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Effect of laser and visible light Irradiation on structural and optical properties of thin films of amorphous selenium and selenium mercury (80:20 composition)

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ABSTRACT

This study is carried out the effect of laser and solar light irradiation on the structural and optical properties of amorphous Selenium and selenium mercury (80:20 composition). Bulk samples were prepared by thermal quenching technique and thin films were papered by thermal evaporation technique on glass substrates. Thin films of a-Se and a-SeHg were irradiated by diode laser (wavelength=405nm, power=100mW) and solar light (energy ~ 0.3MeV calculated by E =hc/λ) for different durations of time. XRD analysis of a-Se and a-SeHg show increase of peak intensity and crystallite size after irradiation indicates the enhancement of crystallinity. Raman analysis of a-Se and a-SeHg also favors this improvement of crystallinity after laser and solar light irradiation. The UV analysis shows the value of optical band gap of a-Se and a-SeHg decreases after irradiation (both laser and solar light). Also it was found that the value of Urbach's energy decreases after laser and solar light irradiation indicates that the disorder has been reduced after irradiation. The other optical parameters like extinction coefficient and absorption coefficient changes accordingly discussed in this study. It was also found that the change of structural and optical parameters is much more in case of solar light irradiation.

Keywords: Thin films of amorphous selenium and selenium mercury; solar and laser irradiation; structural properties; optical properties.

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Introduction

S, Se, and Te based chalcogenide semiconductors have many unique optical properties, they can be used for a wide variety of application **[1]**. These are promising materials for the use of like solar cells, antireflection coating, optical limiting, and manufacture of filters, infrared power delivery, IR emitter, optical rewritable data, IR detector, gratings and optical recording media **[2-8]**. Over many decades Group II–IV materials as possible components in

advanced opto-electronic devices. Amorphous Selenium is one of the promising material for photoconductors used in high-definition television (HDTV) **[9]**, digital radiography (DDR) **[10]**. This is because of low thermal noise, high spatial resolution and high sensitivity against wide variety of wavelengths from visible to ultraviolet **[11]** as well as x-rays **[12]** as compared to silicon based photoconductors. The efficiency of photoconductors will depend on the absorption of incident x-ray photons. In order to obtain maximum absorption coefficient one can increase the thickness of the amorphous selenium layer but the problem is that large voltage is required to capture the electrons. Typically 1000 μm requires 10,000 volts **[13]**. Thus optical properties like extinction coefficient (k), absorption coefficient (α), optical band gap (E_g) and urbach's energy E_u are important parameters and can be utilized from absorption spectra, transmission spectra and reflection spectra. Large number of research groups throughout the world are trying to change the material properties (optical, structural, electrical etc.) for enhancing the device performance by using different techniques such as different deposition techniques, irradiation techniques (laser irradiation, swift heavy ion irradiation, gamma-ray irradiation etc), doping techniques etc. In this study the effect of laser and solar light irradiation on the optical, electrical and structural properties of a-Se and a-SeHg are presented. Optical absorption spectra provide essential information about the optical band gap band structure of semiconductor materials. Optical analysis of the present case was carried out by UV-visible photo spectrometer and it was found that the optical band gap of present system was decreased due to laser light and solar light irradiation and the other optical parameters changes accordingly. Raman analysis gives information about microscopic disorder, molecular vibration and residual stress in the material. XRD pattern of both laser and solar light irradiated thin films of a-Se and a-SeHg (80:20 compositions) shows the grain size increases confirms the crystallinity of the material increases. Further the decrease of urbach's energy and dislocation density supports this improvement in crystallinity. Thus the important thing is that this study shows the value of absorption coefficient increases after laser and solar light irradiation without changing the thickness. Also it was found that this change of absorption coefficient is found much more in mercury doped selenium. Thus the photoconductor based on solar or laser irradiated SeHg alloy may be more efficient than amorphous selenium based photoconductors. It is clear from these optical parameters that has been changed after irradiation, whenever an electromagnetic radiation interacts with the material, it may interacts with the electrons of an atom, may accelerate them and hence in this way energy is absorbed by these electrons. This energy is transferred to the lattice through electron-phonon interaction. On the other hand, in semiconductors the absorption of photons leads to the creation of electron–hole pairs with a certain amount of kinetic energy. These hot carriers thermalize amongst each other forming plasma. Once a common carrier temperature is reached, they transfer their kinetic energy to the lattice via recombination and phonon generation. In both these ways the lattice vibration increases cause heating and melting. This leads to the crystallization of the material. In this way the structure of the sample has been changed and hence the optical properties are modified and hence enhance the industrial optoelectronic applications of chalcogenide semiconductors.

Table 1. Shows the structural parameters of thin films of a-Se and a-SeHg (80:20 composition) before and after irradiation (laser and solar light).

Sample	Irradiation time	2θ	hkl	Grain size, D(nm)	Strain(ε) 10^{-4}lin^{-2}m^{-4}	Dislocation density $\delta \times 10^{15}$(m^{-2})
a-Se	As-deposited	23.42^0	022	22.619	16.005	1.954
	14min laser irradiated	23.43^0	022	30.089	12.104	1.104
	1min solar irradiated	23.45^0	022	34.804	10.402	0.825
a-SeHg	As-deposited	25.16^0	111	25.444	14.223	1.541
	14min laser irradiated	25.40^0	111	28.171	12.853	1.261
	1min solar irradiated	25.39^0	111	47.172	7.671	0.440

Table 2. Represents Indirect band gap (E_g), absorption coefficient (α), extinction coefficient (K), Urbach energy (E_u) and transmittance (T %) for the thin film system of a-Se and a-SeHg (80:20 composition) at 675 nm before and after laser irradiation.

