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The Role of Selenium Concentration in CdTe Absorber Layer for Solar Cell Applications

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Abstract: In this study, we discussed that the Electricity demand is growing globally on a daily basis. Researchers have developed a low-cost alternative to Silicon-based thin film solar cells based on Cadmium. Polycrystalline Cadmium Telluride (CdTe) with Selenium concentrations of 5%, 10%, and 15% ($CdSe_xTe_{1-x}$ at x = 0.5 to 0.15) have been prepared on highly cleaned glass substrates. Further this $CdSe_xTe_{1-x}$ material has been vacuumed sealed and annealed at 250 °C for 1 hour and then for 18 hours at 1150 °C, Thusly CdTe and Se both can melt homogeneously and form $CdSe_xTe_{1-x}$ materials. The thin films of $CdSe_xTe_{1-x}$ material of thickness 1μm have been prepared using thermal evaporation techniques at pressure ~10^{-6} mbar. The optoelectrical parameters have been carried out by applying XRD, UV-Vis-NIR and Current-voltage characterization. The crystallinity size increased with varying concentrations of selenium. The direct energy band gaps have been observed in the range of 1.36 to 1.02 eV. The I-V characteristics of thin films have shown that all thin films are ohmic in nature. All these conditions are required to improve the efficiency of a solar cell.

Keywords: $CdSe_xTe_{1-x}$ thin films; Selenium doping; Thermal evaporation technique; Annealing; Physical properties.

1. Introduction

Solar energy is the need present to fulfill world energy demand. The Cadmium telluride (CdTe) thin film solar cell technology established them in the market of photovoltaic technology. The cheap cost Cadmium telluride based thin film solar cells have represented greater than 22% efficiency, having a 1.45 eV band gap [1-2]. They deposited a thin film of CdTe on a glass substrate by applying thermal to the evaporation deposition process. The laser annealing was performed with the thin films and recognized that 60 J/pulse annealing has shown the best crystallinity properties with low roughness and greater grain size. The Hall Effect measurement indicated the electrical property has found great in a deal at 60 J/pulse annealed thin films.

The Selenium (Se) grading in CdTe increases the carrier lifetime and overall working of the solar cells [3]. They have developed $CdSe_xTe_{1-x}$ /CdTe bilayer and also observed a short circuit value more than 28 mA/cm^2 and a carrier lifetime value of 10-20 ns. They have also observed that the CdSeTe grain sizes are smaller near the pn-junction as compared to the other remaining part of CdTe. The impact of Selenium and chlorine in CdSeTe solar cell has been investigated [4]. In this, the optical band gap of CdTe was reduced below 1.45 eV and the short circuit current has been found to increase with introducing Selenium, resulting in the power conversion efficiency of the CdSeTe/CdTe device has increased. They have also observed a reduction in the mid-gap defects and improved the carrier lifetime.

The doping of Se in CdTe material has been recorded to increase the efficiency of solar cells [5]. In this, the CdSeTe band grading has used and recognized the change in carrier lifetime, alignment of the energy band, transport carrier concentration, recombination losses and overall performance of the solar cell. They have investigated that the Se alloying enhanced the lifetime; resultant efficiency of CdSeTe based on thin film photovoltaic cells. To increase efficiency, the tuning of bandgap and controlled doping technique have been introduced [6]. They have introduced alloying CdTe with CdSe for efficient solar cell application. The device band gap has been observed a little lower, resultant increase in I_{sc} has been recorded.

The impacts of Chlorine and Selenium at the grain boundaries of CdTe have been investigated [7]. These treatments have increased the grain boundaries, and the resultant increase in electron hole charge carrier separation or overall performance of the solar cell device has been recognized. Further by introducing selenium alloy into the CdTe absorber layer a more increase in the efficiency of solar cells has been recorded. The grain CdSeTe and CdTe thin films layer have deposited using close spaced sublimation technique [8]. They have investigated the grain growth mechanism and presented the increase in grain size of 1 mm observed in $CdSe_xTe_{1-x}$ thin films. The colossal grain growth process provides a reduction in grain

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boundary recombination.

