The Optical Properties of Aluminum Doped CdO Thin Films Prepared by Vacuum Thermal Evaporation Technique

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Abstract

In this work, thin films of undoped and Al-doped CdO with (0.5, 1 and 2) wt.% were prepared by using thermal vacuum evaporation on glass substrate at room temperature. The optical absorption coefficient (α) of the films was determined from transmittance spectra in the range of wavelength (400-1100) nm. The spectral transmission and the optical energy band gap decrease from 75% and 2.24 eV to 20% and 2.1 eV respectively depending upon the Al content in the films, also our studies include the calculation of the optical constants (refractive index, extinction coefficient, real and imaginary part of dielectric constant) as a function of photon energy. It is evaluated that the optical band gap of the CdO film could be controlled by Al-doping. The width of localized states in the optical band gap of the films increases with the increase of Al content. The improvement of the optical constant of Al-doped CdO has potential applications as transparent conducting oxide for different optoelectronic device applications.

Keywords: (CdO: Al) thin films, Thermal evaporation, Optical properties, Absorbance, Optical band gap.

Introduction

Pure Cadmium oxide (CdO) has received considerable attention because of its important properties. It is direct band gap energy with 2.22 eV [1]. Because of its high conductivity with high transparency in the visible region, it has been used in several applications including optoelectronic devices and photovoltaic applications [2-5]. Synthesis of CdO films with different types of doping elements such as Cu [6], Ga [7], F [8], Li-Ni [9], Bi [10], Fe [11], In [12] confirms the possibility of tuning their material properties to be utilized in new applications in optoelectronic devices and sensors. Doping of CdO with metallic ions of smaller ionic radius than that of Cd, like Mg, Sn and Al improves its electrical conduction and decreases optical energy gap [13-15].

A survey of literature reveals that several methods are employed on the preparation and characterization of Al doped CdO thin films grown by spray pyrolysis technique, pulsed laser deposition and sol–gel dip coating method [15-17]. In this work, we have prepared Al doped CdO films by thermal evaporation technique. The effects of Al doping on the optical properties of Al-doped CdO films were investigated.

Experimental details

Al-doped CdO films have been deposited on glass substrate by thermal vacuum evaporation using (Edwards – Unit 306) system with $4.5x10^{-5}$ mbar at room temperature. The thickness of films were determined with (Precisa-Swiss) microbalance by using weighing method and found to be about (300∓10) nm, with deposition rate about (1∓0.1) nm/sec**.** The metal bulk Cadmium thin films were obtained from (Fluka A.G company/Germany), were evaporated in vacuum at room temperature onto cleaned glass substrates with (2.5 x 2 x 1)cm^3 size. The distance between the substrate and the boat is (18) cm. A thin layer of CdO is formed on a chemically deposited Cd thin film through reaction with atmospheric oxygen during heating by (VECTOREEN model) thermal oven for one hour at 400 °C. During the heating process, the color of the Cd films changed from silver-grey at room temperature to a black-brown color at a temperature of oxidation. So that Al atomic percentages doping in the films were (0, 0.5, 1 and 2) %wt. Optical transmission measurements were performed with (UV/Visible 1800 spectrophotometer). The band gap (E_g) and optical constants of the transparent films were determined from the optical transmission spectra.

Results and Discussion

Fig. 1 shows the optical transmittance spectra with wavelength from 400nm to 1100 nm of the CdO thin films pure and doped with Al. The optical transmittance decreases with the increase of Al doping concentration. It is seen that the films are transparent in the visible region. All the films demonstrate 20 to 75% transmittance at wavelengths longer than 550 nm. However, the maximum value of transmittance lying in the NIR spectral region exceeds 75%.The transmittance lies in the range of 75-60% for pure CdO and 0.5%wt of Al-doping, 81and 20-55% for 1-2wt% of Al-doping. It suggests that more doping decreases the transparency due to increased absorption by free carriers [18], which is comparable with the values for the $CdO_{1-x}Al_x$ thin films deposited by Maity et al., [19] using sol-gel process method. Below 550 nm there is a sharp fall in the transmittance of the films, which is due to the strong absorbance of the films in this region.

Absorption spectra of pure and Al doped CdO films are shown in Fig. 2. The absorption spectra of CdO thin film expected to depend mainly on three factors: Oxygen deficiency, surface roughness and impurity centers [20]. As seen in Fig. 2, the CdO films exhibit an absorption edge. Also, we can observe that the absorption edge shifts to higher wavelength for higher Al percentage and it changes with Al doping, suggesting a decrease in the band gap due to Al doping.

