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RESEARCH ARTICLE

STUDY THE INFLUENCE OF IRRADIATION BY GAMMA-RAY ON THE STRUCTURAL AND OPTICAL PROPERTIES OF CDO THIN FILMS

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ABSTRACT

Cadmium oxide (CdO) thin films have been deposited on glass substrate at R.T (300 K) by thermal oxidation with exist air. All samples were exposed to Gamma –Ray (γ – rays) source type (Cs^{137}) with energy (0.611MeV) for different irradiation time (2, 4 and 6) hours. In this study determines the effect of irradiation by (γ – rays) at different time on the structural, optical properties of as deposited films. X-ray techniques were used to characterize structural properties which indicate the formation of polycrystalline cubic CdO phase with preferential orientation along (111) plane. However, intensity of all peaks increases after irradiation which indicates that the crystallinity increases with increasing irradiation time. The transmission and absorptions spectrum of the films as a function of wavelengths in the range (300-1100) nm were used to study the optical properties before and after irradiation. The optical constants such as absorption coefficient, refractive index, extinction coefficient, real and imaginary parts of the dielectric constant and the electrical conductivity were investigated and calculated.

INTRODUCTION

Transparent Conductive Oxide (TCO) thin films have received much attention because of their wide applications in the field of thin-film (Khalil Arshak and Olga Korostynska, 2006). Cadmium Oxide (CdO) is one of the most important transparent Conductive Oxide (TCO) materials which is a compromise between low resistivity and high optical transmittance in visible and NIR spectral regionsmainly due to oxygen vacancies (Zaien *et al*., 2013 and Viswanath *et al*., 2012), which make it useful for many application such as photodiodes, phototransistors, gas sensors (Mane *et al*., 2006 and Sinatirajah, 2008), photovoltaic cells, optical communications and other optoelectronic applications. With the development of these optoelectronic devices, the pursuing of novel TCO materials with improved electrical and optical properties which may contribute to better device performances is continuing (Calnan and Tiwari, 2010 and Dakhel, 2013). Cdo films one of the element sensor arrays would enhance the performance of the radiation detection system, the sensitivity of metal oxide films to γ -radiation exposure depends on their composition and thickness (Khalil Arshak and Olga Korostynska, 2006). It is n-type semiconducting having a narrow direct band gap (2.2 -2.7) eV and a high electrical conductivity (10^{-2}-10^{-4}) Ω·cm (Sinatirajah, 2008 and Dakhel, 2013). Therefore many researchers focused on Cadmium oxide in the recent years and used various deposition techniques such as pleased laser deposition (Gupta *et al*., 2008), spray pyrolysis (Viswanath *et al*., 2012 and Amutha *et al*., 2012), sol-gel (Aksoy *et al*., 2009), electron beam evaporation (Ali *et al*., 2009), solid vapor deposition (Zaien *et al*., 2013), RF magnetron sputtering (Saha *et al*., 2008), chemical path deposition (Perumal *et al*., 2012) etc. In the present paper we have prepared (Cd) thin films by thermal evaporation technique then oxidation these films to get CdO films. The structural and optical properties of CdO thin films before and after irradiation by (γ – rays) at different time was studied and investigated.

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Experimental

The pure metal Cadmium thin films have been prepared in a high vacuum system of ($3x10^{-6}$) torr at R.T (300 K) on glass substrate by thermal evaporation technique using Edward coating unit model (E 306) from molybdenum boat, the distance between the substrate and boat was about (18cm). The thickness of these films was about (350±50) nm which determined by using weighing method, the deposition rate (1.1 nm/sec). Then we get CdO thin films by thermal oxidation processes at temperature of (673) K for one hour with exist air flow by using (Kilns Furnaces). These films were exposed to gamma ray form Cs^{137}withenergy (0.611MeV) during different irradiation time (2, 4 and 6) hours. The crystal structure of these films has been examined by X-ray diffraction technique

(XRD) using Siemens X-ray diffract meter system (SHIMADZU Japan XRD 600), by records the intensity in the range of Bragg's angle θ from (10-80). Source Cu K_α radiation of wavelength (λ=1.5405 Å) was employed with generator setting of current (20mA) and Voltage (40kV).