Sample	Irradiation time	E_g(eV)	$\alpha * 10^4 (cm^{-1})$	K	E_u(eV)	T %
a-Se	As-deposited	1.77	0.935	0.050	0.221	52
	2min	1.75	1.928	0.103	0.217	26
	6min	1.74	1.905	0.102	0.216	26
	14min	1.72	1.740	0.093	0.214	30
	1min solar	1.56	2.858	0.153	0.209	26
a-SeHg	As-deposited	1.81	0.658	0.035	0.261	63
	2min	1.75	1.092	0.058	0.228	47
	6min	1.71	2.515	0.135	0.237	45
	14min	1.67	1.142	0.061	0.262	17
	1min solar	1.56	3.021	0.199	0.294	7

Experimental

The samples of a-Se and a-SeHg were prepared by melt quenching method. First of all the composition of selenium of and mercury having purity 99.999% has been taken according to their atomic percentage. The mixtures were put into different quartz ampoules, in which the pressure of the ampoules were kept at 10^{-5}torr and then sealed. These ampoules were kept in the furnace at a temperature of 1000K for 10h. For achieving homogeneous mixture, these ampoules have been continuously rotated in furnace. After 10h these ampoules are taken out from the furnace and quenched in ice cooled water. Thin films of a-Se and a-SeHg were deposited on glass substrates using thermal evaporation technique at room temperature under a vacuum of 10^{-5} torr, achieved through a molybdenum boat. The glass slides was cleaned by ultrasonic bath and then by acetone. The thickness of these films was measured to be 300nm. These films were irradiated with a pulsed diode laser (wavelength 405 nm and power 100mW) and by solar light through magnifying glass for different durations of time. The flux of the solar irradiation was measured to be 150,000 by using digital luxmeter (Model: MS6610). The energy of the focusing beam was ~ 0.3 MeV calculated by using the well-known relation E = hc/λ, where 'h' is the plank's constant, 'c' is the velocity of light and 'λ' is the wavelength (In general the wavelength was taken as 0.5μm) of the visible light and temperature was measured to be 100

°C by thermometer. Raman spectra of as-deposited and laser irradiated thin films ofa- Se and a- SeHg was recorded at room temperature using MiniRaman (BTR111 MiniRaman and diode laser of wavelength 785nm) in the Raman shift range 0-650(cm^{-1}) and laser power 80mW. For structural study, XRD measurements were carried out by using X-ray diffractometer (Burker: D8 ADVANCE Cu$K\alpha$ as X-ray source of wavelength 1.54056A^0), 2θ spectrum from 15^0 to 60^0 was recorded for all thin films of a-Se and a-SeHg. Also the scanning speed was 0.50/min. The characterization of optical properties of these films before and after irradiation have been done by using VU-spectrophotometer (Model: Comspec M550) in the wavelength range 200-1100nm.

Results and discussion

Raman studies

Raman spectra of as deposited and irradiated (laser and solar) thin films of **a**-Se and a-SeHg as shown in **Fig. 1** (a, b). The peak at 231.29 $cm^{-1}$is attributed to the LO phonons of Se **[14]** and the peak at 180.33 cm^{-1} is attributed to the LO phonons of SeHg **[15]** as shown in Fig. 1 (a, b). The peak intensity of a-Se, a-SeHg increases and the full width at half maximum decreases after irradiation (laser as well as solar) indicates the improvement in crystallinity of the material. The observed small asymmetry in the line shape of the LO phonon on lower wave number side indicates the presence of defects and disorder in the crystal structure **[16]**. This asymmetry is reduced due to irradiation (laser and solar) for both a-Se, a-SeHg also confirms the disorder has been reduced in the material.

XRD studies

Fig. 2 (**a** and **b**) shows the x-ray diffraction pattern of a-Se and a-SeHg as deposited, laser irradiated and solar light irradiation thin films respectively. The peaks were indexed according to JCPDS files. All a-Se (as deposited, laser irradiated and solar irradiated) thin films show only one intense reflection peak at 2θ= 23.42^0, 23.43^0, 23.45^0corresponding to (022) plane. The XRD pattern of a-Se thin films is identical to the monoclinic phase with unit cell parameter a = 8.07A^0, b = 9.31A^0, c=12.85A^0 and with space group F21/n **[17]**. Similarly a-SeHg (as deposited, laser irradiated and solar irradiated) also show one intense peak at 2θ= 25.16^0, 25.40^0, 25.39^0corresponding to (111) plane. The XRD pattern of a-SeHg thin films is identical to the cubic phase with unit cell parameter a = 6.084A^0 with zinc blende structure and with space group F43m **[18]**. The peak corresponding to planes (022) and (111) is very clear and abundant for all thin films indicate preferential crystallite growth in this particular direction. It is clear from the XRD pattern of as deposited, laser irradiated and solar light irradiated a-Se and a-SeHg thin films show there is increase in peak height and increase in grain size as shown in **Table 1** due to laser and solar light irradiation indicates the improvement in crystallinity of the material. Also the decrease of dislocation density and urbach's energy supports this increase in crystallinity. The change of structural parameters was found much more in case of solar light irradiation and Hg doped selenium. This may be due to the addition of Hg the bonding arrangement of the material changes.