The energy band structure of $CdTe_{1-x}Se_x$ solutions has been found to decrease with Selenium (Se) concentration in a single exponential function [9]. In this, the Se concentration has been used in the range (0<x>5/16). The energy band gaps have recorded direct-type band gaps with cubic phase structure. The transport free charge carrier concentration has observed an increase with increasing Se concentration. The decrease in the CdSeTe band gap has also been investigated with varying Selenium concentrations [10]. In this the reduction in the CdTe energy band recorded 1.5 to 1.38 eV, resulting in the overall solar cell performance having increased.

The $CdSe_{1-x}Te_x$ thin film using values of 'x' from zero to one has developed onto a cleaned glass substrate by applying a thermal evaporation process [11]. The crystal structure of CdTe has transformed from hexagonal to cubic with increasing concentrations of Se. The optical energy band gaps of CdTe have tuned in the range of 1.67 to 1.51 eV with varying Se concentrations from 0 to 1. The impacts of group V^{th} element on CdSeTe solar cells have been observed [12]. They have used low temperature Cu-free doped material as chloride in CdSeTe solar cell and observed that this process results in both the high hole density of 10^{15} cm^{-3} as well as long carrier lifetime of 22 ns. This is in good condition for an efficient CdSeTe thin film solar cell.

According to the literature survey, finite research has been done on the concentration effect of Selenium in CdTe and thus in this present work, the Se impurity has willfully been introduced with CdTe to make the formation of $CdSe_xTe_{1-x}$ thin films. Thus, the effect of Se concentration at 235^{o} C annealing on morphological, optical and electrical properties to achieve properties of $CdSe_xTe_{1-x}$ for application of photovoltaic cells.

2. Experimental detail

2.1 Material preparation

The Cadmium telluride and Selenium materials, both in powder form with purity of 99.999% have been purchased from Alfa Aesar scientific agencies. Preparing Selenium doped CdTe ($CdSe_xTe_{1-x}$ at x = 0.5, 1, and 0.15) having 5%, 10%, and 15% concentration of Selenium. Firstly, these materials have been weighed according to the atomic weight percentage. Thereafter put into an ampoule at pressure ~ 5 x 10^{-6} mbar for vacuum sealing, using both rotary and diffusion pumps shown in Fig. 1(a). These ampoules had placed into a furnace (Naskar) at the beginning shown in Fig. 1(b); the temperature increased up to 250^{o} C in 40 minutes and then remain at this temperature for 1 hour, thusly Selenium can completely melt (melting point of Se ~ 220^{o} C). Consequently, the temperature of furnace has increased up to 1150^{o}C in two hours then remains on this temperature for the next 18 hours, hence CdTe (melting point ~ 1092^{o} C) could melt and homogeneous melt of both CdTe and Se can occur. Then the ampoule had cooled for a night till the room temperature reached inside the furnace chamber represent in Fig 1(c). So that the CdTe:Se 5%, 10% and 15% have been obtained by cracking the ampoules and source materials have been collected for further deposition of thin films.

(a)

(b)

(c)

Fig. 1: (a, b and c represent the vacuum sealed materials, placed in a furnace and after cooling material in ampoules)

2.2 Substrate cleaning process

Before the further deposition process, the substrate cleaning process is necessary. For this plane and ITO coated glass substrates have been placed into 50° C warmed Deionised water (DI water) for 15 minutes and then placed into Acetone for the next 15 minutes. Later these substrates have put into isopropyl alcohol (IPA) for 15 minutes. Again, these substrates have placed into Acetone for 15 minutes and washed by raining acetone on all substrates, to eliminate contamination of the surface and boost the adhesive nature of the deposited thin films. In last all the substrates have dried using a hot drier. Now the substrates have been ready for the further deposition process.

