Thickness and gamma-ray effect on physical properties of CdO thin

films grown by pulsed laser deposition

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Abstract

Polycrystalline Cadmium Oxide (CdO) thin films were prepared using pulsed laser deposition onto glass substrates at room temperature with different thicknesses of (300, 350 and 400)nm, these films were irradiated with cesium-137(Cs-137) radiation. The thickness and irradiation effects on structural and optical properties were studied. It is observed by XRD results that films are polycrystalline before and after irradiation, with cubic structure and show preferential growth along (111) and (200) directions. The crystallite sizes increases with increasing of thickness, and decreases with gamma radiation, which are found to be within the range (23.84-4.52) nm and (41.44-4.974)nm before and after irradiation for thickness 350nm and 400nm respectively. The dislocation density, microstrain and number of crystallites per unit surface area, decreases with increasing of thickness, while they increases with gamma radiation. From the atomic force microscope (AFM), the grain size of CdO films decrease from 96.69nm before radiation to 89.49 nm after gamma radiation and RMS roughness increases for the irradiated sample from 4.26nm to 4.8nm, increase in the surface roughness is advantages as it increases the efficiency of the CdO solar cells. The optical properties for thin CdOfilms with different thickness before and after gamma irradiation have been determined and reveals direct energy gap. It is decrease with the increase of thickness, while it is increase after gamma irradiation. These films a promising candidate for the window layer in solar cells and other possible optoelectronic application.

Keywords

Cadmium oxides, thin films, pulsed laser deposition, XRD, AFM, optical properties.

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تأثير السمك وأشعة كاما على الخصائص الفيزيائية لاغشية CdO الرقيقة المحضرة بطريقة الترسيب بالليزر النبضي أيمان مزهر ناصر¹، اقبال سهام ناجي¹، هناء ابراهيم محمد² ¹قسم الفيزياء، كلية العلوم، جامعة بغداد، بغداد، العراق ²قسم الفيزياء، كلية التربية للعلوم الصرفة ابن الهيثم، جامعة بغداد، بغداد، العراق الخلاصة

حضرت أغشية أوكسيد الكادميوم (CdO)الرقيقة باستخدام الترسيب بالليزر النبضي على ارضيات زجاجية في درجة حرارة الغرفة بسمك مختلف من (300، 350 و 400) نانومتر، شععت هذه الأغشية بمصدر اشعة كاما السيزيوم 137 (Cs-137). درس تأثير السمك والتشعيع على الخصائص التركيبية والبصرية. لوحظ من خلال نتائج XRD بان الاغشية ذات تركيب متعدد البلورات قبل وبعد التشعيع، وذات تركيب مكعب وتظهر نمو مفضل على طول الاتجاهين (111) و (200). تزداد الحجوم البلوريةمع زيادة السمك، وتتناقص مع زيادة أشعة كاما، ضمن المدى (23.84-4.52) نانومتر، و(4.41.4-4.44) نانومتر قبل وبعد التشعيع لسمك 2000 نانومتر على التوالي، ان كثافة الانخلاعات، الإجهاد المايكروي، وعدد البلورات في وحدة المساحة السطحية يتناقص مع زيادة السمك، في حين أنها تزداد مع تعرضها لأشعة كاما. من خلال مجهر القوة الذرية (AFM)، يقل الحجم الحبيبي للاغشية OCO من 96.69 نانومتر قبل التشعيع إلى 89.49 نانومتر بعد التشعيع باشعة كاما وتزداد الخشونة RMS للعينة المشععة من 4.26 نانومتر إلى 4.8 نانومتر، والزيادة في خشونة السطحية يتناقص مع زيادة السمك، في حين أنها تزداد مع تعرضها لأشعة كاما. من خلال مجهر القوة الذرية (AFM)، يقل الحجم الحبيبي للاغشية OCO من 96.69 نانومتر قبل التشعيع إلى 89.49 نانومتر بعد التشعيع باشعة كاما وتزداد الخشونة RMS للعينة المشععة من 4.26 نانومتر إلى 4.8 نانومتر، والزيادة في خشونة السطح من المزايا التي تزيد كفاءة OCD كخلايا شمسية. وقد تم تحديد الخصائص البصرية لاغشية أوكسيد الكادميوم (CdO) الرقيقة مع سمك مختلف قبل وبعد التشعيع بأشعة جاما والتي بينت فجوة الطاقة المباشرة، والتي تقل مع زيادة السمك، وتزداد بعد تعرضها لأشعة كاما. هذه الأغشية مرشح واعد للطاقة المباشرة، الشمسية و غير ها من التطبيقات الكهروبصرية.

