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Tuning of structural, optical and electrical characteristics of Cu/CdS composite thin films for energy applications

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ABSTRACT: -
This manuscript presents Cu/CdS composite thin films fabricated on glass substrate via spin coating method. The doping of Cu in CdS has been done within the fraction of x= 0.1, 0.3, 0.5 wt%. The structural, optical, electrical and morphological characterization of CdS and Cu doped CdS were characterized by X-ray diffractometer (XRD), UV-Vis spectrometer, Keithley electrometer, FT-IR, Photoluminescence (PL) and Scanning electron microscopy (SEM). XRD spectra confirms cubic phase with particle size distribution of 38-48 nm. The optical direct band gap values have been found in between 2.41 to 1.71 eV for Cu doped CdS composite thin films. Fourier transforms infrared spectrometer (FT-IR) used for bonding analysis and identification of functional groups. The photoluminescence (PL) spectrum emitted a strong peak near band edge emission at approximately 430 nm, attributes better optical quality. A better control in the doping process improves tuning of the band gap, electrical nature for energy applications.

I. INTRODUCTION: -

CdS is a II-VI group semiconductor material with much application such as large area electronic device and solar cells. II-VI group semiconductors with dimension in the nano-meter range have generated considerable interest for researchers and scientific community J.Shadia (2014). CdS is semiconductor material, with direct band gap of 2.40 eVMohammad Afzaal et al (2006). CdS used in transistor W. Wondmagegn et al (2016), FET,LEDs(Molaei M et al 2012), photonics. CdS thin films have been used as a gas sensing material.Copper doped cadmium sulphide (CdS: Cu) quantum particles:topological, morphology and photoluminescence studied Ratnesh Tiwari et al (2013). Cu-doped CdS thin films by chemical bath deposition and ion exchange studied O. I. Diaz‑Grijalva et al (2019). Effect of annealing and dopants on the physical properties of CdS nanoparticles analyzed Rinu Sama et al (2015). Effect of Cu^{+2} doping on structural and optical properties of CdS studiedG. G. Ramtekeet al (2018).Ali Badawi (2021) studied the Effect of Cu-doping on the structure, FT-IR and optical properties of titania for environmental-friendly applications. Effect of Zn content on optical properties and tuning of optical band gap of chemically deposited Cd1-xZnxS thin films by L. S. Ravangave (2013). Efficient red photoluminescence from Al^{3+} and Cu^{+} co-doped CdS QDs embedded silicate glasses studied Kai Li (2021).Investigation of Cu doped cadmium sulphide photoconductive cellsSuchittra Inthong. Microstructure and optical properties of Cu doped CdS nanostructured thin Films by A Sharma et al (2016). Microwave assisted synthesis of undoped and Cu doped CdS nanoparticles and their structural, morphological and optical characterization by K.P. Tiwarya et al (2019).Optical, structural and electrical properties of Cu doped CdS thin films fabricated by SILAR Method D. Pradhabhan et al (2019). Structural properties of copper doped CdS thin films prepared by pulsed laser deposition S.M. Mahdav et al (2008). In this paper a series of Cu^{2+} doped CdS were prepared by a co-precipitation method and characterized by XRD and SEM techniques. The band gap, crystal phase and the morphology of CdS nanocrystals were not found to be affected Cu^{2+} noticeably by Cu doping; there was an optimal Cu

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doping content of wt 5%. The present study mainly incorporated with the, composition; structure, morphology and photoluminescence behaviors of the prepared samples.

II. EXPERIMENTAL DETAILS: -

2.1 Materials: -

Copper (metal) powder (Cu) molecular weight 107.87 g mol^{-1} with purity 99.9 purchased Sigmachemie pvt. Ltd. Cadmium sulphide (CdS) yellow powder of molecular weight 144.47g mol^{-1} with purity more than 99.9% purchased from sigmachemie specialty Pvt. Ltd. (Amaranth West, Maharashtra). Dichloromethane (CH_2Cl_2) Qualikemes Fine Chem. Pvt. Ltd., Propane -2-ol $(CH_3)_2CHOH$, Triton X-100.