The reflectance spectra of the undoped and Al doped CdO films are shown in Fig. 3. It observed that the average reflectance of CdO film increased rapidly in visible region in the range of (400 to 600) nm and then decreases with the increase of wave length from the range of (600 to 1100) nm with the increase in Al doping level. The change in the reflectance of the films suggests that the refractive index of the CdO films is changed with Al doping. The refractive index of the CdO film in the range of (600-1100) nm exhibits the lowest value for 2% Al dopant in the visible range, but the films exhibit the highest value for undoped CdO film. However, the reflectance in NIR spectral region is slightly higher in the doped film compared to the undoped films. The shift of transmittance and reflectance indicates that these are related to the changes in the film characteristics [21].

The ability of a material to absorb light is measured by its absorption coefficient and it is a very strong function of the photon energy and band gap energy [22]. The variation of the optical absorption coefficient with photon energy for various Al doping concentration is shown in Fig.4. The absorption coefficient (α) of a film of thickness (t) can be calculated from the transmittance spectrum using the relation [22]:

$$\alpha = 2.303 \frac{A}{t} \quad \text{......................} (1)$$

Where, A: is the absorbance, which is calculated from the relation {A=log(1/Ʈ)}, (Ʈ) is the transparence. The calculated values of absorption coefficient are in the order of 10^5 cm^{-1}. From Fig. 4 it is shown that in the higher photon energies (shorter wavelengths), the absorption coefficient α exhibits high value which means that there is a large probability of the allowed direct transition, and then α decreases with increase of wavelength. Also, it is shown that absorbance increases with increase of Al-doping concentration and a sharp increase observed near the band gap edge. So, the absorption edge is shifted toward long wavelength region. This variation could be related to the variation of the crystallinity and carrier concentration of CdO film with Al-doping.

The fundamental absorption, which corresponds to electron excitation from valance to conduction band, can be used to determine the nature and value of the optical band gap. The optical band gap of the CdO films is calculated using the expression [23]:

$$(\alpha h\nu) = B(h\nu - E_g)^{\eta} \quad \text{..................} (2)$$

Where, B is constant depending on the type of semiconductor, α [cm^{-1}] is the absorption coefficient, hν is the photon energy and E_g [eV] is the optical band gap. The parameter ɳ is an index related to the nature of the material and which is determined by the optical transition involved in the absorption process, it specifies the allowed direct (ɳ = 1/2) and indirect transition (ɳ = 2) in the electronic band structure. The optical band gap energy E_g was obtained from the intercept on the photon energy axis after extrapolating of the straight line section of the curve of $(\alpha h\nu)^2$ versus (hν) plot as shown in Fig.5 . It was found that the Al doping concentration affects the energy band gap E_g. The direct band gap of pure CdO is 2.24 eV, this value is good agreement with several reports [1, 14, 24], and the film has direct band transitions which is an important characteristic for photovoltaic applications. The shift towards higher or to lower energies depends on the method of film preparation [25]. The Al doped CdO decreases the band gap towards lower energy values. This is due to the increases of the density of localized states in the E_g, or because of the width of localized state in the optical band gap of the films was increased with the increase of Al content, which are consistent with previous report of Al-doped CdO films prepared by sol-gel [18, 19], spray pyrolysis [15] and by r-f sputtering methods [26]. The band gaps for CdO films were found to be 2.2, 2.1 and 2.14 eV for Al with 0.5, 1 and 2 wt.% doped CdO, respectively as shown in Fig. 6 and filled in table (1). The Band gap values are found to decrease with increase of Al doping contents (up to 1%) and then start to increase again with higher Al doping content. Generally one can expect an increase in band gap of CdO thin films when doped with Al due to an increase in carrier concentrations which lead to the Burstein-Moss effect [27]. The direct band gap energy obtained in this study is more consistent to the reported direct band gap values. Therefore, we concluded that both the CdO and Al-doped CdO thin films fall under the class of direct band gap materials.