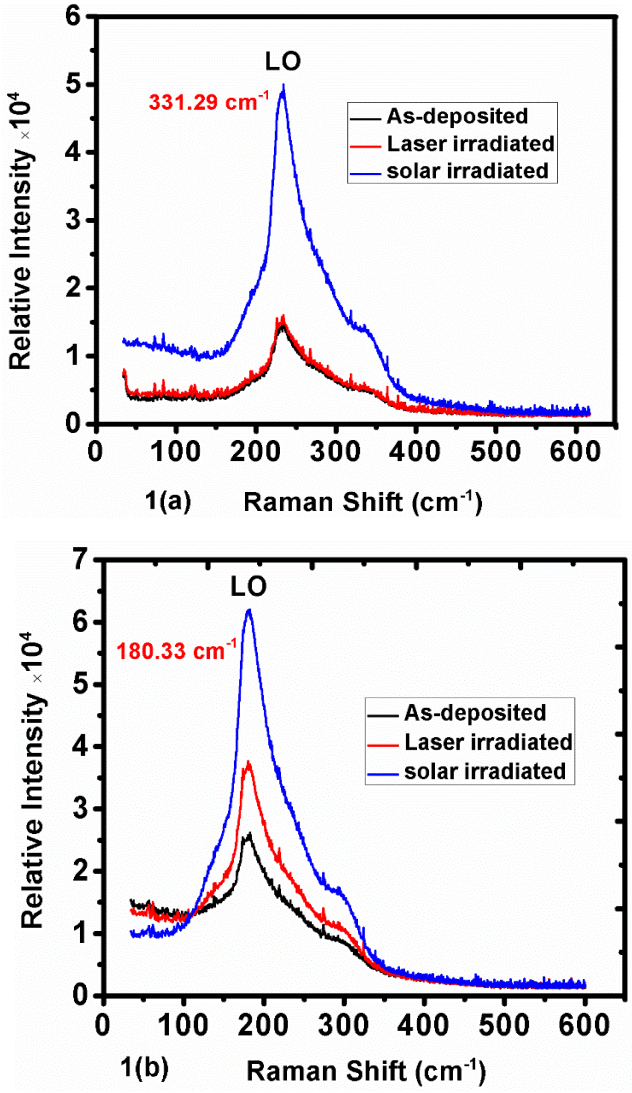


Fig. 1 shows the Raman spectra of thin films of (a) a-Se, (b) a-SeHg before and after laser solar irradiation.

The crystallite size of a-Se and a-SeHg thin films was calculated by using Scherrer's formula **[19]**.

$$D = 0.94\lambda / \beta\cos\theta \quad \text{---------- (1)}$$

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. From **Table 1**, it was found that the crystallite size for a-Se and a-SeHg cubic (100) plane is increased from 22.619 nm to 30.089 nm and 36.202 nm to 37.696 nm respectively due to laser irradiation and for solar light irradiation it is from 22.619 nm to 34.804nm and 36.202 nm to 53.259 nm results that the phase transition occurs in the material from amorphous to crystalline state.

The lattice spacing'd' has been calculated by the use of Bragg's formula

$$d = n\lambda / 2\sin\theta \quad \text{---------- (2)}$$

where 'λ' is the wavelength of the x-rays. The experimental value of d is approximately equal to the standard value. Similar trend was also shown **[20]** by laser irradiation.

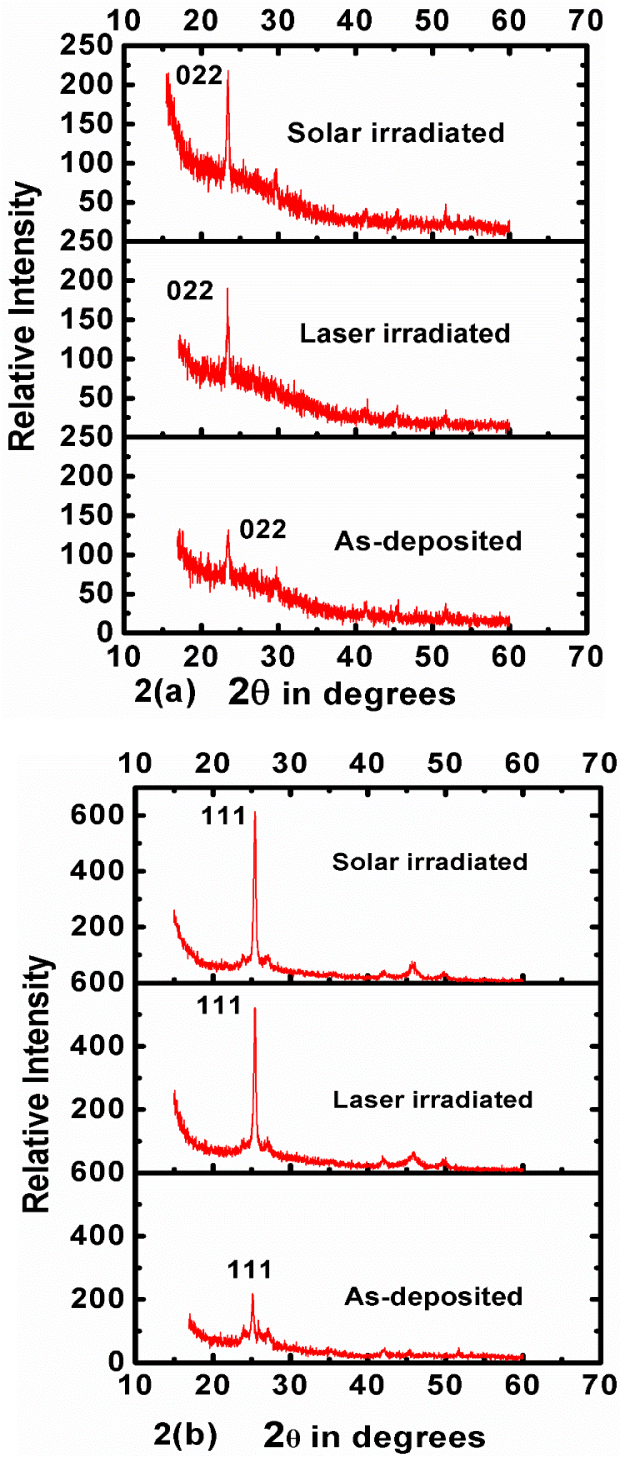


Fig. 2 shows the XRD spectra of thin films of (a) a-Se,(b) a-SeHg before and after laser solar irradiation.

