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Structural, Electrical and Photoresponse Studies of Ni Doped CdS Thin Films for Opto-electronic Applications

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Abstract— The pristine and Ni doped CdS polycrystalline thin films were grown on to glass substrates by Chemical Bath Deposition (CBD) method. The deposition bath contains cadmium chloride (as a source for cadmium ions), thiourea (as a source for sulfur ions), ammonium chloride, EDTA and ammonium hydroxide. The as prepared CdS films were annealed in air at 300 °C for 60 minutes and the as prepared films were characterized structural, morphological, elemental and electric properties. From XRD patterns, all the films exhibit hexagonal phase with (0 0 2) as preferential orientation and the estimated crystallite size were 29 nm and 18 nm for undoped and Ni doped CdS films. SEM images showed that the spherical like grains were formed in the films. Energy dispersive X-ray study confirmed the presence of Cd, S and Ni in the films. From the electrical studies, Ni doped CdS thin films carrier concentrations were increased with the increased Ni concentrations. The higher effective quantum efficiency (EQE) value is obtained for fabricated p-Si/n-Ni:CdS (Ni dopant at 0.08 M) hybrid heterojunctions photo detectors device. Those results could be applied in other CdS-based cells, for more effective PV and photo diode conversion device and other applications that require such properties.

Keywords— *CBD, pristine, opto-electronics, heterojunction and photo response*

I. INTRODUCTION

In recent studies, transparent conducting films have been studied due to low resistivity with high potential application as sensors and other optoelectronic devices. Among transparent conducting films, CdS thin films is an important n-type semiconductor that showed different applications in solar cells, light-emitting diodes and photoconductors [1-3]. The main attention is to improve the electrical resistivity of CdS thin films and the electrical resistivity is deduced by a doping process. This process improves the electrical properties of CdS films. Different elements, like have been doped into CdS films. Therefore, the aim of the study is to examine the impact of Ni doping in the CdS nano films. Various preparation techniques were employed to manufacture the CdS films like vacuum evaporation [4], chemical bath deposition [5], spray pyrolysis [6] and magnetron sputtering [7]. The basic of CdS and Ni doped CdS were obtained through chemical bath deposition technique. Chemical bath deposition technique is simple, low cost and effective method for preparation of nano sized thin films [8]. The previous work reported the Ni:CdS thin films were concentrated on some linear optical properties like the band gap, photoluminescence [9]. The novelty of this research is to deposit Ni doped CdS thin films by CBD method and to investigate the influence of the Ni dopant on CdS systems on the structural, morphological and electrical properties of the films.

II. RELATED WORK

The researchers has focused in the different molar concentrations of Ni dopant in the CdS thin films. M.M. Patidar et al. [10], reported the sensitivity for light is more upon Ni doping and hence is good for tunable applications like photoconductive switches. R. Murugesan et al. [11] reported, the Ni content in the CdS nano structure has induced ferromagnetic property by induced ferromagnetic couplings between doped Ni^{2+} ions and these films can be used in spintronic applications. Doping of Ni gradually induced strain as well as microstructural transformation and which is persist Photocurrent changes in the host CdS nano films, Manju Mishra Patidar et al. [12]. G.G. Ramteke et al. [13] suggested Mn^{2+} doped CdS nanoparticles has a good potential to be used in optoelectronics, solar cell applications.

III. MATERIALS AND METHODS

Substrate used for film deposition is glass slides, which were first cleaned in distilled water in order to remove the impurities and residuals from their surfaces, followed by rinsing in chromic acid for one day. Then the substrates were washed repeatedly in deionized water, and finally put in ultrasonic agitation with distilled water for 15minutes then dried. CdS films were prepared from cadmium chloride and thiourea by chemical bath deposition technique. Our previous work on the preparation of CdS thin films deposited on to glass substrate at different Ni concentrations which showed a similar preparation conditions [9]. As prepared samples analyzed as the structural, morphological, elemental and electric properties has been studied by XRD, SEM and I-V methods. The proposed overall mechanism for the deposition of CdS thin films are given;

