

RESEARCH ARTICLE/ARASTIRMA MAKALESİ

SOME PHYSICAL PROPERTIES OF CDS:SN FILMS OBTAINED BY ULTRASONIC SPRAY PYROLYSIS TECHNIQUE

S. KOSE¹, F. ATAY, V. BILGIN ,I. AKYUZ

ABSTRACT

The most used semiconductor window material in heterojunction solar cells is undoped and doped CdS. In this work, Cd_{1-x}Sn_xS (0≤x≤0.4) films were deposited by ultrasonic spray pyrolysis technique at a substrate temperature of 300±5 °C and the effect of Sn incorporation on the film properties is presented. It was determined that the resistivity values of the produced films are between 3.62×10²-1.76×10⁷ Ω cm in dark and air and the high resistivity of CdS films decreased by Sn doping. The investigations on the electrical properties also showed that CdS and Cd_{0.8}Sn_{0.2}S films have a trapped structure. The forbidden energy gaps were found as 2.43-2,44 eV by using the optical method. The transmittance value of CdS film increased when doped with Sn (x=0.2). The surface morphologies of the films were investigated by Metallurgical Optical Microscope and it was determined that the most homogeneous surface was seen in Cd_{0.8}Sn_{0.2}S films. Sn doped CdS films can be used as window materials like CdS films in photovoltaic solar cells.

Keywords: Photovoltaic solar cells, CdS and CdS:Sn films, Ultrasonic spray pyrolysis technique, Electrical, Optical and morphological properties.

ULTRASONİK KİMYASAL PÜSKÜRTME TEKNİĞİ İLE ELDE EDİLEN CDS:SN FİLMLERİNİN BAZI FİZİKSEL ÖZELLİKLERİ

ÖZ

Heteroeklem güneş pillerinde yaygın olarak kullanılan yarıiletken pencere materyali katkısız ve katkılı CdS' dür. Bu çalışmada, Cd_{1-x}Sn_xS (0≤x≤0.4) filmleri 300±5 °C taban sıcaklığında ultrasonik kimyasal püskürtme tekniği ile çöktürülmüştür ve film özellikleri üzerine Sn katkısının etkisi verilmiştir. Üretilen filmlerin özdirenç değerlerinin karanlıkta ve havada 3.62×10²-1.76×10⁷ Ω cm arasında olduğu ve CdS filmlerinin yüksek özdirençinin Sn katkılanarak azaldığı belirlenmiştir. Elektriksel özellikler üzerindeki incelemeler aynı zamanda CdS ve Cd_{0.8}Sn_{0.2}S filmlerinin tuzaklı bir yapıya sahip olduklarını göstermiştir. Yasak enerji aralıkları optik metot kullanılarak 2,43-2,44 eV olarak bulunmuştur. CdS filminin geçirgenlik değeri Sn ile katkılандığında (x=0.2) artmıştır. Filmlerin yüzey morfolojileri Metalurjik Optik Mikroskop ile incelenmiş ve en homojen yüzeyin Cd_{0.8}Sn_{0.2}S filmlerinde görüldüğü belirlenmiştir. Sn katkılı CdS filmleri fotovoltaik güneş pillerinde CdS filmleri gibi pencere materyali olarak kullanılabilir.

Anahtar Kelimeler: Fotovoltaik güneş pilleri, CdS ve CDS:Sn filmleri, Ultrasonik kimyasal püskürtme tekniği, Elektrik, Optik ve morfolojik özellikler.

¹ Department of Physics, Osmangazi University, 26480 Eskişehir Fax: 0 222 2393578 e-mail: skose@ogu.edu.tr, Osmangazi University Research Fund under the project number of 2001/11.