2.2 Synthesis of samples: -

CdS and Cu powders used for preparation Cu: CdS composite thin films. To synthesis pristine and copper doped CdS thin films by spin coating method.Copper powder was mixed in CdS powder with different wt% composition using sol gel method. Table 1 represents various combinations of Cu doped CdS films.

S.N.	Material	Molecular weight(gm)	Weight (gm)	1% of total weight (gm)	Doping Material	Molecular weight (gm)	Weight (gm)	1% of total weight(gm)
1.	CdS	144.47	144.47 x 100%=144.47	1.4447	Cu	63.54	63.54 x 0%=0	0
2.	CdS	144.47	144.47 x 99% =143.02	1.4302	Cu	63.54	63.54 x 1%=.6354	.006354
3.	CdS	144.47	144.47 x 97%=140.13	1.4013	Cu	63.54	63.54 x 3%=1.9062	0.019062
4.	CdS	144.47	144.47 x 95%=137.24	1.3724	Cu	63.54	63.54 x 5%=3.177	0.03177

Table: -1. Composition representation of copper doped CdS films

CdS andcopper powder were mixed via sonication for 30 minutes. Di-chloromethane, propan-2-ol were mixed at volume ratio of 10:1 and then pure CdS and copper doped CdS mixed powder were dissolved in the mixed co-solvents. Cu doped CdS mixed powder sonicated at 200V, 20min for doping mechanism. The color of the solution was yellow. Then the solution was stirred for 1.30h at 50^0 C temperatures in a magnetic stirrer to completely dissolve the powder. After stirring, the solution was optically transparent and very faint yellow with light black. After that a small amount of Triton X-100 was add in the solution to make uniform and high quality Cu: CdS thin films.

2.3 Deposition process of Cu: CdS thin films:-

The CdS thin film was deposited using the spin coating method on the glass substrate 2x2cm^2. The glass substrates were cleaned by acetone and double distilled water before thin film fabrication. Fig 1 show schematic representation of coating process

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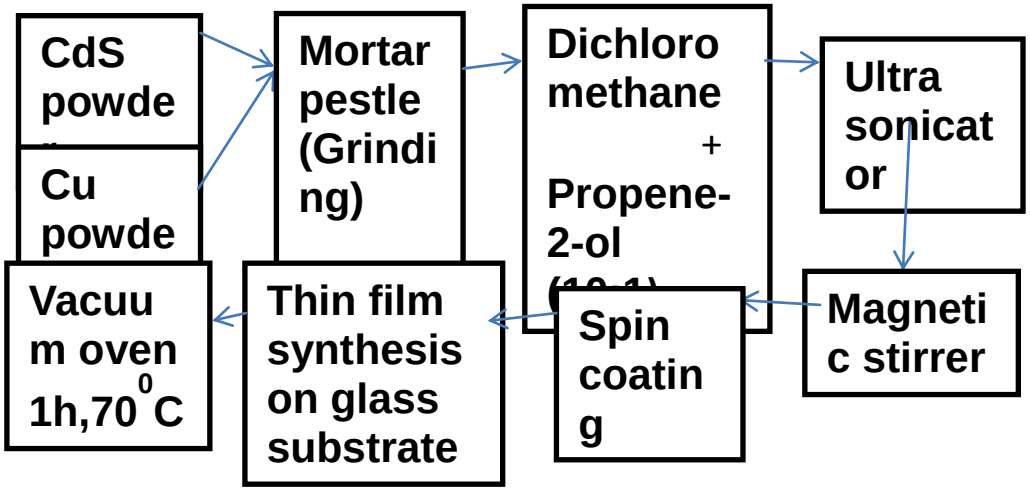

Fig:1 Schematic representation of coating process

The Cu: CdS composite precursor solution was spin coated one time on cleaned substrates at speed 2000 rpm, 60s. This process was completed with pure CdS and different concentration 0.1, 0.3 and 0.5wt% Cu doped CdS. Then the deposited CdS films were dried at 70^0 c at 24 hour in vacuum oven.