2.3 Deposition method

The $CdSe_xTe_{1-x}$ materials at x = 0.5, 1.0 and 1.5 having Se concentrations of 5%, 10% and 15% respectively have been deposited onto cleaned glasses and ITO substrates by applying Hind High Vacuum (HHV) made resistive heating thermal evaporation unit (SMART COAT 3.0) under the pressure of $\sim10^{-6}$ mbar. The inside wall of the vacuum chamber has been cleaned with isopropyl alcohol (IPO) to eliminate the impurity of the last deposition. The source material $CdSe_xTe_{1-x}$ has been filled into a molybdenum boat stacked between graphite crucibles. The glass and ITO coated slides have been stacked with a substrate holder and placed ~17 cm above the source material. Later the chamber evacuated appropriately $\sim10^{-6}$ mbar applying a diffusion pump and rotary pump. Now the deposition rate of 4-5 Å/sec has been maintained by controlling the current 25 to 30 A. A thickness of 1μm has been developed onto substrates, measured using a thickness monitor attached to the coating unit. This process was repeated three times to deposit a thin film of 5%, 10% and 15% of Selenium concentrated materials. Later these thin films were annealed at 235° C for 1 hour using a 'SONAR' muffle furnace to improve properties. Annealing processing has been done on behalf of a feasible literature survey [13]. In this study, annealing has increased the optoelectronic properties of the CdTe.

2.4 Characterization of thin films

The photonic properties such as absorption, energy band gap and extinction coefficients have been investigated by applying a UV-Vis spectrometer (UV-1780) in the 300 to 900nm range. The crystallographic parameters of thin films have been observed using XRD (Rigaku Ultima IV), having 2θ values between $20° < 2\theta > 80°$ with λ = 1.54056 Å at step size 0.05° / second, Rigaku manufactured X-ray diffractometer (Ultima-IV). The electrical properties (I-V measurements) of the thin films have been observed using the Agilent B2901A source meter. For this, the contact on thin films were made using silver paste and kept it dry for two days.

3. Result and Discussion

3.1 crystallographic observations

The structures of $CdSe_xTe_{1-x}$ thin film have been observed by observing the X-ray diffractometer. Fig. 2 represents XRD patterns of all three $CdSe_xTe_{1-x}$ thin films, this states the films having preferred orientation with (111) plane at 2θ = 24.1°C and 24.2°C for 5% and 10% concentration of Selenium respectively, conformed by JCPDS card 75-2086, this clearly states that both the thin films having a cubic structure with cell parameter a = 5.53 Å and 5.51 Å. But the pattern for 15% concentration of Selenium has a more intense peak along (111) at angle 24.1° C and also found three small additional peaks as (100), (101) and (110) at angle 2θ = 21.79, 26.02 and 40.70° C, conformed by JCPDS card 41-1325 for CdSeTe material. That is caused by substituting the Cadmium atom with Selenium, which might show the creation of secondary phases. These small peaks have not been recognized in CdTe thin films with 5% and 10% doping of Selenium. Thus, $CdSe_xTe_{1-x}$ thin film with 15% Selenium concentrated has existed Hexagonal structure with cell parameters a=5.69 Å and c=9.3 Å corresponding to the most intense peak. The cell parameters for other small intense peaks have been shown in table 1. The intensity of 5% and 15% are exactly the same but it's quite low for 10% Selenium doped CdTe thin films which usually indicate the randomization of crystal orientation [14-15]. The $CdSe_xTe_{1-x}$ thin films investigation has also been done [16], with a Selenium concentration of 0 to 0.41%, the thin film has a cubic structure while it has converted into a hexagonal structure for Selenium concentration more than 0.41%. In our case we have also observed the cubic structure for 5% and 10%, while Hexagonal structure was observed for a 15% concentration of Selenium, so this study well matches our result.

