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STRUCTURAL CHARACTERIZATION OF VACUUM EVAPORATION COPPER INDIUM DISELENIDE THIN FILMS

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Copper Indium diselenide thin films have been deposited on to well cleaned glass substrates in a vacuum of 10^{-5} Torr. The thickness of the films have been determined by micro balanced method and confirmed by multiple beam interferometric method. The structural characterization was carried out by X-ray diffraction (XRD) and Scanning electron microscopic (SEM) studies. These studies confirm the polycrystalline nature of the deposited films with chalcopyrite structure. The structural parameters such as lattice constants, grain size, dislocation density and strain have been evaluated.

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1. Introduction

Thin chalcogenide films are of particular interest for the fabrication of large area photodiode arrays, solar selective coatings, solar cells, photoconductors, sensors, etc. [1]. Copper chalcogenide thin films have a number of applications in various devices such as solar cells, super ionic conductors, photo detectors, photo thermal conversion, electro conductive electrodes, microwave shielding coating, etc. [2-5]. The ternary semi conductors of the type I-III-VI_2 have been intensively studied in recent years because of their potential applications in a variety of electro optic devices. Among these, copper indium diselenide is a promising material for the fabrication of thin film solar cells because of its high absorption coefficient, suitable band gap, good thermal, environmental and electrical stability and also because of the potential low cost methods in its preparation [6].Varela et al [7] have deposited $CuInSe_2$ films on to glass substrates by co-evaporation technique. They have observed that $CuInSe_2$ films show both structures chalcopyrite [tetragonal (U)] and sphalerite [cubic (F)] depending on the growth conditions. The sphalerite structure is dominant at low substrate temperatures and the chalcopyrite structure alone appears at high temperatures. $CuInSe_2$ films with chalcopyrite and sphalerite structures were found to have orientation along (112) and (111) directions respectively. The grain size was found to be 0.2 μm. Polycrystalline $CuInSe_2$ films of thickness 1μm have been deposited on glass substrates by Manohar lal et al [8] by solution growth technique. They have found the films to have chalcopyrite structure. Polycrystalline $CuInSe_2$ thin films have been prepared by Dieter schmid et al [9] on molybdenum coated soda lime glass. They have found the Mo-surface to be significantly

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oxidized and the films were found to have chalcopyrite Structure. Zegadi et al [10] have grown $CuInSe_2$ films of thickness 1000-2500 Å by a three source co-evaporation technique and have reported that the films exhibit chalcopyrite structure with a preferred orientation along (112) direction. The present work deals with the structural properties of copper indium diselenide thin films prepared by single source thermal evaporation.

2. Experimental

The $CuInSe_2$ compound was prepared by taking stoichiometrically weighed pure Cu, In and Se.The mass of the elements have been measured using Metler AE 200 digital micro balance. The weighed high purity elemental copper, indium and selenium are taken in a pure cleaned quartz ampoule 65 mm long with 10 mm diameter. A pressure of $2x10^{-4}$ Torr is achieved in the ampoule along with the material. The vacuum sealing is done using oxy indane gas. The sealed ampoule is placed in a rotating furnace and the ampoule is gradually heated at the rate of 100°C/hour step by step up to a temperature of 1100°C. The ampoule is maintained at 1100°C for 24 hours and then cooled slowly to room temperature at a rate of 100°C/hour. The temperature required for synthesisation was determined from the phase diagram of $CuInSe_2$ alloy. During the course of heating and cooling the quartz ampoule is rotated continuously to ensure homogeneity in the molten mixture. After cooling, the alloy is taken by breaking the ampoule. In the present investigation the structure of $CuInSe_2$ compound and $CuInSe_2$ films have been analyzed using a X-ray diffractometer (Model – PHILIPS-PW3710) with a nickel filtered CuK_α radiation (λ = 0.154 nm) at 40KV and 25 mA.