1. INTRODUCTION

The deposition of CdS films has become increasingly important in recent years due to the widened industrial application with a large number of uses (Al Kuhaimi, 1998). Suitable prepared CdS is one of the most sensitive photoconductors known today, especially for detection of visible and near-IR radiation at room temperature (Torres and Gordillo, 1992). It is an n-type direct gap semiconductor ($E_g=2.42$ eV) (Contreras-Puente et al., 2000; Chavez et al., 1997; Lane et al., 2000), and belongs to the II-VI group (Tenne et al., 1992; Nanda et al., 1998; Ruxandra and Antohe, 1998; Su and Choy, 2000). The electrical properties of CdS films are of considerable interest because of its potential use in the fabrication of solar cells and other optoelectronic devices (Valyomana et al., 1990; Pal et al., 1997). CdS is widely used as the window material in heterojunction solar cells together with several narrow band gap semiconductors such as Cu_2S , InP, $CuInSe_2$, CdTe, etc. (Mathew et al., 1995; Ichimura et al., 1999; Özsan et al., 1996; Palafox et al., 1998). CdS thin films were deposited by a variety of techniques, molecular beam epitaxy (MBE), metalorganic chemical vapor deposition (MOCVD), thermal evaporation, chemical bath deposition (CBD), electrodeposition, rf sputtering, vacuum evaporation, spray pyrolysis, etc. (El Assali et al., 2000; Akıntunde, 2000; Moreno et al., 2000; Abd-Lefdil et al., 1998). Generally, in each of these methods polycrystalline, stable, uniform, adherent and hard films are obtained, and their electrical properties are very sensitive to the method of preparation (Vazquez Luna et al., 1999). The spray pyrolysis method (or solution spraying) is a convenient and economical method for the deposition materials (İzci and Köse, 1997). The advantage of this method is relatively easy to scale up for large-area deposition (Choy and Su, 2001; Ajayi et al., 1994). In general the conventional techniques usually end up with more resistive films and indium doping or recrystallization methods are used to improve the conductivity (Günel and Parlak, 1997). Our aim in this work is to see the effect of Sn on the electrical, optical and morphological properties of CdS films.

2. EXPERIMENTAL DETAILS

$Cd_{1-x}Sn_xS$ ($0 \leq x \leq 0.4$) films were prepared onto microscope glass (10×10 mm²) substrates by ultrasonic spray pyrolysis (USP) technique, using a solution of $CdCl_2 \cdot H_2O$ (0.05 M), $SnCl_2 \cdot 2H_2O$ (0.05 M) and $CS(NH_2)_2$ (0.05 M). In the compound $Cd_{1-x}Sn_xS$, the subscript x designates the percentage of the volume of $SnCl_2$ solution in the spraying solution. The substrate temperature was kept at 300 ± 5 °C throughout the spraying. Schematic diagram of the USP set-up is shown in Figure 1. The control of the substrate temperature was done by iron-constantan thermocouple. The spray rate was adjusted to ~ 5 ml/min. The sprayed total solution is 200 ml. Compressed purified air was used as the carrier gas (1.5 bar). Electrical contacts were made by silver-paste. The film was analyzed in a

perpendicular position in the Metallurgical Optical Microscope, and the two different colored sections represent substrate and coated parts. The length of the coated part gives us the thickness of the film. The thicknesses of the films were measured between 0.9-4.6 μm by this way. The thickness of the films increased with Sn amount in the spraying solution. The experimental details of the mixed spraying solutions and the thickness of the films are listed in Table 1. The electrical resistivities of the films were measured by using the two-probe technique (Keithley 485 Auto-ranging Picoammeter and Model 3521 Dual Power Supply). The absorbance spectra of the films were taken by Perkin Elmer UV/VIS Spectrometer Lambda 2S (200-1100 nm) gauge and the surface morphologies were investigated by Metallurgical Optical Microscope.