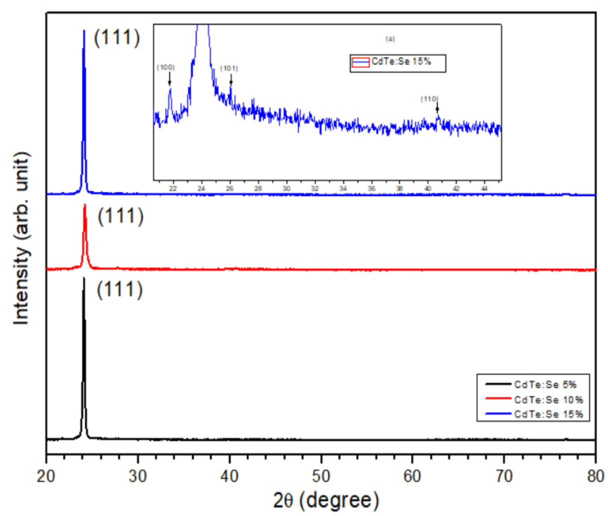


Fig. 2: X-ray pattern of $CdSe_xTe_{1-x}$ 5, 10 and 15% concentration of Selenium

The grain or crystalline size (D) has been evaluated using the Scherrer formula [17], which indicates the growth of the crystal. The other crystal parameter equations (2-6) have been calculated as under [18].

$$D = \frac{0.94\,\lambda}{\beta\cos\theta} \quad (1)$$

Table 1: The cell parameters for calculating various quantities.

Thin films	2θ (°C)	Miller indices	d Å	a Å	b Å	c Å	FWHM β (rad.)* 10^{-3}	D nm	ε* 10^{-4}	δx10^{14} m^{-2}	Nx10^{15} m^{-2}
CdTe:Se 5%	24.1	(111)	3.69	5.53	5.53	5.53	3.67	40.4	9.06	6.13	15.17
CdTe:Se 10%	24.2	(111)	3.67	5.51	5.51	5.51	5.41	27.37	13.23	13.35	48.77
CdTe:Se 15%	24.1 21.79 26.02 40.07	(111) (100) (101) (110)	3.69 4.08 3.42 2.21	5.69 4.07 4.01 3.14	5.69 4.07 4.01 3.14	9.3 6.65 6.65 5.13	3.67	40.4	9.06	6.13	15.17

The Inter planar spacing has been evaluated by applying Bragg's low given as:

$$d = \frac{\lambda}{2\sin\theta} \quad (2)$$

Here λ is the wavelength, θ is an angle, β is the FWHM (full width half maxima).

The dislocation density (δ) shows dislocation lines/volume, which denotes the defects that exist in a crystal given as:

$$\delta = \frac{1}{D^2} \quad (3)$$

The internal strain (ε) indicates the amount of strained energy stored in molecules given as:

$$\varepsilon = \frac{\beta\cos\theta}{4} \quad (4)$$

The crystallites/ area (represented by N) has been calculated as:

$$N = \frac{t}{D^3} \quad (5)$$

The lattice constant (a) has been evaluated using the equation given:

$$\frac{1}{d^2} = \frac{h^2 + k^2 + l^2}{a^2} \quad (6)$$

The crystallite size has been recognized as 40.4 nm for both 5% and 15% and it has value of 27.37 nm for 10% Selenium doped CdTe thin film. The value of FWHM has been observed as 0.21, 0.31 and 0.21 (in degree) for 5%, 10% and 15% Selenium doped CdTe thin films. This implies that the crystallinity has a low value for 10% Selenium concentration while the crystallinity has higher for 15% concentration of Selenium in CdTe thin films. The XRD pattern for $CdSe_xTe_{1-x}$ at x=0.6 has been observed [19], they have investigated that Selenium occupied the place to Telluride in CdTe and become CdSe conformed by XRD pattern. We have also observed the same result in CdSeTe thin film.