2.4 Characterization: -

X-ray diffraction (XRD) spectra of pristine and Cu doped CdS samples have been collected using Bruker AXS Single Crystal X-ray Diffractometer (modal Apex II) in the range (2θ) 10^0-80^0 with step size 1/100 ^{0}. UV-visible spectra have been collected uses shimadzu (UV-2600) wave length range 200-900nm. FT-IR spectra calculated by ALPHA Bruker FT-IR spectrometer (ECO-ATR) in the transmittance mode at room temperature in the wave number range 4000-200 cm^{-1}. Horiba FluoroMax-4 spectrometer used for photoluminescence (PL). The SEM images of nanoparticles were recorded at room temperature by scanning electron microscope EVO MA18 to get information about surface morphology.

III. RESULTS AND DISCUSSION:-

3.1 X-ray diffraction (XRD):

Fig: 3.1 represents XRD spectra of CdS and Cu doped CdS composite thin films prepared by spin coating method. The structural characterization of the undoped and doped CdS samples with different doping concentrations of Cu has been carried out by X-ray diffraction technique using Cu Kα radiation. The XRD pattern of all samples of CdS (Pristine) and Cu/CdS doped with different concentrations of Cu (x=0.0,0.1, 0.3 &0.5wt %) is represented in Fig3. The XRD pattern of Cu/CdS samples indicates three distinct diffraction peaks at three different angles 2θ =26.680, 43.570 and 52.060 corresponding to reflections from (111), (220) and (311) crystal planes respectively which exhibits cubic crystal structure with the help of literature review of CdS well matched with JCPDS card file no.(80-0019) G. G. Ramteke et al. observed similar result 2015**.** The diffraction peaks as obtained in the XRD spectra are significantly broadened with Cu incorporation which reveals the decrease in particle size but not obtained extra peak determined the addition of Cudoping to CdS does not create any change in the CdS matrix. The peak broadening in the diffraction pattern indicates the formation of the particles in nano range. The intense and the sharppeaks reveal the good crystalline of the materials. In addition, the relative intensities of(111), (220) and (311) diffraction are observed to be vary in Cu^{+2} doped CdS nanoparticles. Itcould be due to replacement of $Cd^{+2}$by Cu^{2+} at different lattice sites of CdS. From full width at half maximum (FWHM) of the most intense peak, the average crystalline size for all the samples has been estimated using Debye-Scherer's formula B.E. Warren et.al. (1990).

$$D = \frac{0.9\lambda}{2\sin\theta}$$

Where λ = 0.1541 nm is the wavelength of X-ray diffraction, β is the FWHM in radian of the most IntenseXRD peak and θ is the angle of diffraction. The lattice parameter 'a'for CdS and CdS:Cu nanoparticles is calculated using equation

$$a = \frac{\lambda}{2\mathrm{Sin}\,\theta} \times \sqrt{h^2 + k^2 + l^2}\ \text{Å}$$

The d-spacing for cubic system for 2θ (111)is calculated by using equation

$$d = \frac{a}{\sqrt{h^2+k^2+l^2}}\ \text{Å}$$ and

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The micro strain calculated using equations $\varepsilon = \frac{\beta \cos\theta}{4}$

Samples	Peak position 2θ(111) (degrees	FWHM β(degrees)	$a = \frac{\lambda}{2\text{Sin}\,\theta} \times \frac{}{\sqrt{h^2+k^2+l^2}}$ (Å)	Micro strain $\varepsilon = \frac{\beta \cos\theta}{4}$	$d = \frac{a}{\sqrt{h^2+k^2+l^2}}$ Å
$Cu_{0.0}CdS_{1.00}$	26.0639	0.5628	5.8636	0.12639	3.3853
$Cu_{0.1}Cds_{0.99}$	26.1286	0.4093	5.8771	0.09186	3.3931
$Cu_{0.3}CdS_{0.97}$	26.0219	0.5117	5.8548	0.11495	3.3802
$Cu_{0.5}CdS_{0.95}$	26.0639	0.5117	5.8636	0.11491	3.3853

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Table- 2: The structural parameters of pristine and Cu doped CdS samples