The lattice constant (a) calculated from the relation: (Usharani *et al*., 2013)

$$a=d\,(h^2+k^2+l^2)^{1/2} \quad \ldots\ldots (1)$$

Where d (hkl) is the inter planer distance for different planes which estimated from Bragg's law:

$$2d\sin\theta=n\lambda \quad \ldots\ldots (2)$$

Where n is the reflection order.

The grain size dimension (D) of the films could be calculated from diffraction line broadening using the Scherrere quation (Blackmore, 1974):

$$D=k\lambda/\beta\cos\theta \quad \ldots\ldots (3)$$

Where k is a constant (0.9), and β is full width half maximum (FWHM) of the preferential plane. We can calculate the dislocation density (δ) from the equation (Suryanarayana and Norton, 1998):

$$\delta = 1/D^2 \quad \ldots\ldots (4)$$

While the number of crystallites per unit surface area (N) was calculated using the formula:

$$N = t/D^3 \quad \ldots\ldots (5)$$

The transmittance (T) and absorbance (A) spectrum were recorded in the range of wavelengths (300-1100) nm using a double beam spectrophotometer (UV/VIS).

The incident photon energy (E=hν) was calculated as a function of wavelength (λ) from equation:

$$Eg^{opt}(eV) = 1240/\lambda_{(nm)} \quad \ldots\ldots (6)$$

The absorption coefficient (α) was calculated from equation: (Blackmore, 1974)

$$\alpha = 2.303\,(A/t) \quad \ldots\ldots (7)$$

The absorption coefficient (α) and the incident photon energy (hν) is related by Tauc formulas: (Taus, 1974)

$$(\alpha h\nu) = B\,(h\nu - Eg^{opt})^{r} \quad \ldots\ldots (8)$$

Where B is a constant inversely proportional to amorphousity, r is constant and may take values 2, 3, 1/2, 3/2 depending on the material and the type of the optical transition. We can determined the value of optical energy gap when the straight portion of the plot of $(\alpha h\nu)^{1/r}$ against (hν) is extrapolated to $(\alpha h\nu)^{1/r}=0$.

The extinction coefficient (K) can be determined by using the relation: (Taus, 1974)

$$K = \alpha\lambda/4\pi \quad \ldots\ldots (9)$$

Refractive index (n) can be calculated using equation: (William and Callister, 2003)

$$n = \{[4R/(R-1)^2]-K^2\}^{1/2} - [(R+1)/(R-1)] \quad \ldots\ldots (10)$$

Where R is the reflectance which calculated by using equation:

$$R = 1-T-A \quad \ldots\ldots (11)$$

The dielectric constant were obtained using the formula: (Kasap, 2002)

$$\varepsilon = \varepsilon_1 - i\,\varepsilon_2 \quad \ldots\ldots (12)$$

Where ε_1 is real part of dielectric constant and can be calculated from:

$$\varepsilon_1 = n^2-K^2 \quad \ldots\ldots (13)$$

$\varepsilon_2$is imaginary part of dielectric constant and calculated from:

$$\varepsilon_2 = 2nK \quad \ldots\ldots (14)$$

The optical conductivity (σ) was calculated using the relation: (Pankove, 1975)

$$\sigma = \alpha nc/4\pi \quad \ldots\ldots (15)$$

Where (c) is the velocity of light.

RESULTS AND DISCUSSION

Structural Properties

X-ray diffraction pattern of cadmium oxide films before and after irradiation are shown in Figure (1). This figure shows smooth surface and well adhesive nature of these films with substrate. The peaks are observed due to diffraction from (111), (200) and (222) planes indicate the polycrystalline nature of the CdO films with cubic phase formation as compared with ASTM card No.05-0640 (JCPDS, 1997). The observed lattice constant (a) and d (hkl) values are I n good agreement with standard values taken from ASTM data as shown in Table (1):

We notice from Figure (1) part (B and C), that the intensity of CdO thin films after irradiation becomes stronger and sharper due to the improvement of the crystallinity in the films.