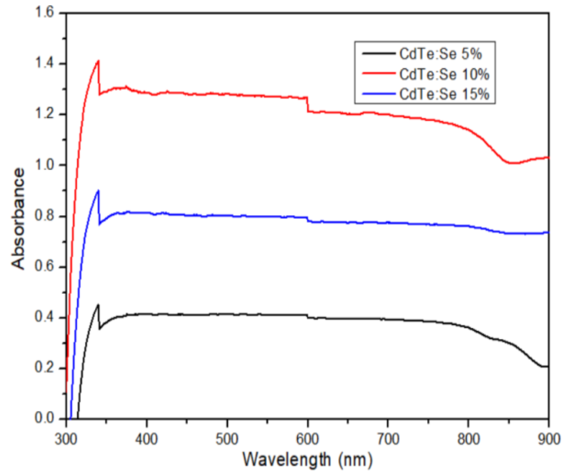
The values of FWHM (β) have been evaluated as 3.67, 5.41 and 3.67 for Selenium concentration as 5%, 10% and 15% respectively. These values clearly state that the crystallinity has been decreased by 10% Selenium concentration while the crystallinity has increased by15% concentration of Selenium in CdTe thin films. Higher value of FWHM indicates smaller grain size and higher dislocation densities [20].

The inter planar spacing values have been observed as 3.69, 3.67 and 3.69 Å for 5%, 10% and 15% Selenium doped CdTe thin films. This clearly states that the interplanar spacing is almost not affected by varying Selenium concentration. The other parameters as dislocation density, micro strain and crystallites/area have also calculated using XRD data calculation, shown in table 1.

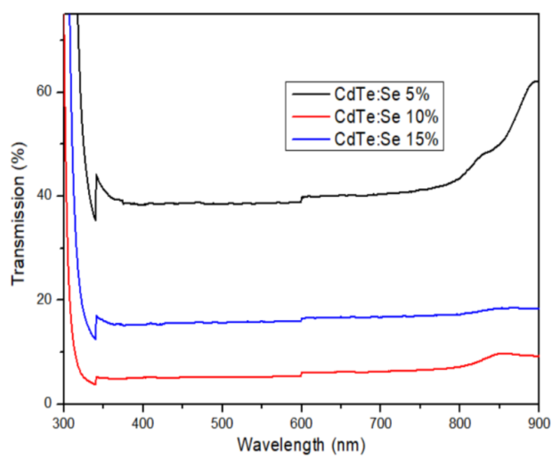
3.2 Optical observations

The observation of optical parameters such as absorbance, transmission, energy band gap, extinction coefficient and refractive index have been observed using UV-Vis. Spectrophotometer. Fig. 3(a) indicates the change in absorbance with a wavelength in the range of 300-900nm of 235° C annealed $CdSe_xTe_{1-x}$ thin films with Selenium concentrations of 5%, 10% and 15%. This figure states that the absorption has increased by 10% of Se concentration and found some lower by 15% of Se concentration (between 5% and 10%). The absorptions have been recognized as high in wavelength range 300 to ~800nm, after the 800nm absorption have found reduced suddenly and as Selenium concentration increased the absorption edge shifted toward the near infrared region (higher wavelength). This may be due to a decrease in the band gap of $CdSe_xTe_{1-x}$ films. The behavior of Copper doped $CdTe_{1-x}Se_x$ alloy has been investigated [21], in this the increase in optical absorption has recorded with increasing copper doping. This is in good agreement with our results and makes it suitable for solar cell application.

The transmission spectrum of all three $CdSe_xTe_{1-x}$ was evaluated in the 300 to 900nm range and illustrated in Fig. 3(b), this indicated the transmission reduced with increasing the concentration of Selenium 10% and further for 15% concentration of Selenium the transmission has found some increase (between 5% and 10%). The transmissions have been observed comparatively low in wavelength range of 300 to ~800nm, but after or above 800nm (near infrared region) a sudden increase have found in transmission with increasing the Selenium concentration in CdTe. The sudden jump in transmission was shifting toward the longer wavelength (>800nm) near the infrared region, it also suggests the change in energy band gap occurs with varying concentration of Selenium. These transmission spectrums are pursuing the contradictory nature of the absorption spectrum. Transmission has been found to decrease with increasing Selenium concentration [22], they have stated that the $CdSe_xTe_{1-x}$ thin film has reduced the transmission as compared to CdTe thin films.



