

## Studies on Sn doped cadmium sulfide thin films as highly selective green light photosensors

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PACS 07.07.Df, 73.50.Lw, 61.10.Nz, 85.60.Gz

**ABSTRACT** Thin films of Tin (Sn) doped Cadmium Sulfide (CdS) have been grown by chemical bath deposition technique. It was observed that all fabricated thin films (doped & undoped) were polycrystalline with nanoscaled crystallites and cubic crystal structure of CdS. SEM micrographs show nanorod structure of CdS and Sn doped CdS. EDS analysis shows the deficiency of sulfur and excess of cadmium in the films. UV-VIS spectroscopy confirms increase in band gap with doping of the films. Microstructural analysis shows that the particle size increases with increasing concentration of the Sn on excitation with 483 nm. The gravimetric analysis shows that the thickness of the pure CdS thin film measured is 134.41 nm, which increases with doping concentrations of tin. Electrical conductivity measurements show that the material switches its Negative Temperature Coefficient (NTC) to Positive Temperature Coefficient (PTC) nature with increase in temperature. TEP measurements show n type semiconducting nature of the films which is highly photo sensitive. The pure CdS thin film was observed to be less sensitive to the green light, however, the Sn (3 wt%) doped CdS thin films exhibit enhanced photo response particularly to green light.

**KEYWORDS** photosensors, thermoelectric, X-ray diffraction, photodetectors

**ACKNOWLEDGEMENTS** Authors thank JNU, Jaipur for generous financial help to complete this research works.

**FOR CITATION** Sharma Y.C., Patil S.D., Nikam H.A., Sharma P., Borse D.B., Patil D.R. Studies on Sn doped cadmium sulfide thin films as highly selective green light photosensors. *Nanosystems: Phys. Chem. Math.*, 2022, **13** (6), 678–687.

### 1. Introduction

Today's world is facing many problems about the availability of energy in sufficient amount. So, the researchers are focusing their attention towards the generation of green and clean energy, particularly the solar energy. Many solar cells are already available in the market, but their cost is very high, so everyone can not afford them. Intensive work on the solar cell fabrication with low cost materials is needed for increasing of the installations availability. Efforts have been made with earth abundant elements like  $\text{Cu}_2\text{ZnSnS}_4$  (CZTS) which have direct band gap ( $E_g \sim 1.5$  eV) and high absorption coefficient [1–3]. The  $\text{Cd}_x\text{Sn}_{1-x}\text{S}_3$  films are used in solar cell as they have a relatively wide band gap as compared to the CdS thin films. The high band gap of ternary Sn doped CdS thin film has less window absorption loss, which can be replaced with CdS thin film in the solar cell systems [4, 5].

Meshram et al. have fabricated the CdS thin film using spray pyrolysis method and studied its structural and optical properties [6]. Furthermore, the Sn doped CdS thin films were fabricated by using chemical bath deposition technique by Roy et al. who have studied their optical and electrical properties [7]. It was reported that the effect of Sn doping on the photoluminescence and electrical properties of the CdS thin film changes the properties significantly. The influence of Sn doping on the CdS thin film prepared by ultrasonic spray pyrolysis had been reported by Eygi et al. and their photoluminescence and electrical properties have been reported [8]. But the studies on synthesis of Sn doped CdS thin films and their photo-sensing performance and needs a further attention. For thin film fabrication, various deposition techniques have been used by various resources viz. spray pyrolysis [9], chemical vapor deposition [10], thermal evaporation [11], molecular beam epitaxy [12], RF sputtering [13], chemical bath deposition [14], etc. Out of these techniques, the chemical bath deposition (CBD) technique has been used extensively to fabricate the thin films because it is a low cost simple technique, which does not require high quality substrates, vacuum system, huge instrumentation, large power handling

and consumption, large area coatings, etc. Therefore, an attempt has been made to deposit the Sn doped CdS thin films by this technique and optimize their properties.

In this study, the Sn doped CdS thin films have been fabricated on the glass substrates using chemical bath deposition (CBD) technique. The thin films were characterized by X-ray diffraction (XRD), scanning electron microscopy (SEM), energy dispersive spectroscopy (EDS), UV-VIS absorption, photoluminescence (PL), gravimetric analysis for film thickness measurement and electrical measurement techniques to study their structural, morphological, optical, physical and electrical properties. The EDS measurements have been carried out to find the presence of different elements in the materials of thin films. The photoluminescence measurement of the Sn doped CdS thin films have been monitored on excitation with 483 nm. The thermo-electrical measurements have been carried out to confirm the semiconducting nature of thin films. The photo response of the thin films has been monitored to see the effect of Sn dopant.

