 eV and 2.25 eV–2.09 eV respectively after gamma irradiation at 75 kGy dose and at 100 kGy, the value of optical band increases again to 2.10 eV and 2.13 eV for $Cd_5Se_{95-x}Zn_x$ (x = 2, 4) respectively as shown in Fig.6. The decrease of optical band gap may be due to the increase of crystallite size after gamma irradiation. However when gamma radiation interacts with the material, two processes occurs at the same time i.e., defect creation and defect annihilation. At higher dose of gamma irradiation the number of defects created becomes more as compared to the number of defects annihilated. Also this change in optical band gap of $Cd_5Se_{95-x}Zn_x$ (x = 2, 4) due to gamma irradiation were also supported by XRD, PL and FT-IR characterization techniques discussed early in this study. The extinction co-efficient of the as-deposited and laser irradiated thin films were calculated by $K = \frac{\alpha\lambda}{4\pi}$, it was found that the value of extinction coefficient increases up to 75 kGy gamma irradiation and decreases at 100 kGy gamma irradiation in all cases of $Cd_5Se_{95-x}Zn_x$ (x = 2, 4)as shown in Fig.7.

The absorption coefficient near the band edge depends exponentially on the photon energy and hence obeys the Urbach's empirical formula [27].

$$\alpha_\nu = \alpha_0 e^{h\nu/E_u} \quad (3)$$

Where α_0 is the characteristic parameter of the material, 'hν' is the photon energy and E_u is the width of the exponential tail known as Urbach's tail energy. The slope of the plot of ($\ln\alpha$) versus (hν) below the fundamental edge gives the Urbach's tail width. Fig.8 shows the Urbach's plot of as deposited and gamma irradiated thin films of $Cd_5Se_{95-x}Zn_x$ (x = 0, 2, 4). It was found that the value of Urbach's energy decreases from 0.438 eV to 0.336 eV, 0.424 eV to 0.309 eV and 0.422 eV to 0.181 eV after 75 kGy gamma irradiation for thin films of$Cd_5Se_{95-x}Zn_x$ (x = 0, 2, 4) respectively indicates the dense defect states are reduced in the forbidden gap due to increases in crystallinity. At higher irradiation dose (100 kGy) the value of Urbach's energy increases in case of Zn doped CdSe samples as shown in Table.1. Indicates that at higher doses of gamma-ray irradiation the number of defects created becomes more as compared to the number of defects annihilated.

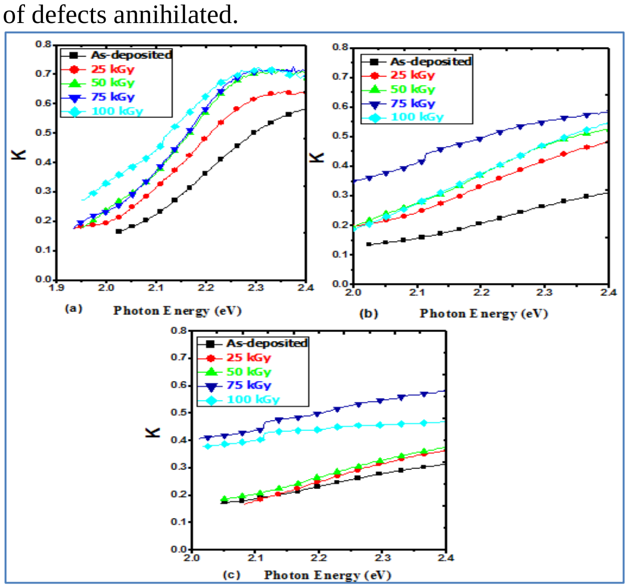


Fig.7 (a, b, c) variation of extinction coefficient (K) with photon energy of as-deposited and gamma irradiated thin films of $Cd_5Se_{95-x}Zn_x$(x=0, 2, 4) respectively.