(a)



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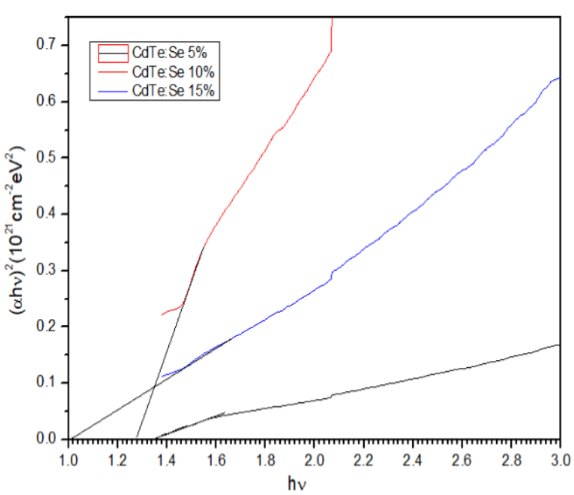

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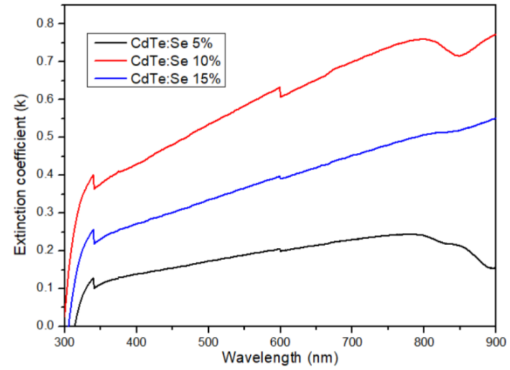

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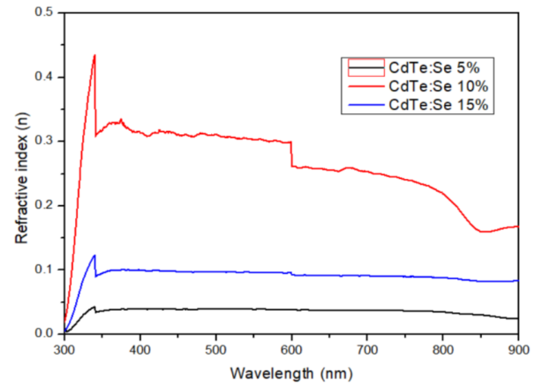

(e)

Fig. 3: (a) Absorption curve of $CdSe_xTe_{1-x}$; (b) Transmission curve of $CdSe_xTe_{1-x}$; (c) Tauc's plot for band gap ; (d) plot for extinction coefficients (k) Fig. 3(e) plot for refractive index (n)

The band gap of the thin film is an essential parameter for investigating optical properties. Materials with suitable energy band gaps are required for higher efficiency solar cells because materials can absorb only the particular photons with energy higher than the energy of the band gap. The energy gap can evaluate by applying Tauc's relation [23].

$(\alpha h\nu) = A(h\nu - E_g)^n$

Here 'α' represents the absorption constant, photonic energy represents by 'hν', E_g is the energy gap and 'n' is a constant quantity (n has the value ½ for direct and 2 for indirect allowed band gap).

Fig. 3(c) is called Tauc's plot drawn among $(\alpha h\nu)^2$ and hν for $CdSe_xTe_{1-x}$. The energy band gap can be calculated, by taking the intercepts of tangents on the (hν) axis. This figure states that the optical energy bandgap value has recorded 1.36, 1.28 and 1.02eV for Selenium concentrations 5%, 10% and 15% in CdTe respectively. Tauc's plot has a linear nature and clearly states that the $CdSe_xTe_{1-x}$ has a direct allowed band transaction. Result the increase in Selenium concentration reduces the optical band gap of $CdSe_xTe_{1-x}$. The effect of Selenium in CdTe has been investigated [24], they deposited a 1.5 µm thin layer of CdSeTe material and observed that the optical energy band gap value decreased with introducing Selenium into CdTe.