## 2. Experimental details

### 2.1. Preparation of Sn doped CdS thin films

The substrates used in this study for fabrication of thin films are borosilicate glass, which were cleaned following the standard procedure. The glass plates were cleaned using double distilled water at temperature 80 °C. For further cleaning, the substrates were kept in chromic acid for 15 min and were washed thoroughly in distilled water to remove the acid. Then the washed substrates were immersed in a soap solution for 5 min and washed thoroughly again in distilled water. The substrates were finally cleaned in ultrasonic cleaner and dried under IR lamp.

In order to fabricate the pure and the Sn doped CdS thin films with different concentrations of Sn, viz. 1, 3, 5 and 7 wt% by chemical bath deposition technique, the 0.1 M aqueous solution of Cadmium Nitrate was prepared and Ammonium Hydroxide was added to this solution slowly drop wise (0.1 ml/min) with a constant stirring on the hotplate stirrer, till the ppt forms at pH equals to 8.9. The ppt vanishes with further progress in reaction and the solution becomes transparent. An aqueous solution of 0.1 M thiourea was added to this transparent solution and thereafter 0.1 M aqueous solution of Tin Chloride was added as dopant in different proportions viz. 0, 1, 3, 5 and 7 wt%. Finally, the ultrasonicated, cleaned and dried substrates were dipped in the beakers at constant temperature of 41 °C for 1.5 hr. The substrates were taken out from solution and washed with double distilled water. These substrates were annealed at 150 °C for 30 min. Thin films thus obtained were used for further investigations.

### 2.2. Characterization of films

The XRD patterns of the thin films have been monitored using X-ray diffractometer D8 Advance (Model no. Bruker AXS GmbH, Germany) to confirm the crystalline nature, phase and to calculate the crystallite size. The morphology of the thin films was recorded using Hitachi field emission scanning electron microscope (FE-SEM Model no. S 4800 Type II). The EDS measurements were carried out using X-Flash 5030 detector (Model no. Bruker AXS GmbH, Germany). The UV-visible absorption measurements were carried out using UV-Hitachi 2900). The PL spectroscopic measurements were carried out on excitation with 483 nm wavelength from a Xenon lamp using Fluoromax-4 spectrofluorometer. Gravimetric analysis was done to measure the film thickness. Thermo-emf measurements were made by TEP setup. Electrical properties of thin films were measured with the help of static gas sensing setup.

## 3. Material characterizations

### 3.1. Structural properties (XRD)

The X-ray diffraction (XRD) patterns of the pure and Sn doped CdS thin films with different concentrations of Sn (viz. 0, 1, 3 and 5 wt%) are shown in Fig. 1. The XRD pattern reveals that the thin films are polycrystalline in nature. The XRD patterns match well with JCPDS File no. 80-0019 and the phases are found to be cubic for the pure and Sn doped thin films. It is evident from XRD analysis that tin doping has not changed the cubic structure of pure CdS even after doping. The lattice parameters of CdS thin films are found to be  $a = 5.811$  and  $\alpha = \beta = \gamma = 90^\circ$  with space group  $F4\bar{3}m$  (206). The indexing of the XRD peaks has been done using the JCPDS File. The average crystallite size ( $d$ ) for each sample has been calculated by Scherer's formula:  $d = 0.9\lambda/\beta \cos \theta$  for the pure and (1, 3 and 5 wt%) Sn doped CdS thin films and their values are found to be as 6.067, 6.589, 8.943 and 21.487 nm, respectively. Thus, the crystallite size of the thin film increases with the increase in the concentrations of Sn.

### 3.2. Microstructural analysis (SEM)

Figure 2 shows the scanning electron micrographs of the pure CdS and (1, 3 and 5 wt%) Sn doped CdS thin films annealed at 150 °C for 30 min. The pure CdS thin film consists of thin nanorods overlying to each other and their size is found to lie in the range of 14.9 – 29.8 nm (Fig. 2a). Fig. 2(b) depicts the micrograph of (1 wt%) Sn doped CdS thin film, which consists large number of nanorods bound with each other associated with the nanosheets and are agglomerated. The film also shows a bulk like structure with porosity in the range of 67.1 – 2.0 nm. Whereas, in 3 wt% Sn doped sample, the particle size is found to be in the range of 22.1 – 40.8 nm with nanosheets (Fig. 2c). Fig. 2(d) depicts the micrograph