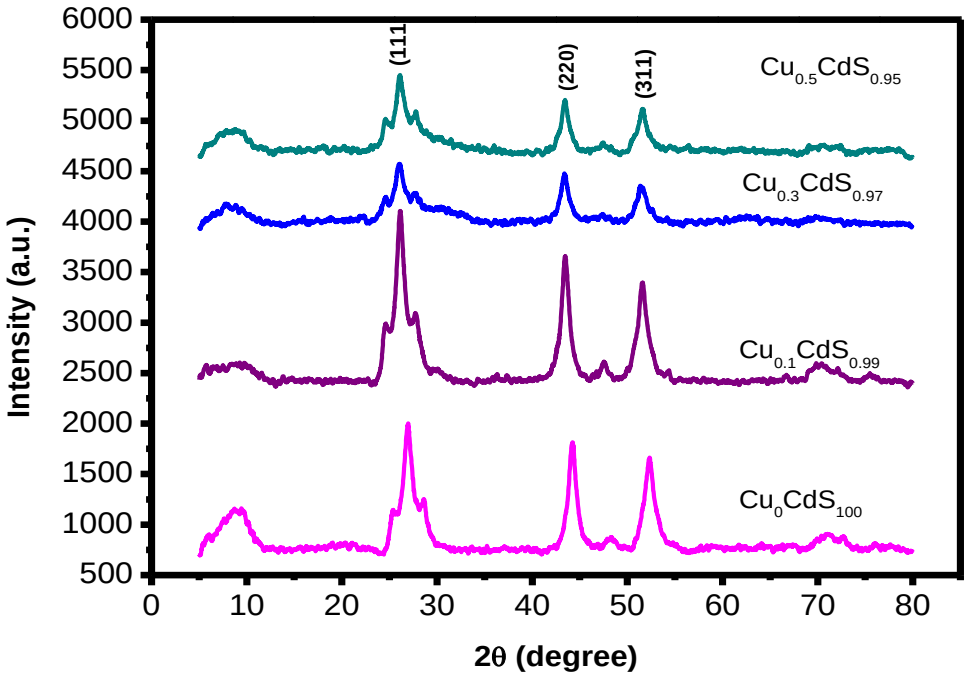

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Fig: 3.1 X-ray diffraction graph intensity v/s 2θ CdS, $Cu_{0.1}CdS_{0.99}$, $Cu_{0.3}CdS_{0.99}$, $Cu_{0.5}CdS_{0.95}$

3.2 Optical spectroscopy (UV-Visible):-

The most remarkable parameter of the optical absorption spectrum of Cu doped CdS thin film show in Fig3.2(a) The optical absorption spectra of the Cu: CdS composite thin films studied using UV-Vis spectrophotometer in the range 200-800nm. CdS is a direct band gap material and absorption edge of the bulk hexagonal CdS is 507 nm which has the band gap energy ~ 2.4 eV R. Elilarassi et al (2010). It can be seen that from 300 to 650 nm spectra present an almost flat absorption region. The excitation wavelength of absorption spectrum observed 480-507nm. Absorption values change with different concentration on 280nm.

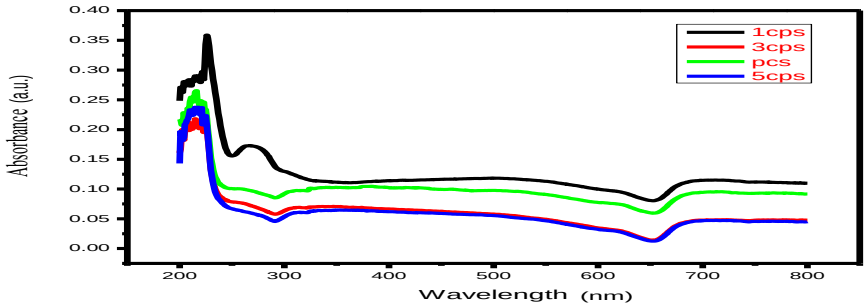

Fig: (a)

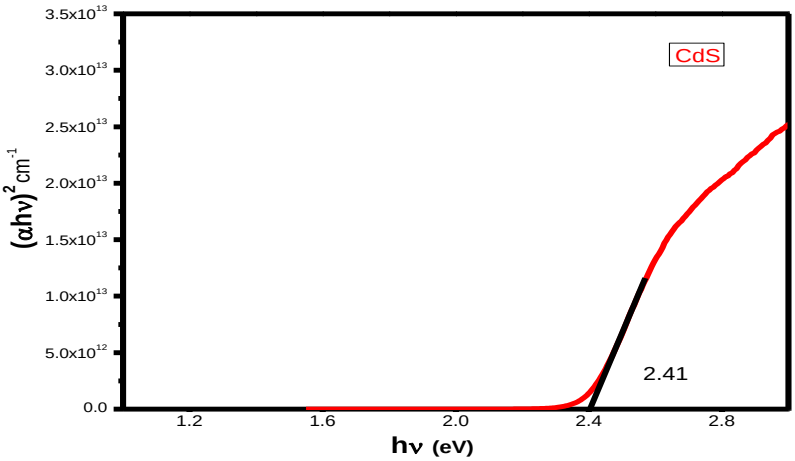

Fig: (b)