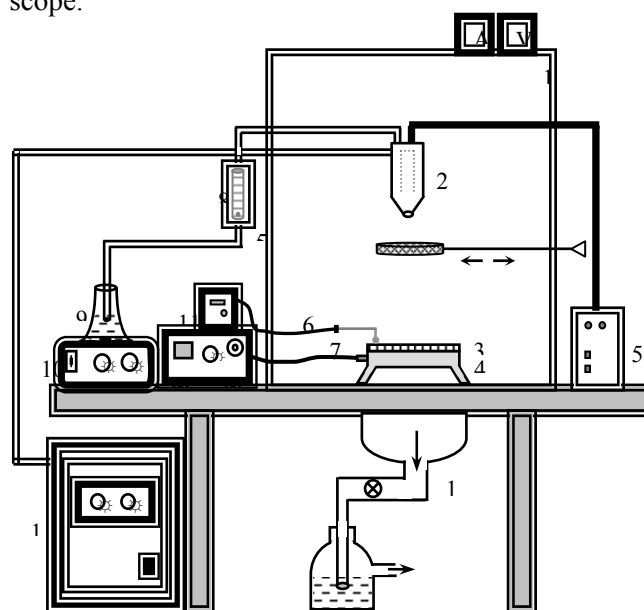


Figure.1. Schematic diagram of USP set-up

- (1) spraying chamber
- (2) ultrasonic atomizer
- (3) pyrex glass substrates
- (4) bronze block
- (5) oscillator
- (6) and
- (7) thermocouples
- (8) flowmeter
- (9) spraying solution
- (10) heater-magnetic mixer
- (11) electrical heater
- (12) air compressor
- (13) fan

Table 1. Amounts of spraying solutions used to deposit films and the thickness of the produced films.

Reactans	CdS	$Cd_{0.8}Sn_{0.2}S$	$Cd_{0.6}Sn_{0.4}S$
$CdCl_2$ (ml)	100	80	60
$SnCl_2$ (ml)	-	20	40
$CS(NH_2)_2$ (ml)	100	100	100
Total (ml)	200	200	200
Thickness (μm)	0.9	3.8	4.6

3. RESULTS AND DISCUSSION

The current-voltage (I-V) plots of $Cd_{1-x}Sn_xS$ films are shown in Figure 2. In the low voltage region there is a slow increase on the current with increasing voltage, while the increase is higher in the high voltage region for CdS and $Cd_{0.8}Sn_{0.2}S$ films. These I-V behaviors suggest that the films have shallow trapped structure. The current changed in the form of $I \sim V^{0.8}$ in the 0.01-0.14 V voltage range for CdS films and in the form of $I \sim V^{1.2}$ in the 0.1-22 V voltage range for $Cd_{0.8}Sn_{0.2}S$ films. The ohmic conduction is dominant these regions. The number of free carriers are more than that of the injected ones in the ohmic region. The Space Charge Limited (SCL) conduction regions come where the current changes in the form of $I \sim V^{1.8}$ for CdS films and $I \sim V^{1.7}$ in for $Cd_{0.8}Sn_{0.2}S$ films. In these regions, the number of injected carriers is higher, while the carriers are being trapped. Hence the current in SCL conduction regions consist of free and injected carriers. For two films the transition voltages between two regions are $V_{tr}=0.14$ V and $V_{tr}=22$ V, respectively. For CdS films, since the current changes in the form of $I \sim V^{1.3}$ after the voltage value of 3 V, the ohmic conduction is dominant in this region and the free charge carriers contribute to the current. Using this result, we can say that the traps loss their effects after this voltage value and their contributions to the current decrease. It was determined from the I-V plot of $Cd_{0.6}Sn_{0.4}S$ films that these films have not trapped structure and the ohmic conduction becomes dominant.

The resistivity values of $Cd_{1-x}Sn_xS$ films were calculated by two-probe method (Schroder, 1990) in the range of 1-10 V and varied from $1.76 \times 10^7 \Omega \text{ cm}$ to $3.62 \times 10^2 \Omega \text{ cm}$ in dark. The resistivity and conductivity values of the films are listed in Table 2. It was seen from this table that the resistivities of the CdS films are very high and these high resistivities were decreased by increasing Sn doping. Therefore the electrical conductivities of CdS films were increased. The conductivity types of the films were determined by the hot-probe method (Seeger, 1982), and the films showed n-type conduction.

Table 2. The resistivity and conductivity values of $Cd_{1-x}Sn_xS$ films.

