Published by :
http://www.ijert.org

International Journal of Engineering Research & Technology (IJERT)
ISSN: 2278-0181
Vol. 5 Issue 09, September-2016

Structural and Electrical Conductivity Studies of Mg Doped $Cd_{0.8}Zn_{0.2}S$ Semiconductor Compounds by Co-precipitation Method

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Abstract **- The bulk $Cd_{0.8}Zn_{0.2}S$ semiconductor compounds doped with different amounts of magnesium(Mg) have been synthesized by controlled co-precipitation method from aqueous solution containing cadmium acetate, zinc acetate and thiourea at 0.02 mol% of Mg_x(x=0~50ml). The solution mixture was made alkaline by adding 25% of liquid ammonia. The structural and electrical conductivity properties of $Cd_{0.8}Zn_{0.2}S$:Mg samples have been studied using X-Ray Diffraction (XRD) and Electrical Conductivity Studies. The samples have polycrystalline nature with hexagonal structure observed in the XRD studies. The average crystalline size varied from 30 to 47 nm and also calculated lattice parameters. The low temperature electrical conductivity measurements of $Cd_{0.8}Zn_{0.2}S$: Mg compounds were studied in the range of 77-300K by Keithley electrometer. It is observed that the increase in Mg doping concentration will increases the electrical conductivity. The values of activation energy were estimated for all the samples and found that the maximum activation energy at lower temperature range is 69.27 meV.**

Keywords - Activation Energy; $Cd_{0.8}Zn_{0.2}S$; Co-Precipitation method; Electrical conductivity; XRD.

1. INTRODUCTION

Wide ranges of modern electronic circuits make use of semiconductor materials, which are doped with impurities to alter electronic properties in a controlled way to suit applications. CdS-ZnS mixed binary semiconductors are most commonly used and studied among binary metal chalcogenides (CdTe, CdSe, ZnS, ZnSe, and CdS). Among varied semiconductor families CdZnS, CdZnSe and CdZnTe are important materials in the field of electronic device fabrication as they provide possibility of tailoring their properties as per requirements [1-4]. One of the ternary semiconductor $Cd_{1-x}Zn_xS$ has novel properties and promising applications particularly in optoelectronics. In $Cd_{1-x}Zn_xS$, which exist in single phase wurtzite structure the value of "x" plays a predominant role in altering and determining their structural, electrical and optical properties. The rapid increase in the resistivity/energy gap of $Cd_{1-x}Zn_xS$ were increases with increase in doping concentration, which leads to more usage of conductivity related applications like near infrared solar cells. So it is interesting to prepare samples of higher energy gap with reasonably good electrical conductivity [5-11].

Recently lots of attention is being directed towards low temperature chemical synthesis of high quality water soluble alloy Nano-crystals. Simple chemical route gives a chance to control the crystalline size, crystalline distribution, to improve crystallinity by altering the concentration of the reagents and their mixing rate at different temperatures. Due to photo defect creation in the lattice, doping will alter electrical and optical properties of semiconductors. The incorporation of alkali metals like Mg into interstitial sites of $Cd_{1-x}Zn_xS$ leads to the formation of Mg^{+2} shallow donor centers. Due to wide technological applications like Alternating Current Thin Film Electro Luminescent (ACTFEL) panels, solar cells, flat television screen, IR detector, sensitive photoconductor, light emitting devices, wide band gap window materials in photoconductive devices and hetero-junction solar cells are widely used CdZnS semiconductor compounds [12-20]. Hence the replacement of CdS with its ternary alloy CdZnS is being attempted in recent years for improvement of CdZnS/$CuInGaSe_2$ solar cell performance. The $Cd_{1-x}Zn_xS$ and related ternary compounds are promising materials for high density optical recording and blue or even UV laser diodes applications based on the structure of $Cd_{1-x}Zn_xS$ [21-26]. The different amounts of Mg doping were done on $Cd_{1-x}Zn_xS$ and their structural and electrical properties were investigated.

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2. EXPERIMENTAL DETAILS

2.1 Sample Preparation:

Co-precipitation method was used to prepare semiconductor powders with different amounts of Magnesium doping of $Cd_{0.8}Zn_{0.2}S$ compound [27-28]. Cadmium acetate [$(CH_3COO)_2$ $Cd.2H_2O$], Zinc acetate [$C_4H_6O_4Zn.2H_2O$], Magnesium Sulphate [$MgSO_4.7H_2O$] and thio-urea [NH_2CSNH_2] chemicals were used for the preparation. 1mol solution of cadmium acetate, zinc acetate and thiourea were prepared proportionally to obtain CdZnS compound with the composition of $Cd_{0.8}Zn_{0.2}S$. 0.02 mol% of Mg_x doped in to the $Cd_{0.8}Zn_{0.2}S$ compound with different amounts (x=0, 5, 10, 15, 20, 25, and 50 ml).