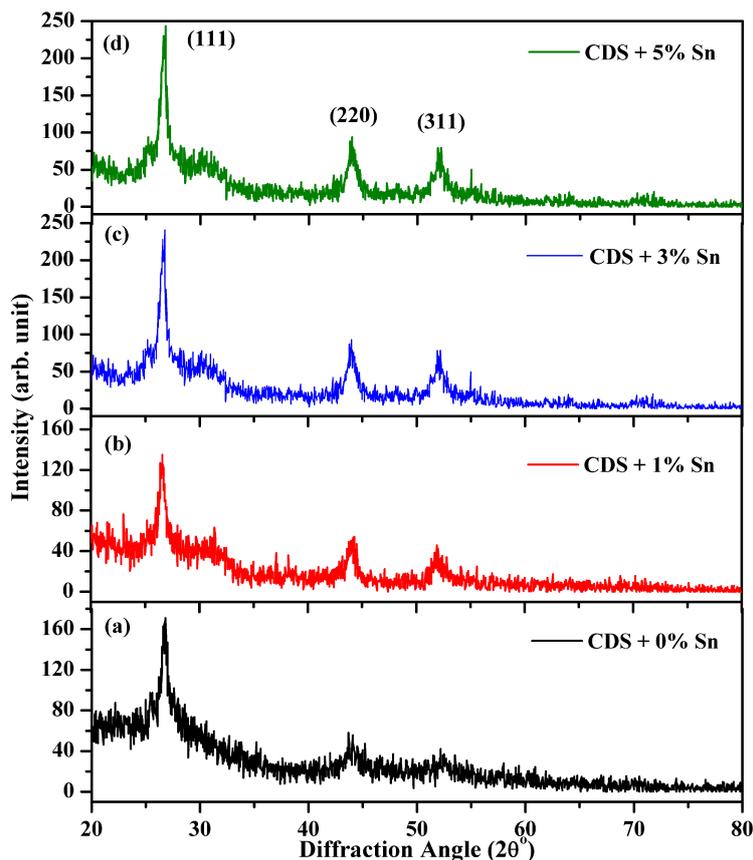


FIG. 1. The XRD patterns of the (a) 0 wt%, (b) 1 wt%, (c) 3 wt% and (d) 5 wt% Sn doped CdS thin films

of 5 wt% Sn doped CdS thin film, which consists of large number of nanosheets bound with each other associated with the crystallite size in the range of 20 – 48.2 nm.

### 3.3. Energy dispersive analysis by X-rays (E-DAX)

The quantitative elemental compositions of the pure CdS and Sn doped CdS thin films with different concentrations of Sn (viz. 1, 3 and 5 wt%) have been analyzed using an energy dispersive spectrometer and the atom % of the Cd, S and Sn elements are summarized in Table 1.

TABLE 1. Elemental compositions in the pure CdS and Sn doped CdS thin films

Elements	Doping Concentrations (wt %)				
	0 (Pure) (Expected)	0 (Pure) (Observed)	1	3	5
Cd	77.80	97.62	87.87	85.06	76.18
S	22.20	02.38	10.69	10.09	17.57
Sn	00.00	00.00	01.44	04.85	06.25
Sn doped CdS	100	100	100	100	100

Stoichiometrically expected mass % of Cd and S in CdS is 77.80 and 22.20, respectively. However, experimentally observed mass % of Cd and S in CdS is 97.62 and 2.38, respectively. Thus, the pure CdS is sulfur deficient thin film, which increases the semiconducting nature of the material. The masses % of Cd and S in the samples (Table 1) are not as per the stoichiometric proportion and all the Sn doped samples are observed to be sulfur deficient CdS, which favors the semiconducting nature. It has been reported by Husham et al. that the sulfur deficient Sn doped CdS thin film shows high photosensing performance of the films [15]. If the film is very thin it contains less sulfur contents [16]. We have also