Material	Resistivity (Ωcm)	Conductivity (Ωcm) ⁻¹
CdS	1.76×10^7	5.68×10^{-8}
$Cd_{0.8}Sn_{0.2}S$	2.41×10^4	4.15×10^{-5}
$Cd_{0.6}Sn_{0.4}S$	3.62×10^2	2.76×10^{-3}

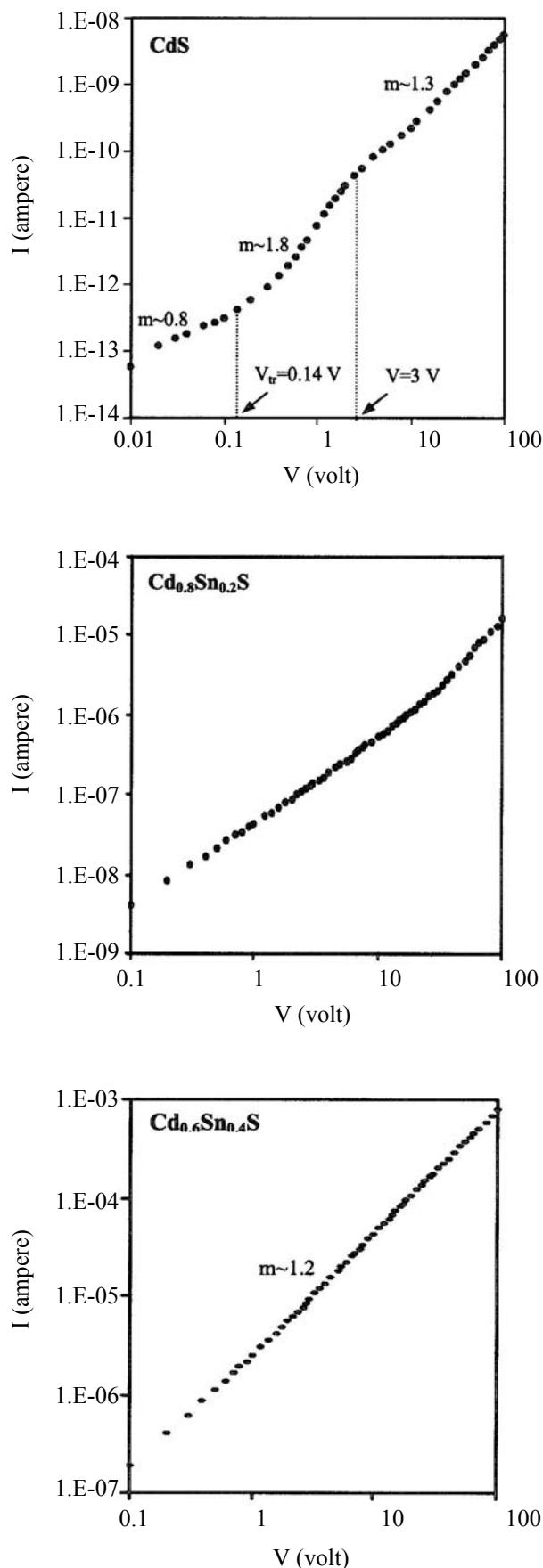


Figure 2. The I-V plots of $Cd_{1-x}Sn_xS$ films.

The fundamental absorbance spectra and $(\alpha hv)^2 \sim hv$ plots of $Cd_{1-x}Sn_xS$ films are shown in Fig.3. It was determined from Figure 3.(a) that all films at long wavelengths are fairly transparent, but as the wavelength decreases a certain threshold is reached, where the absorption coefficient rises rapidly and the materials become opaque. This threshold, referred to as the fundamental absorption edge, corresponds to the onset of optical excitation of electrons from the valance into conduction band. Different methods such as optical and thermal can be used to calculate the energy gap of the materials. The optical method also gives information about the optical transitions. In this method $(\alpha hv)^2 \sim hv$ plot is drawn using the fundamental absorption spectrum ($A \sim \alpha$). The energy of the point that the direction of the linear portion coincide hv axis at $(\alpha hv)^2=0$ gives the energy gap (Nag, 1980; Pal et al., 1997; Koteswara and Ramagrishna, 1998; Ray et al., 1998; Al Kuhaimi, 1998; Ghazali et al., 1998; Padam et al., 1988; Quientero et al., 1992; Bihri et al., 1992). The forbidden band gaps of the films were calculated in between 2.34-2.41 eV by using this method. It was determined that there is a decrease in the forbidden energy gap values with Sn concentration. There are other linear portions in Figure3.(b). We think that the energy values which correspond to these linear

parts represent the band bending or the transitions from the lower states of valance band.