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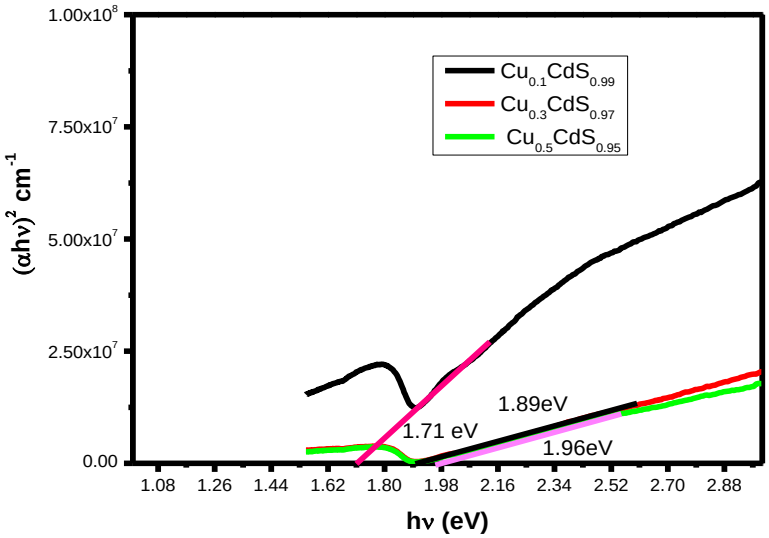

Fig: (c)

Fig: 3.2 (a) Absorption spectra of Cu/CdS thin films (b) Tauc plot forCdS, band gap 2.41eV (c) Tauc plot of Cu/CdS composite x=0.1Cu, band gap 1.71eV, x=0.3Cu, band gap 1.89eV, x=0.5Ag, band gap 1.96eV

The optical band gap of CdS and Cu: CdS composite using Tauc's plot method. The Tauc's equation

$\alpha h\nu = B(h\nu - Eg)^n$ (1)

is used, when α is the absorption coefficient, B is a constant , hν is the photon energy, Eg is the band gap and n equals 1/2 for direct allowed transition. The optical band gap is determine from the parameters obtained of a linear fit of the term $(\alpha h\nu)^2$, O. I. Diaz-Grijalva et al (2019) whose intersection with the x-axis(in α=0) provides the optical band gap. This graphic interpretation starts by transforming the wavelength values to energy level using the equation (1), which comes from: Eg = ch/λ .To obtain absorption coefficient by measured absorbance, assume that sample thickness is 100nm which is measured by cross section of SEM micrograph of the Sample α = 2.303A/t ,A measured by UV-Visible spectrometer and 't' sample thickness. According to Fig 3.2(b), (c),CdS band gap obtained 2.41eV and Cu: CdS composite were 1.71eV, 1.89 eV, 1.96eV with Cu doping percent increases x=0.1, 0.3, 0.5 respectively. Measured band gap values were reduced and then increased with increasing the concentration of Cu doping, which may be due to an increase in film thickness as well as crystalline size S. Rex Rosario et al (2019) obtained same result for Ag:CdS composite with 0, 2,4,6,8 Ag doping percent. Hsu and Shih-Yuan (2008),

3.3 Fourier transforms infrared spectra (FT-IR):

A peak observed at 1530 cm^{-1} is attributed to N-H deformation. A medium peak at 1406 cm-1 is due to C-N stretching. A strong peak at 1113 cm^{-1} occurs due to C-O stretching vibration.