The lattice parameter 'c' is determined for the hexagonal structure by the following expression:

$$1/d = (h^2+k^2+l^2)/c^2 \quad \text{---------- (3)}$$

where h, k and l represents the lattice planes. It has been found that the experimental value of lattice parameter 'c' nearly matches with the standard value.

The strain 'Є' value was evaluated by using the following relation:

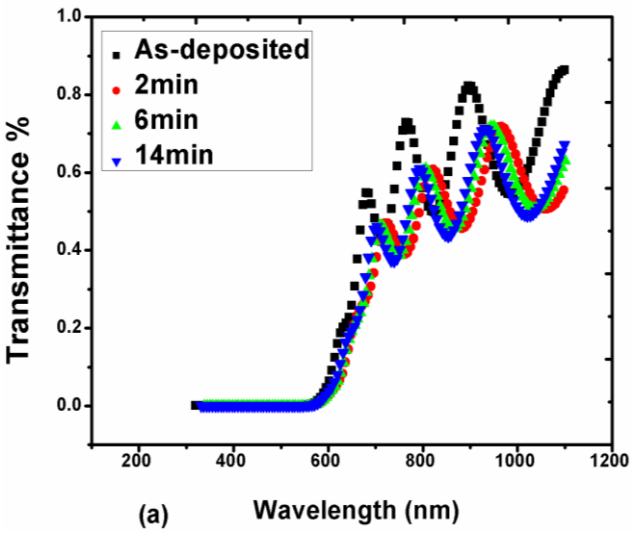
$$Є = \beta\cos\theta/4 \quad \text{---------- (4)}$$

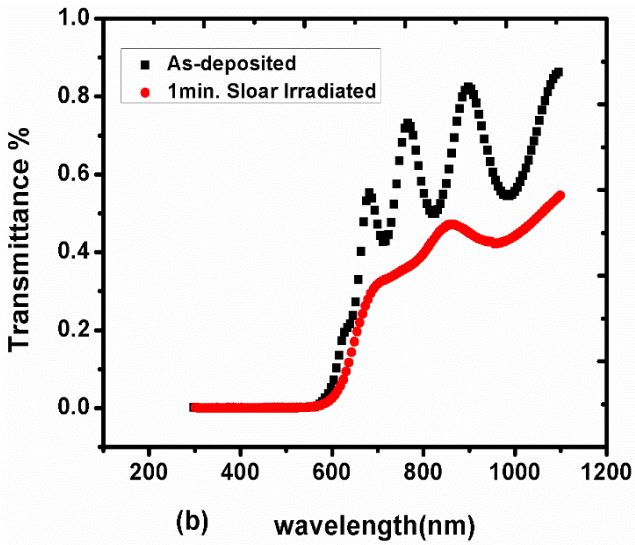
The dislocation density 'δ' (length of dislocation lines per unit volume) has been calculated by using the relation **[21]**.

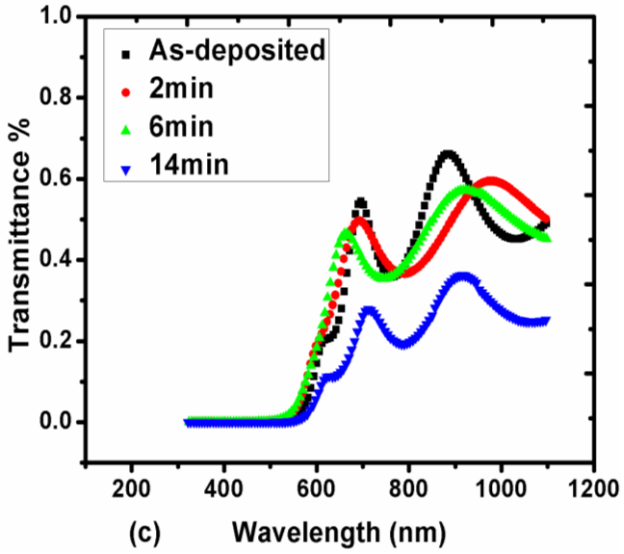
$$\delta = 1/D^2 \quad \text{---------- (5)}$$

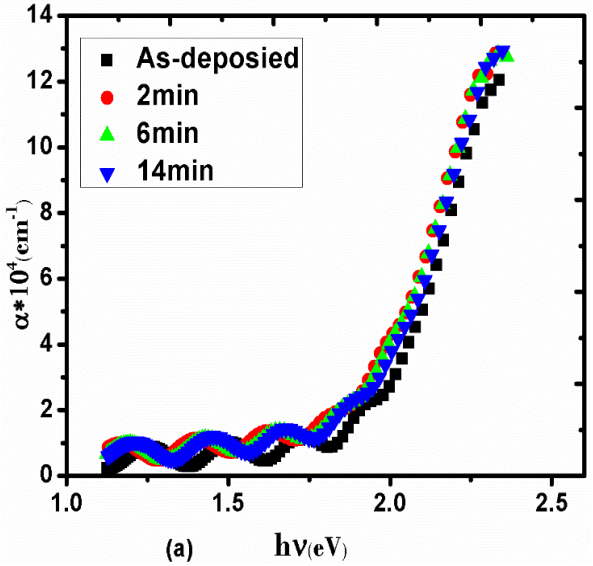
where 'D' is the crystallite size.