The transmissions (T), absorbance (A), linear absorption coefficients (k), reflection coefficients (R), refractive indices (n) and relative dielectric constants (ϵ_r) were calculated for all films using the fundamental absorption spectra and formulas (1-6) mentioned below (Özsan et al., 1996; Kittel, 1996; Rose et al., 1986; Askeland, 1998);

$$\alpha = \frac{A}{w} \tag{1}$$

$$T = 10^{-A} \tag{2}$$

$$R = 1 - \sqrt{Te^A} \tag{3}$$

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

$$k = \frac{\alpha \lambda}{4\pi} \tag{5}$$

$$\epsilon_r = n^2 \tag{6}$$

Where α is absorption coefficient, w is thickness and λ is the wavelength of incident light. These values and the forbidden energy gaps are given in Table 3.

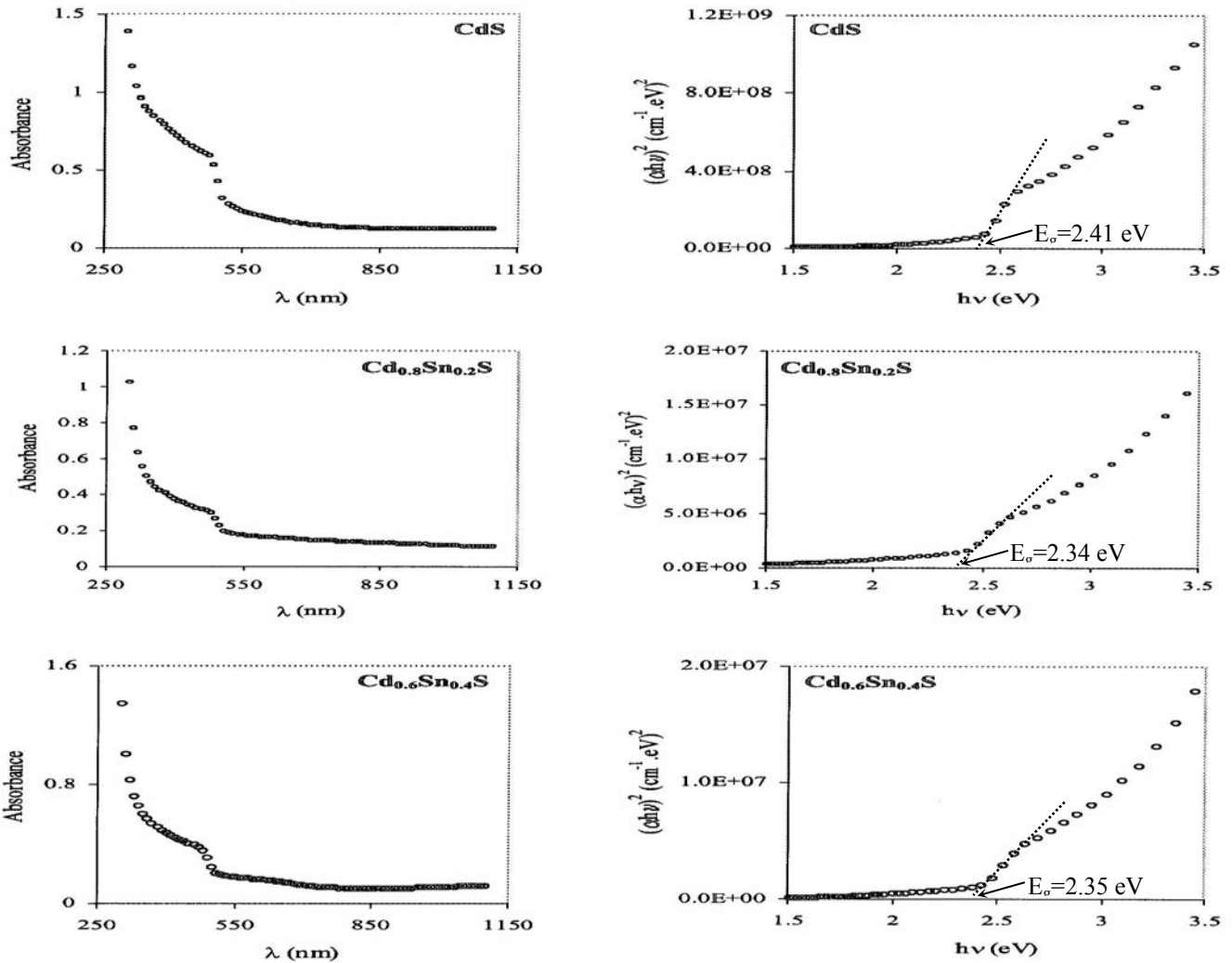


Figure 3. (a) Fundamental absorbance spectra, (b) $(\alpha hv)^2 \sim hv$ plots of $Cd_{1-x}Sn_xS$ films.