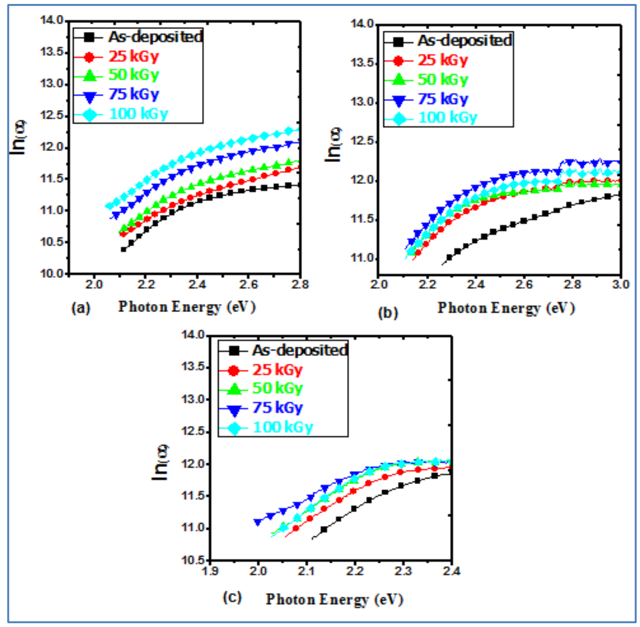


Fig.8 (a, b, c) Shows the variation of ln(α) with photon energy of as-deposited and gamma irradiated thin films of $Cd_5Se_{95-x}Zn_x$(x=0, 2, 4) respectively.

Table.1. Direct band gap (E_g), absorption coefficient (α) and extinction coefficient (K) for as-deposited and gamma irradiated thin films of $Cd_5Se_{95-x}Zn_x$ (x= 0, 2, 4) respectively at 700 nm.

Sample		E_g(eV) ±0.001	$\alpha\times10^4 cm^{-1}$ ±0.3	K	E_u(eV)
Cd_5Se_{95}	As-deposited	2.14	8.028	0.439	0.438
	25 KGY	2.08	10.352	0.566	0.420
	50 KGY	2.07	12.108	0.662	0.341
	75 KGY	2.06	12.368	0.677	0.336
	100 KGY	2.03	12.612	0.690	0.331
$Cd_5Se_{93}Zn_2$	As-deposited	2.19	4.336	0.237	0.424
	25 KGY	2.13	6.908	0.378	0.334
	50 KGY	2.09	7.752	0.424	0.348
	75 KGY	1.99	9.668	0.529	0.309
	100 KGY	2.10	7.780	0.425	0.348
$Cd_5Se_{91}Zn_4$	As-deposited	2.25	4.688	0.256	0.422
	25 KGY	2.18	5.46	0.284	0.207
	50 KGY	2.14	5.192	0.298	0.182
	75 KGY	2.09	9.652	0.528	0.181
	100 KGY	2.13	8.296	0.454	0.263

4. Conclusions

In this study, the systematic analysis of structural and optical properties of a-$Cd_5Se_{95-x}Zn_x$ (x = 0, 2, 4) thin films before and after gamma irradiation was carried out and the following conclusions are drawn:

It has been observed that the values of absorption coefficient (α) are of the order of $10^4 cm^{-1}$ before and after gamma irradiation, and therefore such materials may have an industrial application as optical recording media.

Present study shows the ternary alloy of CdSeZn thin films may be used as beam splitter in FT-IR spectrometer because it absorbs light strongly near mid infra-red range.

The optical band gap of ternary alloy of amorphous CdSeZn thin films allows the rule of direct transition. This change may cause a shift in the Fermi level leading to the reduction in optical band gap from 2.14 eV–2.06 eV, 2.19 eV–1.99 eV and 2.25 eV–2.09 eV of thin films of $Cd_5Se_{95-x}Zn_x$ (x = 0, 2, 4) respectively after 75 kGy gamma irradiation.

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