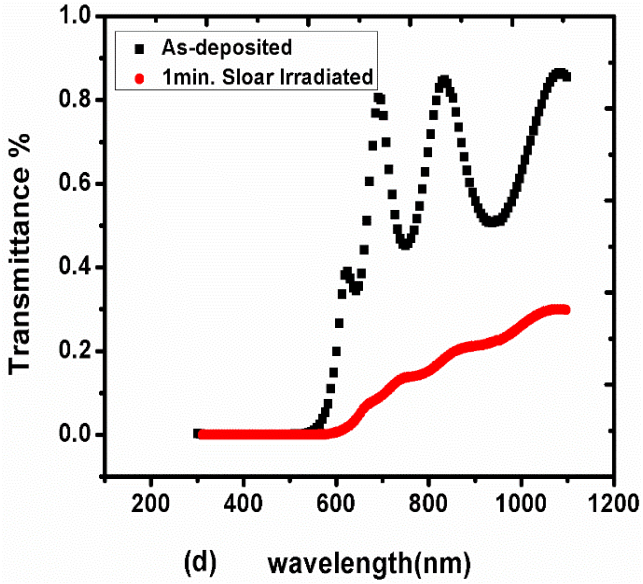
The positive value of residual strain for both laser and solar light irradiated films of a-Se and a-SeHg indicates a tensile strain. This may be due to the difference of thermal expansion co-efficient of the substrate and deposited material or crystallization process during irradiation.

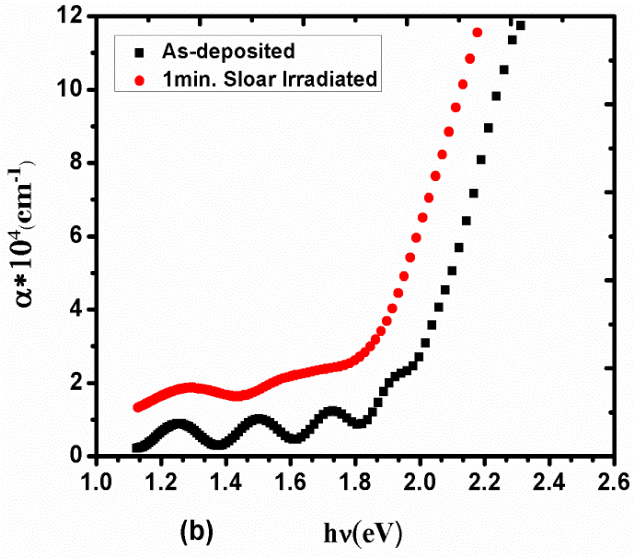












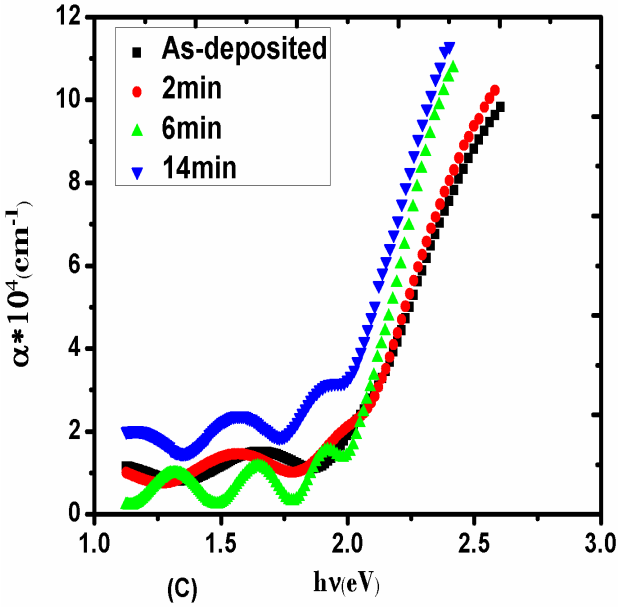


Fig. 3. Shows the variation of transmission with wavelength before and after irradiation of thin films of a-Se (a) laser irradiation, (b) Solar irradiation and a-SeHg (c) laser irradiation, (d) solar irradiation.

Optical studies

The optical properties of thin films of a-Se and a-SeHg has been characterized by UV-spectrophotometer (200-1100nm). **Fig. 3** (**a**, **b**, **c** and **d**) shows the Transmittance spectra of the thin films of a-Se and a-SeHg before and after irradiation (Laser as well as solar). The value of transmittance decreases after irradiation at a wavelength of 675 nm as shown in **Table 2**. The oscillatory behavior of transmission spectra of the all thin film of a-Se and a-SeHg is due to interference of the reflected wave fronts from the two surfaces of the thin films **[22]**. The decrease of transmittance is much more in case of solar irradiation as shown in **Fig. 3**. This may be due to presence of large number of multi-wavelength photons falls in a small space will produce large amount heat inside a system which results the enhancement of crystallinity of the material.