The grain size, dislocation density and number of crystallites per unit surface area were calculated using equations (3, 4, 5) respectively. The grain size increase while dislocation density and number of crystallites per unit surface area showed an opposite trend after irradiation, which was demonstrated the improvement of the crystallinity in the films, as shown in Table (2).

Table 1. XRD results of CdO thin films before and after irradiation

State	2 θ (ASTM)	2 θ Observed	D (A°) (ASTM)	D (A°) Observed	A (A°) ASTM	A (A°) Observed	(hkl) ASTM
Before irradiation	33.00	32.98	2.7120	2.713	4.695	4.699	111
	38.283	38.27	2.3490	2.3499		4.698	200
	69.284	69.28	1.3550	1.3551		4.694	222
after irradiation (2houre)	33.00	33.02	2.7120	2.7105	4.695	4.6947	111
	38.283	38.3	2.3490	2.3482		4.696	200
	69.284	69.29	1.3550	1.3549		4.6937	222
after irradiation (6houre)	33.00	33.04	2.7120	2.7089	4.695	4.6919	111
	38.283	38.34	2.3490	2.34579		4.6915	200
	69.284	69.32	1.3550	1.35447		4.6920	222

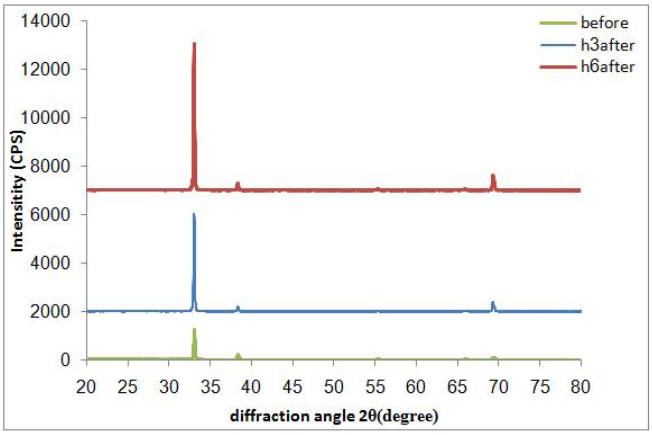

Figure 1. X-ray diffraction pattern of CdO thin films before and after irradiation

Table 2. results of grain size,δ and N of Cdo thin films for the (111) preferred peak

State	D (nm)	$\delta^* 10^{14}$ (m^{-2})	N^*10^{15} (m^{-2})
Before irradiation	58.37	2.9346	1.508
after irradiation (2houre)	60.956	2.691	1.324
after irradiation (6houre)	63.773	2.4588	1.1566

Optical Properties

Fig.(2) shows the variation of absorbance (A) as a function of wavelength range (300 – 1000) nm for CdO thin films before and after irradiation, from this figure we can notice that the absorbance values decreased generally as the wave length increases and has low values in the NIR region, we can also observe that these films exhibit an absorption edge and the absorption edge shifts to lower wavelength after irradiation. It is clear from this figure that the value of the absorbance of CdO thin films decreased with increase their radiation time for all values of the wavelength due to improvement of the crystallinity in the films after irradiation leads to growth small grain and decrease grain boundaries; it yields to decrease the absorbance values. Also we can see a strong absorption at wavelength range of (300-500) nm, which is made Cadmium Oxide films suitable for solar cell. The transmittance (T) versus wavelength of CdO thin films before and after irradiation is shown in Fig(3), we can see from this figure that the transmittance value increase with increase irradiation time. Also we can see from this figure the transmittance values increase with increase wavelength and have high values in the NIR region, all samples demonstrate (20-80) % transmittance at wavelengths longer than 500 nm which makes these films