A complexing agent of 1mole solution of tri-ethanol-amine [$C_6H_{15}NO_3$] was added to the prepared solution. This solution was made alkaline with 25% of liquid ammonia [NH_3] to maintain pH of 10 and heated at 80°C with a constant stirring process for 1 hr. The colour of the solution changes from white to dark yellow, which indicates the formation of precipitation. The bath was heated further for 3 hrs to complete the reaction. The precipitate was filtered through whattman filter paper No.40 and repeatedly rinsed with de-ionized water. The final precipitate was dried at room temperature. The sample was placed into a high temperature tubular furnace (1000°C) with facility to pass inert gas through the tube. The precipitate was pre-sintered for 2 hrs at 300°C in nitrogen atmosphere. After slow cooling (2° C/min), the heat treated precipitate was made to fine powder and pelletized to10 mm diameter with 2 mm thickness under the pressure of 10 ton/cm^2. The pellets were sintered at 800°C for 6 hrs in nitrogen gas maintained uniform pressure of 0.2 kg/cm^2[13]. The pellets of $Cd_{0.8}Zn_{0.2}S$ compound containing different amounts of 0.02 mol% of Mg_x (x =0, 5, 10, 15, 20, 25 & 50 ml) were prepared and these pellets used for XRD and electrical conductivity studies.

2.2 Measurements:

X-ray Diffraction (XRD) spectra's studies were performed using SHIMADZU XRD-7000 X-Ray diffractometer with Cu-Kα radiation (λ=1.5418A°) and recorded at room temperature in the angular range of 20°≤2θ≤80° at a scan speed of 0.02°/s. Two-probe method [6] with high grade Eltecks 1228C silver paste as electrodes were used to measure the low temperature electrical conductivity. Potential drop across the sample were measured using a Keithley 182 Sensitive digital voltmeter and multimeter (Keithely, Model 2000) was used to measure the output of the temperature sensor. The electrical conductivity measurements were performed for different temperatures ranges (77-300 K).

3. RESULTS AND DISCUSSIONS

3.1 X-ray diffraction studies:

Structural studies are carried out by X-ray diffraction of undoped and Mg_x (x in ml) doped $Cd_{0.8}Zn_{0.2}S$ samples. XRD patterns of undoped and Mg_x(x=0~50 ml) doped $Cd_{0.8}Zn_{0.2}S$ samples were showed in Fig.1 and it is confirm the formation and composition of all $Cd_{0.8}Zn_{0.2}S$:Mg_x samples with x=0~50 ml in XRD patterns. The diffraction peaks in all samples found good polycrystalline nature of the compounds. The observed peak positions (2θ) and d-values were compared with the Joint Committee on Powder Diffraction Standard (JCPDS-ICDD card No.49-1302) data of $Cd_{0.8}Zn_{0.2}S$ with hexagonal phases are shown in Table 1 and found that they matched within the experimental limitations and also allotted miller indices. It implies that standard values of undoped and Mg doped $Cd_{0.8}Zn_{0.2}S$ possess hexagonal structure and further, no other impurity phase is present in all the samples. The $Cd_{0.8}Zn_{0.2}S$ compound samples of hexagonal structure were allotted with major peaks occurring due to the reflections from (100) (002) (101) and (110) planes. The samples peaks were matched with the values reported by other researchers [29-36].

Table 1: XRD data of JCPDS-ICDD $Cd_{0.8}Zn_{0.2}S$ (H) File (49-1302) and Observed data of undoped and Mg_x(x=10ml) doped $Cd_{0.8}Zn_{0.2}S$ samples

JCPDS-ICDD data of $Cd_{0.8}Zn_{0.2}S$ (H) (#49-1302)			Observed X-ray diffraction data of undoped $Cd_{0.8}Zn_{0.2}S$		Observed X-ray diffraction data of Mg(10ml) doped $Cd_{0.8}Zn_{0.2}S$	
2θ	d(A°)	hkl	2θ	d(A°)	2θ	d(A°)
25.522	3.49	100	25.5620	3.48197	25.511	3.48881
27.103	3.28	002	27.2413	3.27102	27.1840	3.27778
28.799	3.09	101	28.9486	3.08187	28.8812	3.08891
37.472	2.40	102	37.4437	2.39988	37.3800	2.40382
44.870	2.019	110	44.5625	2.03163	44.4536	2.03636
48.943	1.86	103	48.7252	1.86735	48.9200	1.86037
52.273	1.75	200	52.7526	1.73388	52.3600	1.74595