Introduction

Transparent conducting oxides have been intensively investigated in recent years owing to their high optical transmittance and low resistivity. According to these properties it has application spacious such as transparent conducting electrodes, gas sensors, surface acoustic wave device, varistors, etc. [1]. On the other hands, CdO is a candidate material for solar cell applications due to their high quality electro-optical properties in the visible solar spectrum part [2]. In addition, it has a great potential use for optoelectronic devices. Cadmium oxide is a transparent oxide in the visible and NIR spectral region. It is a degenerate, n-type semiconductor with a high electrical conductivity $(10^{-4}-10^{-2})$ $\Omega \cdot cm$), and with a direct and indirect band gap of 2.5, 1.98 eV respectively [3]. Undoped CdO thin films have been prepared by different methods such as molecular-beam epitaxy, spray pyrolysis, radio frequency magnetron sputtering, pulse laser deposition, the sol-gel technique, etc.[4-9]. Highenergy radiations, such as γ -rays, change the physical properties of the materials they penetrate, and these changes are strongly dependent on the structure of the absorbed substances. It is thinks that the exposure to ionizing radiation (γ -ray) causes defects in structure (called color centers or oxygen vacancies in oxides). The effect of radiation based on both the

parameters of the films including their thickness and the dose: the degradation is more severe for the thinner films and the higher dose. The absorption of γ radiation in the thin films depends upon their electronic structure which in turn alters by the interaction with photons [10]. In this paper, the pulsed laser deposition technique was used. The main advantage of pulsed laser deposition (PLD) is the ability to create high-energy source particles, permitting the growth of high quality film at relatively low substrate temperature. Hence, the aim of the present work is to study the effect of thickness and Gamma radiation on the structural, morphology and optical properties of cadmium oxide thin films growth by PLD.

Experimental

High purity (99.99%) Cadmium Oxide powder was used as a source for deposition of CdO films on glass substrates using pulsed laser deposition using Nd:YAG laser with λ =1064 nm, average frequency 6 Hz and pulse duration 15 ns at different thickness of 300, 350, 400nm. All samples were irradiated by Cs-137 source which emitted gamma ray with energy of 661.66 eV and intensity of 85.01,the total doses was 3.164 Sv, where the time of irradiation is two days. The structure of the prepared thin CdO films was obtained using the X-ray diffraction techniques using radiation from CuKa radiation target and using XRD 6000 SHIMADZU diffractometer. The grain size and average roughness of CdO thin films been measures have by AFM (AA3000), (Scanning probe microscope (SPM)) at thickness of 400nm before and after gamma irradiation. The optical transmission spectra of the deposited thin films measured **UV-VIS** were by

spectrophotometer (Optima-3000), the optical constant was calculated at the wavelength range 300-1100 nm.

Results and discussion X-ray diffraction

X-ray diffraction of the CdO thin films at different thicknesses (300, 350, 400) nm before and after irradiation by gamma radiation are display in Fig. 1(a-f).

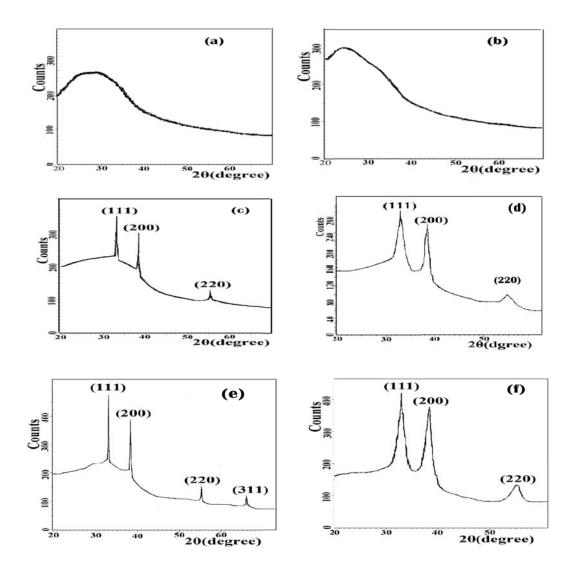


Fig. 1: X-ray diffraction pattern of the CdO thin films at: (a) 300nm before radiation (b) 300 after radiation (c) 350 before radiation (d) 350 after radiation (e) 400 before radiation (f) 400 after radiation.