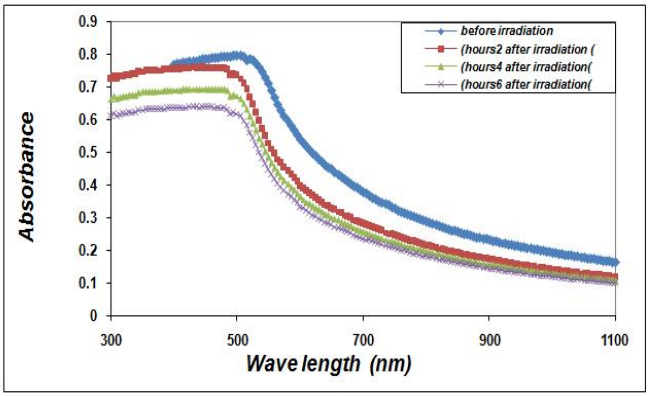

Figure 2. The variation of absorbance versus wavelength for cadmium oxide thin films

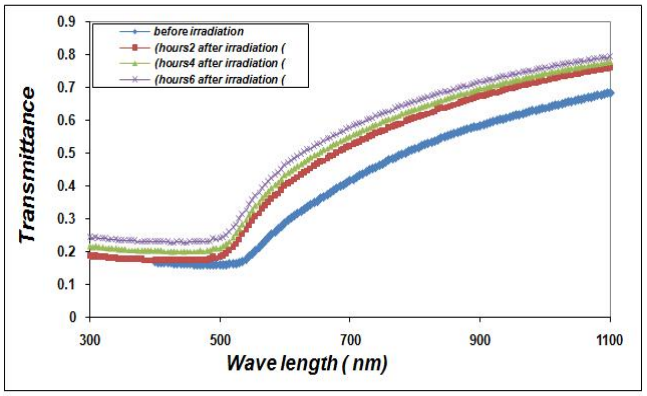

Figure 3. The variation of transmittance versus wavelength for cadmiumoxide thin films

suitable for solar energy collection. The shift of absorbance and transmittance due to the fact that these are related to the changes in the films characteristics (Dakhel *et al*., 2003). The variation of the absorption coefficient values (α) of CdO films as a function of photon energy before and after irradiation were calculated from equation (7) and shown in Fig (4).We can notice from this figure that all the films have high values of absorption coefficient ($\alpha > 10^4$ cm^{-1}) this means that the direct transition is possible occurs. This result is in agreement with refs (Majid H. Hassouni *et al*., 2013 and Mahaboob Beevia *et al*., 2010). Itis clear from this figure thatthe absorption coefficient values decrease with irradiation time within the whole range of the spectrum and shifts toward higher energy. This behavior may be due to the changes in crystal structure of these films after irradiation, the absorption is not attributed to the free carriers only, but to impurities or

localized electronic states. We plot $(\alpha\ h\nu)^2$ versus photon energy ($h\nu$) to find the type of the optical transition for (CdO) films before and after irradiation, which describes the allowed direct transition as shown in Fig.(5) and calculated the optical energy gap (Eg^{opt}) values from Tauc equation (8)by select the optimum linear part, which determined by the extrapolation of the portion at (α =0). It is clear from Fig. (6) and Table (3) the optical energy gap increased from (2eV) to (2.2eV) when the irradiation time increased. This behavior can be attributed to the decrease of the density of localize states in the Eg after irradiation which caused the energy gap seems large as well as the reduction in the number of defects in films and the increases in stoichiometric composition, might also lead to the increase in the optical band gap. The value of the optical energy gap is agreed with refs (Mahaboob Beevia *et al*., 2010 and Mohamed and Mahmoud Ali, 2008). It can be seen from the Figure (7) the variation of the refractive index values (n) versus wave lengths for (CdO) films before and after irradiation, which calculated from equation (10). It is obvious from result that the refractive index values increases with increasing photon energy followed decrease these value at higher photon energy.