All sample lattice parameters were calculated using the equation 1, based on hexagonal phase [36].

$$\frac{1}{d^2}=\frac{4}{3}\left(\frac{h^2+hk+l^2}{a^2}\right)+\frac{l^2}{c^2} \quad \text{for Hexagonal} \qquad (1)$$

The calculated lattice parameters with x=0 ml are a=0.435 and c=0.534 nm and for the doped compound with x=15 ml is a=0.437 nm and c=0.534 nm respectively.

The average crystalline sizes of undoped and Mg doped $Cd_{0.8}Zn_{0.2}S$ compounds were calculated using Scherer equation 2 [6, 37].

$$D=\frac{K\lambda}{\beta\cos\theta} \qquad (2)$$

From the above equation, D is the average crystalline size, K is constant, taken value as 0.94, λ is X-ray wavelength, β is Full Width at Half Maximum (FWHM) of the peak and θ is the Bragg's angle. It is found that average crystalline size of the compounds varies between 30 nm and 47 nm. Lattice parameters and average crystalline sizes of undoped and Mg doped $Cd_{0.8}Zn_{0.2}S$ compounds are shown in Table 2.

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Table 2: Average crystallite sizes and Lattice parameters of undoped and Mg doped $Cd_{0.8}Zn_{0.2}S$ samples

Sample CdZnS:Mg_x x in ml	Average crystallite size D(nm)	Lattice parameters	
		a(nm)	c(nm)
x=0	42	0.435	0.534
x=5	38	0.437	0.536
x=10	47	0.436	0.535
x=15	36	0.437	0.534
x=20	46	0.443	0.543
x=25	44	0.436	0.535
x=50	30	0.435	0.532

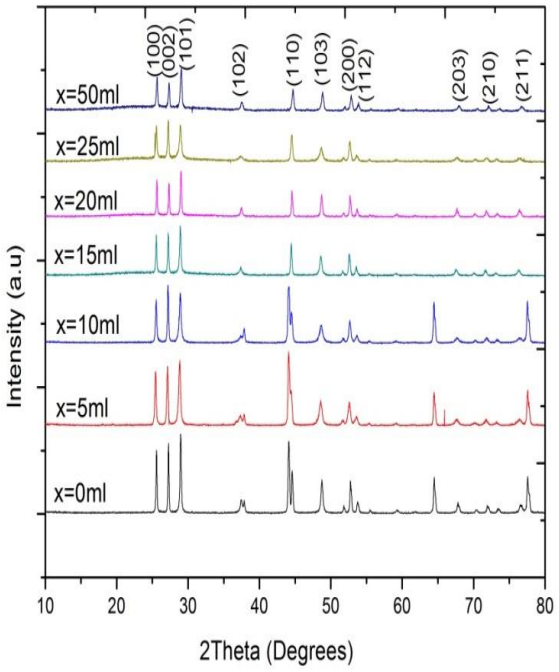

Fig.1. XRD Difractograms of $Cd_{0.8}Zn_{0.2}S:Mg_x$ (x=0~50 ml) semiconductor compounds

3.2 Electrical conductivity studies:

The measurements of dc electrical conductivity of undoped and doped with different amounts of Mg_x(x= 0, 5, 10, 15, 20 and 25 ml) in $Cd_{0.8}Zn_{0.2}S$ compounds were performed in the low temperature region (77-300 K), to understand basically the effect of Mg on charge carrier related movements with temperature in $Cd_{0.8}Zn_{0.2}S$ compounds. Most commonly, semiconductors and metals show σT variation with temperature as exponential function relating to their activation energies. Arrhenius plots of undoped and magnesium doped $Cd_{0.8}Zn_{0.2}S:Mg_x$(x=0 and 10 ml) samples are shown in Fig.2a and 2b. It shows that the conductivity increases with the increase in temperature and with Mg doping concentrations. In all the samples increase in electrical conductivity is governed by thermally activated process. The increase of electrical conductivity with temperature is according to the typical activation law [38]. Graphs of ln(σT) vs $10^3/T$ are drawn and used to calculate the activation energy. These graphs showed two linear regions of conductivity in each sample. Above two regions have been attributed to transitions from a sub band of the valence band to the conduction band. A similar type of variation was also observed by H.A Zayed et al [39] and D.Pathinettam Padiyan et al [40]. In sample with x=0 ml and 10 ml, the plots shown in Fig.2a and 2b exhibits Arrhenius behavior in two different temperature regions (i) (77-125 K) and (ii) (125-300 K).