$$CdCl_2 + (NH_2)_2CS + 2OH^- \rightarrow CdS + H_2CN_2 + 2Cl^- + 2H_2O$$

the thickness of the films were calculated by using weight gain method and these values were tabulated in the Table - 1. XRD study was carried out using an X'Pert PRO (PANalytical) diffractometer with Cu K_α radiation (K = 1.5405 Å) in steps of 0.1° over the 2θ range from 20° to 80°. The surface morphology and film composition were carried out by X-ray setup attached with scanning electron microscope (JEOL JSM 840). The electrical study is used to measure the conducting properties of the semiconductor films**.** The photovoltaic characterization of CdS/p-Si and Ni:CdS/p-Si hetero junctions have been instigated by measuring the current-voltage characteristics in dark and under light illumination. A 200W halogen lamp was used for optical illumination of the device. A Keithley 4200 semiconductor parameter analyzer was used to record the photocurrent of the device.

IV. RESULTS AND DISCUSSION

A. X-ray diffraction Analysis

The X-ray diffraction patterns of $Cd_{1-x}Ni_xS$ (x = 0, 0.04 and 0.08 M) films are shown in Figure 1 (a) and (b), respectively. The diffraction peaks showed that the films have polycrystalline nature of hexagonal phase, with (0 0 2) preferred orientation, irrespective of the Ni doping level, which indicates that the incorporation of Ni into Cd sites has not altered the preferential growth [14]. The other peaks observed at 2θ = 24.8°, 26.5° and 28.2° can be indexed to (1 0 0), (0 0 2) and (1 0 1) planes of $Cd_{1-x}Ni_xS$ (x = 0, 0.04 and 0.08 M) films which is indexed as standard pattern (JCPDS card No. 89-2944). No diffraction peaks corresponding to Ni metal or other impurity phases were observed in diffraction patterns. This may confirms the formation of the $Cd_{1-x}Ni_xS$ films. The predominance of the (0 0 2) plane in all the films clearly shows that the growth of the crystal was such that the c-axis was perpendicular to the surface of the substrate. The formation of hexagonal phase CdS thin films with preferential orientation along the (0 0 2) plane was prepared by Wilson et al. [15]. The crystallite size (D) of CdS films is calculated using the Scherrer formula [16],

Average crystallite size, $$D = \frac{k\lambda}{\beta Cos\theta}$$

Where λ is the wavelength of the X-ray used (1.5406 A°), β is the full width at half maximum of the strongest peak, and θ is the Bragg angle. The crystallite size decreased with increased Ni content, this is due to the lowest ionic radius of Ni (0.74 Å) compared to Cd (0.97Å) ions. The crystallite size of the pure and Ni doped CdS films were tabulated in the table - 1. The dislocation density is defined as the length of dislocation lines per unit volume of the crystal [17]. This were calculated from the following formula,

Microstrain, $$\varepsilon = \frac{\beta Cos\theta}{4}$$

Dislocation density, $$\delta = \frac{1}{D^2}$$

The variation of dislocation density (δ) and strain (ε) values of undoped and Ni doped CdS thinfilms with respect to Ni concentration. The crystallite size gradually decreased from 29 nm to 18 nm with the increased Ni concentration [18]. The strain gradually increased with increase of Ni content from X = 0 to 0.08 M. Similarly, the dislocation density values increased due to the increased dopant Ni concentrations [19].

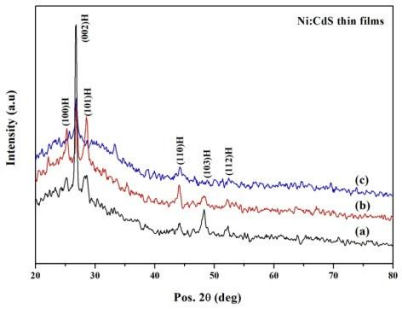

Figure 1: X – ray diffraction pattern of $Cd_{1-x}Ni_xS$ thin films obtained at various Ni concentrations (a) x = 0, (b) x = 0.04 and (c) x = 0.08 M.

