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Synthesis $CuInSe_2$ (CISe) Thin Films Prepared from Metal-Ethanolamine Complex Compound

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Abstract. $CuInSe_2$ (CISe) thin film was successfully fabricated from copper and indium salts with ethanolamine as precursors. All of these precursors were dissolved and formed complex compounds with ethanolamine simultaneously which deposited on soda lime glass by spin coating at 200 rpm, followed by heat treatment in the ambient atmosphere at 200^{o}C for 120 minutes and finally selenization at 550 ^{o}C using selenium pellets under Ar (95%) + H_2 (5%) for 120 minutes to fabricate CISe thin film. Reaction mechanism, structure, morphology and chemical composition also reported in this work.

Keyword: CISe, thin film, ethanolamine, complex compound, spin coating, selenization.

1. Introduction

$CuInSe_2$ (CISe) is an ideal material for thin film solar cell application due to has optimum band gap (1.01 eV) and high absorption coefficient (10^5 cm^{-1}) [1,2]. CISe thin films could be prepared by vacuum and non-vacuum methods. However, non-vacuum process such as chemical process is preferable due to low cost but still get high efficiency [3]. Although chemical process is preferable, this method using harmful solvents such as hydrazine and pyridine [4,5]. Besides that, chemical process could leaves carbon in the thin film as residue [6]. In this work, CISe thin film has been fabricated by chemical process using ethanolamine as a solvent, deposited by spin coating and followed by selenization.

2. Experimental Detail

2.1 Materials and Methods

In this work, the precursor solution was made from 0.1 M copper from copper(II) acetate and 0.1 M indium from indium(III) chloride 0.1 M from Aldrich were dissolved in 30% ethanolamine (ETA) which dissolved in ethanol (from both from Aldrich) and exhibited a blue color with 1:1 Cu/In ratio. Moreover, 0.05 mL of precursor solution was deposited at 2000 rpm for 10 seconds on soda lime glass (SLG) which cleaned with acetone, ethanol, deionized water using ultrasonic for 15 minutes respectively and dried by nitrogen gas in order to eliminate contaminants further. Subsequently, deposited precursor was dried in tubular furnace at 200 ^{o}C for 120 minutes until black shinny color was appeared. Repeat the deposition and the heat treatment appropriately until get desirable film thickness. Finally, the CISe thin film obtained by selenization in tubular furnace at 550 ^{o}C using selenium pellets as selenium source under Ar (95%) + H_2 (5%) atmosphere for 120 minutes. After selenization, the sample was cooled to room temperature under natural cooling condition.

2.2 Characterization of CISe thin film

The morphology was observed by scanning electron microscopy (SEM, Hitachi S4800, Japan). The chemical composition of the films was analyzed by energy dispersive x-ray spectrometry (EDS, Horiba, Japan). The crystal structures were determined by x-ray diffraction (XRD, PANalytical MPD, Netherlands). The absorption spectra were recorded using a UV-Vis-NIR spectrophotometer (Varian Carry 5000, U.S.A). The electrical properties were examined by Hall-Effect measurements using a Van der Paw method (HMS-3000, Ecopia U.S.A.).

3. Results and Discussion

In this work, ethanolamine (ETA) and metal ions such as copper and indium could form a complex compound due to ethanolamine has free electron pair which donated to metal ions and form a coordination or complex bonding where the ETA acts as ligands and the metal ions act as a center atom. Since the unique structure of ETA, thus ETA could act as a chelating agent which illustrated as figure 1.

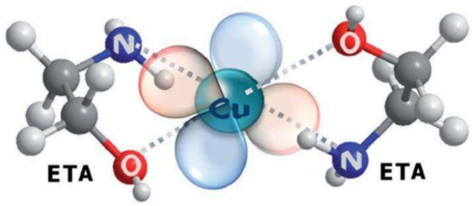

Figure 1. Illustration of Cu-ETA complex compound [7].

Moreover, at drying process, Cu-ETA and In-ETA complex compounds were decompossed under air atmosphere into metal oxide

$$C_2H_7NO\ (aq) + M^{x+}\ (aq) \rightarrow M[C_2H_7NO]_x\ (aq)$$

$$M[C_2H_7NO]_x\ (aq) + O_2\ (g) \rightarrow MO\ (s) + CO\ (g) + C\ (s) + H_2O\ (g) + NO\ (g)$$

M = Cu, In

After annealing at 550 ºC under Ar (95%) + H_2 (5%) atmosphere for 120 minutes, selenium was changed into vapor phase and then, some of Se vapor was react with metal oxide and H_2 simultaneously. Se vapor react with metal oxide and H_2 form metal selenide and H_2Se respectively. Further, H_2Se could react with metal oxide and form metal selenide either. Meanwhile, carbon as residue was reacted with H_2 became methane gas and removed from thin film. Finally, metal selenide such as CuSe and InSe were coalesced into $CuInSe_2$ as a final product. In other hand, this reaction mechanism shows that H_2 acts as catalyst and as carbon remover simultanously.