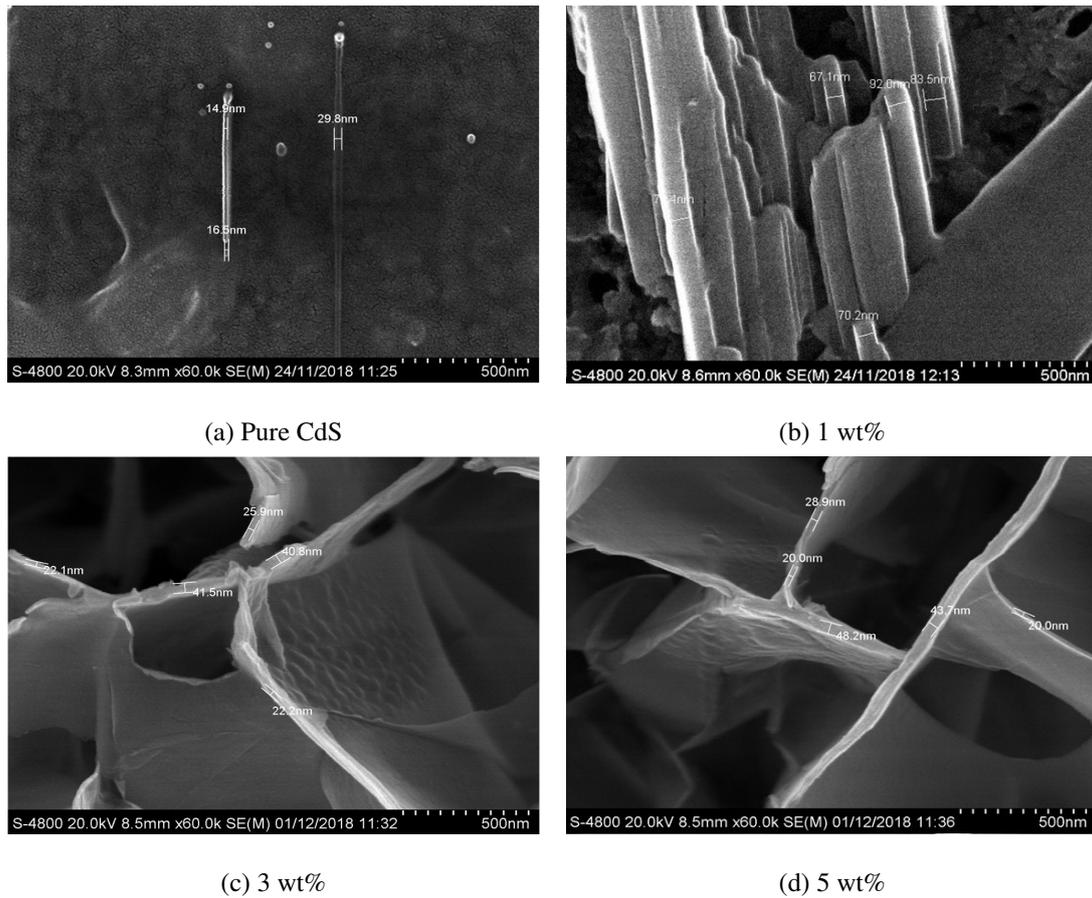


FIG. 2. SEM Micrographs of (a) Pure CdS, (b) 1 wt%, (c) 3 wt % and (d) 5 wt % of Sn in CdS thin film

observed that the sulfur deficient Sn doped CdS thin film (3 wt%) exhibits the crucial photosensing performance. The E-DAX analysis shows the presence of Sn, Cd and S elements in the thin film.

### 3.4. Gravimetric analysis

Thin film is a 2D solid film whose third dimension is restricted to nanoscale and whose properties could be changed in accordance with the change in the third dimension, irrespective of the size of the remaining two dimensions. Thus, electric, electronic and optical properties of the materials of the film can be tailored by varying the film thickness, which can be measured by gravimetric analysis as per the eq. (1) [17, 18]:

$$t = \frac{M}{A \cdot \rho}, \quad (1)$$

where  $t$  is the thickness of the thin film,  $M$  is the mass of the deposited material (film) on the substrate ( $M$  is the mass of the substrate with deposited film minus the mass of the substrate without deposition),  $A$  is the area of the film,  $\rho$  is the density of the material.

Film thicknesses of pure CdS and Sn doped CdS thin films with different concentrations of Sn (viz. 1, 3 and 5 wt%) were analyzed using gravimetric analysis in the laboratory which leads to conclusion that the film thickness increases with doping concentrations of Sn. The results are summarized in Table 2.

### 3.5. Optical properties

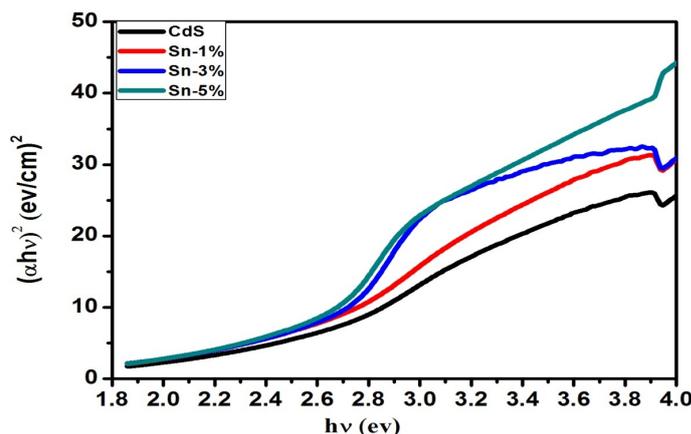
3.5.1. *UV-visible measurements.* The optical band gap of the pure CdS and Sn doped CdS thin films with different concentrations of Sn (viz. 1, 3 and 5 wt%) has been calculated by using the Wood and Tauc formula [19]:

$$\alpha = \frac{B(h\nu - E_g)^m}{h\nu},$$

where,  $E_g$  is the optical band gap energy,  $h\nu$  and  $B$  are an energy of the incident photon and the band tailoring constant, respectively. The value of  $m$  was chosen as 1/2 for direct band gap allowed transitions [20]. Fig. 3 depicts the variation of  $(\alpha h\nu)^2$  versus  $h\nu$  for the pure CdS and the Sn doped CdS thin films with different concentrations of Sn (viz. 1, 3 and 5 wt%). The optical band gap of the pure CdS thin film was found to be 2.32 eV, which matches to the earlier reported value [21–24]. The optical band gaps of Sn doped CdS thin films are found to be 2.35, 2.41 and 2.43 eV for the 1, 3 and