When electromagnetic radiation stakes a surface, some part is reflected, some is absorbed and some is transmitted. The optical constants fully describe the optical behavior of materials; they are important fundamental properties of matter [28, 29]. The optical properties of an evaporated film depend strongly on the technique of evaporation [30]. Optical constants included refractive index (n), extinction coefficient (k), real part (ε_1) and imaginary parts (ε_2) of dielectric constant. The refractive index (n) can be calculated using following equation[23]:

$$n = \left[\frac{4R}{(R-1)^2} - k^2\right]^{1/2} - \frac{(R+1)}{(R-1)} \quad \text{………………} \textbf{(3)}$$

Where, R: is the reflectance and given by the equation [22]:

$$R + A + T = 1 \quad \text{…………………..} \textbf{(4)}$$

The refractive index dispersion plays an important role in the research for optical materials, because it is a significant factor in optical communication and in designing devices for spectral dispersion. The refractive index values of the films were calculated using Eq (3). The behavior of refractive index is nearly similar to the reflectance spectra, we can observe from Fig. 7. Refractive index increases with the increase in photon energy (decreases in wavelength) indicating that all the films exhibit a normal dispersion behavior in the range (1.127-2) eV corresponding to the wavelength (620-1100) nm range. It is clear that the refractive index increases with the increase of photon energy followed by decreases at the higher photon energy. At lower photon energy, n decreases for all the films reaching the lowest value of ‘n’. The values of ‘n’ for films in NIR region (at λ=1100 nm) was found to be equal to 2.3, 4.1, 4.7 and 7.2 for undoped, 0.5, 1 and 2% Al respectively, ‘n’ increases with the increase of Al dopants in NIR region. The value of ‘n’ obtained for CdO thin film is slightly higher than the value obtained by Manjula et al. [31] for CdO thin film fabricated by a simplified spray technique and it is exactly matches with the value obtained by Mohamed et al. [32] for CdO buffer layer formed by electron beam evaporation technique. Also we can see from Fig. 7 that the refractive index decreases with the increase of the incident photon energy above the E_g value, indicating that all the films exhibit anomalous dispersion. As seen in plotted figure of refractive index, the refractive index decreases with Al dopant in the visible region.

The Extinction coefficient, k (imaginary part of the refractive index), which is related to the exponential decay of the wave as it passes through the medium can be determined by using the equation [33]:

$$\text{…………………..} \textbf{(5)} \; k = \frac{\alpha\lambda}{4\pi}$$

Where, λ: is the wavelength of the incident radiation.

Fig. 8 shows the variation of extinction coefficient (k) as a function of photon energy. The behavior of (k) is nearly similar to the corresponding absorption coefficient (α) because of the extinction coefficient depends mainly on (α) according to the Eq (5); for this reason, we notice the increase of extinction coefficient with the increase of photon energy due to the increase of the absorption coefficient. This means that direct electronic transition happens in these films [34]. Also, it shows that before the E_g values of the films, *k* changes strongly with Al dopant due to the structural changes in the films.

Free electron in metals and free carriers in semiconductors absorb light and alert dielectric functions [33], the fundamental electron excitation spectrum of the films was described by means of a frequency dependent of the complex electronic dielectric constant(ε), and it is defined as [22]:

$\varepsilon = \varepsilon_1 - i\,\varepsilon_2$ ………………. (6)

The real and imaginary parts of the dielectric constant (ε_1 and ε_2) are related to the n and *k* values, and can be calculated by using the equations [35]:

$\varepsilon_1 = n^2 - k^2$ …………..... (7)

$\varepsilon_2 = 2nk$ ………………… (8)

It can be seen in Fig. 9 that the behavior of the real dielectric constant (ε_1) is nearly similar to the corresponding refractive index (n) and that is clear from Eq.(7) because of the small value of (k^2). The curves increase to maximum peak and then they start decreasing as the photon energy increases. All the films show same trend. It is also evident from the figure that ε_1 decreases with the increase of Al dopant in films in visible region, while it increases with Al in the NIR region. Also, the peaks of ε_1 where shift to the lower photon energy with the increase of the Al dopant.

The behavior of (ε_2) with photon energy is nearly similar to the corresponding extinction coefficient, because of ε_2 depends mainly on the extinction coefficient values which are related to the variation of absorption coefficient and that is clear in Fig.10. The peaks of ε_2 for all films shift to the lower photon energy with the increase of Al doping.

The complex dielectric constant (ε) is a fundamental intrinsic material property. The (ε_1) associated with the term that, how much it will slow down the speed of light in the material and (ε_2) which showed how a dielectric absorb energy from electric field due to dipole motion. (ε_1)is the normal dielectric constant and (ε_2) represents the absorption associated of radiation by free energy [33].

The variation of (ε_1) with photon energy indicates that some interactions between photons and electrons in the films are produced in this energy range. (ε_2) is directly related to the density of states within the forbidden gap of semiconductor materials [32].

