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# The Effects of Different Ga Doping on Structural, Optical and Electrical Properties of CdO Films

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Electrical properties

**Abstract:** CdO and CdO:Ga films were produced using the ultrasonic spray pyrolysis method. The structural, optical and electrical properties of produced films were defined by x-ray diffraction pattern (XRD), UV–VIS spectrophotometer and electrical measurements, respectively. The XRD patterns show that all the films deposited at 275 ± 5 °C are in face center cubic polycrystalline structure. The optic band gaps of Ga doped films were obtained with the optical method and observed to change from 2.50 to 2.66 eV with the increase of Ga doping. Additionally, the resistivity, conductivity and I–V characteristics of the samples are investigated with four point and two point probe methods. It is clearly seen that the electrical properties of CdO have improved with the appropriate Ga doping. The best conductivity was obtained with 4% and 8% Ga doping of CdO films.

# CdO Filmlerinin Yapısal, Optiksel ve Elektriksel Özellikleri Üzerine Farklı Ga Katkılamasının Etkileri

**Anahtar Kelimeler**
Kadmiyum oksit,
Galyum katkılama,
Ultrasonik sprey piroliz,
Optiksel özellikler,
Elektriksel özellikler

**Özet:** CdO ve CdO:Ga filmleri ultrasonik sprey piroliz metodu kullanılarak üretilmiştir. Üretilen filmlerin yapısal, optiksel ve elektriksel özellikleri sırasıyla, x ışını kırınım deseni (XRD), UV-VIS spektrofotometresi ve elektriksel ölçümleri ile tanımlanmıştır. XRD desenleri 275 ± 5 °C sıcaklığında biriktirilen bütün filmlerin yüzey merkezli kübik polikristal yapıya sahip olduğunu göstermektedir. Ga katkılı filmlerin optik band aralıkları optik metod ile elde edildi ve Ga katkısının artması ile 2.50 eV'den 2.66 eV'a değiştiği gözlenmiştir. Bunlara ek olarak, örneklerin özdirençleri, iletkenlikleri ve I – V karakteristikleri dört nokta prob ve iki nokta prob metodları ile incelendi. Ga'un uygun katkısı ile CdO'nun elektriksel özelliklerini geliştirdiği açıkça görülmüştür. En iyi iletkenliğin CdO filmlerinin %4 ve %8 Ga katkısıyla elde edildi.

## 1. Introduction

CdO, InO and ZnO thin films showing promising perspectives in modern optoelectronic and photovoltaic devices are known as transparent conductive oxides (TCO) [1]. In recent years, the transparent conductive oxides are attracted attention because of excellent carrier concentration, conductivity close to metallic properties and simple cubic rock salt crystal structure [2,3,4,5]**.** Zn, In, Sn and Cd oxide thin films commonly use photovoltaic solar cells, gas sensors, transparent electrodes, architectural windows, flat panel displays, smart windows, polymer-based electronic [6,7]. The use of indium oxide in TCO applications is limits due to the low avability and high cost of indium, relatively low conductivity and chemical instability [8]. In addition, extensive works have been done on indium, tin, zinc oxides [8,9,10,11]. Due to the relatively low band gap and toxicity, CdO has not been studied as extensively

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as other oxides [12,13]. TCO materials have been studied as binary and multi junction enhanced mobility and the conductivity of materials [5,14].

Although some invesigations have stated that it is not toxic [15], in recent years, the works on the toxicity of Cd have been made [10,16,17]. In these studies, the toxicities of the nanoparticles have been identified as having toxic effects on the biological systems according to the ratio of the increased surface area to the volume of the nanoparticles. Sreekanth et al. [16] reported that the nanoparticles smaller than 100nm easily enters the cells, below 40nm infiltrate into the nuclei and below 35nm the blood brain barrier. Although it is toxic, recently studies on CdO are continuing [16,17]. Because, when CdO is coated with carbon, its toxicity decreases and if CdO applications are industrial waste, it should be ensured that they do not come into contact with people by physical and respiration [16,17].