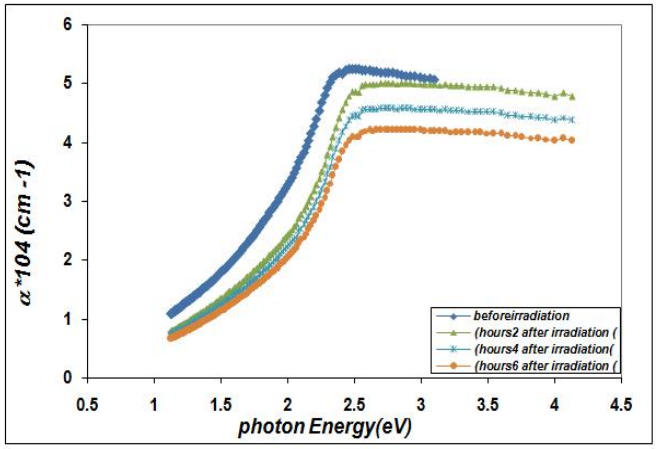

Figure 4. Absorption coefficient behavior as a function of photon energy for cadmium oxide thin films

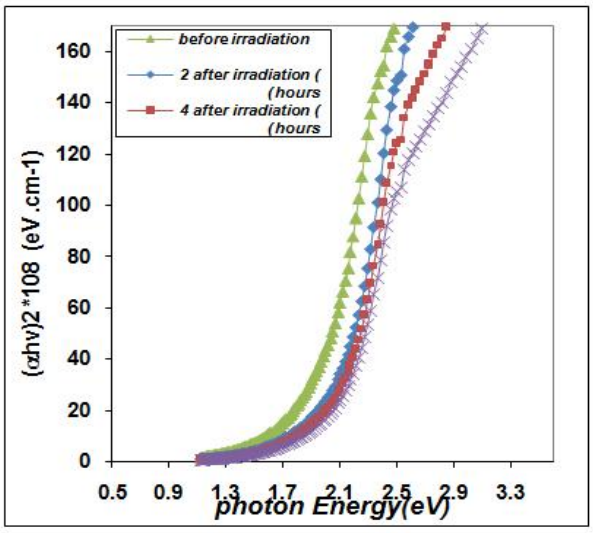

Figure 5. Variation $(\alpha\ h\nu)^2$ & photon energy for cadmium oxide thin films

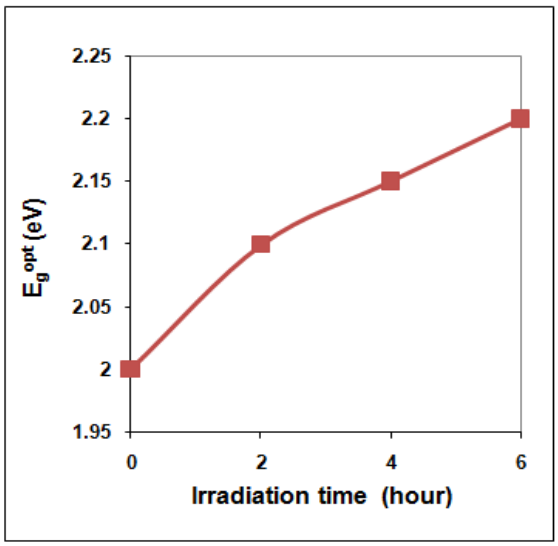

Figure 6. Optical energy gap as a function of Irradiation time for cadmium oxide films

Table 3. The optical constant of CdO thin films before and after irradiation

State	Eg^{opt} (eV)	Optical constant at λ =550 nm				
		α x 10^4 cm^{-1}	n	k	ε_1	ε_2
Before irradiation	2	4.675	1.889	0.2047	3.5269	0.7735
after irradiation (2houre)	2.1	3.497	2.433	0.1531	5.9007	0.7455
after irradiation (4houre)	2.15	3.1978	2.526	0.1400	6.3650	0.7076
after irradiation (6houre)	2.2	2.945	2.586	0.1289	6.6710	0.6670