B. SEM and EDX Analysis

Figure 2 (a) and (b) are the surface morphologies of undoped and Ni doped CdS thin films, which depicts the deposited films are less dense with observable voids or pinholes. The SEM pictures under 10,000 magnification which depict the spherical like grains due to the controlled growth conditions during the formation of CdS thin films. Small spherical grains starts to joined together in between them and formed the aggleromated big spherical grains under the doping Ni contents.

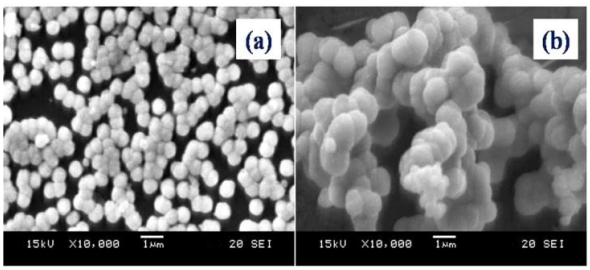

Figure 2 (a, b): SEM images of $Cd_{1-x}Ni_xS$ (x = 0 and 0.08 M).

The EDAX spectra of undoped and Ni doped CdS films are given in Figure 3 (a) and (b), which shows that pure CdS films contains Cd and S elements whereas the doped films contain the Cd, S and Ni elements as it has been expected. The other elements (Ca, Na and Si) that are not expected to be in the deposited films may stem from the glass substrates [20]. From 3 (b), the increment of sulfur content with the increment of Ni concentration (0.08 M) which reduced the sulfur deficiency in Ni doped CdS films.

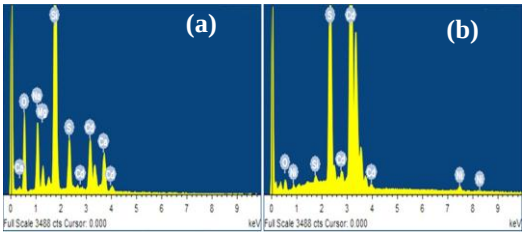

Figure 3 (a, b:) EDX spectra of $Cd_{1-x}Ni_xS$ (x = 0 and 0.08 M).

C. I - V Studies

The electrical study is used to measure the conducting properties of the semiconductor films**.** Keithley interactive digital source-meter (Model-2450) was used for the Hall measurements of pristine annealed and Ni doped CdS thin films. Table 1 shows the Hall measurement results of the undoped and Ni doped CdS thin films. The carrier concentration and mobility of the films were calculated using the Hall coefficient and Four point probe method [21]. Hall measurement for undoped CdS film has lower mobility and carrier concentration due to the high resistivity of film. Hall mobility has increased to a maximum value for $Cd_{0.92}Ni_{0.08}S$ films with the Nickel dopant. The resistivity of the films was calculated by four probe method [22] in the ranges of 0 to 10 V. The ohmic conduction is dominant in this region and in this region the numbers of injected carriers are so high compared to undoped films. The current-voltage (I-V) characteristics of the undoped and Ni doped CdS films are shown in Figure 4 (a - c). All the films exhibit semiconducting behavior at room temperature. While the carrier concentration has increased with Ni dopant concentrations and these values are tabulated in Table 1, the resistivity decreased with increased Ni concentrations due to the increased carrier concentrations of the doped films with the substitution of Cd^{2+} ions by Ni^{2+} ions [23]. The doping materials might have induced the sulphur deficiencies in the doped CdS thin films which might have increased the carrier concentration and also reduced the resistivity of the films [24]. From the experimental results it was noticed that the carrier concentration reaches saturation as heavy doping is carried out that is for concentration of Ni was as 0.08 M. i.e, the remarkable doping effect of Ni doped in CdS thin films is that the resistivity is decreased by the increased electrical conductivity (σ) with the presence of the dopant concentration [25].