CdO from transparent conductive oxides is a promising photovoltaic material [1,18,19]. Even if pure, this material has the high electrical conductivity, and the high optical transmittance in the solar spectrum [1,20]. Reddy et al [21] have point out that the optical transmittance and an electrical conductivity of CdO films could be 85% and up to $5 \times 10^4$ $(\Omega\cdot cm)^{-1}$, respectively. CdO can be characterized by very low resistivity ($\sim 10^{-4}$ Ωcm) [22]. The energy band gap values of CdO, a n type degenerate semiconductor, ranges from 2.3 to 2.7 eV [23,24,25]. Two indirect band gaps of these materials are ranges of 1.18 to 1.2 eV and 0.8 to 1.12 eV [25]. CdO possesses the high refractive index ($n_o$ = 2.49) because of high electrical conductivity [26,27]. The optical and electrical properties of CdO with the atomic radius (0,097 nm) and the ion–storey properties can be adjusted.

CdO film can be produced with vacuum evaporation [28,29], dc reactive magnetron sputtering [30], pulse laser deposition (PLD) [31,32], chemical bath deposition [33], metalorganic chemical vapour deposition (MOCVD) [34,35] spray pyrolysis [36,37] and sol–gel technique [38,39]. Among these techniques, the spray pyrolysis technique has lower cost and suitable properties [40]. The spray pyrolysis method is a simple and economic method, and allows the deposition on the large area without vacuum [41]. It's also provides the production of materials in the different band gaps during deposition processes. However, the spraying process is influenced by the changes in the droplet size due to the thermal behaviour depending on the droplet mass. The ultrasonic spray pyrolysis technique possesses the higher deposition rate, more homogeneous particle composition with controlling of particle size and better thickness uniformity deposition over a large area than the spray pyrolysis technique [42]. For these reasons, the ultrasonic spray pyrolysis technique is preferred in this work.

Pure CdO film generally shows low resistivity due to the native defects of oxygen vacancies and the cadmium interstitials. High ionic radius impurities, such as F, In, Ga, while producing solar cells and diodes are suitable for reducing impurity diffusion in the pn junction [43]. Cadmium $Cd^{2+}$ ion (0.097 nm) have a ionic radius longer than that of $Ga^{3+}$(0.062 nm). The Ga dopant compresses the CdO lattice lighter than other dopants and also allows it to be preferred because of its low concentration [43]. Eventually CdO with doping Ga can reduce its resistivity, thereby increasing its conductivity. Moreover, very little work has on gallium-doped CdO films [44,45]. Therefore, the aim of this work has been tried to research the characteristic of CdO films by Ga doping.

## 2. Material and Method

The ultrasonic spray pyrolysis technique were used to depositing the films on microscope glass substrates (10×12×1mm$^3$). Undoped CdO sample was obtained aqueous 0.1 M $Cd(CH_3COO)_2.2H_2O$ and 0.1 M gallium nitrate hydrate $GaN_3O_9.xH_2O$ was used as Ga sources. Doping was achieved by adding $GaN_3O_9.xH_2O$ at different concentrations (2%, 4%, 6% and 8%) to spraying solution. Substrates were heated by an electrical heater formed quartz lamps and up to 275±5 °C.

The optimized experimental parameters such as spray nozzle–substrate distance, spray times, precursor solutions and the pressure of carrier gas were taken stationary as 30 cm, 10 min, 50 ml and 0.2 kg/cm$^2$, respectively. When the spraying process is finished, after the precursor solution, carrier gas and heater were closed, and the glass substrates were left to cooling for 4-5 hours in air atmosphere.

For each doped concentration, the produced films are researched to characterize. The film thicknesses are determined in the range of 178 to 235 nm using PHE-102 spectroscopic ellipsometer (250–2300 nm). The D8 Advance Bruker model x–ray spectrophotometer with CuK$_\alpha$ radiation (1.5406 Å) were used to analyse the crystal structures of samples. The crystallinity levels of films were determined the lattice constant, the texture coefficient and the crystallite size. Shimadzu UV–VIS 1200 Model spectrophotometer is used to analyse the absorption and transmittance spectrums of samples, and used to determine the band gap values of films. I–V measurements are investigated at room temperature under light conditions by four point probe method using Keithley Model 2400 dc device. Hewlett Packard 4140B Model pA Meter/DC voltage source is used to determine the DC conductivities by two point probe method.