The ε_1 and ε_2 values of the films change with incident photon energy and Al dopant. Al dopant decreases both ε_1 and ε_2 of the dielectric constant of the films in the visible region, which are consistent with Ilican et al. of Al-doped CdO films prepared by sol-gel [36].

At last, we see that both Fig. 9 and Fig. 10 imply that the values of (ε_1) are higher than that of (ε_2) and follow the almost same pattern. Also, some of the optical constants values are shown in table (1).

Conclusion

The undoped and Al-doped CdO films were deposited by Physical Vapor Deposition in vacuum technique. The optical properties of the CdO films were influenced by Al doping at room temperature. It was observed that absorbance in the visible region is high. Absorption coefficient had been calculated from transmission spectra taken within the wavelength of (400 to 1100) nm. The absorption coefficient obtained of the order of 10^5 cm^{-1} in the higher energy region and the rate of absorption is max near the absorption edge, which is around 550 nm.

The optical band gap values were found to decrease from 2.24 to 2.1 eV with Al doping. The optical constants of the films had been investigated in this work, and they depend on the Al content. Finally, it may be concluded that Al-doped CdO films may be a good candidate for suitable application in various optoelectronic devices as photocell and photodetector.

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Table No.(1):Energy gap, extinction coefficient, refractive index and ε_1, ε_2.

Sample	E_g (eV)	λ (nm)	*k*	n	ε_1	ε_2
CdO (Pure)	2.24	553.571	0.621	6.228	38.402	7.735
0.5 % Al	2.2	563.636	0.607	5.927	34.761	7.195
1 % Al	2.1	590.476	0.597	5.614	31.160	6.703
2 % Al	2.14	579.439	0.573	4.782	22.531	5.546

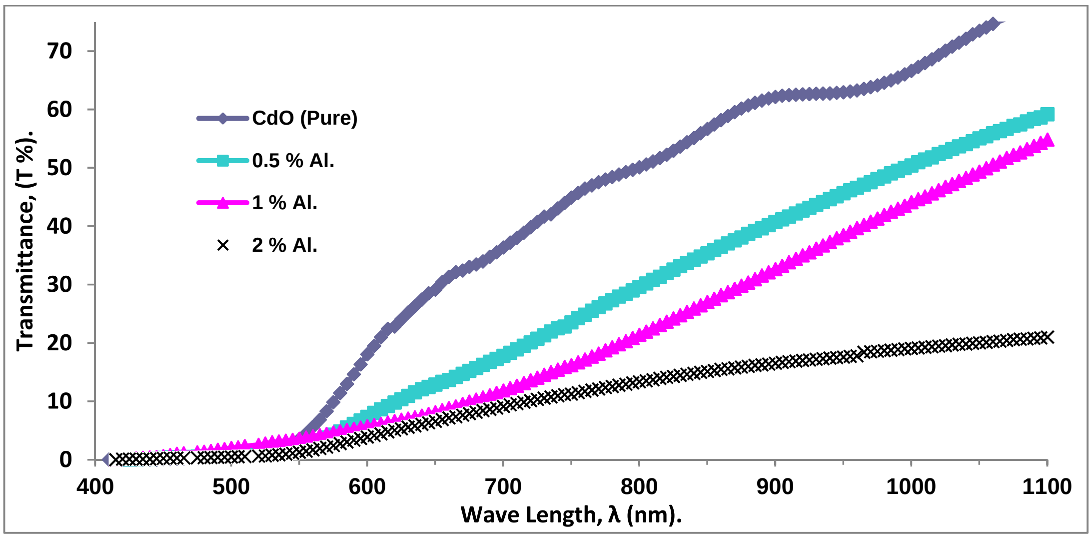


Figure No. (1): Transmittance spectra of Pure and Al doped cadmium Oxide thin films