XRD pattern for prepared CdO thin film of thickness 300nm, indicate that the film was amorphous before and after irradiation as in Fig.1 (a-b), and films become polycrystalline the before and after irradiation, with an fcc cubic structure typical of CdO (PDF card No: 005-0640) with increasing thickness. All the films indicate a preferred orientation along (111) and (200) planes as in Fig. 1(c-f). XRD studies obvious that the structure of these films was became nanostructure on irradiation with a gamma radiation. It can be also observed that the

diffraction patterns exhibits peaks represent to (220) and (311) plane which appear with low intensities as compared to that of (111) and (200) planes. The intensity of the (111), (200) and (220) peaks decreases, and all peaks are more broadened with increasing gamma radiation, as shown in Fig.1(a-f), and this is an agreement with other literatures [1,9]. The shift in the diffraction peak position towards lower diffraction angle compared to CdO films before irradiations as in Table 1.

 Table 1:XRD results for CdO thin films at different thickness before and after irradiation.

Thickness (nm)	2θ(exp.) (degree)	2θ(stand.) (degree)	hkl (stand.)	d(exp.) (Å)	d(stand.) (Å)	D(nm)	δx10 ¹³ line/m ²	εx10 ⁻⁴	N _o *10 ¹¹ atom/cm ²
350 before	33.5	33	111	2.672	2.712	23.84	175.865	13.121	29
radiation	38.5	38.283	200	2.336	2.349				
	55.4	55.256	220	1.657	1.661				
350 after	33	33	111	2.712	2.712	4.52	4892.5	69.23	4328
radiation	38	38.283	200	2.365	2.349				
	54.8	55.256	220	1.662	1.661				
	33	33	111	2.712	2.712	41.44	58.212	7.551	7
400 before	38.3	38.283	200	2.348	2.349				
radiation	55.2	55.256	220	1.662	1.661				
	65.9	65.908	311	1.416	1.416				
400 after	33.1	33	111	2.704	2.712	4.97	4040.7	62.918	4061
radiation	38.3	38.283	200	2.347	2.349				
	55.2	55.256	220	1.662	1.661				

For all samples, which indicates a slight lattice shrink attributed to the decreasing the diameter of crystal, and the displacement of diffraction peaks is indication increase an to the dislocation density and strain as in Table 1. Also the intensity of peaks increases with increasing of thickness, and decreases with gamma radiation. This increase in intensity is indication of improvement in crystallinity, while the decrease in intensity is due to creation or annihilation of defects and recrystallization.

The average grain size (D) of the CdO structures before and after irradiation

by gamma radiation was calculated from the full width at half maximum (FWHM) of the prominent peak, using Debye- Scherer equation [9]:

$$D = \frac{K\lambda}{\beta\cos\theta} \tag{1}$$

where $\lambda = 1.54$ Å is the wavelength of the X-ray radiation, θ is the angle of diffraction and β is the FWHM in radians of the XRD peak. The crystallite sizes increases with increasing of thickness, and decreases with gamma radiation, which are found to be within the range (23.84-4.52) nm and (41.44-4.97) nm before and after irradiation for thickness 350 and 400

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respectively, and these are summarized in the Table1. The increasing in grain size is attributed to improvement of crystal structure, while the decreasing in grain size is due to the gamma radiation induced recrystallization and transform the structure of these films to the nanostructure.

The dislocation density (δ), microstrain (ϵ) and the number of crystallites per unit surface area (N) of the prepared CdO thin films before and after irradiation can be calculated from the following equations respectively [11-13]:

$$\delta = \frac{1}{D^2} \tag{2}$$

$$\delta = \frac{15\varepsilon}{Ad} \tag{3}$$

$$N = \frac{t}{D^3} \tag{4}$$

The dislocation density, microstrain and number of crystallites per unit surface area, decreases with increasing of thickness, while they increases with gamma radiation which was demonstrated the improvement of the crystallinity in the films with thickness, and increases the disordered with radiation that means the radiation creates defects in the structure of films and this is indicates the change in the microstructure of the film due to gamma radiation, as shown in Table 1, and this is agreement with researches [12-14].

Atomic force microscope (AFM)

The grain size and average roughness of CdO thin films prepared by pulse laser deposition have been measures by atomic force microscope (AFM).

Fig. 2 shows the two and three dimension AFM images (of area 2060-2041nm) for CdO films before and after gamma irradiation at thickness 400nm. The results showed that the thin films exhibit a uniform surface morphology over the complete substrate and that the films are of good quality. The grain size of CdO films decrease from 96.69nm to 89.49 nm after gamma radiation as shown in Table 2.