**Table 1.** The data obtained from XRD patterns.

| Material | 2θ | d(Å) | a(Å) | TC(111) | D(nm) | $\delta \times 10^{-3}(nm)^{-2}$ |
|---|---|---|---|---|---|---|
| PDF 03-065-2908 | 33.042 | 2.70899 | 4.69180 | - | - | - |
| CdO | 33.1098 | 2.7034 | 4.68935 | 1.67154 | 22.603 | 1.957 |
| CdO:Ga 2% | 33.1683 | 2.69879 | 4.65885 | 1.66324 | 22.607 | 1.957 |
| CdO:Ga 4% | 33.0709 | 2.70652 | 4.68783 | 1.7897 | 24.886 | 1.615 |
| CdO:Ga 6% | 33.0514 | 2.70807 | 4.6905 | 1,638 | 22.599 | 1.958 |
| CdO:Ga 8% | 33.0514 | 2.70807 | 4.6905 | 1.77541 | 24.885 | 1.615 |

## 3. Results

### 3.1. Structural Properties of CdO:Ga Films:

The structural properties of films are investigated with XRD patterns in 2θ range of 20–70°, shown in Fig. 1. It is seen from Fig.1 that the polycrystalline nature of CdO and Ga doped CdO has cubic structure basis of PDF Card No: 03–065–2908 data. Fig. 1 also indicates that the face center cubic CdO and on the CdO:Ga films have the planes of (111), (200), (220) and (311). The preferential orientation in the previously reported works for Ga doped CdO films is indicated as preferential growth along (200) plane. [43]. However in this work, XRD patterns obtained for samples are seen that films have preferential growth along (111) direction. The preferential growth orientation may vary with the nature of doped, minimum surface energy plane and experimental parameters [43]. No peaks of any phase were observed in the films due to the addition of gallium to the calcium oxide films. The shift of 2θ value in the XRD patterns indicates the presence of Ga in the CdO films (Tablo1). Furthermore it is observed that the intensities of all plane were increased by increase of gallium doping up to 4 at%, which then decreased in 6% and again increased in 8%.

For the comparison of the observed and standard d and a values of the deposited CdO and CdO:Ga films are given in Table 1. As shown in Table 1, the lattice constants of pure and Ga doped CdO are very small different from PDF card and decreased with increase in Ga concentration. The very small change in lattice parameter is related to input $Ga^{3+}$ ions into the crystalline structure of CdO. Because $Ga^{3+}$ ionic radius is the smaller than that of $Cd^{2+}$ ion. Thus it is considered that $Ga^{3+}$ ions changes with $Cd^{2+}$ ions in the lattice at lower Ga concentrations. When Ga concentration exceeds at 4%, most of the doped Ga ions are in interstitial of CdO lattice rather than lattice sites [43,45].

The crystalline size is identified by using the following Scherer's formula

$$D = \frac{0,9\lambda}{\beta \cos\theta} \tag{1}$$

where $\beta$ is the full width at half–maxima (FWHM) of a diffraction peak expressed in radians and $\lambda$ is the x–ray wavelength (1.5406 Å). For different Ga concentrations, the crystallite sizes and dislocation densities are tabulated in Table 1. The crystallite size is influenced by the experimental parameters: substrate temperature, spray rate, growth atmosphere, concentration of the solution etc. Therefore, it is difficult to obtain the uniform crystallite size of the films. In Table 1, it is observed that the crystallite size increases by increasing of Ga

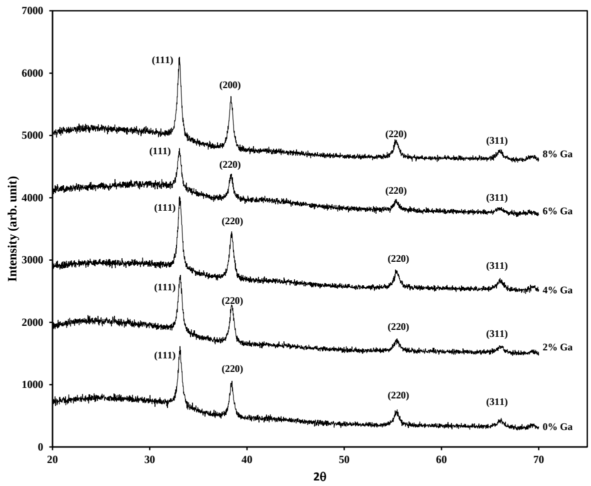


**Figure 1.** XRD patterns of CdO:Ga films.

doped except for CdO film with Ga doped at 6%. The deviation of texture coefficient (TC) from unity indicates the preferred growth. The preferential orientations of films for all the planes are examined using below equation [5]:

$$TC(hkl) = \frac{I(hkl)/I_o(hkl)}{\frac{1}{N}\sum I(hkl)/I_o(hkl)} \tag{2}$$

where TC(hkl) is the texture coefficient of (hkl) plane, I is the measured intensity, $I_o$ is the standard intensity of plane (hkl) taken from the PDF card and N is the number of reflections observed in x–ray diffraction pattern. Fig. 2 is illustrated the variation of TC(hkl) with the gallium concentration for obtained planes. Although TC(200) and other values show nearly stability in the complete range, TC(111) indicates the increase up to 4% and in 8%. If it is in 6%, TC(111) shows quite remarkably decreasing. This indicates the effect of gallium concentration on crystal orientation. TC(111) values for Ga concentrations in range of 0–8% are given at Table 1.

The dislocation density ($\delta$) defines the amount of defects in films. The formula suggested by Williamson and Smallman is used to obtain the dislocation density of films [46];

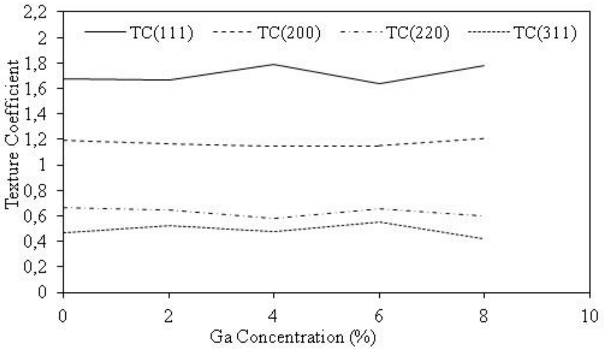


**Figure 2.** Variation of texture coefficient with the gallium concentrations.

$$\delta = \frac{\tau}{D^2} \qquad (3)$$

where $\delta$ is dislocation density and $\tau$ is dislocation factor. For the minimum value of $\delta$, it is taken as $\tau$ =1 [34]. The smaller $\delta$ value, the better crystallization shows [47]. It is also observed that the 4% and 8% Ga doped CdO films have better crystallization on comparison with other (Table 1).

### 3.2. Optical Properties of CdO:Ga Films

The transmission spectra of CdO and CdO:Ga films in Figure 3 is seen that the their transmittances are low at short wavelengths and high at long wavelengths. It is seen in Fig. 3 that the transmittance of CdO film increased from 50% to 72% with Ga–doping. Thus the films behave as an opaque material. This considers that the obtained films exhibit a good optical quality because of low scatter or absorption losses.

The energy values of photons of incident light are enough to exceed the energy band gap, if they equal to or bigger than the band gap values of CdO films. It can be utilized to determine the optical band gap of semiconductor films from the absorbing spectrum. The absorption coefficient obtained from absorbance spectra is used to have the detailed information about energy band gaps. Considering that this transition remains stable at the absorption side, the absorption coefficient can be taken as a function of photon energy of the incident light as;

$$\alpha h\gamma = B(h\gamma - E_g)^n \qquad (4)$$

where α is the absorption coefficient, hγ is the photon energy, $E_g$ is the optical band gap and B is the slope of Tauc edge called the band tailing parameter.

In Equation (4), n =1/2; 3/2; 2; 3 are for a direct allowed transition, a direct forbidden transition, an indirect allowed transition and an indirect forbidden transition, respectively [48,49]. The band gap of film is determined as direct from linearity of the plots of $(\alpha h\gamma)^2$ versus (hγ) and as indirect from linearity of the plots of $(\alpha h\gamma)^{1/2}$ versus (hγ). From plots the band gap is obtained by extrapolating the linear portion of curve to the zero of $(\alpha h\gamma)^2$ or$(\alpha h\gamma)^{1/2}$. Fig. 4 shows

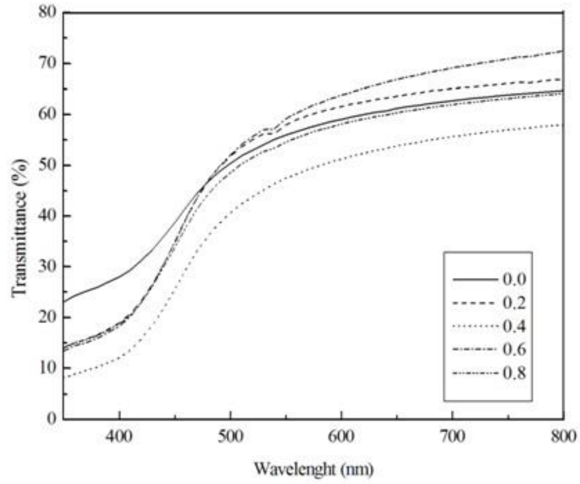


**Figure 3.** The transmittance spectra of CdO:Ga films.

the typical $(\alpha h\gamma)^2$ versus (hγ) graph of cadmium oxide and Ga doped cadmium oxide films, obtained as direct band gap, n=1/2.