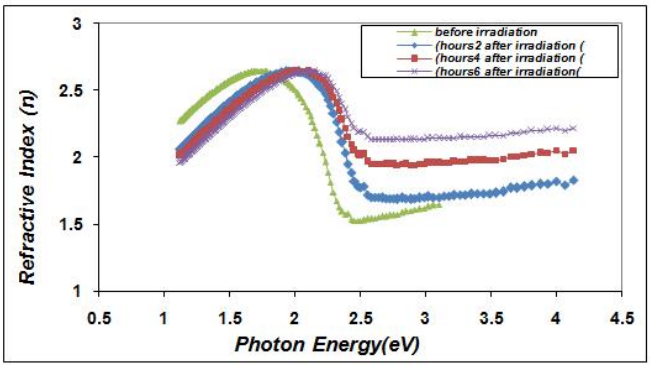

Figure 7. Variation refractive index & photon energy for cadmium oxide thin films

At lower photon energy refractive index values decrease with increasing irradiation time while at higher photon energy after 1.9eV showed an opposite trend these values increases with increasing irradiation time, this behavior may be due to change in the films structure after irradiation. We can notice all samples have a peak refractive index which increase from (1.88 to 2.58) after irradiation and this peak shift to higher photon energy. Fig. (8) shows a plot of the extinction coefficient (K) versus photon energy for CdO thin films as a function of irradiation time, we can notice from this figure that the extinction coefficient behavior nearly similar for all the

range of the wavelength spectrum to that of the absorption coefficients, this means the extinction coefficient values decrease with irradiation time within the whole range of the spectrum for the same reasons as we mentioned before and have peak shifts toward higher energy. The variation of the real (ε_1) and imaginary (ε_2) parts of the dielectric constant values as a function of photon energy for CdO thin films before and after irradiation was seen in Figures (9), (10) respectively.

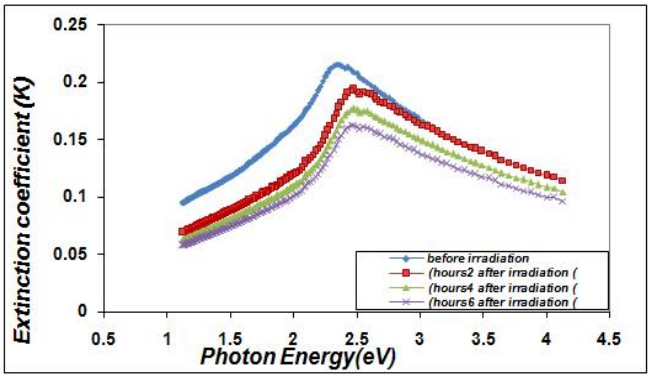

Figure 8. Variation extinction coefficient & photon energy for cadmium oxide thin films

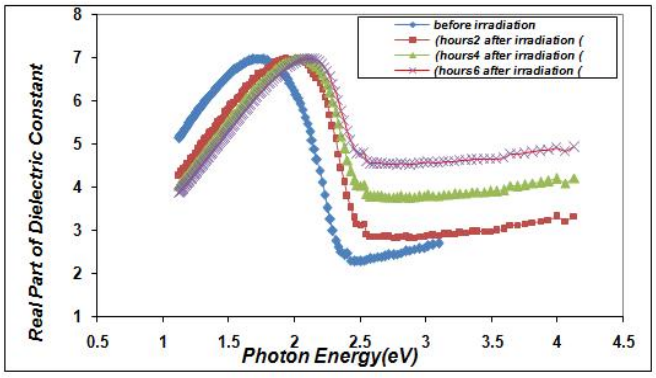

Figure 9. Variation real part of the dielectric constant & photon energy for cadmium oxide thin films