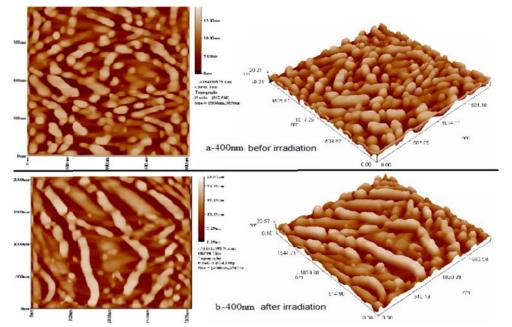


Fig. 2: The two and three dimension AFM images for CdO films (a-before and b-after) gamma irradiation at thickness 400nm.

		10011111		
Thickness (nm)	Average diameter(nm)	RMS(nm)	Peak to peak (nm)	Average roughness
400 before	96.69	426	15.8	3.68
radiation				
400 after	89.49	4.8	18.4	4.16
radiation				

Table 2: The effect of gamma radiation on the AFM parameters of CdO films at thickness400nm.

This may be due to gamma radiation induced recrystallization process in these films. RMS roughness increases for the irradiated sample from 4.26-4.8nm, due to the increase in cracks dislocations by the and gamma irradiation. The grains appeared more packed after irradiation, because the grains melt and then re-growth, which is clearly seen from Fig.2b. The efficiency of the CdO solar cells increases with increasing the surface roughness, because the rougher surface

will promote multiple surface reflections by reducing the energy loss due to reflectance. The results were agree with the literature references[1,15].

Optical properties

The transmittance of the thin CdO films deposited by pulsed laser deposition with different thickness (300, 350and 400) nm before and after gamma irradiationare shown in Fig. 3.

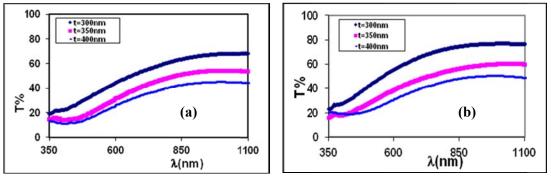


Fig. 3: The transmittance of thin CdO films at different thickness (300, 350, 400)nm:(a) before and (b) after gamma irradiation.

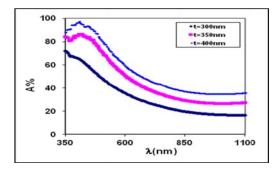
All spectra show good transparency. The transmittance decreases at the spectral region of fundamental absorption. In this region the incoming photons have sufficient energy to excite electrons from the valence band to the conduction band and thus these photons are absorbed within the material to decrease the transmittance. The transmittance pattern of all deposited thin films increased with increasing wavelength (λ). On the other hand the transmittance decreases with the increase of thickness and increases slightly with gamma irradiation as in Table 3.

Table 3: The effect of gamma radiation and thickness on the T, A and R of CdO films.

Before radiation: λ =800nm				After radiation: λ =800nm			
Thickness (nm)	Т%	A%	R%	Т%	A%	R%	
300	60.88	21.55	17.56	72.61	13.90	13.48	
350	48.39	31.52	20.08	54.06	26.71	19.22	
400	40.55	39.20	20.24	46.29	33.45	20.25	

The decrease in transmittance means increase in the reflection and absorption. It is obvious from the figures that the transmittances decrease in the range of the wavelength (600-800) nm and reaching the maximum value at the range (800-1100) nm. On the other hand the transmittance decreases with thickness which means increase in the reflection and absorption. For 300nm thickness the transmittance equal to 60.88% and the lowest transmittance at 400 thicknesses is 40.55% before irradiation and 72.61% and 46.29% after irradiation, and this agreed with [12-15].

The Absorbance for the thin CdO films deposited by pulsed laser deposition with different thickness (300, 350 and 400)nm before and after gamma irradiation are shown in Fig. 4.



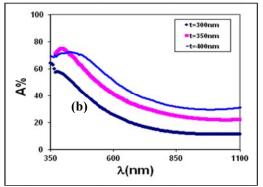
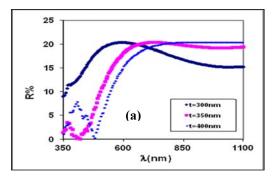


Fig. 4: The absorbance of thin CdO films at different thickness (300, 350, 400) nm: (a) before and (b) after gamma irradiation.