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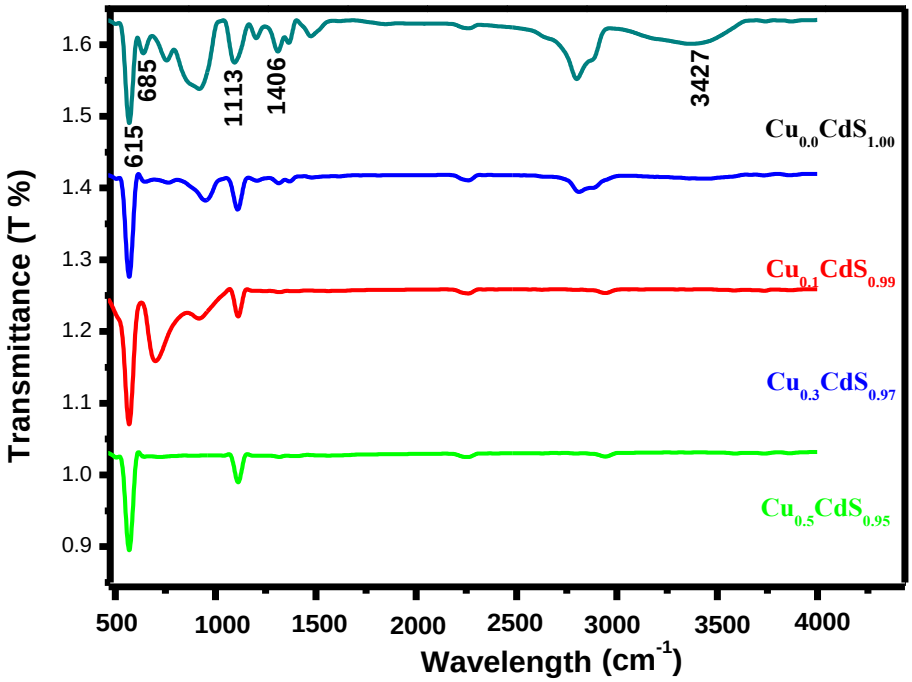

Fig 3.3 shows FT-IR spectra of pristine CdS and Cu/CdS composite thin filmswith different concentration

The peak at 610 cm^{-1} and 685 cm^{-1} which have been attributed to vibrations of CdS bond Vijay Kumar et al (2015), V. J. Fulari, (2015) et al.Figure 3.3 shows that the FT-IR spectra of CdS and Cu/CdS doped CdS thin film for conforming to the existence of constitutional elements functional group and chemical bonding between Cu and CdS. FT-IR spectra were carried out from 400 to 4000 Cm^{-1} at ambient temperature. FT-IR spectra of undoped CdS and x=0.1, 0.3, 0.5 Cu concentration added in CdS shows in below diagram. A broad peak is observed in the higher energy region at 3427 cm^{-1} is assigned to O-H stretching vibration of absorbed water on the surface of the sample.The absorption bands of FT-IR spectra on wave number 1030-1070 cm^{-1} represents the strong stretching due to S=O sulphide group. The sharp bending at the Infrared active spectrum due to absorption band 1365cm^{-1}-1465 cm^{-1}, it is indicated by C–H bond.

3.4 PL(photoluminescence):-

Photoluminescence (PL) is a non-destructive optical technique used for the characterization, investigation, and detection of point defects or measuring the band-gaps of materials. Photoluminescence involves the irradiation of the crystal to be characterized with photons of energy greater than the band-gap energy of that material. In the case of a crystal scintillator, the incident photons will create electron-hole pairs. When these electrons and holes recombine, this recombination energy will transform partly into non-radiative emission and partly into radiative emission. PL Spectra were measured to obtain quality of the Sample. The PL emission spectra of Cu doped CdS samples show that Cu related orange-red emission is centered at 638 nm Ratnesh Tiwari et al (2013).