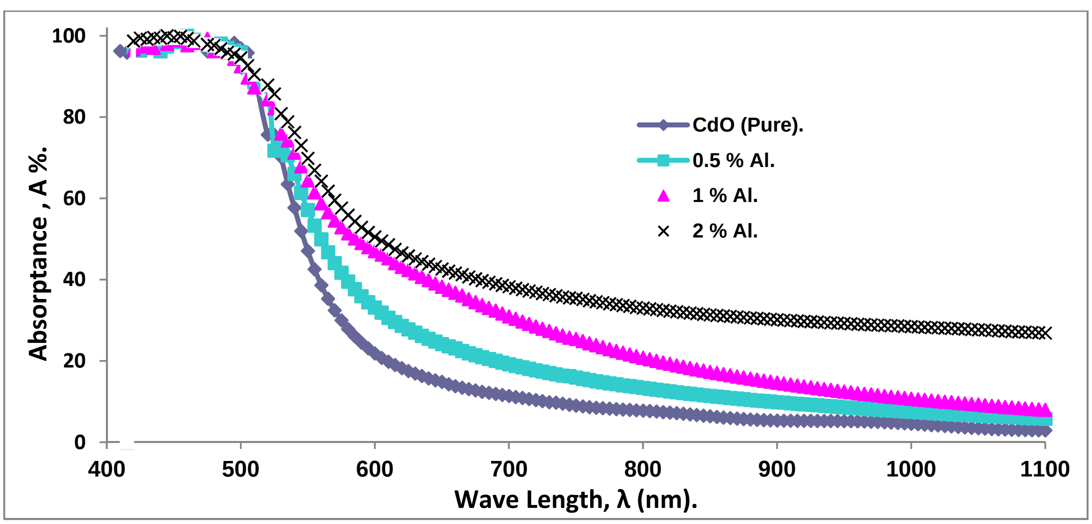


Figure No.(2): Absorption spectra of Pure and Al doped cadmium Oxide thin films

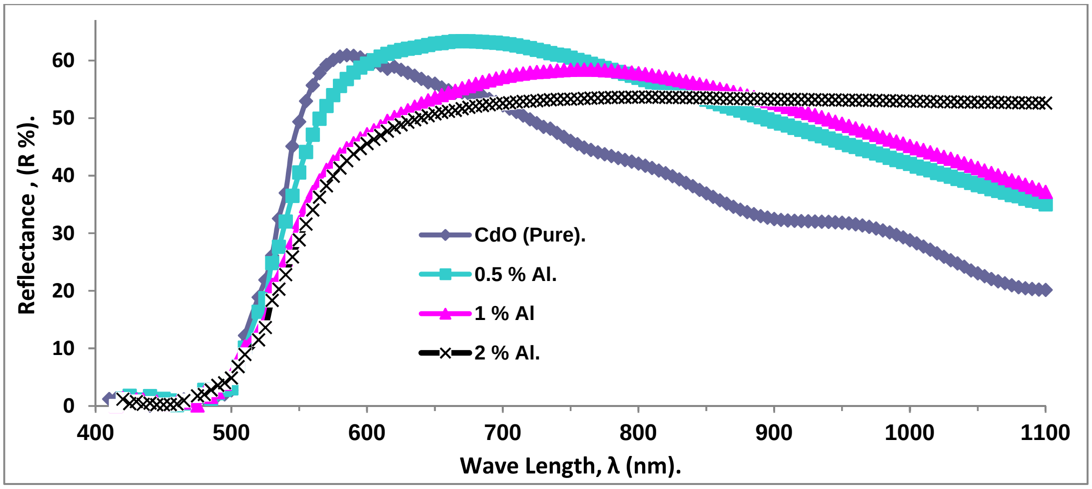


Figure No.(3): Reflectance spectra of Pure and Al doped cadmium Oxide thin films.

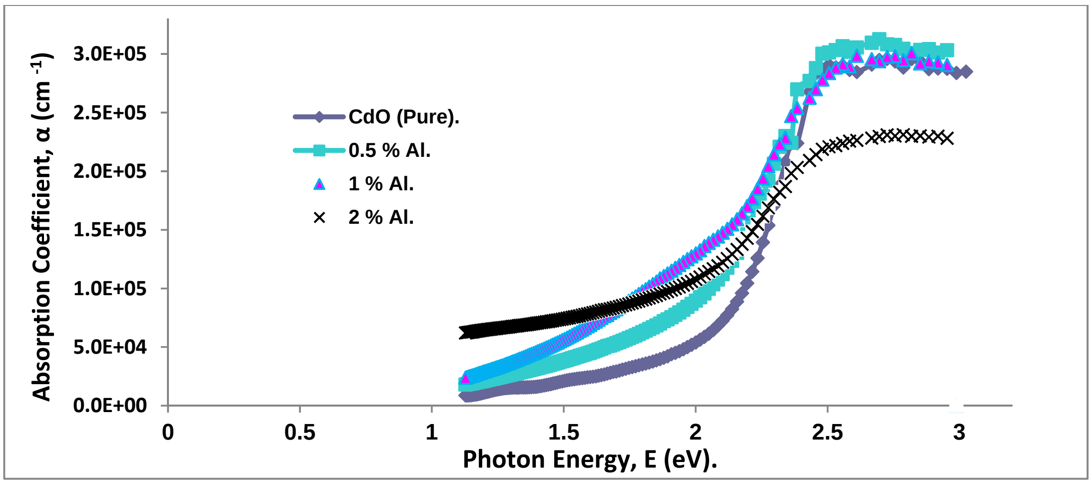


Figure No.(4): Variation of absorption coefficient as a function of photon energy of pure and Al doped CdO thin films