3. Results and discussion

3.1 Structural Properties of $CuInSe_2$ Thin Films

The X-ray diffractogram of the prepared $CuInSe_2$ compound is shown in Fig. 1. The presence of sharp peaks in the X-ray diffractogram shows that the prepared $CuInSe_2$ compound is polycrystalline in nature. The $CuInSe_2$ compound is found to exhibit chalcopyrite structure with a preferred orientation along certain crystallographic planes which have been identified as (101),(112),(211),(220),(312),(400) and (316) planes. This is in good agreement with Senthil et al [11]. The lattice parameter values a and c of the bulk $CuInSe_2$ compound has been found to be 5.784Å and 11.617Å respectively and they are in good agreement with the ASTM values= 5.782 Å and c = 11.619 Å. The X-ray diffraction pattern of $CuInSe_2$ thin films of different thicknesses is shown in Fig. 2.

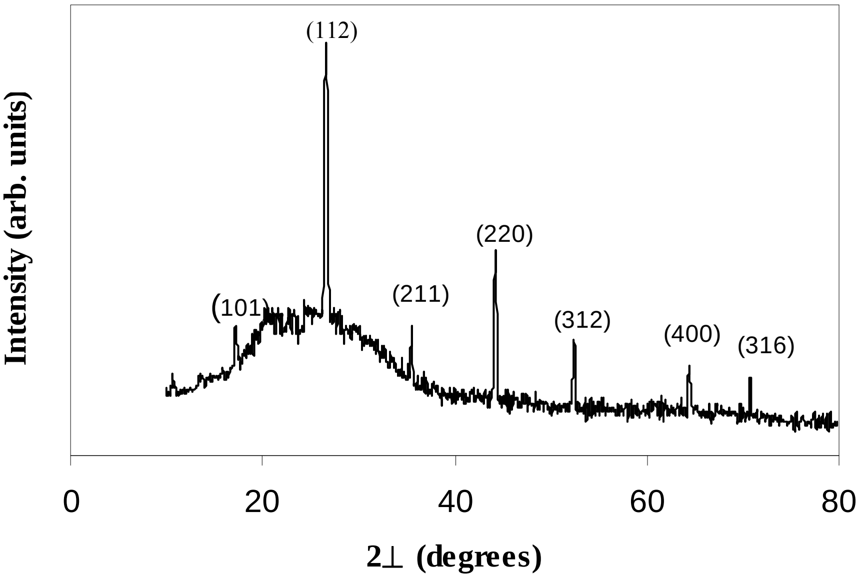

Fig. 1. X-ray diffractogram of the bulk CuInSe$_2$

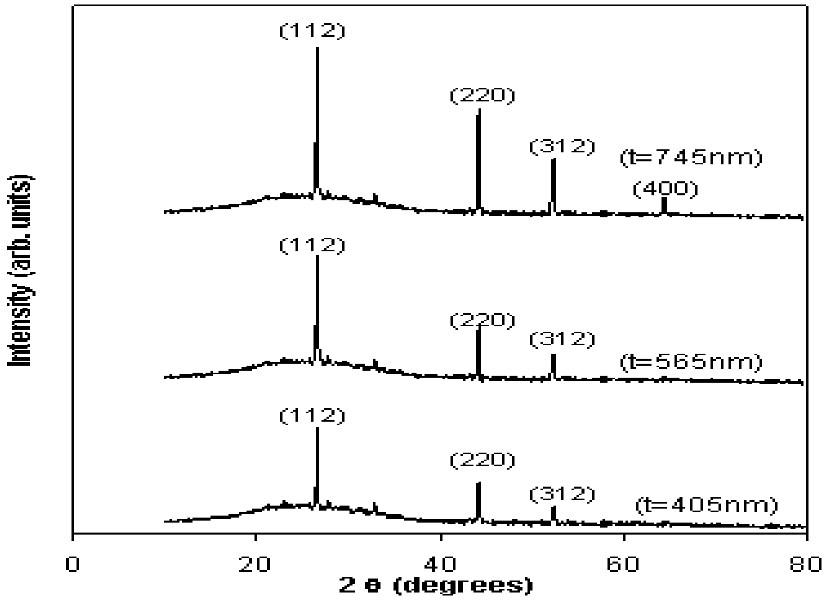

Fig. 2. X-ray diffractograms of CuInSe$_2$ thin films of different thicknesses

All the deposited CuInSe$_2$ films are found to be polycrystalline in nature exhibiting chalcopyrite structure. The chalcopyrites are found to have orientation along (112), (220) and (312) directions. The (400) orientation is seen only in the film of thickness 745 nm. The structural parameters of the deposited CuInSe$_2$ films like lattice parameter values (a, c), c/a ratio, tetragonal distortion, grain size, dislocation density and strain have been evaluated and are shown in Table. I. It is observed that the grain size increases but the strain and dislocation density decrease with the increase of film thickness. A similar dependence of grain size, strain and dislocation density on film thickness has been reported by Dhanam et al [12] for chemical bath deposited CuInSe$_2$ films.

Table .I. Structural Parameters of $CuInSe_2$ Thin Films

Film thickness (nm)	Lattice parameter		c/a ratio	Tetragonal Distortion 2-c/a	Grain size D (nm)	Dislocation Density $\times 10^4$ lin/m^2	Strain ε $\times 10^{-4}$
	a Å	c Å					
405	5.785	11.597	2.0046	-0.0046	79.82	1.569	4.752
565	5.786	11.618	2.0079	-0.0079	150.98	0.438	2.668
745	5.779	11.590	2.0055	-0.0055	208.76	0.229	1.929