TABLE 2. Film thickness of pure CdS and Sn doped CdS thin films

S. No.	Material Composition of Thin Film	Film Thickness (in nm)	Average Crystallite Size (in nm)
1	Pure CdS	134.41	6.067
2	Sn (1 wt%) CdS	158.07	6.589
3	Sn (3 wt%) CdS	162.34	8.943
4	Sn (5 wt%) CdS	193.22	21.487

FIG. 3. Variation of  $(\alpha h\nu)^2$  versus  $h\nu$  of pure CdS and the Sn doped CdS thin films

5 wt% concentrations, respectively. Thus, the optical band gaps of the thin films are found to increase with increase in the concentrations of Sn.

**3.5.2. Photoluminescence measurements.** The photoluminescence spectra of the pure CdS and the Sn doped CdS thin films with different concentrations of Sn (viz. 1, 3 and 5 wt%) recorded in the range of 500 – 550 nm on excitation with 483 nm are shown in Fig. 4. The pure CdS thin film does not show any photoluminescence. However, the Sn doped CdS thin films exhibits weak emission in the green region centered at 520 nm. The intensity of the green emission increases on increasing the concentrations of Sn contents. The 1 wt% Sn doped CdS thin film emits the green color and enhances up to 3 wt% doped thin film. Above 3 wt%, the emission intensity is found to decrease due to concentration quenching as the concentration was increased to 5 wt%. This is due to the fact that at higher concentrations, the inter-nuclear separation between the atoms becomes smaller than the critical distance and the excitation energy migrates to the energy killing centers [25]. Thus, the emission intensity is maximum for the 3 wt% concentration of Sn.

There are several factors responsible for an increase in the emission intensity. When the thin film is annealed, the surface defects are reduced significantly. This also reduces the impurity elements present in the thin film [26]. As discussed earlier, when the Sn is doped in the CdS thin film the crystallinity of the film is improved and the crystallite size was increased slightly from 74 to 108 nm. It is also clear from SEM analysis that the particle size of the thin film was increased from 20 to 52 nm after doping the Sn element. This can enhance the photoluminescence intensity. Wang et al. have observed an intense photoluminescence for larger particle size due to high volume to surface area ratio [27]. Recently, Yadav et al. have also reported that the larger particles can easily absorb more incident photons and give an intense photoluminescence [28]. They have also annealed the samples and found significant enhancement in the emission intensity due to an increase in crystallinity and particles size. They observed a reduction in the impurity ions on annealing the samples. In the present case, the increase in the emission intensity has been observed due to the improvement in the particle size of the thin film after Sn doping. As a result, the Sn doped CdS thin film may be used in the optoelectronic devices.

## 4. Electrical properties

### 4.1. I–V characteristics

The photocurrent ( $I$ ) of the pure CdS and Sn doped CdS thin films with different concentrations of Sn (viz. 1, 3 and 5 wt%) has been measured at different input voltages ( $V$ ) and the plots thus obtained are shown in Fig. 5. It has been

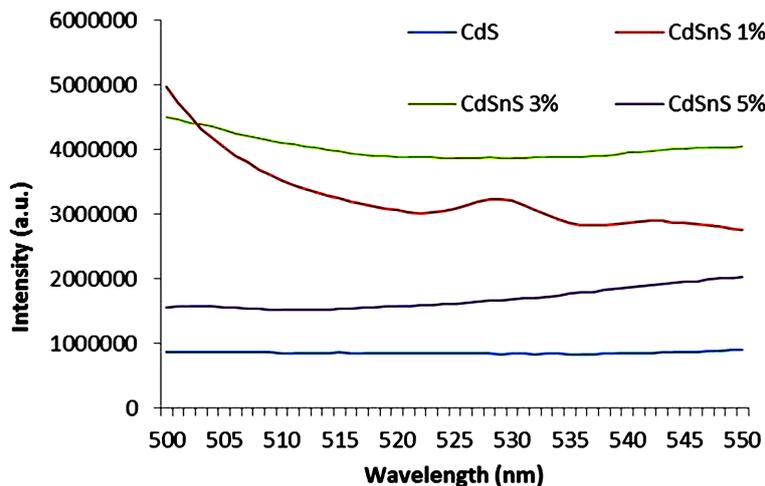


FIG. 4. Photoluminescence spectra of the as-deposited pure CdS and the Sn doped CdS thin films with different concentrations of Sn (viz. 1, 3 and 5 wt%)