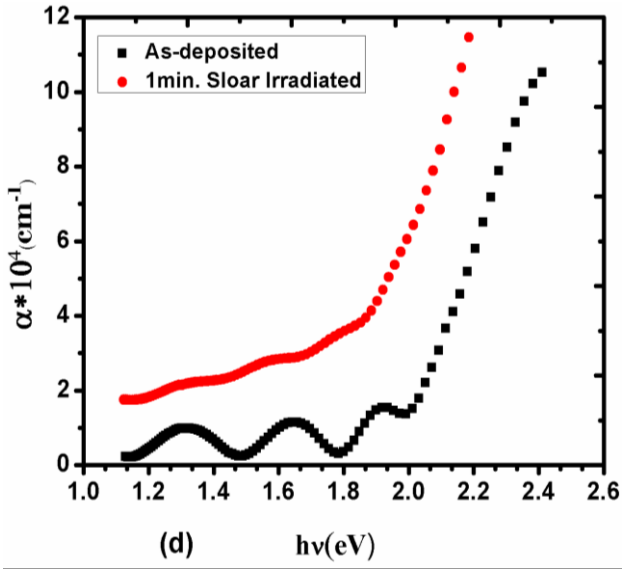


Fig. 4 shows the variation of absorption coefficient with photon energy before and after irradiation of thin films of a-Se (a) laser irradiation, (b) Solar irradiation and a-SeHg (c) laser irradiation, (d) solar irradiation.

The absorption co-efficient 'α' of the thin films of a-Se and a-SeHg has been calculated by using the relation **[23, 24]**.

$$\alpha = A / d \qquad \text{----------} \quad (6)$$

where 'A' is the absorbance and 'd' is the thickness of the film. **Fig. 2** (**a**, **b** and **c**) shows the variation of absorption co-efficient with photon energy of thin films of a-Se and a-SeHg before and after laser and solar light irradiation. It is clear from **Fig. 3** (**a**, **b**) and **Fig. 4** (**a**, **b**), the absorption co-efficient increases after irradiation in both cases given in **Table 2**. Similar results were also obtained by different workers **[20, 24]** by laser irradiation. The increase of absorption (or shift) on the shorter wavelength or higher energy side indicates that the concentration of defects decreases due to laser and solar light irradiation.

The absorption coefficient near the band edge depends exponentially on the photon energy and hence obeys the Urbach's empirical formula **[25]**.

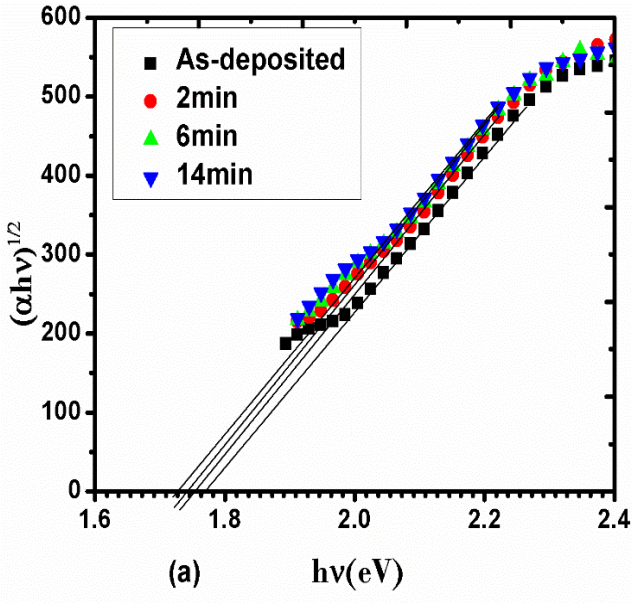
$$\alpha_{\upsilon} = \alpha_{\upsilon} e^{h\nu/Eg} \qquad \text{----------} \quad (7)$$

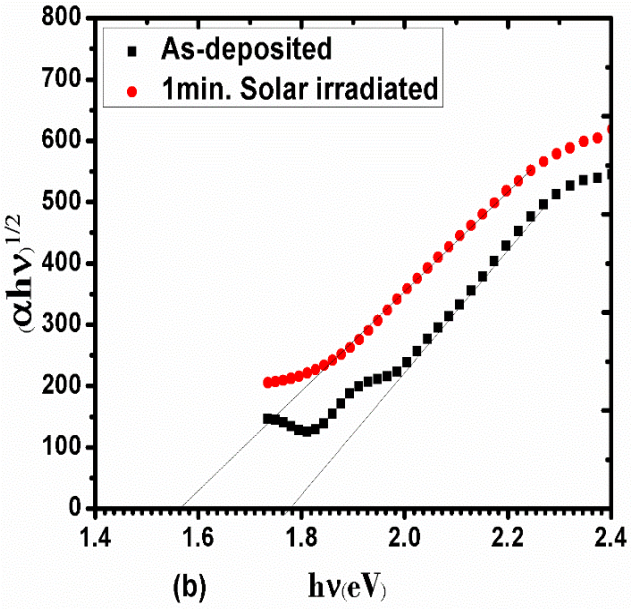
where 'α' is a constant, 'h' is the plank's constant and 'E_u'is the energy width of the band tails of the localized states. In case of amorphous semiconductors the Urbach's energy represents the degree of disorder. The exponential behavior of the absorption edge represents the value ofUrbach's energy of the thin films of a-Se and a-SeHg before and after irradiation. The value of Urbach's energy decreases due to irradiation indicates that the defects are decreased upon laser and solar light irradiation.