It is obvious from this figure that as the wavelength increases the absorbance decrease and also as the thickness increased the absorbance increased, and it is decreased with gamma radiation as in Table 3, and this is agreement with [3]. Fig. 5 represent the reflectance of thin CdO films deposited by pulsed laser deposition with different thickness (300, 350 and 400)nm before and after gamma irradiation.



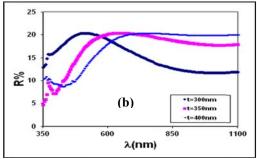


Fig. 5: The reflectance of thin CdO films at different thickness (300, 350, 400) nm: (a) before and (b) after gamma irradiation.

The band gap energy values for CdO samples have been calculated by using Tauc equation. We have examined $(\alpha hv)^2$, $(\alpha hv)^{3/2}$, $(\alpha hv)^{1/2}$ and $(\alpha hv)^{1/3}$ versus photon energy (hv) and select the optimum linear part from the equation[15]:

$$\alpha h \upsilon = A (h \upsilon - E_g)^r \tag{5}$$

where hv, A and E_g are photon energy, constant and optical band gap, respectively.

It is found that the first relation yields linear dependence, which describes the allowed direct transition. Then E_g is determined by the extrapolation of the portion at α =0. Fig. 6 reveals that the values of direct optical energy gap, in

general, and before irradiation it is decrease with the increase of thickness from 2.2-1.9eV for thickness 300 to 400nm, whereas after irradiation, the energy gap decreases from 2.4-1.98eV with increasing thickness from 300 to 400nm. These values are tabulated in Table 4.

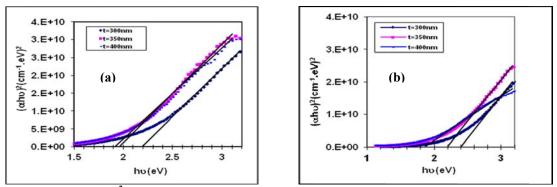


Fig. 6: The $(ahv)^2$ as a function of hv for thin CdO films at different thickness (300, 350, 400)nm :(a) before (b) after gamma irradiation.

Table 4: The optical parameters of thin film of different thickness (300, 350, 450)nm:before and after gamma irradiation.

Before radiation: λ =800nm							
Thickness	$\alpha \times 10^4$	Eg	n	k	ε _r	ε	
(nm)	$(cm)^{-1}$	(eV)					
300	1.65	2.20	2.437	0.105	5.930	0.513	
350	2.07	1.95	2.616	0.132	6.828	0.691	
400	2.26	1.90	2.627	0.143	6.881	0.754	
After radiation: λ =800nm							
300	1.07	2.40	2.158	0.067	4.653	0.293	
350	1.76	2.20	2.556	0.111	6.520	0.571	
400	1.93	1.98	2.630	0.122	6.900	0.644	

In generally, the values of energy gap were increase after gamma irradiation as in Table 4. These films a promising candidate for the optoelectronic applications and window layer in solar cells [13-15].

The absorption coefficient α is calculated using the formula [3,15]:

$$\alpha = \frac{2.303A}{t} \tag{6}$$

where A and t are absorption and film thickness, respectively. The variation of absorption coefficient α with wavelength before and after irradiation is shown in Fig.7.

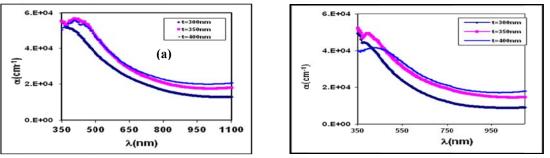


Fig. 7: The absorption coefficient as a function of wavelength for thin CdO films at different thickness (300, 350 and 400)nm: (a) before and (b) after gamma irradiation.

Both films show higher absorption on the shorter wavelength side, this is attributed to increase the defect states which lead to increase absorption coefficient. The values of absorption coefficient increase are with incresesing thickness and decrease with gamma radiation as in Table 4. Refractive index (n) is one of the fundamental properties for an optical material, because it is closely related to the electronic polarizability of ions and the local field inside materials. The evaluation of refractive index of optical material is important for many

applications especially in optical devices, and the refractive index can be determined from the reflectance (R) and extinction coefficient (k) uses the relation [16]:

$$n = \left(\frac{4R}{(R-1)^2} - k^2\right)^{\frac{1}{2}} - \frac{(R+1)}{(R-1)}$$
(7)

The variation of refractive index with wavelength for thin CdO films at different thickness and gamma irradiation are shown in Fig. 8 and Table 4.