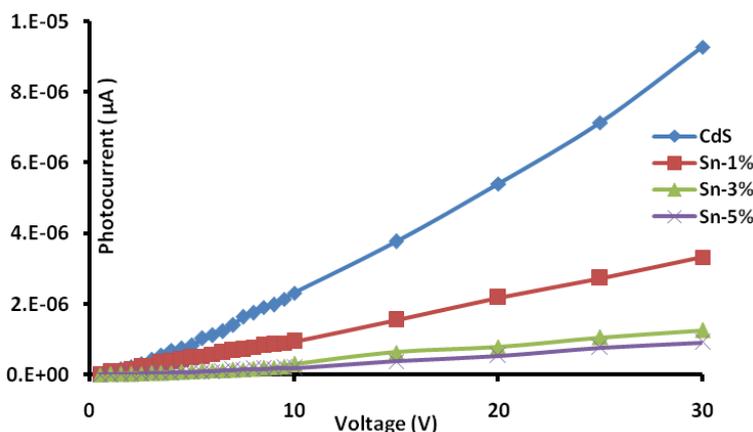


FIG. 5. I–V characteristics of the pure CdS and Sn doped CdS thin films

observed that the photocurrent generated in the pure CdS thin films is maximum as compared with the Sn doped CdS thin films, which may be attributed to the fact that as the doping concentration increases the energy band gap also increases. The nature of the I–V plot is found nearly linear for the pure and Sn doped CdS thin films.

#### 4.2. Electrical conductivity

Figure 6 shows a variation of logarithm of the electrical conductivity with reciprocal of operating temperature of the pure CdS and the Sn doped CdS thin films with different concentrations of Sn (viz. 1, 3 and 5 wt%). At higher temperature, i.e. at 150 °C, the conductivity was high, which decreases with the decrease in the operating temperature, i.e. up to 130 °C. Thus, the film exhibits the negative temperature coefficient of resistance up to 130 °C. The conductivity of the film increases further with the decrease in operating temperature, exhibiting the positive temperature coefficient of resistance. This proves the semiconducting nature of the Sn doped CdS thin films. The electrical conductivity is found to be maximal for the 3 wt% Sn doped CdS thin film.

### 5. Photo response of the thin films

#### 5.1. Time based performance

Photo response (*S*) of the photo sensor is defined as the ratio of the change in photo currents generated upon exposure of the light rays and in dark to the photocurrent in light rays. Fig. 7 depicts the variation of photo response of the pure CdS and the Sn doped CdS thin films with different concentrations of Sn (viz. 1, 3, 5, and 7 wt%) with respect to exposure time for different colors (wavelengths) of light. It is observed from the Fig. 7(a) that the photo response of pure CdS thin film increases with the increase in time of exposure of green, blue and yellow colors of light. However, the negligible photo response was observed for the red and ordinary light. These films are relatively less sensitive for the blue and yellow light.

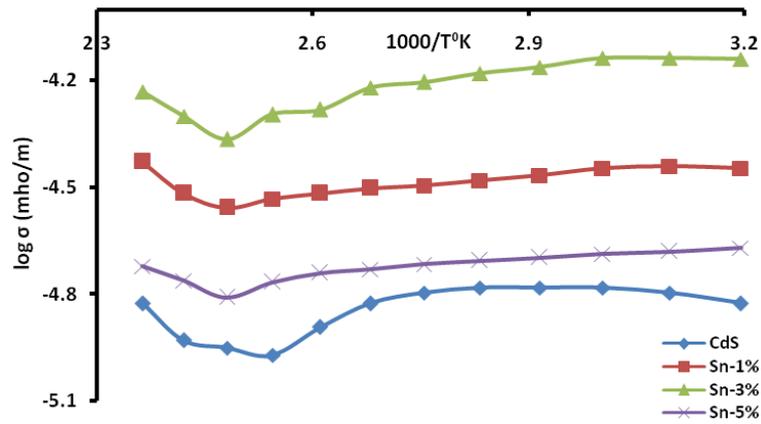


FIG. 6. Conductivity-temperature profile of pure CdS and the Sn doped CdS thin films with different concentrations of Sn (i.e. 1, 3 and 5 wt%)