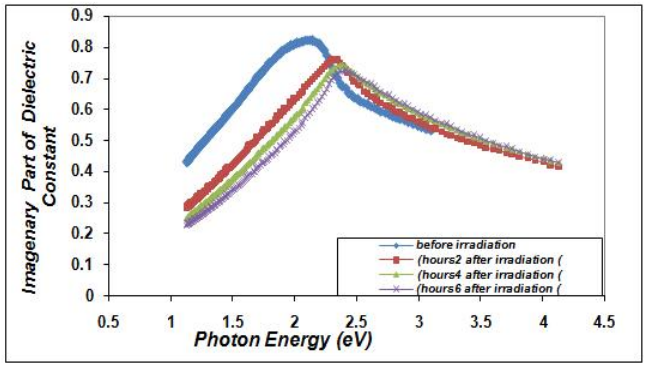

Figure 10. Variation imaginary part of the dielectric constant& photon energy for cadmium oxide thin films

From this figure we can deduce that the values of the real part are higher than those of the imaginary part, the real part of the dielectric constant behavior nearly similar for all the range of the wavelength spectrum to that of the refractive index because of the real part depends mainly on refractive index according to the equation (13), while the imaginary part depend on the extinction coefficient values which are related to the variation of absorption coefficient, we can see from Fig.(10) the imaginary part value decrease with increasing irradiation time at lower photon energy followed increase this values with increasing irradiation time at higher photon energy and the peak shift tohigher photon energy. This behavior is due to the imaginary part is directly related to the density of states within the forbidden gap of semiconductor materials (Mohamed and Mahmoud Ali, 2008). Some of the optical constants values are shown in Table (3). The variation of optical conductivity as a function of photon energy of CdO films before and after irradiation is shown in Fig.(11).

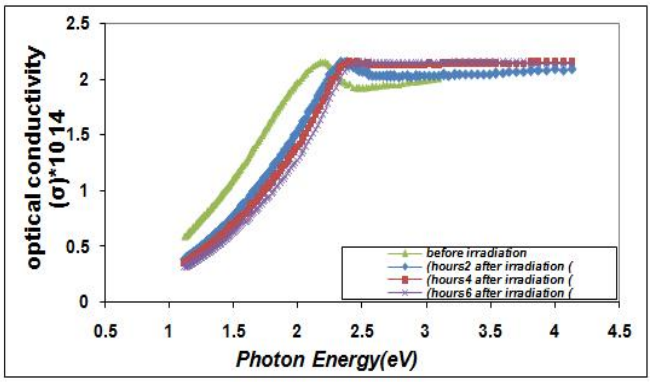

Figure 11. Variation optical conductivity & photon energy for cadmium oxide thin films

We can notice from this figure that the optical conductivity value decreases after irradiated with gamma ray at lower photon energy followed increase this values with increasing irradiation time at higher photon energy. This behavior is due to the optical conductivity value depends mainly on refractive index and absorption coefficients according to the equation (15). The value of the optical conductivity is a good agreement with refs. (Majid H. Hassouni *et al*., 2013).

Conclusion

The present study determines the effect of Gamma –Ray irradiation on the structural and optical properties of CdO films.XRD pattern confirms the formation of CdO phase with preferential orientation along (111) plane. Structural analysis results demonstrate that increase the degree of crystallinity and grain size as we exposed the samples to Gamma –Ray. The Optical studies carried out on the films reveal that the absorbance values decrease while the transmittance values increases after irradiation, the average transmittance is more than 70% for wavelengths in the (500–1100) nm range, indicating these films have high transmittance in the NIR region, which makes it suitable for solar energy collection. It also shows that all films prepared before and after irradiation have high values of absorption coefficient ($\alpha > 10^4$ cm^{-1}) and band gap increases with increase irradiation time. The values of all optical constant (absorption coefficient, refractive index, extinction coefficient, dielectric constant, and optical conductivity) affected after irradiated with gamma ray.

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