The extinction coefficient indicates the attenuation and scattering behavior of light from $CdSe_xTe_{1-x}$ illustrate in Fig. 3(d). The relation of the extinction coefficient is given as [25]

$$k = \frac{\alpha\lambda}{4\pi}$$

Here α represent the absorption coefficient

$$\alpha = \frac{2..303\ A}{t}$$

The figure states that the extinction coefficient value has been increased as well as wavelength shifted toward the higher wavelength (near infrared region), also the value of extinction coefficients has been increased with increasing the Selenium concentration from 5% to 10%. But extinction coefficient value has decreased with increasing Selenium concentration from 10% to 15%. The increase in extinction coefficient indicates the $CdSe_xTe_{1-x}$ thin film with a Selenium concentration of 10% makes it applicable as an absorbing layer in solar cells.

Fig. 3(e) shows the relation between the refractive index and wavelength, the refractive index indicates the electrical polarizability of the materials. This has been calculated using the Swanepoel envelope method (SWEM) [26]. The refractor index increased by increasing the Se concentration from 5% to 10%, while the refractive index value has some low value with increasing the Se concentration from 10% to 15%. This may be because of the reduction in the energy gap value of materials.

3.3 Current-Voltage (I-V) characteristics

The I-V curve has been observed by applying two-probe source meters for $CdSe_xTe_{1-x}$ thin films with all three concentrations and annealed at temperature 235° C in the voltage range -2.5 to +2.5 Volts. The nature of thin films was shown in Fig. 4.

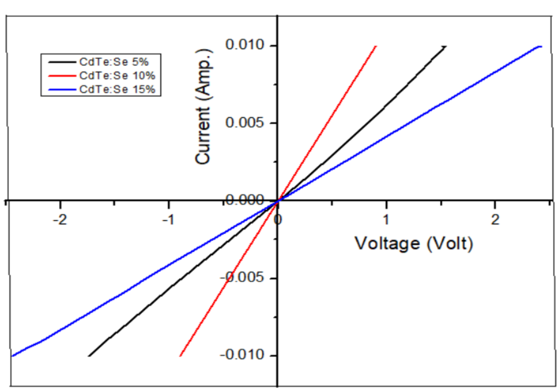

Fig. 4: Current voltage characteristics of $CdSe_xTe_{1-x}$

This indicates the linear behavior for all $CdSe_xTe_{1-x}$ thin films on both sides of reverse and forward bias, so all thin films have shown ohmic nature, such ohmic behavior is the mandatory condition for efficient thin film solar cells [27]. This figure states that the conductivity has been increased with increasing concentration of Se from 5% to 10%, but an enormous decrease in conductivity or increase in resistivity has been recorded with increasing Se concentration from 10% to 15%. This may be undertaken based on free charge carrier concentration in CdSeTe [28]. In this study, the $CdTe_{1-x}Se_x$ thin films have been deposited with varying the value of x=0 to 0.2 and recognized that the resistivity has decreased for x=0.1, while the resistivity has a maximum value at x=0.2. This might be due to a change in the charge carrier concentration.

4. Conclusion

We have successfully injected Selenium from 5% to 15% into CdTe using a vacuum sealing method, and then annealed it at 235°C. It was observed that the size of crystallites, interplanar spacing, and lattice constant differed with the concentration of Selenium. The observed CdTe thin films with 5% and 10% Selenium have a cubic structure with a preferred orientation (111) and cell parameters a= 5.53Å and 5.51Å. While with 15% Selenium has a hexagonal structure with preferred orientation along (111) and cell parameters a=5.69Å and c= 9.3Å. The crystalline size from the Scherrer formula is recorded within the range 27-40 nm. As the Selenium concentration increased from 5% to 15%, the direct energy band gap decreased from 1.36 to 1.02 eV. This indicates that the wavelength range of energy absorption has shifted to a higher wavelength range (red shift). The I-V characteristic of thin films indicates the ohmic nature of the films and this is a required condition for solar cells. The study has all these qualities and characteristics, making it very useful for the application of solar cells.

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