In Fig. 4, it is seen that the band gap of cadmium oxide film increases with doping gallium. The band gap of CdO film are slightly smaller than that of CdO:Ga films. The band gap value of pure CdO film is 2.50 eV and further it increases up to 2.66 eV with increasing Ga concentration form 2% to 8%. The increase of band gap of films with Ga contribution can be stated as the blue shift of absorption edge defined by Burstein–Moss (B–M) effect. This shift is that in degenerated n-type semiconductors, the optical absorption edge is shifted to higher energy. But it is seen that the value of band gap decreases in doping 8% in comparison the other doped films (Table 2). The trend of decreasing on $E_g$ may be explained to the band shrinkage effect. This effect occurs as a result of electron–dopant interactions, electron–electron Coulomb interactions and exchange interactions in the conduction band by virtue of increasing of carrier concentration [50,51].

### 3.3. Electrical Properties of CdO:Ga Films

To investigate the electrical properties of films, I–V characteristics, conductivity and resistivity values of films are measured using four–point probe and the two–probe methods at room temperature in light condition, respectively.

It can be injected to semiconductor sufficiently electrons with an applied electric field to contact.

Space charge limited (SCL) current is observed in metal–semiconductor–metal structures with the ohmic contact property. It should be sent to enough carriers for the occurring of SCL current in semiconductors. SCL current varies according to the crystalline defects and impurities. The energy levels of impurities locate in the forbidden band gap, and act as a trap. Traps above and below Fermi energy level are called by the deep trap and shallow trap, respectively. The SCL current behaves differently according to the shallow and deep traps. I–V characteristic of a semiconductor, depending on the expression $I \propto V^m$, shows m = 1 for the ohmic transmission and m = 2 for the SCL transmission characteristic [24]. Figure 5 shows I–V plots of CdO films for different Ga concentrations at room temperature in light condition. I–V plots of films shown in Fig. 5 are achieved with two–probe method. In the two–probe method, the contact is made with silver paste, and the contact–distance and the distance between two contacts for all films are measurement as ~1 mm and as ~5 mm, respectively. It is determined the current values

**Table 2.** The energy band gap values of CdO and CdO:Ga films.

| Films | Doping (%) | $E_g$ (eV) |
|---|---|---|
| CdO | 0 | 2.50 |
| CdO:Ga | 2 | 2.62 |
| CdO:Ga | 4 | 2.63 |
| CdO:Ga | 6 | 2.66 |
| CdO:Ga | 8 | 2.60 |

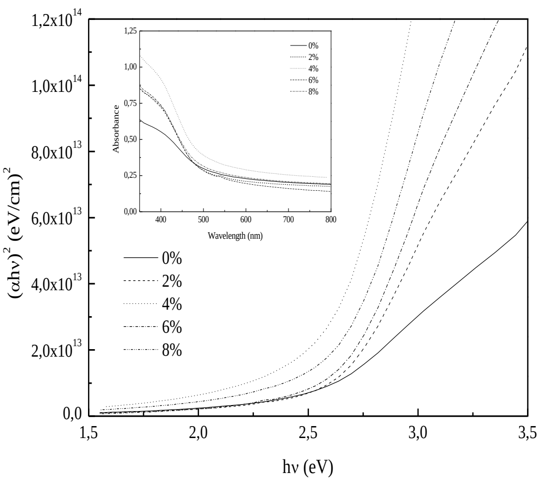


**Figure 4.** $(\alpha h\gamma)^2 \sim (h\gamma)$ plots of CdO:Ga films: Inset: Absorbance spectra of CdO:Ga films.

corresponding to the voltage applied between contacts material with two metal contacts in semiconductor. As can be seen from this figure, the ohmic conduction is dominant in 1–10V voltage range because the slope is smaller than 1. I–V slopes of CdO, 2%, 4%, 6% and 8% Ga doped CdO films possess about to 1. In this ohmic region, more free carriers is injected from metal contact to semiconductor. It was seen from I–V characteristic that these films have not got trapped structure.