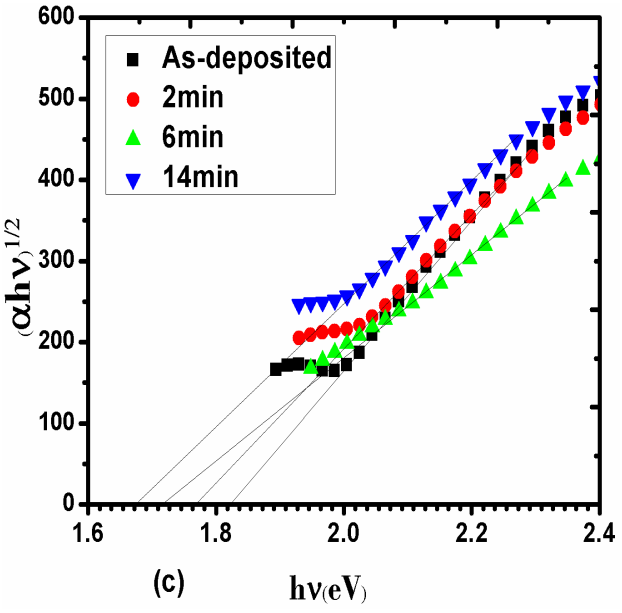
The optical band gap of thin films of a- Se and a- SeHg were determined with the help of absorption spectra. According to the well-known relation of Tauc **[26]**.

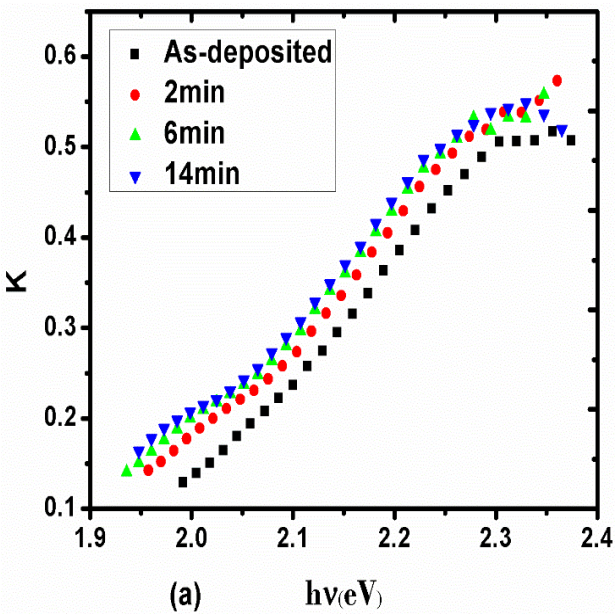
$$\alpha h\nu = A (h\nu - E_g)^m \qquad \text{----------} \quad (8)$$

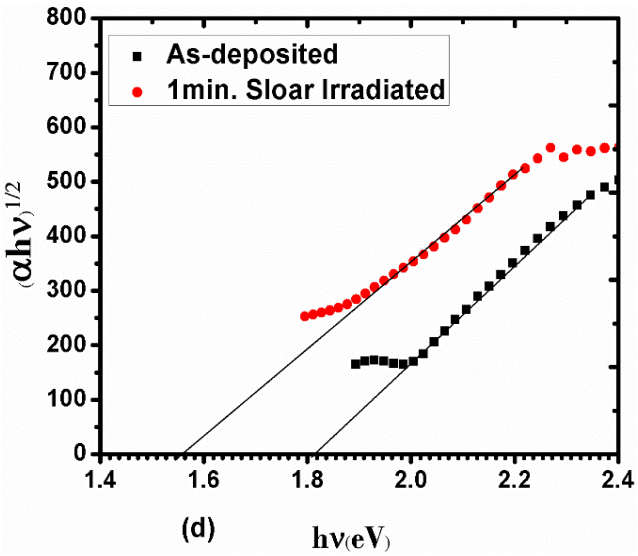
where 'A' is a constant and 'E_g' is the band gap of the semiconductor material. The value of 'm' decides the type of transition, where m = 1/2, 2, 3, 3/2 for direct allowed, indirect allowed, indirect forbidden, and direct forbidden transitions respectively. The present system obeys the rule of indirect transition (i.e., m = 2). **Fig. 5** (**a**, **b**, **c**, **d**) shows the variation of $(\alpha h\nu)^{1/2}$ with photon energy of the thin film system of a-Se and a-SeHg. The intercept on the x-axis determines the value of indirect band gap. From table (II), it has been found that the value of optical band gap is decreased on irradiation. This reduction of optical bang gap can explained on the basis of Mott and Davis **[27]** model of density of states. Laser and solar irradiation gives sufficient energy to system a-Se and a-SeHg in the form of heat reduces the number of defect states in the material that results the decrease in optical band gap and there may be transition from amorphous to crystalline state.