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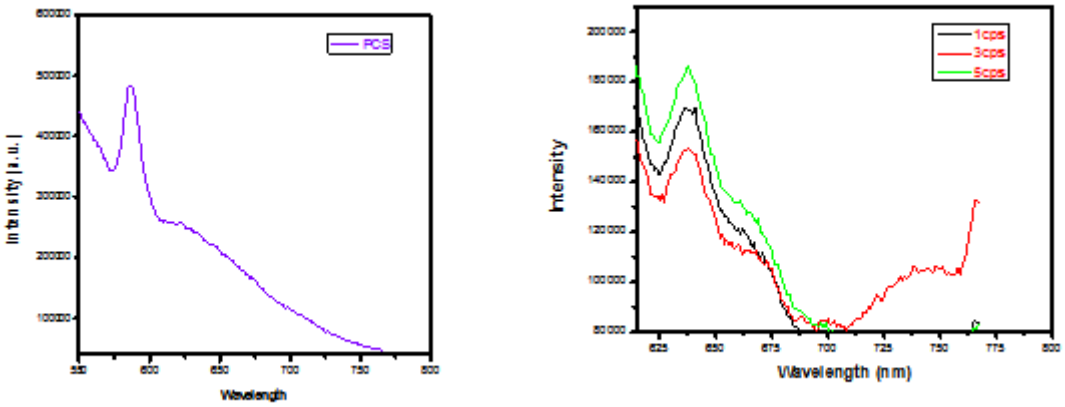

Fig 3.4 PL spectra of (a) pristine CdS (b) Cu/CdS composite thin films with different concentration

3.5 Scanning electron microscopy (SEM):-

SEM is one of the promising techniques used for the topography study of prepared samples, which reveals the information regarding growth mechanism, shape and size of the particles. The morphological study of the prepared pristine and Cu doped CdS thin film at different concentration was done using SEM analysis. Fig.5 shows the SEM image of pure and Cu doped CdS thin films. The images obtained give us indication of considerable amount of change in morphology and porosity on doping. A SEM result shows that the prepared system was found to be agglomerated as synthesized sample. The average particles size obtained from the SEM result is analyzed by histogram curveFig 4.K.P. Tiwarya (2019) obtained similar result for CdS and Cu doped CdS sample.

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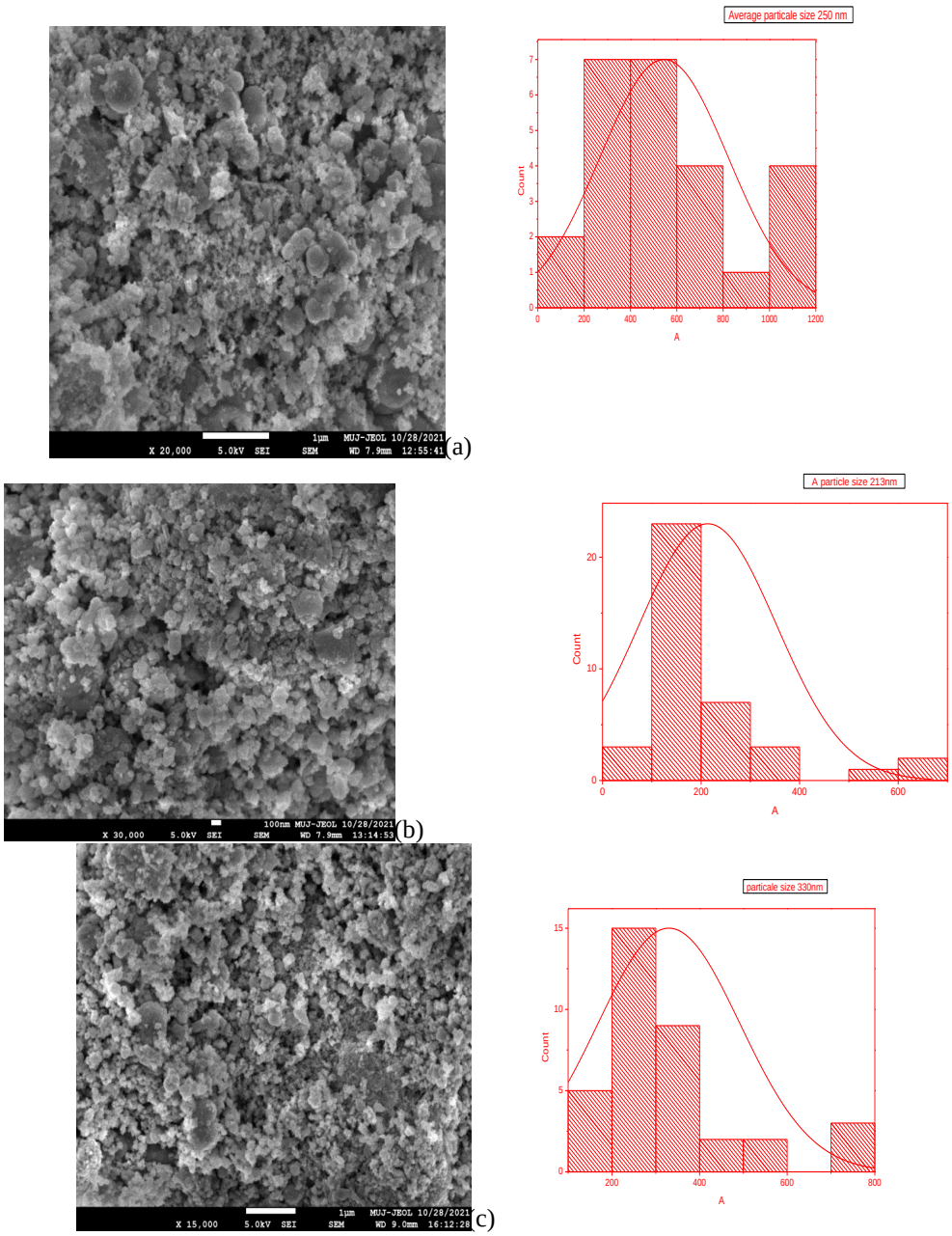