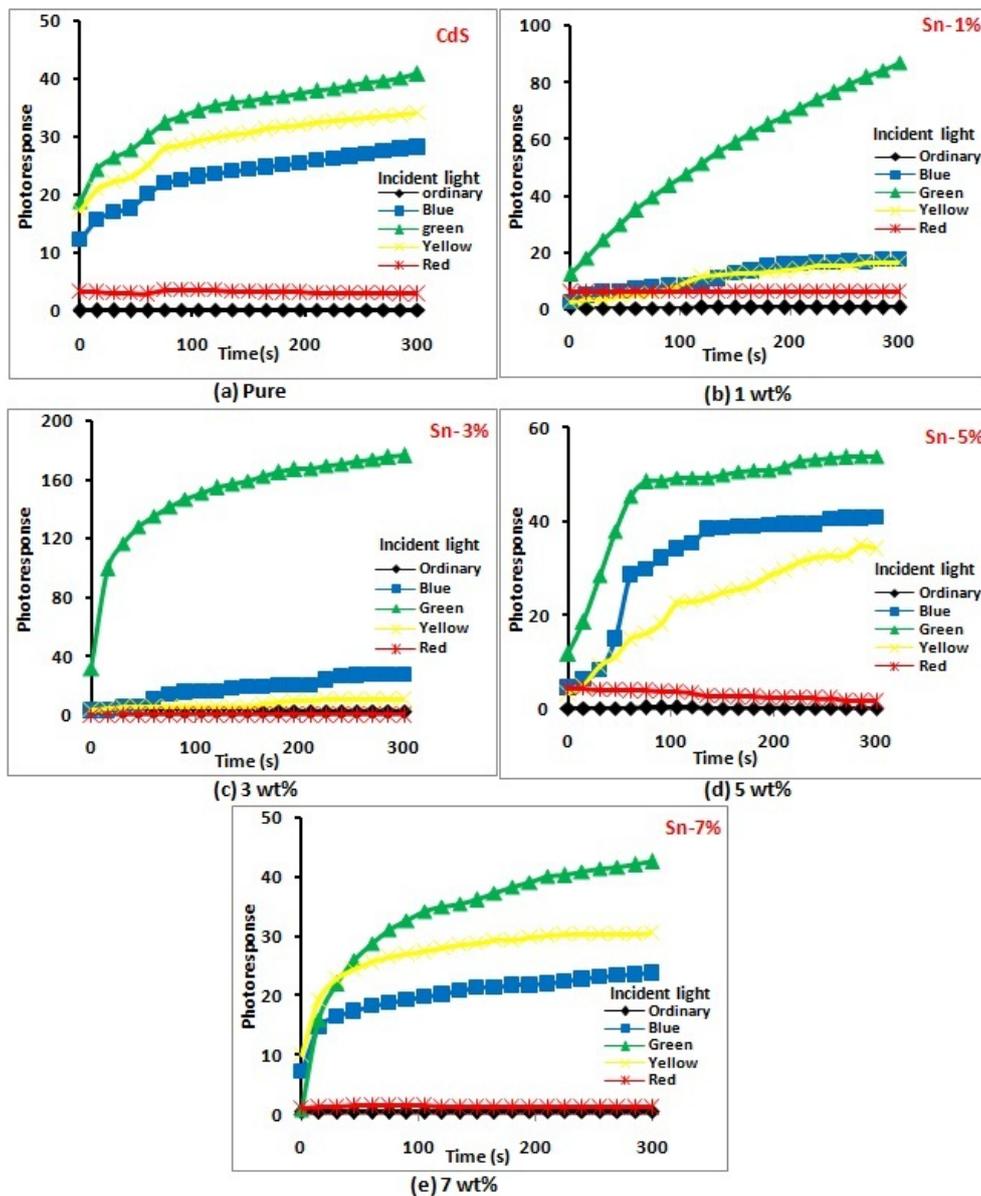


FIG. 7. Variation of photo response of the pure CdS and Sn doped CdS thin films with different concentrations of Sn (i.e. 1, 3, 5, and 7 wt%) with time

From Figs. 7(b–e) it is clear that the Sn doped CdS thin films are highly sensitive and selective to the green light while less responsive to the other wavelengths, viz. blue, red, yellow, etc. Further, this is found to increase with the increase in the Sn doping concentration up to 3 wt% and it is decreased considerably for 5 and 7 wt% of Sn concentrations. This may be attributed to the excitation of the material by photon of the wavelength in between 520 to 560 nm. Upon exposure of the photon of wavelength in this region, the free electrons move to the conduction band and constitute to enhance the photo current. The Sn doped CdS thin films exhibit crucial response to only green light among all, which may be attributed to the fact that, due to doping of Sn atoms, some periodic arrangements of the crystals disturbs, though in smaller extent and crystal structure does not gets changed even after doping by Sn. This arrangement modifies the energy levels of the crystals, results in shifting of absorbance towards green wavelength.

## 5.2. Doping concentration based performance

Figure 8 shows the green light photo response as a function of doping concentrations of Sn in the CdS thin films. It has been observed that, the pure CdS thin film was less sensitive to green light and the photo response of the Sn doped thin film increases up to 3% Sn doping concentration and then decreases further for 5 and 7 wt% concentration.