It is observed by four–point probe method that the resistivity of all CdO films decreases with doping gallium. It is seen that as gallium is incorporated to CdO films, the electrical resistivity of films generally decreases. This decrease in resistivity with Ga doping is due to increase in grain size, reduction in grain boundary scattering, and thereby increased conductivity. Hence a decrease of donor sites trapped at dislocations and grain boundaries was lead to an enhanced crystallinity. The reason for this can be attributed to the increase in free–electron concentration with gallium doping in CdO film [44]. We obtained that the best conductivity was at 4% and 8% Ga doping of CdO films (Table 3). This can be explained by possessing lower dislocation densities and better crystallizations of these two films than other films. Dakhel was observed the better conductivity in 6% CdO:Ga film [25]. We suggest that this difference is due to the use of different production methods. Also, produced films in Dakhel's study [25] were annealed and so films may be lose some oxygen vacancies, and this can change the conductivity of CdO.

**Table 3.** The calculated values of DC conductivities and resistivity of films using four–point probe method.

| | Four–Point Probe Method | |
|---|---|---|
| Films | ρ(Ωcm) | σ(Ωcm)$^{-1}$ |
| CdO | 3.668x10$^{-2}$ | 0.2276x10$^{1}$ |
| CdO:Ga 2% | 1.123x10$^{-2}$ | 0.8904x10$^{2}$ |
| CdO:Ga 4% | 5.209x10$^{-3}$ | 1.8680x10$^{2}$ |
| CdO:Ga 6% | 6.190x10$^{-2}$ | 1.6136x10$^{1}$ |
| CdO:Ga 8% | 7.986x10$^{-3}$ | 1.2521x10$^{2}$ |

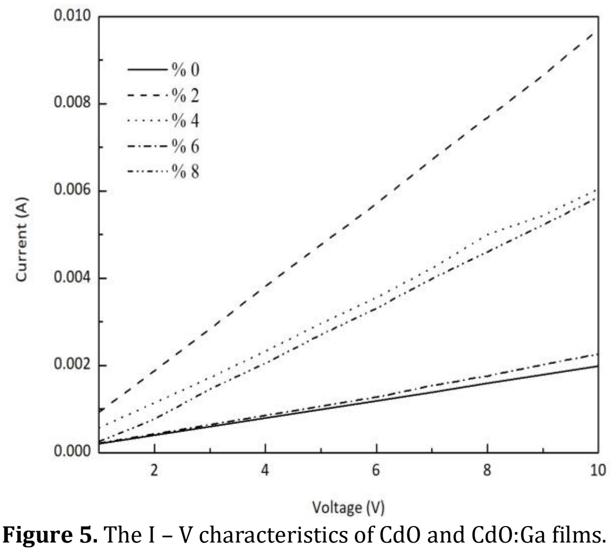


**Figure 5.** The I – V characteristics of CdO and CdO:Ga films.

## 4. Discussion and Conclusion

Ultrasonic spray pyrolysis method was used to produce the pure and CdO:Ga samples. The structural properties of produced films were determined by examining the XRD patterns of films. It has been seen that all films possess the cubic polycrystalline structure. The preferential growth of all films have along the (111) direction and no extra peaks due to the addition of gallium were observed. However it was seen that the intensity of (111) peak increased with increasing Ga concentration doped to CdO. Nevertheless, we observed that the intensities of all peaks decrease on 6 % gallium doping (Fig.1). Ga might also be occupying lattice along with interstitial sites.

The grain size of CdO doped 6% Ga is smaller than others and accordingly it possesses the highest density. In response to the highest transmittance, it was observed that this film (CdO: Ga 6%) has the highest energy band gap. In the analysis of electrical properties of films, gallium doping decreases the resistivity of CdO and correspondingly increases the conductivities of the films. Because this increase in the conductivity can be explained to a decrease in grain size and an increase in grain boundary scattering. The best conductivity was obtained with 4% and then 8% Ga doping of CdO films (Table 3). The obtained results, CdO films doped Ga at 4% and 8% are preferably available as a transparent conducting oxide layer for optoelectronic and photovoltaic devices.

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