$$\mathrm{Se\ (s) \rightarrow Se\ (g)}$$

$$\mathrm{Se\ (g) + 2\ MO\ (s) \rightarrow MSe\ (s) + O_2\ (g)}$$

$$\mathrm{Se\ (g) + H_2\ (g) \rightarrow H_2Se\ (g)}$$

$$\mathrm{H_2Se\ (g) + MO\ (s) \rightarrow MSe\ (s) + H_2\ (g)}$$

$$\mathrm{C\ (s) + 2H_2\ (g) \rightarrow CH_4\ (g)}$$

$$\mathrm{MSe\ (s) + MSe\ (s) \rightarrow M_2Se\ (s)}$$

Before annealing, x-ray defraction pattern (XRD) shows a broadened peak from 13.75º to 38.48º which shows amorphous phase belong to carbon as dominant compound, while metal oxides due to in amorphous phase couldnot detected by XRD. After annealed at 550 ºC under Ar (95%) + H_2 (5%) atmosphere for 120 minutes, $CuInSe_2$ has been formed with four peaks at 26.76 º, 35.55 º, 44.25º, and 52.49º which indexed as (112), (211), (200/204), and (116/312) respectively which matched with $CuInSe_2$ standard (ICDD: 04-005-3912) as tetragonal crystal system.

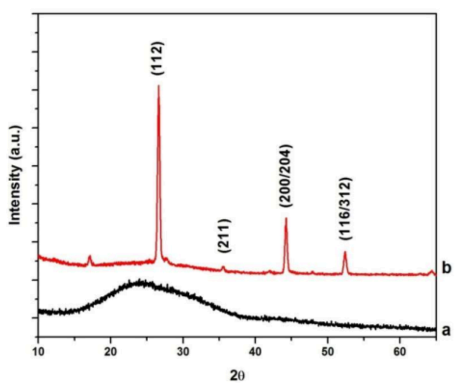

Figure 1. XRD pattern of $CuInSe_2$ thin film before (a) and after (b) annealing.

Moreover, from scanning electron microscope (SEM) shown that the morphology of precursor has smooth and tiny particles spread around the surface. Meanwhile, after annealing, shows the morphology, became larger and compact without shown hexagonal shape from copper was seen. It seems the reaction was going completely.

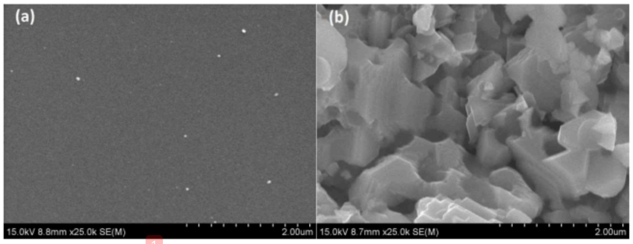

Figure 2. SEM image of CISe before (a) and after (b) annealing.

Organic residue was represented by carbon which existed in the thin film and could be determined by energy dispersive spectrometry (EDS). The spectrum shows the proof of carbon existence in the thin film before annealing. In addition to carbon, also could be seen the spectrums of chloride from metal salts, while oxygen could represent of either organic residue or metal oxide. However, after annealing, the EDS spectrums of carbon, chloride and oxygen spectrums were no longer seen in the thin film. It seems they have been removed successfully during annealing.

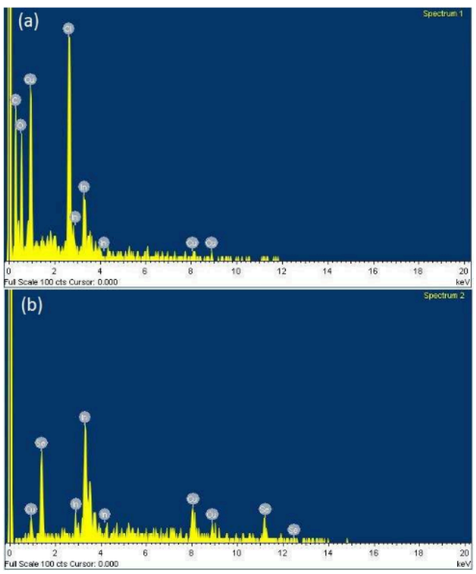

Figure 3. EDS spectrums of CISe before (a) and after (b) annealing.

EDS also could be used for determining the chemical composition of CISe thin film before and after annealing. Before and after annealing Cu/In ratio was 1.20 and 1.35 respectively. The slight enhancement of ratio probably due to diminished of indium in the thin film during annealing considering the melting point of indium (156 ^{o}C), which is below the annealing temperature. In other hand, copper is a stable material, thus during annealing it was not much diminished.

Table 1. Chemical composition of CISe thin film before and after annealing

	% Atomic				Ratio	
	Cu	**In**	**Se**	**C**	**Cu/In**	**(Cu+In)/Se**
Before Annealing	19.61	16.34	0.00	64.05	1.20	-
After Annealing	15.68	11.60	24.58	0.00	1.35	1.11

4. Conclusion

$CuInSe_2$ (CISe) thin film has been synthesized successfully from metal salts and ethanolamine complex compounds by spin coating and followed by annealing method. Synthesis reaction mechanism of CISe also proposed in this work in order to explain whole process of synthesis from metal salts, complex compounds to CISe thin film. The structure of CISe thin film shows a tetragonal and matched with CISe standard. Moreover, CISe thin film has a suitable chemical composition with no more carbon or organic residue in the thin film.

Acknowledgement

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