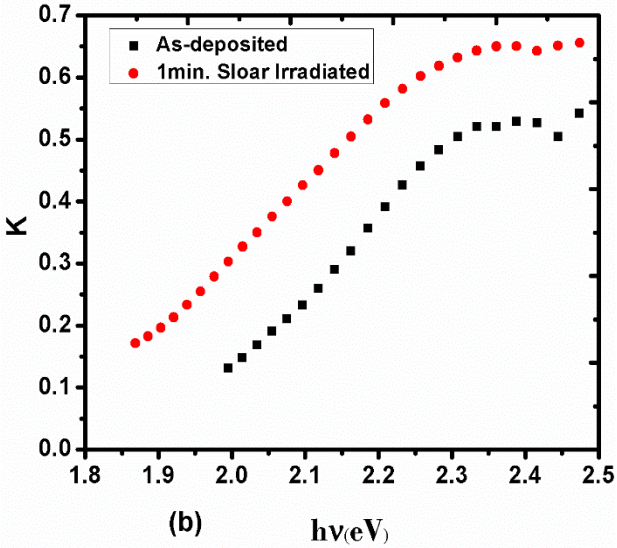


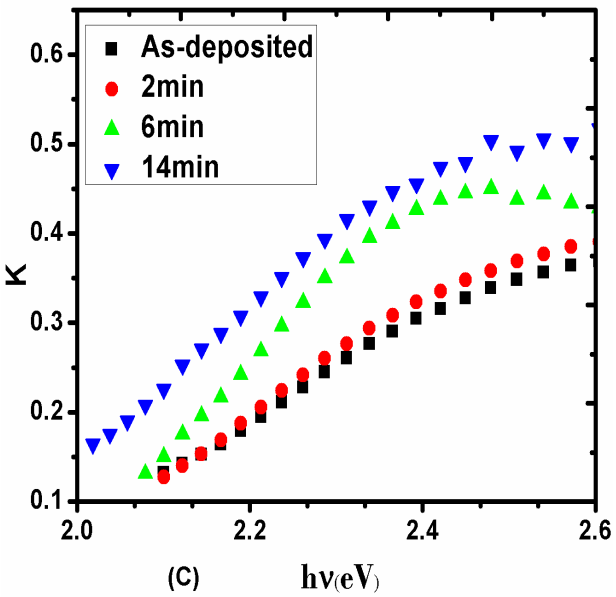
Fig. 5. Shows the variation of $(\alpha h\upsilon)^{1/2}$ with photon energybefore and after irradiation of thin films of a-Se (a) laser irradiation, (b) Solar irradiation and a-SeHg (c) laser irradiation, (d) solar irradiation.

The extinction co-efficient and 'K' has been calculated by using the relation **[28]**.

$$K = \alpha \lambda/4\pi \quad \text{---------- (9)}$$

Where 'α' is the absorption coefficient and 'λ' is the wavelength of the photon.

Fig. 6 (a, b, c, d), shows the variation of extinction coefficient with photon energy. It is found that the value of extension coefficient 'K' is increased after irradiation of the thin film system of a-Se and a-SeHg by laser and solar light. Similar results have been observed by Mott and Davis **[27]** for the thin films of various other amorphous semiconductors.



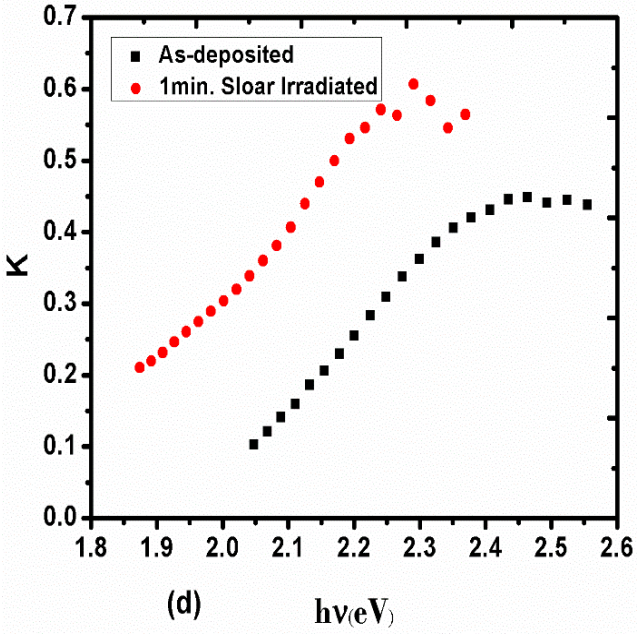


Fig. 6. Shows the variation of extinction coefficient with photon energy before and after irradiation of thin films of a-Se (a) laser irradiation, (b) Solar irradiation and a-SeHg (c) laser irradiation, (d) solar irradiation

Conclusion

In this study, it is shown that just like laser irradiation has tendency to change the properties of the material similar results were obtained also by solar light irradiation. It is found that that the Raman and XRD analysis of thin films of a-Se and a-SeHg shows the phase transition from amorphous to crystalline state occurs after laser and solar light irradiation. The grain size of a-Se increases from 22.619 nm to 30.089nm (due to laser irradiation) and from 22.619nm to 34.804nm (due to solar irradiation). In case of a-SeHg the grain size increases from 25.444nm to 28.171nm (laser irradiation) and from 25.444nm to 47.172nm (solar irradiation). It was found that the value of optical band gap of a-Se decreases from 1.77 eV to 1.72 eV (laser irradiation) and from 1.77 eV to 1.56 eV (solar irradiation). In case of a-SeHg it decreases from 1.81 eV to 1.67 eV (Laser irradiation) and from 1.81 to 1.56 eV (solar irradiation). The decreasing value of dislocation density and Urbach's energy due to irradiation indicates the disorder or defects are decreased in the material system. The important thing found from this study is the value of absorption coefficient of a-Se increases $0.935\times10^{4}cm^{-1}$ to $1.74\times10^{4}cm^{-1}$ (laser irradiation) and it increases to $2.858\times10^{4}cm^{-1}$. Similarly the absorption coefficient of a-SeHg increases $0.685\times10^{4}cm^{-1}$ to $1.142\times10^{4}cm^{-1}$ (laser irradiation) and it increases to $3.042\times10^{4}cm^{-1}$. Thus the photoconductor based on solar or laser irradiated SeHg alloy may be more efficient than amorphous selenium based photoconductors.

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