Fig: 3.5. The SEM image and particle size distribution histogram graph of (a) CdS (b) $Cu_{0.1}CdS_{0.99}$(c) $Cu_{0.5}CdS_{0.95}$ composite thin films

3.6I-V characteristics: -

The Current-voltage(I-V) curves of the undoped and Cu/CdS composite thin film obtained Keithley electrometer. The curves of the filmshowed straight lines, indicating that current(I) varies as the voltage (V) in Fig 3.6. Moreover, at thesame applied voltage, the higher concentration sample showed a higher current but obtained straight line also. The differences were small.The resistances of the samples were calculated by ohm's law and tabulated in Table 3.The resistance of the undoped CdS sample was about 3.55 $\times 10^6$ Ω. The resistance of each film slowly dropped from $2.37\times10^6\Omega$ down to 1.89×106 Ω. Suchittra Inthong et al (2019)and Alka Sharma (2016) obtained similar result for CdS thin film.

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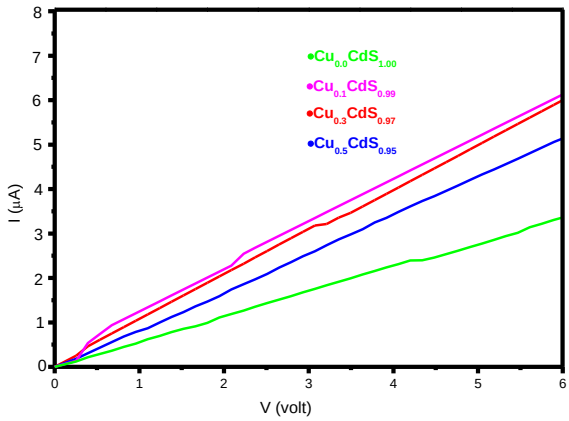

Fig: 3.6 shows the I-V curve of undoped and Cu/CdS composite thin film with different concentration

Samples	Resistance (x 10^6 Ω)
$Cu_{0.0}CdS_{1.00}$	3.55
$Cu_{0.1}Cds_{0.99}$	1.89
$Cu_{0.3}CdS_{0.97}$	1.99
$Cu_{0.5}CdS_{0.95}$	2.37

Table: 3 Resistance values of pristine and Cu/CdS composite thin films

IV. CONCLUSION: -

In the present study it has been concluded that x-ray diffraction spectra show particle size distribution 38-48 nm in cubic to hexagonal plane shift of Cu_x $(CdS)_{1-x}$ thin films. Optical absorption spectra show variation in band gap values with Cu content upto maximum 2.41eVand minimum 1.71eV. FT-IR spectra confirms sharp and strong bonding of composition. PL spectra showing strong visible emission band positioned at about 630 nm in all the prepared Cu doped CdS composite thin films. SEM images show effectively change of microstructure due to Cu content in CdS. The resistance of each film slowly dropped from 2.37×10^6 Ω down to 1.89×106 Ω.

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