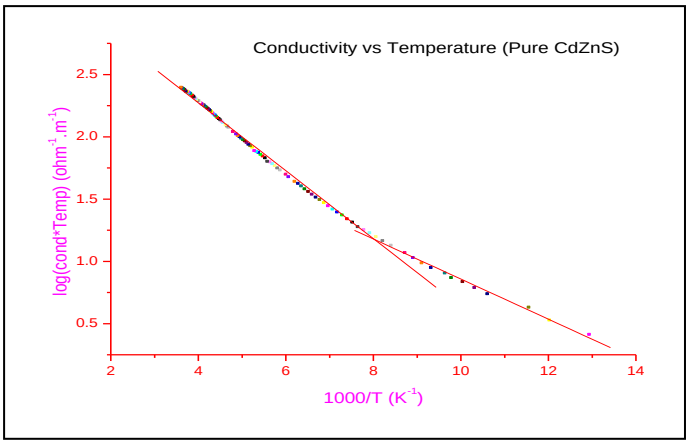

Fig.2a. Graph of ln (σT) versus $10^3/T$ for undoped $Cd_{0.8}Zn_{0.2}S:Mg_x$(x=0 ml) composition

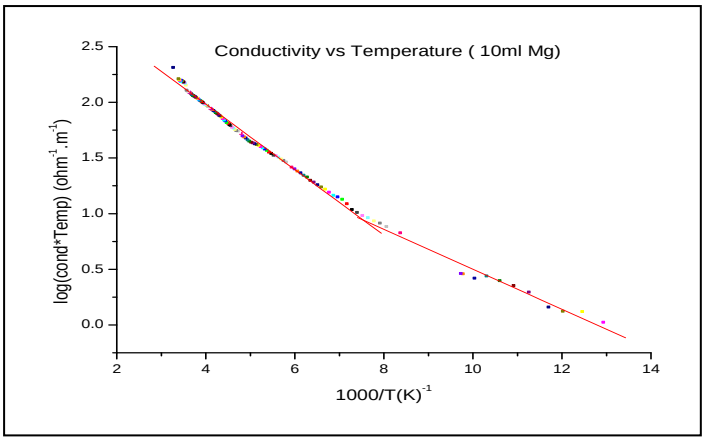

Fig.2b. Graph of ln (σT) versus $10^3/T$ for doped $Cd_{0.8}Zn_{0.2}S:Mg_x$(x=10 ml) composition

The activation energy for all the two regions is found by the equation 3.

$$\sigma = \sigma_0 \exp\left(\frac{-Ea}{kT}\right) \qquad (3)$$

From above equation, σ_0 is the pre exponential factor, Ea is activation energy, k is the Boltzmann constant and T is temperature in Kelvin.

The linear regions observed in ln (σT) vs $10^3/T$ graphs of all the compounds are fitted with the linear equations and the activation energies corresponding to them are computed. The activation energy values of two temperature regions calculated for all the samples are given in Table 3. From the table one can understand easily that, there are two activation energies in each compound, one corresponding to lower temperature region (77-125 K) and

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ISSN: 2278-0181
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the other at slight by higher temperature region (125-300 K). These regions are corresponding to two extrinsic conductivity regions, most probably occurring due to shallow impurity levels having in both activation energy regions. The existence of various defects such as dislocations, structural disorders, surface imperfections, also plays a role in the variation of conductivity. The behavior of electrical conductivity in semiconductors follows different mechanisms at below room temperature like [41],

(i) Due to the drifting of charge carriers, the ordinary conductivity applies at higher temperature (above room temperature),

(ii) Electrical conductivity owing to hopping of charge carriers due to existence of localized states around E_F and