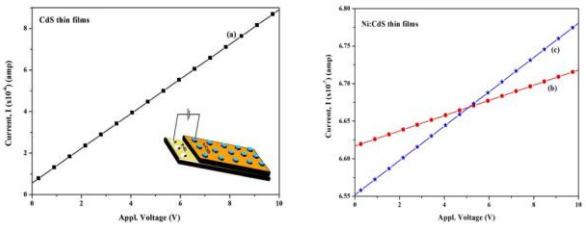

Figure 4: The I-V plots of $Cd_{1-x}Ni_xS$ thin films for (a) x = 0, (inset: schematic view of the device) (b) x = 0.04 and (c) x = 0.08 M

D. Photoresponse studies

Figure 5, shows the room temperature I-V characteristics curves of the Al/Ni-CdS/p-Si/Al based heterojunction diode thin film for undoped and 0.08M of Ni doped CdS thin films. The value of the photocurrent density reveals the photo response behavior of measured in dark and under illumination intensity at 100 mW/cm^2. The inset in this figure displays the scheme of the assembled n-CdS/P-Si photodiode. The I-V curve measured in dark shows the current rectification characteristic of diode devices. Under illumination, both, the reverse and forward current increased with increased illumination intensity, evidencing the conversion of light energy into electrical current by the photodiode device. The minimum of each I-V curve, around at 0 V, shifts to higher voltage with illumination intensity to 0.2, due to the photovoltaic effect. Under illumination, the photo generated electron-hole pairs are separated more effective at 0 voltage than either at forward or reverse bias. This means that, at this condition, in the illuminated diode, the electrons are transferred from the valence band to the conduction band resulting in a photocurrent. Also remarkable is the difference of the photocurrents at the different polarizations (reverse bias, forward bias and 0 voltage). From this, it is clear that the diode showing a photodiode behavior [26]. It can be observed from I-V plot for the forward current is slightly superior compared to that in the forward one under illumination. Such behavior can be explained as the reduction region of semiconductor separates the photo-generated electron-hole pairs under high electric field. The illumination current increases with increasing doping level as predicted by the thermionic emission (TE). This increase in

current can be attributed to the drift of generated electrons and holes of the host. TE theory defines the current through a Schottky diode as given below [27];

$$I = I_o \exp\left(\frac{qV}{nKT} - 1\right)$$

where V is the applied voltage, K is the Boltzmann's constant, n is the ideality factor, q is the charge of an electron, and T is the room temperature. The ideality factor (n) is calculated from the following equations,

$$n = \frac{q}{KT}\left(\frac{dV}{d(\ln I)}\right)$$

and reverse bias saturation current (I_o) are calculated through equations [28]:

$$\emptyset = \frac{KT}{q} \ln\left(\frac{AA^* T^2}{I_o}\right)$$

here all the used symbols are commonly used and have usual meanings. We have measured the current in reversible mode up to 2 V from 0. Turn-on voltage for the fabricated heterojunction is obtained about ~0.43 V, which shows the constructive retort to light [29]. The semilogarithmic curve of the device revealed that the characteristics with the slope of linear region of the forward bias in IV curves. This enables to evaluate the value of (n) and observed to be 4.31. It is known that this value should be unity for an ideal diode; however, we have observed higher value that may be due to interface states, non-uniform film deposition, and series resistance. The y-intercept of semi-logarithmic curve give the value of Io around 6.7034×10^{-7}. The value of Ø is calculated around 0.73 eV for 0.08 M Ni doped CdS thin films (see Table 1). It can be seen that the value of photocurrent enriched up to a certain level under illumination as during this the number of free-charge carriers increase. This indicates that the constructed device have photo conducting nature. Moreover, the spectral responsivity (R_s) is determined using the equation below [30]:

$$R_s = \frac{\Delta I_{ph}}{P_L A}$$

where ΔI_{ph} is the current difference between the I_{photo} and the I_{dark} at a specific monochromatic light illumination, P_L is the intensity of monochromatic light, A is the effective illuminated area of diode. The value of the R_s of Al/n-Ni:CdS/p-Si/Al hybrid heterojunction diode is found to be 329mA/W at a bias of 5 V (see Table 1). The equation for effective quantum efficiency (EQE) is calculated as in the following equation [31]:

$$EQE = \frac{R_s hc}{e\lambda}$$

where R_s is the responsivity of the diode, h is the planck's constant, c is the velocity of light, q is the charge of an electron and λ is the wavelength of the source light. The higher value of the EQE of p-Si/n-CdS (Ni dopant at 0.08 M) hybrid heterojunctions was found to be 68.3% at a bias of 5 V. This confirming the potential of the Ni doped CdS thin films is suitable for multicolor detection applications.