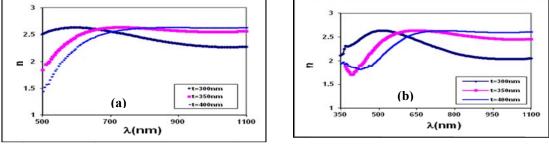


Fig.8: The refractive index as a function of wavelength for thin CdO films at different thickness (300, 350 and 400)nm: (a) before and(b) after gamma irradiation.

It is observed that n increases with increasing thickness and decreases with irradiation as a result to reflectance increasing with thickness and decreasing with irradiation. The peak of refractive index shifts to high wavelength with increasing thickness.

The extinction coefficient (k) represents the imaginary part of complex refractive index and it can be defined as the amount of energy losing

as a result of interaction between the light and the charge of medium, and it can be determined by using the equation [13,14]:

$$k = \frac{\alpha \lambda}{4\pi} \tag{8}$$

The relation between the extinction coefficient and wavelength is shown in Fig. 9.

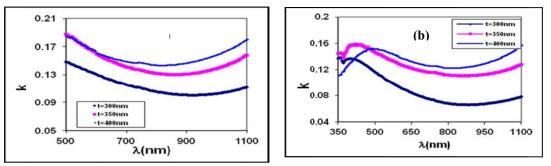


Fig. 9: The extinction coefficient as a function of wavelength for thin CdO films at different thickness (300, 350 and 400)nm: (a) before and (b) after gamma irradiation.

It is found that the extinction coefficient increases with increasing thickness and decreases with radiation as in Table 4, because it is behavior depend on absorption coefficient.

Two parts of the dielectric constant, real and imaginary parts (ε_r , ε_i), respectively were obtained using the formula [17]:

$$\varepsilon_{\rm r} = n^2 - k^2 \tag{9}$$

$$\varepsilon_i = 2nk \tag{10}$$

The variation of ε_r and ε_i parts as a function of wavelength in the range of 300–1100 nm before and after gamma irradiation are shown in Figs. 10 – 11, respectively.

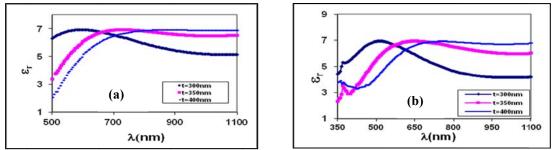


Fig. 10: The (ε_r) as a function of wavelength for thin CdO films at different thickness (300, 350 and 400) nm: (a) before and (b) after gamma irradiation.

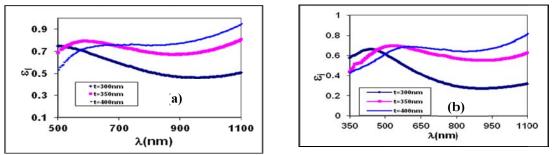


Fig. 11: The (ε_i) as a function of wavelength for thin CdO films at different thickness (300, 350 and 400) nm: (a) before and (b) after gamma irradiation.

The behavior of ε_r is similar to that for the refractive index because of the smaller value of k^2 compared with n^2 according to above equation. The imaginary part of dielectric constant (ε_i) reveals same behavior of extinction coefficient (mainly depends on the k values) with the variation of wavelength. thickness and gamma radiation. All values optical of constants are tabulated in Table 4.In general, both value of ε_{r} and ε_{i} increased with increasing of thickness. Also, it is clear from these figures and Table 4, that both the real and the imaginary parts are decreased after irradiated by gamma ray, also the values of the real part are higher than those of the imaginary part. The same explanation of n and k can be given to ε_r and ε_i respectively. From these figures, it is clear that the peak of ε_r and ε_i of all the prepared samples shift slightly to the higher wavelength (lower energy) with increasing of thickness.

Conclusions

CdO thin films prepared using pulsed laser deposition were irradiated with gamma radiation for different thickness. There is a significant change in the material characteristics because of influence of thickness and gamma radiation. The study highlights the change in the structural and optical properties due to variation of thickness and irradiation. Changes in the morphology of the films were also noticed in the irradiated thin films. The morphology of the irradiated films reveals increase in surface roughness, which is more desirable to enhance the solar cell efficiency. Direct energy gap has been noticed by studied the optical properties and these values of direct optical energy gap is decrease with the increase of thickness, while it is increase after gamma irradiation. Higher band gaps are always required for TCO films used in solar cell applications. Also it can conclude that thickness and gamma irradiation were affecting all the parameters under investigation.

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