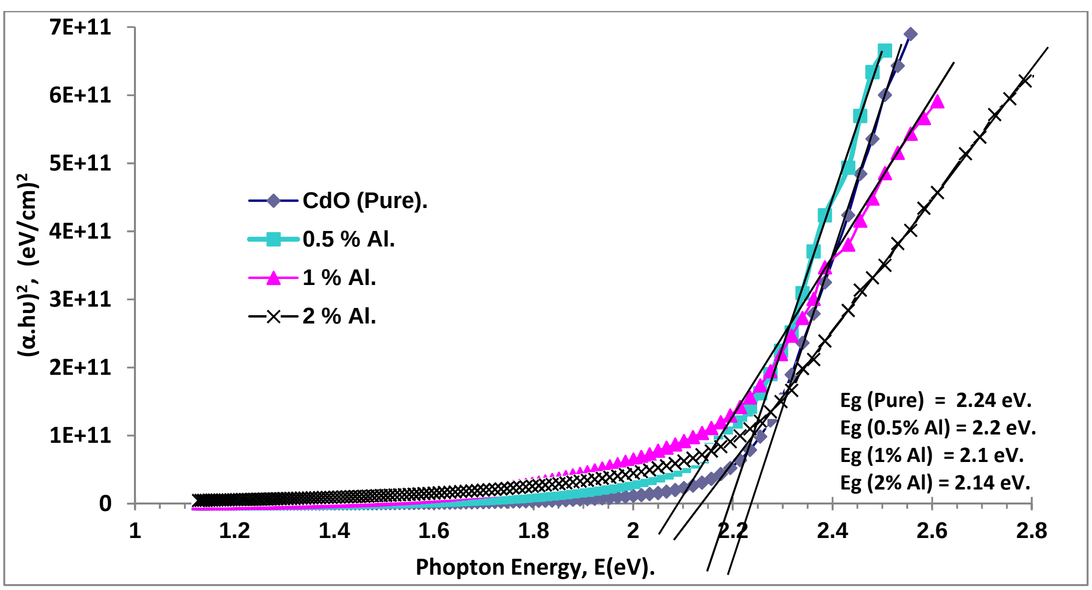


Figure No.(5): Variation of $(\alpha h\nu)^2$ with photon energy of pure and Al doped CdO thin films

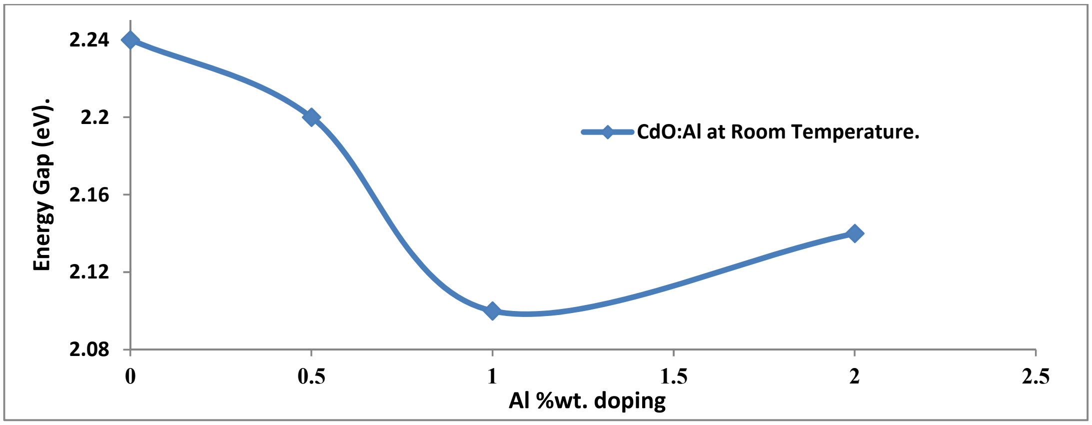


Figure No.(6): Variation of energy gap of CdO:Al as a function of Al concentration