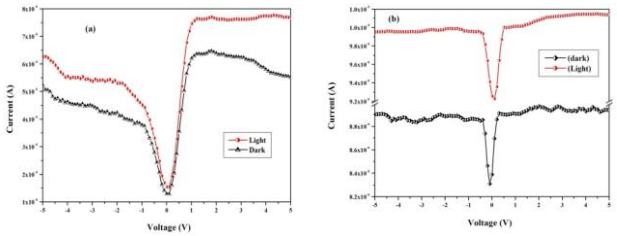

Figure 5: Forward and reverse bias I -V measurements of (a) x = 0 and (b) x=0.08 M, Ni doped CdS hybrid heterojunction photodiodes under dark and under illumination.

Table 1. Structural and Electrical parameters of $Cd_{1-x}Ni_xS$ thin films for (a) x=0, (b) x=0.04 and (c) x=0.08 M.

Ni Concent-rations (x) (M)	Crysta-llite size D (nm)	Micro Strain ε (× 10^{-3}) (lines^{-2}m^{-4})	Hall Mobility (×10^2) (cm^2/Vs)	Carrier Concent-ration (×10^{15}) (cm^{-3})	Resisti-vity ρ [Ω.cm]	Conducti-vity σ (×10^{-3}) (Ω.cm)$^{-1}$
(a) (x=0)	29	2.1	1.38	0.68	141.56	7.064
(b) (x=0.04)	22	2.6	1.83	1.09	122.98	8.131
(c) (x=0.08)	18	2.9	2.32	1.51	104.19	9.598

V. CONCLUSION AND FUTURE SCOPE

Undoped and Ni doped CdS thin films were deposited onto non conducting glass substrate using chemical bath deposition technique. From XRD data, it is confirmed that all samples are in the hexagonal CdS structure. The crystallite size of the films has been calculated by Debye Scherer's equation. It is observed that the average crystallite size decreases with increased Ni content. SEM images showed that spherical like grains are formed on to the substrate well. Energy dispersive X-ray study confirmed the presence of Cd, S and Ni in their respective films. From the electrical studies, Ni doped CdS thin films carrier concentrations were increased with the increased Ni concentrations. The photo response studies gives the higher value of the EQE of p-Si/n-CdS (Ni dopant at 0.08 M) hybrid hetero-junctions photo detectors for Ni-CdS thin films. Those results could be applied in other CdS-based cells, for more effective PV and photo diode conversion device and other applications that require such properties.

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Dr. R. Chandramohan, a specialist in Low Cost Nanomaterials Synthesis and Thin films, More than 35 years of academics, 25 years of research experience in physico-chemical synthesis, characterization and device design with post doctoral experience. To indulge in multi

disciplinary research leading to an explosion of low cost products that may enhance the living conditions of the deprived. To strive hard for creating a world class research team at any University in the emerging fields of Nano Science & Nano Technology. To promote the ethics of science and nano ethics among the emerging researchers. Besides above I have developed interest recently in Gravity, Soil Physics and Environmental Sciences. Looking for Science in Ancient Tamil Literature is also my Passion.

Dr. J. Jebaraj Devadasan is working as Associate Professor in Physics, Pope's College, Sawyerpuram. (TN) India. He has more than 20 years of teaching experience and 4 years of Research Experience. He has published more than 25 research papers in national & international reputed journals. His area of research is Material Science, semiconductor thin films and Nano materials. Currently, six students are perusing Ph. D.