Table 3. The forbidden energy gaps and some optical properties of $Cd_{1-x}Sn_xS$ films.

Material	E_g (eV)	T	A	K	R	n	ϵ_r
CdS	2.41	0.57	0.31	1.44×10^{-2}	0.17	2.55	6.50
$Cd_{0.8}Sn_{0.2}S$	2.34	0.62	0.21	2.60×10^{-3}	0.14	2.49	6.20
$Cd_{0.6}Sn_{0.4}S$	2.35	0.64	0.24	2.20×10^{-3}	0.13	2.23	4.97

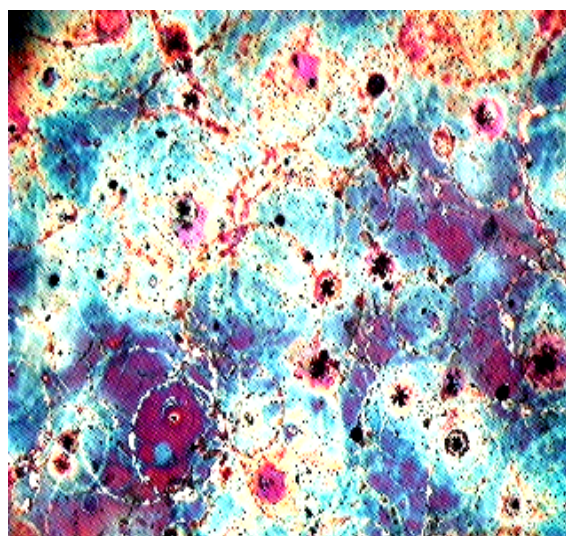
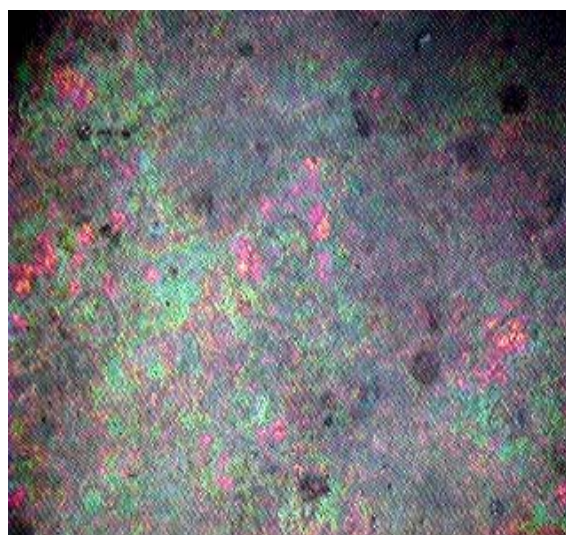
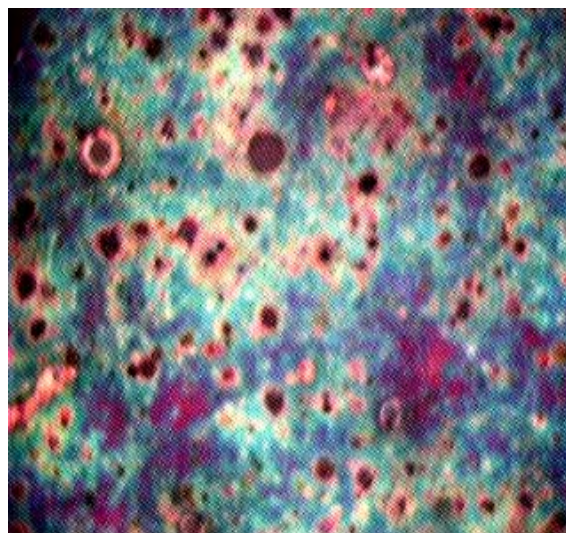
The surface morphologies of all films were investigated by Metallurgical Optical Microscope. The micrographs of the films are shown in Fig.4. Islands with different sizes and colors were seen on the surfaces, and the distribution of them is not uniform for all films. However the most homogeneous surface was seen in $Cd_{0.8}Sn_{0.2}S$ films. This undesired formation may probably occur during deposition itself or after deposition resulting from the thermal difference between the film and the substrate or the high stress in the films. It is very difficult to obtain microscopically smooth and uniform films by USP technique.