(iii) Electrical conductivity due to thermally assisted hopping

The activation energies of compound in the temperature region 77-125K varies from 56.71 meV to 69.27 meV, which referred to the conduction due to hopping of charge carriers owing to existence of localized states around E_F and in other temperature region 125-300K it changes from 31.57 meV to 57.44 meV, referred to the electrical conductivity is due to thermally assisted hopping. The type of defects that are responsible for these levels cannot be inferred from the conductivity studies alone. However, the literature reports certain vacancy interstitial complexes having activation energies of range 75 meV. Since the maximum activation energy observed in lower temperature range is 69.27 meV, it may be the Mg^{+2} interstitial is getting complexed with the vacancies of the constituent ions and are responsible for higher activation energy levels. Whereas the lower activation energy levels may be due to Mg^{+2} interstitial defects. The possibility for the formation of Mg^{+2} interstitial is quite good for the reason that the size of Mg (Atomic No.12) is much less than Cd (A.No.48), Zn (A.No.30) and S (A.N0.16). It was observed that from the Table 3 the shallow activation energy increases with increase in doping concentration. The graphical variation of activation energies of two shallow level with doping concentration are shown in Fig.3 and 4.

Table 3: The activation energy (Ea) values of $Cd_{0.8}Zn_{0.2}S:Mg_x$ (x=0~25ml) semiconductor compounds

Sample $Cd_{0.8}Zn_{0.2}S:Mg_x$ (x in ml)	Activation Energy(meV)(1) (77-125K)	Activation Energy(meV)(2) (125-300K)
0	56.71	31.57
5	60.30	33.10
10	61.41	35.60
15	64.45	40.19
20	67.62	42.54
25	69.27	57.44

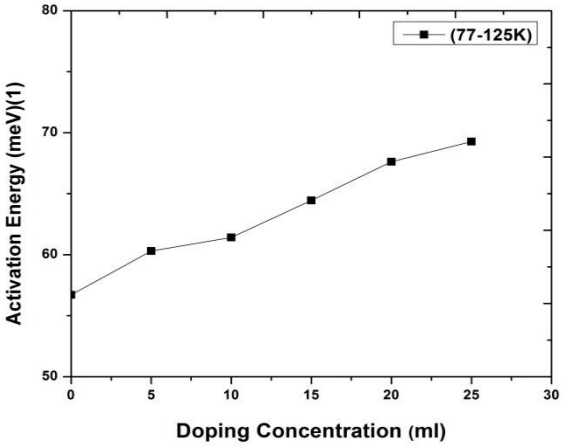

Fig.3. Activation Energy (meV) (1) versus Doping concentration (ml)

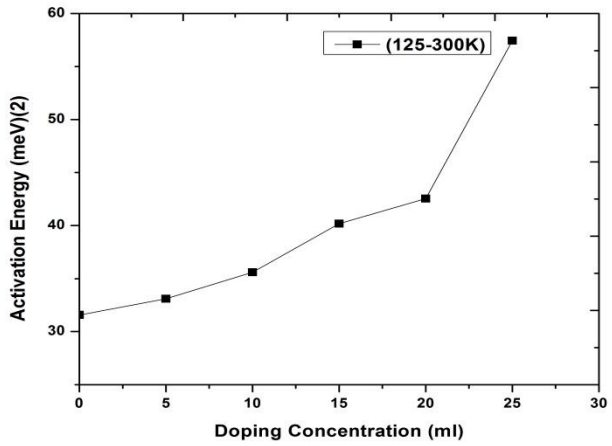

Fig.4. Activation Energy (meV) (2) versus Doping concentration (ml)

4. CONCLUSIONS

1. Undoped and Mg_x(x=0~50ml) doped $Cd_{0.8}Zn_{0.2}S$ semiconductor compounds are prepared by controlled co-precipitation method.
2. In the XRD studies, the samples have polycrystalline nature with hexagonal structure and the average crystalline size varied from 30 to 47 nm.
3. Lattice parameters of undoped and Mg doped $Cd_{0.8}Zn_{0.2}S$ are determined.
4. The increase in temperature leads to increase in electrical conductivity, which confirms the semiconductor nature of the samples.
5. It is observed that the activation energies below room temperature in temperature regions (77-125 K) and (125-300 K) are increase with the increase in the doping concentration of Mg.
6. $Cd_{0.8}Zn_{0.2}S$ semiconductor compounds finds more useful in Opto-electronic devices, because of increase in the conductivity with the increase of Mg doping concentration.

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ISSN: 2278-0181
Vol. 5 Issue 09, September-2016

ACKNOWLEDGEMENTS

One of the authors thanks the UGC-RGNF, New Delhi for providing research fellowship. Authors thank to University College of Technology, Osmania University, Hyderabad and the Department of physics and also thank Prof. D.JayaPrakash for permitting to use the analytical facilities.

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