## 6. Thermo electric power (TEP)

Figure 9 depicts the variation of thermo emf as a function of temperature difference of the pure and modified samples of CdS thin films. It is observed from Fig. 9 that the thermo emf decreases with increase in the temperature of hot end keeping the temperature of cold end constant at 32 °C, i.e. at room temperature.

In semiconductor, temperature gradient yield thermoelectric effect in which phonons travels preferentially from the hot end to the cold end due to phonon-electron interaction. It is seen that TEP measurements, thermal gradient establish change of density of charge defect states by capturing electrons and holes which can take place due to the process of diffusion.

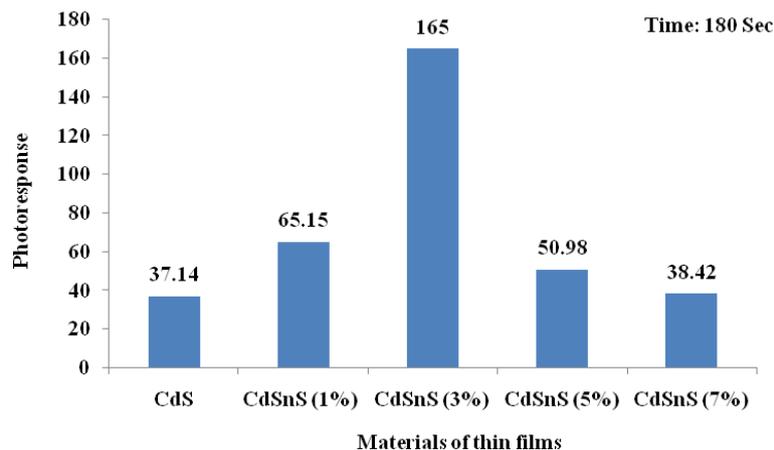


FIG. 8. Variation of the photo response of green light with Sn doping concentrations in the CdS films

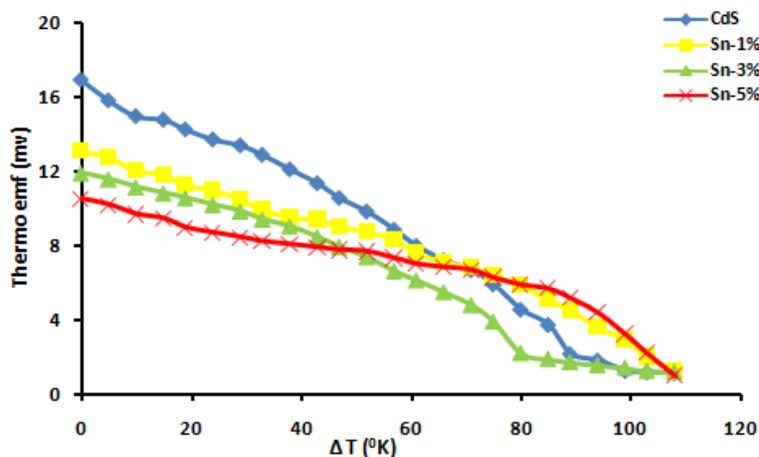


FIG. 9. Variation of thermo emf with temperature difference for Sn doped CdS thin films with different concentrations (0, 1, 3 and 5 wt%)

Thermoelectric power measurement is useful tool for measurement of carrier type. The developed thermoelectric voltage due to temperature difference across ends of material is directly depending on carrier concentrations. It is called as Seebeck effect. Fig. 9 shows the increase in the conductivity of material attributes to the decrease in mobility of charge carrier and carrier concentrations. From the above discussion, it is clear that as the polarity of thermally generated voltage at the hot end is positive one conclude that the material has the n-type nature.

## 7. Conclusions

The Sn doped CdS thin films have been grown successfully using chemical bath deposition technique. Gravimetric analysis was used to measure the film thickness. It was confirmed from XRD analysis that the cubic structure of pure CdS has not been changed even after doping CdS with Sn, as sulfur atoms in the crystal replaces Sn. The microstructural analysis shows the increase in the particle size and the film thicknesses with increasing concentration of the Sn on excitation with 483 nm. It was observed from the UV spectroscopy that the band gap increases with dopant concentration, particularly Sn, increase. The Sn doped CdS thin films show the n-type semiconducting nature. The pure CdS thin film was observed to be less sensitive even to the green light; however, the Sn doped CdS thin films exhibit crucial photo response to the green light. Thus, the Sn doped CdS thin films may be useful in the fabrication of optoelectronic devices and solar cells.

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*Submitted 13 June 2021; revised 10 October 2022; accepted 12 October 2022*

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*Conflict of interest:* the authors declare no conflict of interest.