4. CONCLUSIONS

In this work the effect of the Sn concentration on some physical properties of CdS films was investigated. It was seen that the thickness of the films varied between 0.9-4.6 μm and increased with Sn amount in the spraying solution. After the investigations of dc current mechanisms of the films, it was determined that the materials have ohmic and SCL current mechanisms. The resistivity values of CdS thin films are very high in dark, while Sn doping made a dramatical change in resistivity values. However, these values are still high for solar cell applications. So, the films which have lower resistivity can be obtained by annealing in H_2 , N_2 and metal atmosphere of the films. It was seen that Sn doping made a noticeable change on band gap of CdS films. It was determined that the most homogeneous surface was seen in $Cd_{0.8}Sn_{0.2}S$ films. In this work, the high resistivity of CdS films was decreased by increasing Sn doping and also the transmittance values (for 200-1100 nm) were increased. Hence it was determined that Sn-doped CdS films can be used as window material in photovoltaic solar cells.

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Figure 4. The surface micrographs of $Cd_{1-x}Sn_xS$ films.

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Salih Köse, 04.02.1956'da Konya'da doğdu. 1980'de Hacettepe Üniversitesi Fizik Mühendisliği Bölümünden mezun oldu. 1983'de Anadolu Üniversitesi Fizik Bölümünde Uzman olarak göreve başladı. 1986'da Anadolu Üniversitesi Fen Bilimleri Enstitüsünde Katıhal Fiziği Bilim dalında yüksek lisans eğitimini tamamladı. 1993'de Osmangazi Üniversitesi Fen Bilimleri Enstitüsünde Katıhal Fiziği Bilim dalında doktora eğitimini tamamladı.



Ferhunde Atay, 05.07.1973'de Eskişehir'de doğdu. 1994'de Anadolu Üniversitesi Fen Edebiyat Fakültesi Fizik Bölümünden mezun oldu. 1994'de Osmangazi Üniversitesi Fizik Bölümünde Araştırma Görevlisi olarak göreve başladı. 1996'da Osmangazi Üniversitesi Fen Bilimleri Enstitüsünde Katıhal Fiziği Bilim dalında yüksek lisans eğitimini tamamladı. 2002'de Osmangazi Üniversitesi Fen Bilimleri Enstitüsünde Katıhal Fiziği Bilim dalında doktora eğitimini tamamladı.



Vildan Bilgin, 29.10.1975'de Lapseki'de doğdu. 1996'da Osmangazi Üniversitesi Fizik Bölümünden mezun oldu. 1997'de Osmangazi Üniversitesi Fizik Bölümünde Araştırma Görevlisi olarak göreve başladı. 1998'de Osmangazi Üniversitesi Fen Bilimleri Enstitüsünde Katıhal Fiziği Bilim dalında yüksek lisans eğitimini tamamladı. 2003'de Osmangazi Üniversitesi Fen Bilimleri Enstitüsünde Katıhal Fiziği Bilim dalında doktora eğitimini tamamladı.



İdris Akyüz, 28.01.1978'de Ankara'da doğdu. 1998'de Anadolu Üniversitesi Fizik Bölümünden mezun oldu. 1998'de Osmangazi Üniversitesi Fizik Bölümünde Araştırma Görevlisi olarak göreve başladı. 2000'de Osmangazi Üniversitesi Fen Bilimleri Enstitüsünde Katıhal Fiziği Bilim dalında yüksek lisans eğitimini tamamladı. 2000'de Osmangazi Üniversitesi Fen Bilimleri Enstitüsünde Katıhal Fiziği Bilim dalında doktora eğitimine başladı ve halen devam etmektedir.