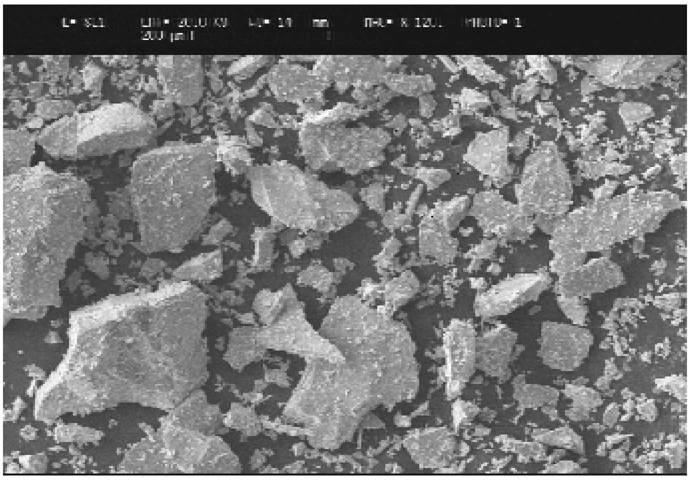

Fig. 3. SEM micrograph of $CuInSe_2$ bulk powder

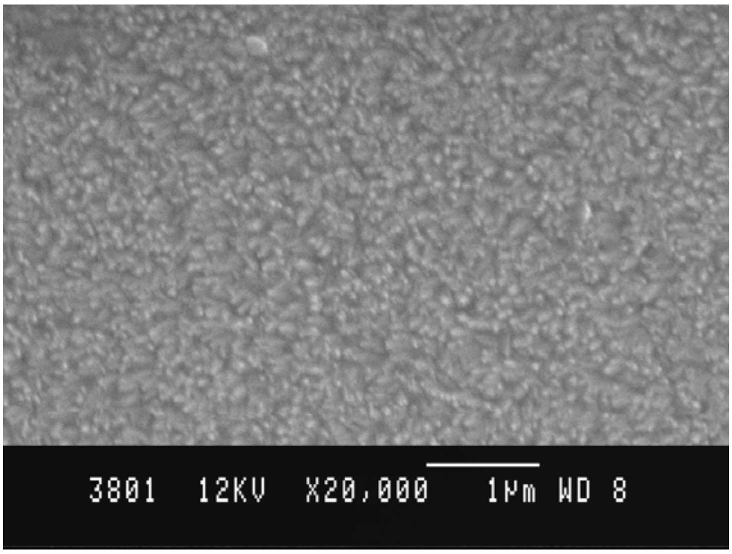

Fig. 4. SEM micrograph of the deposited $CuInSe_2$ thin film of thickness 405nm

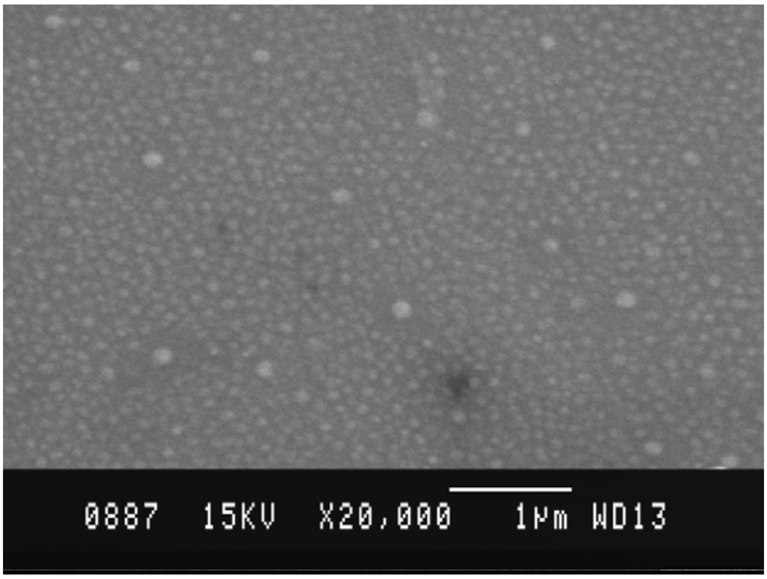

Fig. 5. SEM micrograph of the deposited $CuInSe_2$ thin film of thickness 565nm

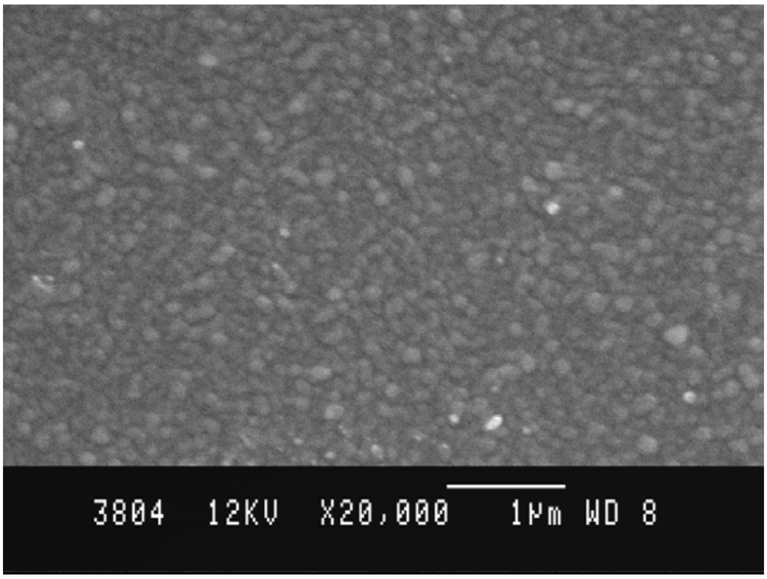

Fig. 6. SEM micrograph of the deposited $CuInSe_2$ thin film of thickness 745nm

The SEM photographs of $CuInSe_2$ compound and as deposited $CuInSe_2$ thin film of thicknesses of 405 nm, 565 nm and 745 nm are shown in figures 3-6 respectively. The grain size of CIS is strongly affected by Cu: In ratio of the film. The SEM micrographs confirm the crystalline nature of the film. The grain size is found to increase with increase in film thickness.

4. Conclusion

$CuInSe_2$ alloy has been prepared using high purity elemental Cu, In and Se taken according to their weight percentage. The $CuInSe_2$ compound is found to exhibit Chalcopyrite structure.$CuInSe_2$ thin films of different thicknesses have been deposited on well cleaned glass substrates by thermal evaporation technique. The films are found to be polycrystalline in nature, single phase exhibiting chalcopyrite sructure. The crystallites are found to have preferred orientation along the (112) direction. As the Substrate temperature is increased the films are found to exhibit a highly preferred orientation along the (112) direction like a single crystal. The SEM micrograph of the deposited $CuInSe_2$ films confirms the crystalline nature of the films.

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