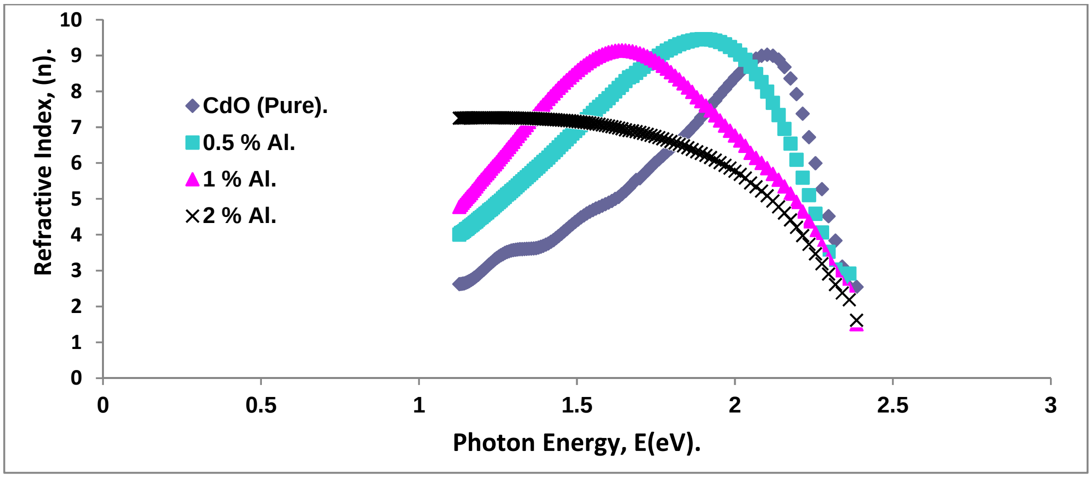
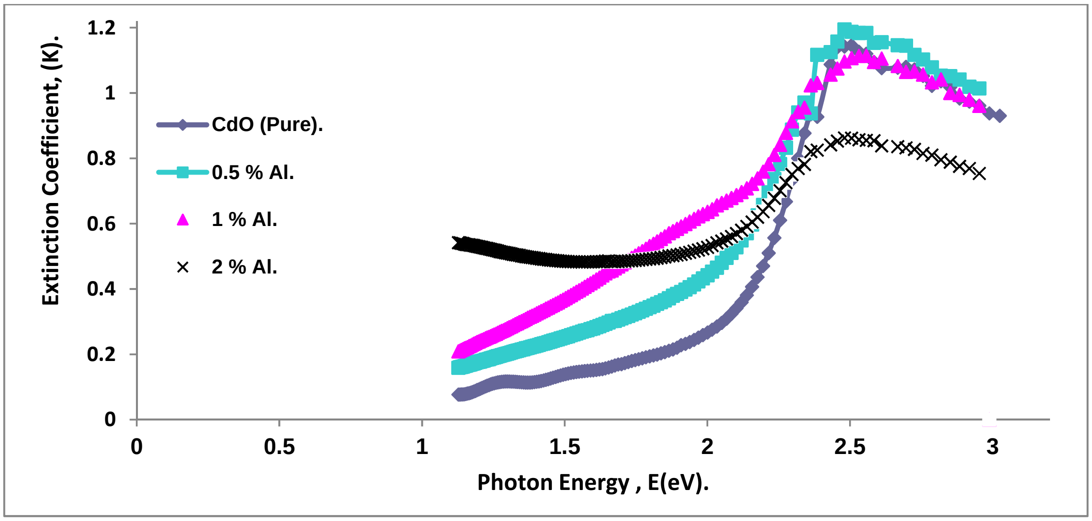


Figure No.(7: Variation of refractive index a function of photon energy of pure and Al doped CdO thin films



8: Variation of extinction coefficient as a function of photon energy of pure and Al doped CdO thin films

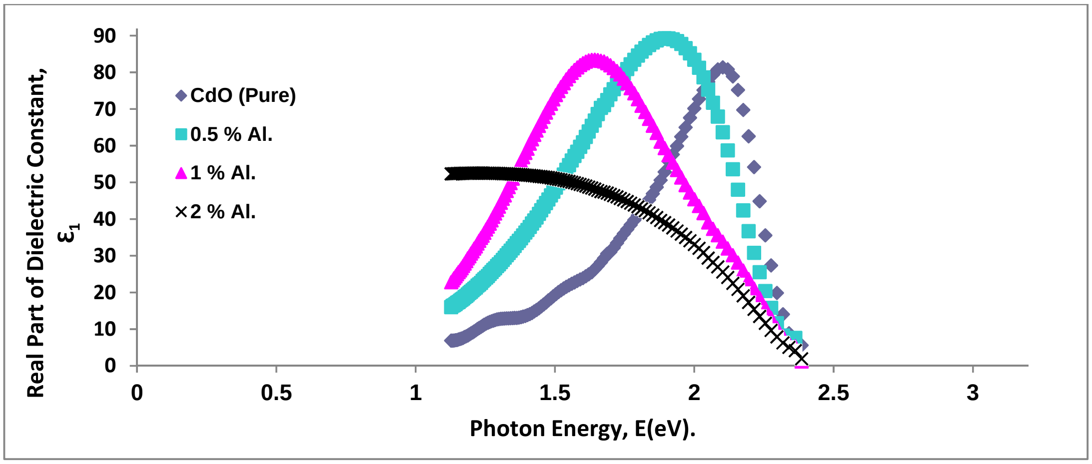


Figure No.(9): Real part of dielectric constant as a function of photon energy of pure and Al doped CdO thin films

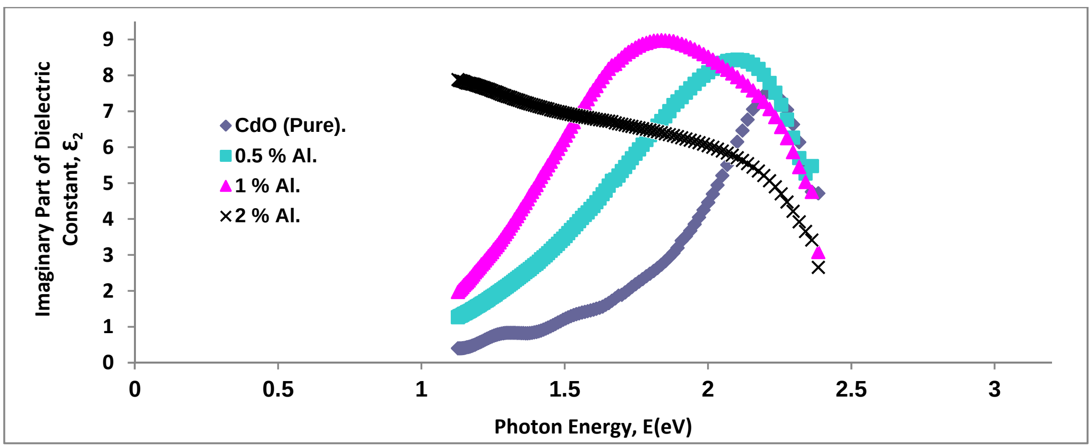


Figure No.(10): Imaginary part of dielectric constant as a function of photon energy of pure and Al doped CdO thin films

الخواص البصرية لأغشية(CdO) النقية و المشوبة ب(Al) والمحضرة بتقنية التبخير الحراري في الفراغ

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الخلاصة

حضرت أغشية أوكسيد الكادميوم (CdO) الرقيقة النقية والمشوبة ب(Al) بنسب تشويب %(0.5, 1, 2) وبإعتماد تقنية التبخير الحراري الفراغي على أرضيات زجاجية بدرجة حرارة الغرفة, وحُسِب معامل الإمتصاص البصري(α) للأغشية الرقيقة من خلال طيف النفاذية بصفته دالة للطول الموجي ضمن المدى nm (1100-400). ومن خلال النتائج وُجِد أن كِلا قيمتي النفاذية وفجوة الطاقة البصرية تتناقصان من 75% و2.24eV إلى 20% و2.1eV على الترتيب, وبالإعتماد على نسبة تشويب الألمنيوم (Al) في الأغشية المحضرة. وكذلك أحتوت الدراسة على حسابات الثوابت البصرية (معامل الإنكسار, معامل الخمود, وثابتي العزل الحقيقي والخيالي) بصفتها دالة لطاقة الفوتون الساقط على الغشاء. وقد وجد ان قيم فجوة الطاقة لغشاء CdO يمكن التحكم بها عن طريق نسبة التشويب بالألمنيوم بسبب زيادة عرض المستويات الموضعية في منطقة فجوة الطاقة البصرية للأغشية. وأظهرت النتائج أن تحسين الثوابت البصرية لأغشية CdO:Al يجعل هذه الأغشية واسعة التطبيقات بصفتها أكاسيد موصلة شفافة في مجال النبائط الالكتروضوئية.

الكلمات المفتاحية: أغشية رقيقة (CdO:Al), التبخير الحراري الفراغي, الخواص البصرية, الامتصاصية, فجوة الطاقة البصرية